

1 **Full set of replies to referee's and mark-up of changes for Monks et al. (acp-2014-881)**

2
3 **Reply to Referee #1**

4
5 We are grateful to the referee for their comments and insights and recognising the substantial
6 challenge in preparing this review.

7
8 *This article represents a significant effort towards summarizing the state of knowledge regarding*
9 *tropospheric ozone and its impacts. It is thematically comprehensive, and generally well-written. It*
10 *suffers from some redundancies and hiccups typical of an article with so many contributing authors; I*
11 *encourage the authors to pay particle attention to editing for this before final publication (some*
12 *specific examples are included below).*

13
14 *My primary critique is that the article is quite Euro-centric, some sections more than others. Given the*
15 *author affiliation and the association with the ACCENT project, this is not surprising, but needs to be*
16 *addressed prior to publication. In present-form the article requires some caveat ("with a focus on*
17 *Europe") in the title, abstract, and throughout the text. Alternatively, to accurately reflect the title*
18 *and goals of the paper the authors need to address this issue on two fronts.*

19
20 In revision, we have made strenuous attempts to meet this criticism head on! We have reviewed
21 each section in order to ensure balance. There has been more explicit mention of N.American/Asian
22 locations in some sections. On reflection, the failing, if any, maybe that some of the examples are
23 from Europe and I think the perceived bias in references is influenced by this. Closer examination
24 does not hold this to be true uniformly.

25
26 *The first is including discussion of results from non-European projects. Two examples of this: in the*
27 *context of biomass burning (4.2), there was no discussion of the ARCTAS project which provided*
28 *significant insights into the role of boreal fires on atmospheric composition. Similarly the megacities*
29 *section (4.1) made no reference to the MILAGRO campaign in Mexico City, a tremendously*
30 *comprehensive megacity field campaign.*

31
32 We have revised the text to add mention of the MILAGRO campaign, which was indeed an oversight
33 on our part considering the amount of work published about Mexico City. References have been
34 added where appropriate, e.g.:

35 P32764, L15-19: "Other megacities in which vehicles are cited as the top or among the top sources of
36 ozone precursors are Cairo, Delhi, Istanbul, Los Angeles, New York, Mexico City, Sao Paulo, and the
37 Ruhr-Rhein metropolitan region (Bon et al., 2011; Chelani, 2012; de Fatima et al., 2012; Parrish et al.,
38 2011; Khoder, 2009; Im and Kanakidou, 2012; Melkonyan and Kuttler, 2012)."

39 as well as text:

40 P32765, L26 – 32766, L2: "The emissions from megacities also have an effect on the surrounding
41 region. In many cases the urban areas have been identified to be VOC-limited, where high NOx
42 concentrations in the cities suppress ozone concentrations (Im and Kanakidou, 2012; Tie et al., 2013).
43 For example, a modeling study conducted in the context of the MILAGRO measurement campaign in
44 Mexico City identified that reductions in VOC emissions led to decreases in maximum ozone
45 concentrations while NOx emission reductions led to increased maximum ozone concentrations,
46 demonstrating that the urban core of the Mexico City metropolitan area was VOC-limited – a
47 conclusion supported by the in-situ measurements. The surrounding mountain/rural areas were

48 identified as mostly NO_x-limited, although the range of these areas was meteorologically dependent
49 (Song et al., 2010). This suppression of ozone by high NO_x has been identified as generally more
50 characteristic of extratropical megacities in the Northern Hemisphere (despite the Mexico City
51 example), relative to tropical cities, because of differences in transport patterns.”

52 Details of ARCTAS were already included in the Arctic section (Alvarado et al. (2010), Wespes et al
53 (2010) and Singh et al (2010)), explicit mention of ARCTAS has been added. With respect to the
54 biomass burning section, ARCTAS and a cross-link to the Arctic section has been added.

55
56 *Section 4 is also poorly organized – it seems a grab bag of topics. Topics such as lightning (4.7) and*
57 *biomass burning (4.2), halogens (4.4) appear to belong in Section 2.3 (precursors). I suggest a re-*
58 *organization.*

59
60 Following referee#2's advice an explanation as to the rationale for this assortment has been added to
61 the text and it has been retitled.

62 63 Minor Comments

64
65 I include some additional minor comments below.

66
67 1. Overall: inconsistency of units; suggest using ppb throughout (and converting all
68 references using ug/m³)

69
70 Done

71
72 2. Pg. 32712, lines 14-29: This content doesn't seem to fit particularly well in the introduction, and is
73 a bit redundant with the review in section 2.1. I suggest merging this paragraph into that section. It is
74 also worth noting that the end of this paragraph is a bit misleading as it could leave the reader with
75 the impression that this debate is still alive.

76
77 Removed from introduction and merged with section 2.1

78
79 3. Pg 32714: The tropospheric ozone budget is quite precisely quantified here (3 significant figures on
80 the burden!), but a discussion of uncertainty and variability in model budgets (beyond the regionality
81 discussed in Figure 2) is lacking.

82
83 The uncertainty has been added as suggested to the budget terms here: STE 550+/-140 and burden
84 335+/-10 for the observationally-derived terms, and Dep 1000/-200 and Chem 450+/-300 for the
85 model terms. Variability between models in the burden is of the order of 10%; interannual variability
86 in a single model is far less (probably 5-10 Tg). The variability and uncertainty is discussed in the
87 papers cited.

88
89 4. Section 2.1.1: It would be useful if the authors could include some discussion of the relevant
90 timescales of processes in this section.

91
92 A reference to typical timescales for reactive chemistry has been added.

93
94 5. Pg 32720, line 9: language needs to be corrected.

95
96 Section has been rewritten.

97

98 6. Section 2.1.2d: Obvious missing reference/discussion: Kurpius and Goldstein, GRL,
99 2003
100
101 Reference added
102
103 7. Page 32729 lines 26-30 & 32730 lines 1-3: Specify whether these are surface, profile or column
104 concentrations
105
106 Clarified - surface concentrations.
107
108 8. Page 32731, lines 1-3: This list of factors should also include meteorology beyond transport (rain,
109 radiation, temperature, etc), as well as changing surface cover (e.g. vegetation).
110
111 Text amended to reflect this.
112
113 9. Page 32733, lines 19-22: This sentence requires a reference – unclear if the citation from the
114 previous sentence applies here.
115
116 Wilson et al, (2012) and Colette et al (2011) references added.
117
118 10. Page 32745, lines 15-17: Sindelarova et al. is not an appropriate reference here
119 (an application, not the model description). MEGAN v2.1 also includes an algorithm for CO₂ inhibition
120 of isoprene emissions.
121
122 Reference amended and text added
123
124 11. Page 32758, lines 16, 21: Remove the repetition of line 16 of line 21. The specific reference to rice
125 is also odd, there are many more crops affected by ozone...a list of vegetation types affected and
126 some references would be useful.
127
128 Removed odd sentence on rice and overhauled section to make it cover crops and vegetation
129 affected by ozone.
130
131 “Typical ozone effects on plants include reduced growth, less seed production, lower functional leaf
132 area and earlier leaf senescence. Data compilation studies have shown that many species of plants
133 are sensitive to ozone, including: agricultural crops such as wheat, tomato, soybean and rice and
134 salad crops such as lettuce, spinach and onion (Mills et al., 2007a); grassland species such as clover
135 species, buttercup and harebell (Hayes et al., 2007, Mills et al., 2007b); and tree species such as
136 beech, birch and Holm oak (Karlsson et al., 2007). These effects impact on the important ecosystem
137 services provided by plants, including food security, carbon sequestration, timber production, and
138 protection against soil erosion, avalanches and flooding.”
139
140 12. Page 32760, line 2: Tai et al. show that climate reduced global yields by 11% (not
141 > 20%)
142
143 Modified (10%)
144
145 13. Page 32762, line 8-9: What does “the fundamental processes involved” mean?
146 Please elaborate.
147

148 Sentence added "For example, the radiative forcing from aerosols has a larger uncertainty because
149 we are less sure of the changes in aerosol since the pre-industrial, both in terms of their magnitude
150 and geographical distribution, but also because the aerosol forcing originates from changes in
151 multiple different aerosol types, including mixtures of different aerosol species, with highly uncertain
152 optical properties (Fuzzi et al., 2015)"
153

154 14. Page 32763, line 1-2: Needs reference.
155
156 Reference added (Zhu et al, 2012)
157

158 15. Page 32763, line 28: also domestic biofuel use?
159
160 Added
161

162 16. Page 32764: lines 25-29: needs a reference (particularly for the role that evaporative
163 emissions played in this event)
164
165 Elansky et al. reference details this. New Zvyagintsev et al. reference added
166

167 17. Page 32770: lines 15-17: needs reference.
168

169 Section deleted as unpublished. Figure 30 changed to one from Parrington et al. (2013)
170

171 18. Section 4.3: This might also be a good place to note the potential impacts of
172 biofuels on ozone concentrations, via changes to vegetation and BVOC emissions (e.g.
173 Ashworth et al., 2012; Porter et al., 2012).
174

175 Sentence added on biofuels and ozone/AQ.
176

177 19. Section 4.5: This section does not seem to be particularly relevant to a review of tropospheric
178 ozone. The key elements of the role of NO_x have been discussed elsewhere.
179
180 The aim of this section was to demonstrate the wider linkages of ozone chemistry through Nr to the
181 nitrogen cycle. The premise being that these are often treated in isolation.
182

183 20. Section 4.7: The basic chemistry described in this section is redundant with earlier descriptions of
184 ozone formation. Please harmonize.
185
186 Chemistry in this section has been cut-down
187

188 21. Section 4.7: the discussion of how lightning relates to aerosols is not relevant to this review.
189
190 Removed and section restructured.
191

192 22. Page 32794, lines 1-11: In addition to several studies which show high methane leakage from
193 fracking operations, the authors should note the Allen et al., PNAS, 2013 study which provides the
194 counter-example of low measured leakage rates.
195
196 Added Allen reference and also recent PNAS Kang (2014) reference
197

198 23. Section 4.10: This section is quite long, and should be edited to re-focus on implications for
199 ozone.

200
201 There has been much debate as to the effect on oxidation chemistry of the radical pathways detailed
202 on section 4.10. The authors feel there is significant value in the section in full.
203
204 24. Page 32803, lines 20-25: A number of studies identified the role of intercontinental transport of
205 ozone in ozone exceedances prior to the publication of the HTAP report in 2010. As written, this
206 sentence is a bit misleading.
207
208 Changes made to remove incorrect implication.
209
210 25. Page 32805, lines 14-18: unclear what scenario this mortality reduction corresponds to – is this
211 the result of reductions of European emissions only?
212
213 Yes, this only applies to European emission's. Text clarified.
214
215 26. Section 5.3, 5.2, and to some degree 5.1 need to be merged and redundancies eliminated.
216
217 These sections have been reviewed and there is some blurriness around the edges, they have been
218 edited to drive greater self-consistency.
219
220 27. Section 5.4: Tai et al., GRL, 2013 recently showed the "climate penalty" has been substantially
221 overestimated. The discussion should be modified in light of this.
222
223 Discussion modified to include "There is some debate as to the magnitude of any climate penalty e.g.
224 Tai et al. (2013) indicates that in the presence of CO₂-isoprene interaction, the projected change in
225 isoprene emission by 2050 will be largely offset or even reverse in sign, leading to much reduced
226 sensitivity of ozone and SOA (by >50%) to climate and natural vegetation."
227
228 28. Page 32808, lines 14-16: It is not clear why emissions that were constructed for radiative forcing
229 purposes would be inappropriate for air quality purposes. Please justify any concerns about quality of
230 these emission inventories, and why they might therefore still be appropriate for climate projections.
231
232 We have aimed to clarify this point in the text, added two sentences to the referenced lines. The
233 revised text now reads:
234
235 "Pioneering studies relied on projections originally designed for climate projections (SRES,
236 Nakicenovic et al., 2000 or RCPs, van Vuuren et al., 2011), but the ozone precursor information in
237 such scenarios was solely provided to assess radiative forcing and their use for surface air quality
238 projections constitutes a deviation from their original purpose. The various scenarios make differing
239 assumptions for future air pollution emissions and therefore, describe a wide range of future
240 emissions over large world regions. Any downscaling in regions that exhibit large spatial
241 heterogeneities could be problematic leading to inaccurate results (Amann et al., 2013). The use of
242 emission projections relying on policy relevant emissions factors such as the Global Energy
243 Assessment (Riahi et al., 2012), the ECLIPSE (Klimont et al., 2013a, b) or PEGASOS datasets are more
reliable."
244
245 29. Page 32810, lines 10-22: duplication with Section 5.4.

245 While this is true, we have tried to make all sections self-contained and this is an important part of
246 the AQ & CC discussion, as embedded in this forward look. My preference is to keep it.

247
248

249 **Reply to Referee #2**

250

251 This is a very interesting review paper. At first I was slightly daunted by its length, but it actually was
252 a very easy and enjoyable read. It provides a very nice up-to-date review of what is a complex topic,
253 simultaneously providing a good introduction to the subject, whilst exploring some of the latest
254 research findings. I expect to use it as a recommended paper for my students (UGs and PGs).

255

256 I think the paper should be published subject to minor revisions.

257

258 *We are grateful to the referee for the comments and insights on the paper. We have dealt with all the*
259 *comments and list below (in brief) the changes made.*

260

261 I thought the title of the paper was rather cumbersome and not quick to understand. I would suggest
262 something more concise.

263

264 The title has been chosen to reflect scales and roles of ozone.

265

266 Throughout the paper, but particularly towards the front, ensure that all acronyms and chemical
267 formula are defined.

268

269 Noted and checked.

270

271 P. 32712, lines 26-30. I believe that the reference to Bloss et al (2005) refers to the high OH
272 concentrations in the tropical lower troposphere rather than the greater ozone production from
273 long-lived HCs there. As it stands, this is not clear. Whilst it is expected that there will be greater
274 oxidation of the long-lived HCs in tropical lower troposphere, the production of ozone does rather
275 depend on the amount of NO_x present. Fig. 2 relates to this. Whilst the plots indicate high ozone
276 production in the tropical lower troposphere, the non-linear scale for the legends tends to hide some
277 of the variation at the higher rates. Note axes labels are missing from the upper panels.

278

279 The Bloss paper shows fraction of methane loss by atmospheric region which is referred to in the
280 context of the highlighted sentence. We agree that ozone production is a combination with NO_x
281 availability as mentioned.

282

283 P. 32716, lines 21-23. "Considering : : :". This is not a sentence.

284

285 Sentence rewritten.

286

287 P. 32717, lines 5-6. I suggest including a reference to Reaction R3, for example, for the
288 regeneration of OH.

289

290 Added

291

292 P. 32717, lines 6-8. This says that production of O₃ is shown schematically in Fig.
293 3. Although Fig. 3 was not what I had expected which was a figure that would show schematically the
294 reactions involved in the formation of ozone. I think this would be a useful additional figure.

295

296 New figure 3 added for methane oxidation cycle.
297
298 P. 32717 discusses Fig 3 including the transition from point A to B and from point C to B. I also think it
299 is useful to discuss moving from point A towards the origin and from point C towards the origin.
300
301 Text has been revised to highlight the points and the transitions in differing ozone
302 formation/destruction regimes.
303
304 P. 32718, line 12, suggest "which can be readily".
305
306 Done
307
308 P. 32718 discusses the impact of deposition in rural areas in such a way that it seems to suggest that
309 deposition is not important in urban areas. Of course in urban areas there is the added impact of the
310 NOx titration effect, but that doesn't mean that deposition isn't still an important ozone sink.
311
312 Having looked at this, we are not sure that we get the same inference from the text.
313
314 P. 32729, line 9. I didn't fully understand what was meant by "by treating the vegetation to fully close
315 stomata". Please clarify.
316
317 This section of text has been rewritten.
318
319 Structure of section 2.1.2. This seemed a bit odd, with the end of section b introducing sections c, d
320 and e, which all seemed to be sub-sections of b. Consider restructuring the headings.
321
322 These are all separate features of the uptake, though obviously interrelated. Sections kept as was.
323
324 P. 32722, line 14, suggest "deposition rates to water"
325
326 Done
327
328 Structure of section 2.1.3. I suggest giving the sub-headings letters a, b, etc. I also suggest perhaps
329 separating stratospheric-tropospheric exchange and summer monsoons.
330
331 I don't see the need for them to be together.
332
333 Separate section headings made and STE separated from seasonal transport patterns as suggested.
334
335 P. 32733, lines 14-18. "this behaviour", which I take to be the decreasing trend in high percentiles is
336 said to be in part due to increasing hemispheric ozone levels, but this would lead to reduced ozone.
337 Please clarify.
338
339 Sentence modified to "This behaviour has been interpreted as resulting from the combined effect of
340 regional pollution controls since 1990 (Vautard et al., 2006) and increasing hemispheric ozone levels
341 (Derwent and Hjellbrekke, 2013)".
342
343 P. 32734, line 6, "non-significance of the trends". Urban or rural background?
344
345 Rural background added
346
347

342 Section 2.3.2 relates to “evolution” of emissions and yet, quite a lot of the text at the end of this
343 section is on uncertainties, which might fit better in the following section 2.3.3. in uncertainties.
344
345 We feel though while this might be correct, that the benefits from such a major reorganisation might
346 be counterproductive.
347
348 P. 32738, line 5, suggest “among these inventories”.
349
350 Done
351
352 P. 32738, line 7, suggest “lowest and highest”.
353
354 Done
355
356 End of section 2.3.2.a is about shipping emissions. These do not seem to specifically
357 relate to Europe or N. America, so I suggest some restructuring.
358
359 Section restructured and shipping pulled out as a separate section.
360
361 P. 32739 lines 21-22, suggest “For India, the range of values proposed by the different
362 groups is even larger, : :”
363
364 Done
365
366 P. 32739 line 26, suggest “Figures 7 and 10”
367
368 Done (with new Figure numbers)
369
370 P. 32740 line 13. When you say “deterioration” of emissions, do you actually mean
371 “varying deterioration”?

372 Change made

373 P. 32740 line 21. “most countries”.
374
375 Deleted “the”
376
377 P. 32741 line 1. “on a few”.
378
379 Added “a”
380
381 P. 32741 line 6. “information on”.
382
383 Added “on”
384
385 P. 32741 line 8. “uncertainties in”.
386
387 Changed
388
389 P. 32741 line 20, says that Figure 14 also highlights the “growing” importance of emissions from
390 Africa. It doesn’t actually highlight this as it only shows 2005 data. It does however show that in
391 2005, African emissions are comparable to emissions from other regions.

392
393 Referee is correct removed "growing".
394
395 P. 32742 line 24-26. This is confusing as it says an example of "variability" is : : : :the
396 "constant" high fire episodes.
397
398 Sentence rewritten
399
400 P. 32743 line 9. I would also suggest that Fig. 17 shows large differences for breal
401 regions as well.
402
403 Agreed.
404
405 P. 32743 line 12. "limit in".
406
407 "limit to the accuracy" is OK
408
409 P. 32743 line 29. "remain in".
410
411 Done
412
413 P. 32744 line 6. "taken as constant".
414
415 Done
416
417 P. 32744 lines 18-19. "fire plumes".
418
419 Changed to from fires
420
421 P. 32744 line 23. "fire emissions".
422
423 Done
424
425 P. 32746, line 11, suggest "lowest and highest".
426
427 Done
428
429 P. 32747, line 16, should refer to Fig 21b not 22b.
430
431 Done
432
433 P. 32750, lines 8-9, suggest "during a 2 year period".
434
435 Done
436
437 P. 32750, lines 13-15, I am not sure of the relevance of this within this section.
438
439 As this section deals with a climatological view, the impact of Saharan dust seemed to fit best here.
440
440 P. 32751, line 13, suggest "than would be".
441
441 Deleted "what"

442
443 P. 32752, line 1, refers to Figure 26. As well as ozone, this figure includes PM10, which
444 is not discussed in the text and is actually not relevant.
445
446 Agreed (as per referee#3) - PM10 is present owing to source of data, thought best not to doctor
447 figure. The figure caption has been expanded to explain significance of result.
448
449 P. 32752, line 10, suggest "coupling at the".
450
451 Left as is – two things can be a couple
452
453 P. 32752, line 20, "emission".
454
455 Done
456
457 P. 32753, lines 22-24. It is not clear why there is an interim target and the "High Level"
458 maximum daily mean (one being 40 ppb than the other). Please clarify.
459
460 These are nomenclature used by WHO to indicate high and significant health impacts from ozone.
461 Text added to indicate this.
462
463 P. 32756, line 18. It would be helpful to provide the definition of SOMO here. It is given
464 later on page 32801.
465
466 Cross referenced to section 5.
467
468 P. 32756, line 22. Surely this should be "calculated" increases in health effects?
469
470 Agreed and changed
471
472 P. 32757, lines 1-13. Again the comments here seem to refer to actual increases in
473 ozone impacts depending on the choice of SOMO. Surely these should be calculated impacts.
474
475 Yes, these are indeed calculated impacts based on measurements and models.
476
477 P. 32758, lines 12-13. This could be defined as AOT40 as again this used later in the
478 paper.
479
480 Added AOT40 and cross link to section 5.
481
482 Section 4 Topics. It is not clear to me why the various topics are ordered in the way
483 they are. I would suggest ordering the topics something along the lines of emission
484 sources, chemistry, specific regions, general topics (such as modelling and nitrogen
485 cycle).
486
487 A new introduction has been added here to overview the selection of topics (as per referee #3).
488
489 P. 32763, lines 23, suggest "concentrations have been observed".
490
491 Done
492

493 P. 32765, Up until now ozone levels have been given as mixing ratios. I realise that here they are
494 given as mass densities because of the units of the thresholds, but I think it would be helpful to be
495 consistent with the earlier text to give the values in mixing ratios (even if this means giving the values
496 in both units).
497
498 Units have been standardised.

499 P. 32765, line 20. The 3 needs to be a subscript.

500 Done

501
502 P. 32765, line 22, suggest "avoided by such policies".
503
504 OK, as is
505

506 P. 32765, line 24. What is meant by "small but substantial"?
507
508 It means low fractions which have large health impacts.
509

510 P. 32768, lines 3-4, "Tropical biomass burning dominates : : :". It would be good to
511 give the percentage contribution from this biomass to burning emissions to compare to
512 boreal fires.
513

514 Page 32768, states in terms of carbon that only 9% is boreal, therefore 91% in tropical.
515

516 P. 32768, line 17. "boreal".
517
518 done
519

520 P. 32768, line 21. "BORTAS was to".
521
522 done
523

524 P. 32769, line 4. "drivers of the OPE".
525
526 done
527

528 P. 32769, line 26. "algorithms"
529
530 done
531

532 P. 32769, line 29 – p. 32770, lines 1-3. I am confused by the comment that boreal fires emit products
533 up to 10 km, and then the subsequent comments which limit emission to below 4 km. Please clarify.
534

535 Text modified to "Depending on the fire radiative power (FRP) and size of the fire, Freitas et al.,
536 (2007) have shown that plumes from fires are likely able to reach 10 km altitude. Based on a
537 statistical analysis of 5 years of satellite observation by MISR over North America, Val Martin et al.,
538 (2010) have shown that the median altitude of plumes is found below 3 km altitude for boreal forest
539 fires. A significant fraction (4-12%) of those plumes are thus injected above the boundary layer and
540 are more spread-out vertically depending on the stability conditions. In comparison, tropical biomass
541 burning plumes are mostly limited to the first km of the atmosphere."
542

543 P. 32770, line 16. "have a strong impact on meeting air : : :".
544
545 Section removed owing to unpublished results.
546
547 P. 32770, line 17. "Figure 30".
548
549 Figure changed
550
551 P. 32770, line 18. "July 2012 in the frame"
552
553 Removed
554
555 P. 32770, lines 23-24. "and given plume age : : :..pathway apparently)." I really don't
556 understand the point being made here. In fact I am really not very sure what message
557 is trying to be conveyed with respect to Figure 30.
558
559 Removed
560
561 P. 32772, line 20. "of its key".
562
563 Done
564
565 Section 4.4 Halogens, first paragraph. I would prefer to
566 see the reactions given on separate lines rather than integrated within the text.
567
568 Done
569
570 P. 32777, line 20. "to O3".
571
572 Done
573
574 P. 32777, line 28. "Bromine is twice as important as chlorine as an ozone sink".
575

576 Done

577 P. 32782, lines 8 and 9. "reported for the tropics".

578 Done
579
580 P. 32785, line 1. "At the continental".
581
582 Done
583
584 P. 32786, line 8. "can be a significant".
585
586 OK
587
588 P. 32787, lines 22-23. If the power is given for over the land, why not for over the ocean?
589 Having said that, the rest of the paragraph suggest that other parameterisations may be better, so
590 perhaps there is no need to give any values (i.e. remove the bit in brackets).
591

592 Done
593
594 P. 32798, line 16. What is the Advanced Light Source?
595
596 Added synchrotron - <http://www-als.lbl.gov/>
597
598 P. 32799, line 23. "in its excited".
599
600 Done
601
602 P. 32802, line 2. "100s".
603
604 Done
605
606 P. 32805, lines 2-5. "even if : : : . penalty". This is confusing. Please clarify.
607
608 Sentence rewritten.
609
610 P. 32805, line 16. "deaths".
611
612 Done
613
614 P. 32806, first paragraph. Comments relate to 20% emission changes. The emissions of exactly which
615 chemical species were changed?
616
617 In all precursors
618
619 Section 5.4. Most of the material on page 32807 is about the science of the impact of climate change
620 on ozone and it is not until the next page that this section really deals with the policy context, which
621 is what section 5 is about.
622
623 Agreed, there are some blurry lines between these sections. It was aimed that one would look at
624 policy and the second at the science of the situation.
625
626 P. 32808, line 9. "impacts".
627
628 Done
629
630 P. 32812, last paragraph. I think it would be good to mention personal exposure monitors
631 here.
632
633 References and a sentence added for personal exposure methods.
634
635 P. 32813, lines 3-6. It seems a bit odd to add this bit about the whitefly in the conclusions when it
636 hasn't been mentioned before.
637
638 The aim here was to give a broad perspective on future directions and new slightly left-field
639 directions, hence the inclusion of the whitefly.
640
641 Throughout the paper, but particularly towards the front, ensure that all acronyms and
642 chemical formula are defined.
643

644 Noted and reviewed throughout manuscript.
645
646 Be consistent with the use of upper case letters, particularly when referring the specific regions, e.g.
647 polar regions, boreal regions, tropics. Also stratosphere-troposphere exchange, chlorophyll, dissolved
648 organic matter, background ozone

649 Noted and reviewed throughout manuscript.
650
651 The text on a number of figures was rather small. Please make sure it is legible in the
652 final paper.
653
654 Noted
655
656 Fig. 4. Although the RC terms are defined in the text, none of the other terms are.
657 They should either explained in the text or figure caption.
658
659 Added link to nomenclature in text
660
661 Fig. 7. According to the figure headings some of these emissions are for the USA and
662 others for N. America. The caption should reflect this.
663
664 Figure redrawn to ensure consistency.
665
666 Fig. 8. "data for Canada".
667
668 Done
669
670 Fig. 17. "see Fig. 17-1b" makes no sense.
671
672 Cross reference updated
673
674 Fig. 18. "of the mass"., "from Sofiev et al. (2013)."
675
676 Corrected
677
678 Fig. 21. In the caption please indicate for the fight hand panel whether "This study" is
679 a bottom up or top down study.
680
681 The "This study" from the Tohjima paper is a top-down estimate, figure amended.
682
683 Fig. 27. The two panels would be more comparable if they were both either daily maxima or annual
684 means.
685
686 This may well be true, but this is as they are published in the paper.
687
688 Fig. 29. Define MMM and T which are used in the title.
689
690 Done
691
692 Fig. 32. States that the values plotted are losses. However as the values are negative, they are
693 actually negative production terms. The x-axes labels are rather unclear.

694
695 The negative terms represent loss terms.
696
697 Fig. 35 need more explanation.
698
699 More commentary has been added to the figure legend.
700
701 Fig. 37. "emission increases", "leakage rates".
702
703 Done

704 I didn't see Figure 39 referred to in the text.

705 Added

706

707
708

709 **Reply to Referee #3**

710

711 *The authors have clearly invested much effort in compiling an interesting discussion on tropospheric*
712 *ozone. The level of detail in the different sections, and even within sections, is not always well*
713 *balanced, and the authors may wish to consider narrowing the scope in some places to ensure that*
714 *the key messages are clearly distilled, with specific suggestions provided below. In general, it would*
715 *be stronger if the authors can provide where possible a more critical assessment of the studies they*
716 *are reviewing, or reconcile disparate findings. I generally agree with the major suggestions of the*
717 *other reviewers.*

718

719 We thank the referee for this comments and insights. We accept that the paper has
720 inhomogeneities and have tried the upmost to harmonise them. The paper is substantial and in line
721 with the other referee's wishes a more critical tone has been taken with respect to the outcomes of
722 the review.

723

724 General comments

725 The abstract could be misleading by emphasizing the challenges of reducing surface level ozone
726 without acknowledging the substantial improvements in air quality that have been achieved due to
727 air pollution regulations; peak ozone pollution levels have responded to local and regional emission
728 reductions. As an example: Simon, H., A. Reff, B. Wells, J. Xing and N. Frank (2014). "Ozone Trends
729 Across the United States over a Period of Decreasing NO_x and VOC Emissions." Environmental
730 Science & Technology 49(1): 186-195. The comment on ozone as a short-lived climate pollutant is a
731 slightly awkward in that the mitigation would involve the precursors, and the net impact on climate
732 depends on which precursors are controlled. A stronger abstract would provide more specific
733 conclusions, or possibly recommendations. For example, what is the knowledge across scales needed
734 for addressing air quality and climate, and does it exist or remain to be generated?

735

736 The abstract has been rewritten to draw some conclusions in terms of what is required.

737

738 When prior reviews are mentioned, why not give a 1-2 sentence summary of their main conclusions
739 (e.g., Monks et al. 2009 P32718 and throughout the text)?

740

741 This is quite challenging, as they do cover some breadth. Clear pointers have been added to text to
742 signpost the elements that can be found in these reviews.

743

744 The level of detail given for deposition seems deeper than that provided for chemistry (some is later,
745 but that's not obvious here). It would help to give a rationale for the relative attention given to these
746 processes. The scope seems very broad for the seasonal transport patterns section, some of which
747 might fit with the climate variability section, which also seems connected to discussion in 2.4.

748

749 New figures have been added to balance the chemistry section.

750

751 As per the suggestion of referee#2 and this referee the section on transport has been restructured to
752 focus the seasonal transport questions.

753

754 Can anything be said in Section 2.3.3 about which regional emission inventories are most accurate in
755 their representation of trends or total amount of emissions? Are there top-down constraints from
756 satellites on in situ measurements that can distinguish between the various estimates? A
757 recommendation would be very useful.

758

759 This would be a review in itself. There is an undoubted need for what you are asking. Some of this
760 is covered in the framework on GEIA (<http://www.geiacenter.org/>), but there is a need for more
761 critical evaluation.
762 Sections 3 and 5 might be best focused solely on Europe, but if the U.S.A. is discussed, it should be
763 noted that the criteria for the ozone standard are reviewed periodically, and the most recent
764 assessment was just completed, e.g. <http://www.epa.gov/ncea/isa/>
765

766 The links and context of the USA NAAQS ozone work has been added to the paper, as it provides
767 needed wider context for the reader. Details have been added to section 3 and 5.

768 Section 4 should open with a rationale for why the selected topics are the most pressing ones to
769 discuss here. A revised title, rather than "Topics", should also strive to convey their importance.
770 Section 4.2 overlaps earlier discussion.

771 A rationale has been added and the section title changed to recent advances.

772 Agreed, there is a measure overlap, the aim of the earlier section is to look at emission inventories
773 and the later one experimental evidence.

774 Specific comments:
775
776 p. 32712 The Intro here fails to communicate what the current paradigm is for what controls
777 tropospheric ozone.
778
779 At the suggestion of referee#1 the introduction was restructured and this material moved to section
780 2.1. This has now been integrated to give a better overview of the required paradigm.
781
782 P32715. The phrasing of something well understood but remains a challenge seems a bit
783 contradictory.
784
785 Agreed, added in practice to give the correct emphasis to the challenge.
786
787 P32718 L24-25. Is this specific to deposition? Might be worth illustrating how the diurnal variability
788 varies with geographic location, at the surface vs. at altitude to emphasize the importance of
789 multiple spatial scales mentioned in the abstract.
790
791 The climatological view of ozone is dealt with in section 2.4. There is clear merit in this idea, but it
792 probably is beyond the scope of what can be achieved in this review.
793
794 P32721 L3. What does "positive and negative effects" mean here? Is deposition ever reversing and
795 becoming a source?
796
797 Sentence modified to "For example, surface water has been found to both enhance and reduce
798 deposition rates."
799
800 P32724. Can any conclusions be drawn here about the sign of the change of climate change on long-
801 range transport?
802
803 HTAP (2010) says "Changes in climate will affect meteorological transport processes as well as the
804 chemical environment and lifetime of the transported pollutants and hence the concentrations of
805 pollutants arriving at downwind continents." Of the papers mentioned Glotfelty et al (2014) suggest

806 a larger impact on the US from East Asian Emissions and Doherty et al (2013) showed stronger
807 chemistry than transport positive climate feedbacks on ozone. This has been added to text.

808
809 P32727. 30-year periods may yet be a bit short for screening out the influence of low
810 frequency climate variability (e.g., PDO, AMO)?

811
812 The following sentence can be added to the end of line 13 on page 32727

813 "The influence on ozone of low frequency climate variability on time scales longer than 30 years has
814 not yet been assessed due to continuous ozone data sets being limited to durations shorter than 30-
815 40 years."

816
817 P32732. What other factors could be at play besides those put forth in the Parrish et al. "untested
818 hypothesis"? Might the question be better posed as quantifying the relative importance of these
819 factors at the individual locations of the measurements?

820
821 The purpose of Section 2.2 is merely to give a brief history of changes in tropospheric ozone. The
822 Parrish et al. study gives very recent insight into current and developing research on ozone
823 changes. Mentioning their hypothesis allows the reader to understand where research on this topic
824 might head in the future, but in this particular section we don't want to further this discussion by
825 adding our own hypotheses on top of those of Parrish et al.

826
827 A related point is on P32735, another possibility is that the measurements may reflect natural
828 climate variability internal to the climate system that a climate model would not represent.

829
830 This is a valid point and we now mention this possibility in lines 3-6 on page 32735.
831 "These differences may reflect poor representation of emission changes or ozone changes due to
832 natural sources, or they may reflect more fundamental weaknesses in resolving key chemical or
833 dynamical processes or climate variability over continental regions."

834
835 P32733. Shifting seasonal cycles over the U.S.A. have recently been discussed by:

836
837 Simon, H., A. Reff, B. Wells, J. Xing and N. Frank (2014). "Ozone Trends Across the
838 United States over a Period of Decreasing NO_x and VOC Emissions." Environmental Science &
839 Technology 49(1): 186-195.

840 Bloomer, B. J., K. Y. Vinnikov and R. R. Dickerson (2010). "Changes in seasonal and
841 diurnal cycles of ozone and temperature in the eastern U.S." Atmospheric Environment
842 44: 2543-2551.

843
844 Clifton, O. E., A. M. Fiore, G. Correa, L. W. Horowitz and V. Naik (2014). "Twentyfirst
845 century reversal of the surface ozone seasonal cycle over the northeastern United
846 States." Geophysical Research Letters 41(20): 2014GL061378.

847
848 This advice is most useful and the references have been added to the discussion on shifting seasonal
849 cycles.

850
851

852 P32471 L20. How does Figure 14 show growth of emissions? The caption suggests that we are only
853 shown 2005 here?
854
855 Text has been changed (as per referee#2) to note magnitudes rather than “growth”
856
857 P32749. Some explanation for why the global satellite data do not provide a complete picture would
858 be useful. It might be worth noting that there are direct tropospheric ozone retrievals, e.g.: Liu, X., et
859 al. (2006), First directly retrieved global distribution of tropospheric column ozone from GOME:
860 Comparison with the GEOS-CHEM model, J. Geophys. Res., 111, D02308, doi:10.1029/2005JD006564.
861
862 The section is dealing with ozone at a climatological scale. Details of the Liu et al reference has been
863 added but a longer discourse on satellite ozone seems to detailed within this context. References are
864 present to guide reader on this topic.
865
866 P32762. The scientifically dubious statement seems overly strong, since it is only fairly recently that
867 computer models have included stratospheric and tropospheric ozone chemistry to enable a
868 combined estimate of RF from ozone.
869
870 This has been changed to “fraught”.
871
872 P32767 “surprisingly small”. If one considers the lower ozone production efficiency in urban plumes,
873 is this so surprising?
874
875 Removed surprising, it is often a truism as to what one finds “surprising” .
876
877 P32773 “factor of five difference”. Are these models all using state-of-the-art isoprene oxidation
878 schemes? This reference may also be relevant: Ito, A., S. Sillman, and J. E. Penner (2009), Global
879 chemical transport model study of ozone response to changes in chemical kinetics and biogenic
880 volatile organic compounds emissions due to increasing temperatures: Sensitivities to isoprene
881 nitrate chemistry and grid resolution, J. Geophys. Res., 114, D09301, doi:10.1029/2008JD011254.
882 How certain is the temperature-driven increase in biogenic emissions in light of CO2-driven
883 suppression?
884
885 This is a good question, a recent paper by Squire et al, (ACP, 2015) has highlighted this as an issue. A
886 comment has been added to this section. The Ito work was already discussed in this section.
887
888 P32803. How important are plumes from one continent to the next versus diffuse background such
889 as resulting from global methane and NOx?
890
891 The best answer to this questions is given in HTAP (2010) - Part D 'Answers to Policy-Relevant Science
892 Questions'
893 (http://www.htap.org/publications/2010_report/2010_Final_Report/HTAP%202010%20Part%20D%20110407.pdf)
894
895
896 "To quantify the relative importance of emission changes outside each of these regions, as compared
897 to emission changes inside each of these regions, we defined the Relative Annual Intercontinental
898 Response (RAIR) metric. RAIR is defined as the sum of the changes in the annual, regionally-averaged
899 concentration within a region due to a 20% decrease in emissions in the three other regions divided
900 by the sum of the changes in concentration within a region due to a 20% decrease in emissions in all
901 four regions. The value of the metric ranges from 0%, indicating no intercontinental influence, up to
902 100%, indicating that air quality in a region is completely dominated by intercontinental sources."
903

904 For O₃, Table 3.1 of HTAP (2010) gives RAIR values for North America (32%), Europe (43%), East Asia
905 (40%) and South Asia (32%).
906

907 Therefore, the diffuse impacts are generally assumed to dominate; additionally enhanced plumes do
908 occur but make a relatively small contribution on the whole, depending on receptor location and
909 meteorology.
910

911 P32805. The U.S. context seems out of place since the previous discussion was on future climate
912 change impacts versus emission changes. There have been several U.S. studies on that topic and
913 reviewing those seems more appropriate here.
914

915 The section referred to deals with the “background” ozone contribution to the future ozone
916 exceedances which is both an issue in the literature for the US and Europe. In the main this details
917 current observations and model attributions of this.
918
919

920 P32808. Not all projections assume air pollution declines. See for example Prather,
921 M., M. Gauss, T. Berntsen, I. Isaksen, J. Sundet, I. Bey, G. Brasseur, F. Dentener, R. Derwent, D.
922 Stevenson, L. Grenfell, D. Hauglustaine, L. Horowitz, D. Jacob, L. Mickley, M. Lawrence, R. von
923 Kuhlmann, J.-F. Muller, G. Pitari, H. Rogers, M. Johnson, J. Pyle, K. Law, M. van Weele and O. Wild
924 (2003). "Fresh air in the 21st century?" *Geophysical Research Letters* 30(2): 1100.
925

926 Reference and discussion added on future directions.
927

928 P32809. Why is IPCC 2007 cited rather than 2013?
929

930 Reference updated and changed to chapter lead.
931

932 The conclusions section reads more as a continuation of discussion, including mentioning tomato
933 volatiles, which doesn't seem to fit. Seems better to focus on robust conclusions that can be drawn
934 from the studies reviewed in the paper, or provide recommendations for tackling some of the
935 challenges outlined on P32811.
936

937 In the conclusions, an attempt to summarise in brief the position but also look to the future as to
938 what some of the new avenues and challenges might be. In some senses the challenge as to the
939 robust conclusions of this paper is a fair one and a few summary sentences have been added.
940

941 The number of figures could probably be reduced, and it would help if figure captions could
942 communicate the relevance of the figure to our understanding of tropospheric ozone. For example,
943 why is PM10 (Fig 26), the nitrogen cascade (Fig 33), methane from fracking (as opposed to other
944 sources; Fig 37), RO2 isomerisation to QOOH (Fig 38) highlighted here?
945

946 Given the length of the paper, the authors did not feel there was an overabundance of figures. It is
947 fair that the captions could be sharper, and they have all been reviewed to more give the relevance
948 as requested. In particular
949

950 Figure 27 (old 26) – PM10 is present owing to source of data. The figure caption has been expanded
951 to explain significance of result.
952

953 Figure 34 (old 33) – more commentary on nitrogen sources and ozone added to caption
954

955 Figure 38 (old 37) – shown as dramatic example of methane emissions. Text details other work on
956 NMVOC emissions from fracking.
957
958 Figure 39 (old 38) – is to help the reader with the chemistry.
959
960 Figure 2. Why not use the more recent ACCMIP models?
961
962 Figure 2 is a good representation to match the text. There is good referencing to the ACCMIP
963 models.
964
965 Fig 3. What is assumed here for BVOC? How sensitive is this picture to assumptions in the UKCA
966 mechanism? Why is this referred to as a ‘schematic’ in the text (p 32717)? Please explain the
967 significance of A/B/C labels in the caption.
968
969 It is not schematic – text changed.

970 A/B/C –added caption

971 As mentioned earlier the Squire et al (2015) reference has been added to the text that explicitly deals
972 with this good point.

973 Fig 7. Can the same colors or symbols be used for the same inventories across the different panels?
974
975 Figure redrawn and colours and symbols matched.

975 Fig 11. What are LDGVs? What are “real driving conditions”? How important are these differences to
976 the ozone distributions?
977
978 Caption amended to explain these are light-duty good vehicles under real world driving conditions
979 rather than test-cycle. In relationship to the importance, in the UK it has been shown that the failure
980 of these standards on the older vehicles has impacted measured NO₂.
981
982 Fig 13. Is this the best estimate, or the only available one? How does this differ in other world
983 regions?
984
985 Figure 13 is an example (as it says in text). Caption has been expanded to include key point re. VOC
986 speciation.
987
988 Fig 18. Is there a diurnal cycle in the fire emissions, and if so, how does that combine with the
989 injection height variation in terms of the impact on tropospheric ozone?
990
991 There is indeed a study of the diurnal variation of the fires injection height in the Sofiev et al. paper.
992 They indicate for example: "Diurnal variation of the injection height is huge: one can practically
993 consider two independent datasets – one for daytime and one for nighttime, with transition during
994 morning and evening." This has been added to the text.

995
996 Fig 41. Is this deemed by the authors to be the best approach to attributing ozone?
997 There are numerous studies attempting to do so over the USA and elsewhere.
998
999 Figure 42 (old 41) is illustrative of an ozone measurement system. There is no implicit endorsement
1000 as to the approach.

1001
1002 Table 2. How were these megacities selected? It's hard to know what to take away with the different
1003 statistics being used for comparison.
1004

1005 The megacities in the table were selected based on data that was available from peer-reviewed
1006 papers or otherwise reliable sources, for a somewhat longer time period (so as to not present ozone
1007 episodes), while still trying to present a geographic mix of locations. A table with comparable data
1008 (e.g., annual average hourly data or daily 8hr maximums) could have been created, however this
1009 then limits the data to one world region, as truly comparable data would likely only be possible for
1010 Europe or the US. Global comparisons of ozone data definitely exist, but tend to focus on rural sites,
1011 and not urban areas. The aim was to give an impression of O₃ values in megacities that do not
1012 represent extreme episodes.

1013 Does Table 3 repeat information in Figure 40?

1014 Table 3 is more complete than Figure 41 (old Figure 40)

1015

1016

1017

1018 **Tropospheric ozone and its precursors from the urban to the global scale**
1019 **from air quality to short-lived climate forcer.**

1020

1021 P.S. Monks,¹ A.T. Archibald,² A. Colette,³ O. Cooper,⁴ M. Coyle,⁵ R. Derwent,⁶ D. Fowler,⁵ C. Granier,
1022 ^{4,7,8} K.S. Law,⁸ G.E. Mills,⁹ D.S. Stevenson,^{9,10} O. Tarasova,^{10,11} V. ~~Thouret~~¹¹, Thouret,¹² E. von
1023 Schneidermeyer,^{12,13} R. Sommariva,¹ O. ~~Wild~~¹³, Wild¹⁴ and M.L. ~~Williams~~¹⁴, Williams¹⁵

1024

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1043 London SE1 9NH, UK.

Formatted: Tab stops: 10.99 cm, Left

1044 **Abstract**

1045

1046 Ozone holds a certain fascination in atmospheric science. It is ubiquitous in the atmosphere, central
1047 to tropospheric oxidation chemistry, yet harmful to human and ecosystem health as well as being an
1048 important greenhouse gas. It is not emitted into the atmosphere but is a by-product of the very
1049 oxidation chemistry it largely initiates. Much effort is focussed on the reduction of surface levels of
1050 ozone owing to its health and vegetation impacts but recent efforts to achieve reductions in
1051 exposure at a country scale have proved difficult to achieve ~~due~~owing to increases in background
1052 ozone at the zonal hemispheric scale. There is also a growing realisation that the role of ozone as a
1053 short-lived climate pollutant could be important in integrated air quality climate-change mitigation.
1054 This review examines current understanding of the processes regulating tropospheric ozone at global
1055 to local scales from both measurements and models. It takes the view that knowledge across the
1056 scales is important for dealing with air quality and climate change in a synergistic manner. The
1057 review shows that there remain a number of clear challenges for ozone such as explaining surface
1058 trends, incorporating new chemical understanding, ozone-climate coupling as well as a better
1059 assessment of impacts. There is a clear and present need to treat ozone across the range of scales, a
1060 transboundary issue, but with an emphasis on the hemispheric scales. New observational
1061 opportunities are offered both by satellites and small sensors that bridge the scales.

1062

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1095		

1096 1. Introduction

1097

1098 Ozone is central to the chemistry of the troposphere owing to its role in the initiation of
1099 photochemical oxidation processes *via* direct reaction, photolysis and the subsequent reactions of
1100 the photoproducts to form the hydroxyl radical (~~Monks, 2005~~)(Monks, 2005). Tropospheric ozone is
1101 also recognised to be a threat to human health (WHO, 2003;Lim et al., 2012), to have a deleterious
1102 impact on vegetation (WHO, 2003;Lim et al., 2012)(Fowler et al., 2009), to have a deleterious impact
1103 on vegetation (Fowler et al., 2009) and, through plant damage it impedes the uptake of carbon into
1104 the biosphere (Sitch et al., 2007). It is also an important tropospheric greenhouse gas and, through
1105 plant damage it impedes the uptake of carbon into the biosphere (Sitch et al., 2007) as well as
1106 impacting built infrastructure (Kumar and Imam, 2013). It is also an important tropospheric
1107 greenhouse gas (IPCC, 2007;Stevenson et al., 2013)(IPCC, 2007;Stevenson et al., 2013) and is referred
1108 to as a short-lived climate pollutant (~~Shindell et al., 2012~~)(Shindell et al., 2012).

1109 Ozone-related deaths are estimated to make up about 5-20% of all those related to air pollution e.g.
1110 (Silva et al., 2013;Anenberg et al., 2009;Lim et al., 2012;Brauer et al., 2012)(Silva et al.,
1111 2013;Anenberg et al., 2009;Lim et al., 2012;Brauer et al., 2012). The OECD (OECD, 2012)(OECD,
1112 2012) have stated that “without new policies, by 2050, air pollution is set to become the world’s top
1113 environmental cause of premature mortality.” The report goes on to state that “Because of their
1114 ageing and urbanised populations, OECD countries are likely to have one of the highest premature
1115 death rates from ground-level ozone”.

1116 While ozone has a relatively short atmospheric lifetime, typically hours, in polluted urban regions
1117 where concentrations of its precursors are high, its lifetime in the free troposphere is of the order of
1118 several weeks (Stevenson et al., 2006;Young et al., 2013)(Stevenson et al., 2006;Young et al., 2013),
1119 sufficiently long for it to be transported over intercontinental scales. Thus in addition to its role as a
1120 priority pollutant on an urban scale, it may influence air quality on a hemispheric scale (Akimoto,
1121 2003;HTAP, 2010)(Akimoto, 2003;HTAP, 2010). There is little doubt that ozone is a multifarious
1122 molecule. Recently, Simpson et al (Simpson et al., 2014)(Simpson et al., 2014) described ozone as
1123 the “persistent menace.”. Figure 1 shows some of the key interactions that drive ozone
1124 concentrations in the troposphere and some of the feedbacks.

1125

1126 ~~Historically, tropospheric ozone was thought to be rather unimportant in atmospheric chemistry~~
1127 ~~terms, being controlled by its main source in the stratosphere and subsequently transported by~~
1128 ~~synoptic scale subsidence and turbulent mixing to the surface, where it is destroyed (Junge, 1962). If~~
1129 ~~tropospheric ozone photochemistry was important at all, then this was limited to highly polluted~~
1130 ~~locations such as to the Los Angeles basin (Leighton, 1961). On this basis, long-range transport~~
1131 ~~appeared to be the dominant process with the tropospheric ozone budget controlled by~~
1132 ~~stratosphere-troposphere exchange and surface destruction. The spring-time ozone maximum which~~
1133 ~~occurred simultaneously with the annual influx of radioactive tracers from the stratosphere,~~
1134 ~~confirmed the importance of transport processes. Crutzen (Crutzen, 1973) proposed the contrary~~
1135 ~~position that photochemical ozone production involving hydrocarbons and oxides of nitrogen may~~
1136 ~~take place throughout the unpolluted troposphere. Furthermore, Chameides and Walker (Chameides~~
1137 ~~and Walker, 1973) suggested that ozone in the lower troposphere was entirely controlled by~~
1138 ~~atmospheric chemistry. These latter suggestions were strongly contested (Chatfield and Harrison,~~

1139 1976; Fabian and Pruchniewicz, 1977) and have been the subject of much debate over the intervening
1140 years (Monks, 2000).

1141 This review examines current understanding of the processes regulating tropospheric ozone at global
1142 to local scales from both measurements and models. It takes the view that knowledge across the
1143 scales is important for dealing with air quality and climate change in a synergistic manner. It would
1144 be a herculean task to review all the literature on ozone, therefore much of the focus of this review is
1145 on the recent findings and discoveries relating to tropospheric ozone. It builds on earlier integrative
1146 reviews such as Brasseur et al (Brasseur et al., 2003)(Brasseur et al., 2003) and the ACCENT reviews
1147 (Fowler et al., 2009; Isaksen et al., 2009; Monks et al., 2009; Laj et al., 2009)(Fowler et al., 2009; Isaksen
1148 et al., 2009; Monks et al., 2009; Laj et al., 2009).

1149 The review has four major sections. The first reviews the control of ozone across the scales, looking
1150 at the interplay of chemistry, transport and deposition, and includes a brief climatological picture of
1151 ozone. The second major section details the major impact of ozone with respect to health,
1152 ecosystems and climate, while the third section highlights several current topics in relation to ozone
1153 in the troposphere. The final section briefly overviews some of the policy context and issues in
1154 relation to tropospheric ozone. In the conclusion, some future directions and issues with relation to
1155 ozone are discussed.

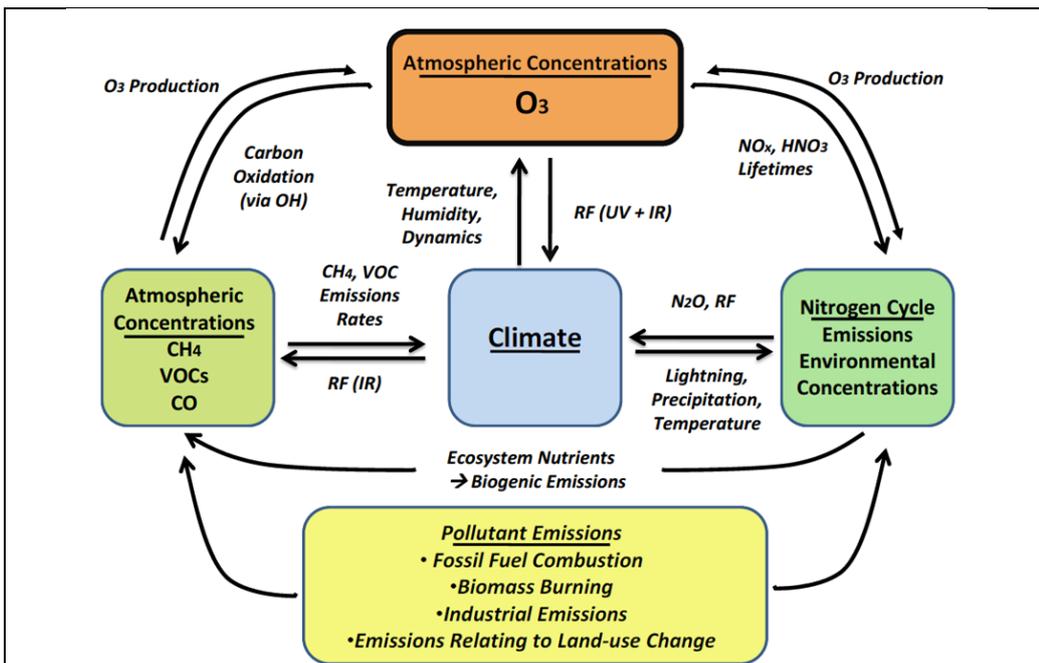


Figure 1 – Schematic representation of the interactions of ozone in the earth system (EPA, 2009)(EPA, 2009).

1156

1157

1158 2 Ozone – control, precursors and climatology

1159 **2.1 What controls ozone?**

1160

1161 The annual variation in ozone concentration at any given spatial scale depends on a number of
1162 factors, such as the proximity to large sources of ozone precursors, geographical location and the
1163 prevailing meteorological conditions (~~Logan, 1985~~)(~~Logan, 1985~~). The tropospheric ozone budget at
1164 a given location is dependent on both photochemical processes and physical processes, including
1165 photochemical production and destruction of ozone, transport from upwind sources and removal at
1166 the Earth's surface (~~Monks, 2000;Lelieveld and Dentener, 2000~~)(~~Monks, 2000;Lelieveld and~~
1167 ~~Dentener, 2000~~).

1168

1169 Given the substantial stratospheric ozone concentrations, transport from the stratosphere was long
1170 thought to be the dominant source of ozone in the troposphere (~~Danielsen, 1968;Junge, 1962;Fabian~~
1171 ~~and Pruchniewicz, 1977~~)(~~Danielsen, 1968;Junge, 1962;Fabian and Pruchniewicz, 1977;Chatfield and~~
1172 ~~Harrison, 1976~~). Photochemical mechanisms for the generation of ozone were first identified in work
1173 carried out in California in the 1950s (~~Haagen-Smit, 1952~~)(~~Haagen-Smit, 1952~~) and until the 1970's
1174 high ozone was thought to be a local phenomenon associated with air pollution- (~~Leighton, 1961~~).
1175 Early in the 1970's Crutzen (~~Crutzen, 1973~~)(~~Crutzen, 1973~~) and Chameides and Walker (~~Chameides~~
1176 ~~and Walker, 1973~~)(~~Chameides and Walker, 1973~~) suggested that tropospheric ozone originated
1177 mainly from production within the troposphere by photochemical oxidation of CO and hydrocarbons
1178 catalysed by HO_x and NO_x. Since then, improved understanding of the importance of both natural
1179 and anthropogenic sources of ozone precursors has highlighted the dominance of this ozone source,
1180 and recent model assessments suggest that chemical production contributes about 5000 Tg/yr to
1181 global tropospheric ozone (~~Stevenson et al., 2006;Young et al., 2013~~)(~~Stevenson et al., 2006;Young et~~
1182 ~~al., 2013~~). In contrast, the net global source of ozone from stratosphere-troposphere exchange (STE)
1183 is estimated to be about 550±140 Tg/yr based on observational constraints (~~Olson et al.,~~
1184 ~~2001;McLinden et al., 2000~~)(~~Olson et al., 2001;McLinden et al., 2000~~). The removal of ozone through
1185 dry deposition to vegetation and other surfaces at ground level remains poorly constrained on the
1186 global scale, although model assessments driven by observed ecosystem fluxes, vegetation types and
1187 meteorology suggest net removal of about 1000±200 Tg/yr (~~Stevenson et al., 2006~~)(~~Stevenson et al.,~~
1188 ~~2006~~). Balancing the global tropospheric ozone budget therefore requires that there is net chemical
1189 production of about 450±300 Tg/yr (~~Stevenson et al., 2006~~)(~~Stevenson et al., 2006~~). Note that this
1190 term is constrained by the estimated STE and deposition terms and by the global tropospheric ozone
1191 burden of about 335±10 Tg, and that uncertainty in the absolute magnitude of gross ozone chemical
1192 production and destruction remains relatively large (~~Wild, 2007~~)(~~Wild, 2007;Wu et al., 2007~~).

1193

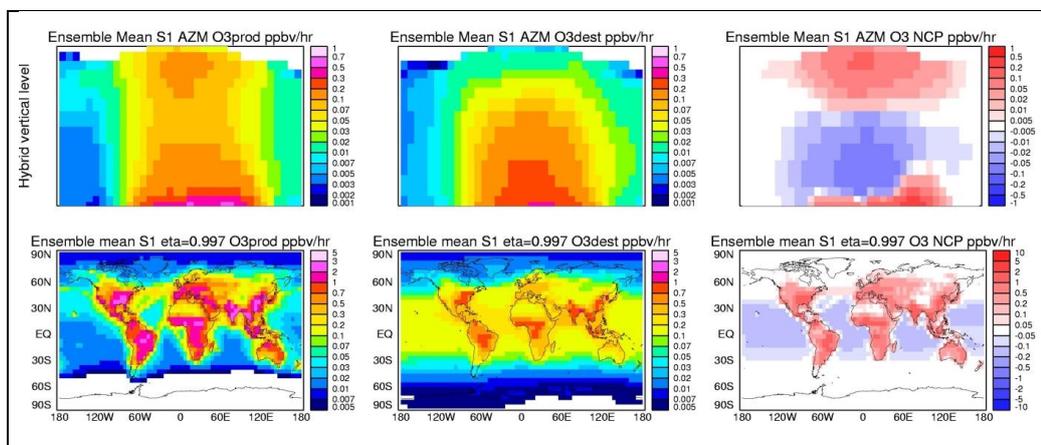


Figure 2 - Multi-model mean (20 models) year 2000 ozone budgets (units: ppbv/hr). Top row: annual zonal mean (AZM) chemical production; chemical destruction; net chemical production. Vertical scale is from surface to approximately 100 hPa. Bottom row: maps of same quantities in the surface model layer (Stevenson et al., 2006)(Stevenson et al., 2006).

1194

1195 These global budgets mask substantial regional variation (see Figure 2) which spans the wide range
 1196 of chemical environments present in the troposphere. Troposphere-wide chemical production of
 1197 ozone from long-lived hydrocarbons such as methane is greatest in the tropical lower troposphere
 1198 where OH radical concentrations are high (Bloss et al., 2005)(Bloss et al., 2005), and in continental
 1199 regions and parts of the free troposphere with fresh NO_x emissions. Chemical destruction of ozone is
 1200 largest in the lower troposphere where water vapour concentrations are high and in highly polluted
 1201 regions where there is direct removal by reaction with NO. This latter condition is commonly referred
 1202 to as chemical titration of ozone, and is typically short-lived as the NO₂ formed subsequently
 1203 catalyses further ozone formation once it has been transported into cleaner environments. Localised
 1204 halogen-catalysed ozone destruction may also affect surface ozone in polar regions during
 1205 spring time (see Halogens section). Consequently, net chemical production occurs over most of the
 1206 continental boundary layer where precursor concentrations are high and in the upper troposphere,
 1207 where destruction is slow (Wild and Palmer, 2008; von Kuhlmann et al., 2003b, a)(Wild and Palmer,
 1208 2008; von Kuhlmann et al., 2003b, a). Net chemical destruction occurs in the mid-troposphere and
 1209 marine boundary layer where precursor concentrations are low and in localised urban and Polar
 1210 Regions under titration (via reaction with NO) and halogen destruction conditions, respectively.
 1211 Despite large changes in the emissions of ozone precursors and in net chemical production, the
 1212 general location of these net production and loss regimes is thought to have changed little since
 1213 preindustrial times (Wild and Palmer, 2008)(Wild and Palmer, 2008). However, spatial heterogeneity
 1214 of anthropogenic precursor emissions and of biogenic hydrocarbons (Zare et al., 2014)(Zare et al.,
 1215 2014), along with strong diurnal variability in chemical processes, deposition and meteorology,
 1216 suggests that local ozone production and loss regimes in the boundary layer may be much more
 1217 diverse than indicated by these coarse, regional-scale assessments.

1218

1219 2.1.1 Role of Chemistry

1220

1221 The basic chemistry that leads to the production and destruction of ozone has been detailed
 1222 elsewhere (~~Monks, 2005;Ehhalt, 1999;Jenkin and Clemitchaw, 2000~~)(~~Monks, 2005;Ehhalt,~~
 1223 ~~1999;Jenkin and Clemitchaw, 2000~~). Ozone (O₃) photochemical production in the troposphere occurs
 1224 by hydroxyl radical oxidation of carbon monoxide (CO), methane (CH₄), and non-methane
 1225 hydrocarbons (generally referred to as NMHC) in the presence of nitrogen oxides (NO_x). The
 1226 mechanism of ozone formation in the troposphere, although very well understood, remains a
 1227 challenge ~~that will, no doubt, require many more years of research to fully unravel in practice that~~
 1228 ~~will, no doubt, require many more years of research to fully unravel. Typical photochemical~~
 1229 ~~timescales in the atmosphere are quite short (in the order of seconds to tens of minutes) (Seinfeld~~
 1230 ~~and Pandis, 2006)~~. In brief, the production of ozone in the troposphere relies, fundamentally, on the
 1231 photolysis of NO₂ (reaction ~~1R1~~) and the subsequent association of the photoproduct O(³P) (the
 1232 ground electronic state oxygen atom) with O₂ *via* reaction 2 through a termolecular reaction with a
 1233 third body (M being used to represent any third co-reactant i.e. N₂):



1236 The difficulty with understanding the production of O₃ in the troposphere comes about through the
 1237 manifold ways in which NO and NO₂ are interconverted. The rapid interconversion between the two
 1238 compounds allows them to be treated as a chemical family – NO_x. (NB photolysis of NO₂ is fast, and
 1239 approximately altitude independent – ~~1.1~~ $\approx 10^{-2} \text{ s}^{-1}$). For example, the reactions



1242 both convert NO into NO₂. Considering reactions 1,2 and 4 one can derive a ratio of [NO]/[NO₂] that
 1243 depends on the local concentration of O₃ and the rate coefficients for reaction (4) and the photolysis
 1244 frequency for reaction (1) (at the surface the pseudo first order rate constant k_2' ($k_2 \cdot [\text{O}_2] \cdot [\text{M}]$) is
 1245 sufficiently fast that it is not a rate limiting step). This ratio is termed the Leighton ratio ~~(Leighton,~~
 1246 ~~1961)(Leighton, 1961)~~ and also allows for an expression to be derived for the equilibrium
 1247 concentration of O₃. Whilst this chemistry is important, particularly in urban areas, it does not
 1248 represent a mechanism for the net production of O₃ in the troposphere. ~~Considering~~With the
 1249 addition of volatile organic compounds, such as CO: ~~viz~~



1251 ~~We are able~~It is possible to write a mechanism for the formation of O₃ that is propagated *via* VOC
 1252 (~~Volatile Organic Compound~~) and NO_x (reactions (5), (3), (2) and (1)). In this series of reactions O₃ is
 1253 used as a source of the hydroxyl radical (OH) through:



1256 where O(¹D) is the electronic excited state atomic oxygen formed through photolysis at wavelengths
 1257 <320 nm. However, the presence of VOC (CO) and NO_x, allow OH to be regenerated (~~R3~~) and
 1258 promote formation of O₃ through NO₂ photolysis. The production of O₃ in the troposphere, mediated

1259 | through the reactions involving VOC (CH₄) and NO_x can be shown schematically in Figure 3: and in
1260 | relationship to changing precursor concentrations in Figure 4.

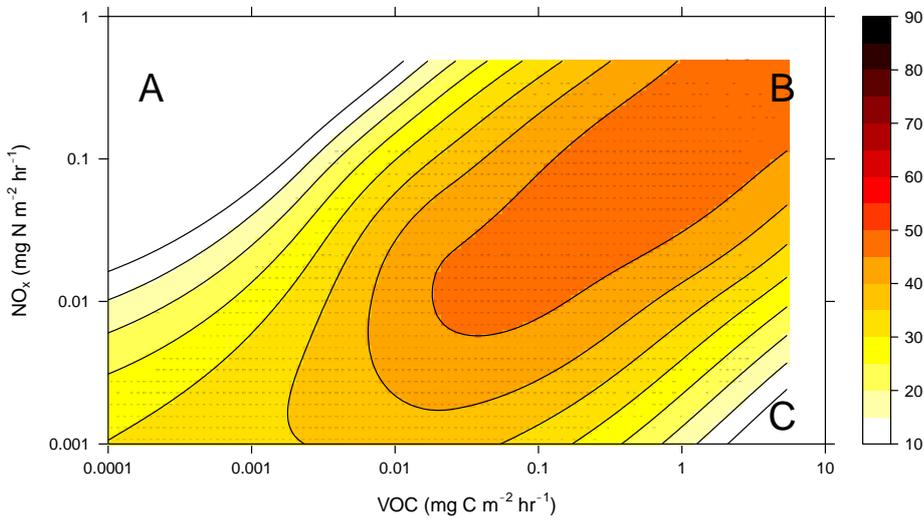
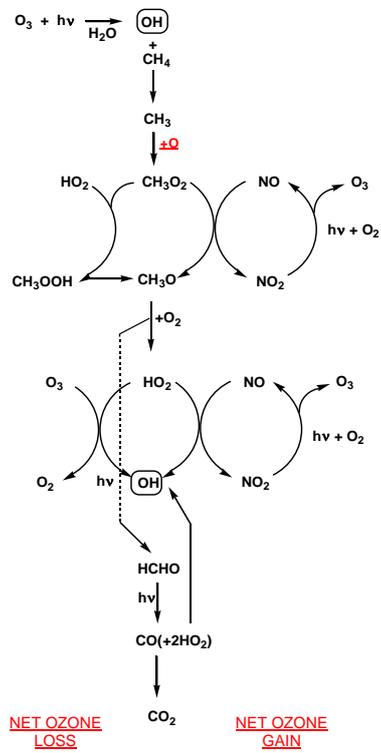


Figure 3 – O_2 -mixing ratios (ppb) as a function of VOC and NO_x emissions as computed using the UKCA model of atmospheric chemistry (Archibald et al., 2011). Three main regions are identified. Top

left corner: region of NO_x saturation and O₃ titration. Bottom right corner: region of VOC saturation and O₃ destruction. Diagonal elements: efficient conversion of NO-NO₂ and hence O₃ production increasing with increasing VOC and NO_x emissions (NB log₁₀ scales for emissions). **Figure 3** - Simplified mechanism for the photochemical oxidation of CH₄ in the troposphere (Lightfoot et al., 1992).

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1261

1262 **Figure 3**

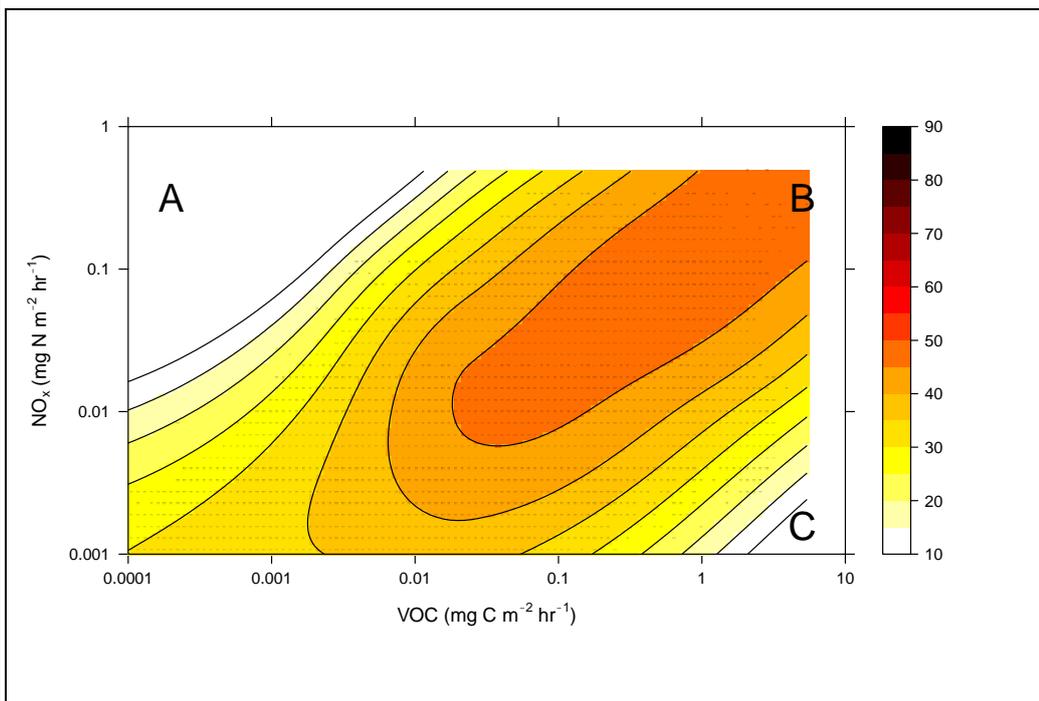


Figure 4 - O₃ mixing ratios (ppb) as a function of VOC and NO_x emissions as computed using the UKCA model of atmospheric chemistry (Archibald et al., 2011). Three main regions are identified. Top left corner (A): region of NO_x saturation and O₃ titration. Bottom right corner (C): region of VOC saturation and O₃ destruction. Diagonal elements (A-C, B-C): efficient conversion of NO-NO₂ and hence O₃ production increasing with increasing VOC and NO_x emissions (NB log₁₀ scales for emissions).

1263

1264 **Figure 4** highlights the non-linearity of the O₃-VOC-NO_x system. The O₃ mixing ratios presented in
1265 Figure 34 are generated from ~~photo-chemical~~photochemical modelling output generated using the
1266 UKCA model (O'Connor et al., 2014;Archibald et al., 2011)(O'Connor et al., 2014;Archibald et al.,
1267 2011). Regions in Figure 34 where there is net O₃ destruction (top left (A) and bottom right corners)
1268 (C) are typically referred to as the VOC limited and NO_x limited regimes. VOC limited refers to the
1269 fact that the production of O₃ is limited by the input of VOC (see e.g. (Zhou et al., 2014)(Zhou et al.,

1270 | [2014](#)). This can be rationalized by moving from point A to point B along a line of constant NO_x
1271 | emissions and increasing VOC emissions. Moving in this direction it is evident from Figure 34 that O₃
1272 | production increases. A similar case applies for the NO_x limited regime where this time moving from
1273 | point C to point B requires increasing NO_x emissions and results in increasing O₃ mixing ratios.

1274 | This general mechanism of OH initiated O₃ formation can be extended to more complex VOC such as
1275 | alkanes:



1278 | Where C₂H₅O₂ represents a member of a group of radicals termed organic peroxy radicals (RO₂ –
1279 | where R is used to represent alkyl, ally, or aryl groups), all of which possess the ability to convert NO
1280 | to NO₂. The fate of the alkyl oxy radical (C₂H₅O) is more complex and can result in the formation of
1281 | HO₂ and other VOCs. It is the organic atmospheric chemistry of RO₂ that remains the great challenge
1282 | to our full understanding of the production of O₃ in the troposphere. Recent advances in our
1283 | understanding of the fate of RO₂ in the atmosphere are reviewed in section 4.10.

1284 | There are added complexities to the chemistry outlined above owing to the variety of sources of OH
1285 | e.g. from the photolysis of HONO (~~Kim et al., 2014~~)([Kim et al., 2014](#)), HCHO and the reaction of ozone
1286 | with alkenes. The Monks et al review (~~Monks et al., 2009~~)([Monks et al., 2009](#)) looked at much of the
1287 | variety of ozone photochemistry in more detail.

1288 | [2.1.2 Role of Deposition](#)

1289

1290 | The concentration of ozone in the surface boundary layer is regulated by three processes:
1291 | atmospheric transport, chemical production/destruction and losses to surface by dry deposition.
1292 | Ozone is physically transported to the surface by atmospheric turbulence which ~~is can be~~ readily
1293 | measured or modelled using well established methods. The rate of ozone removal at the surface
1294 | strongly influences the exposure of vegetation and the human population to ozone and is responsible
1295 | for much of the nocturnal decline in surface ozone in rural areas as deposition to the surface
1296 | consumes ozone beneath a nocturnal temperature inversion. During the day, vertical transport of
1297 | ozone to the surface layers is generally sufficient to maintain mixing ratios within 10% of the
1298 | boundary layer mean values, except in urban areas or near major roads, where local nitric oxide
1299 | sources remove ozone by titration (~~Colette et al., 2011~~)([Colette et al., 2011](#)). Thus the interplay
1300 | between dry deposition of ozone in the surface layers and mixing from higher levels in the
1301 | atmosphere plays a major role in regulating ozone exposure of ecosystem and the human
1302 | population. The mixing in surface layers is largely driven by wind and its interaction with frictional
1303 | drag at the surface. The large diurnal variability in ozone mixing ~~rationsratios~~ in rural areas and its
1304 | variability with altitude has been used to quantify the spatial variability in ozone exposure at the
1305 | surface.

1306

1307 | Ozone dry deposition has been widely measured using micrometeorological methods, which average
1308 | the flux to the surface at the field to landscape scale, over typically (10⁴-10⁶ m²) (~~Fowler et al.,~~
1309 | [2009](#))([Fowler et al., 2009](#)). The controlling processes in ozone deposition are generally simulated

1310 numerically using a resistance analogy (Erisman et al., 1994)(Erisman et al., 1994), as illustrated in Fig
 1311 4Figure 5. Correct parameterisation in models is critical to the determination of ozone budgets
 1312 (Val Martin et al., 2014)(Val Martin et al., 2014).
 1313

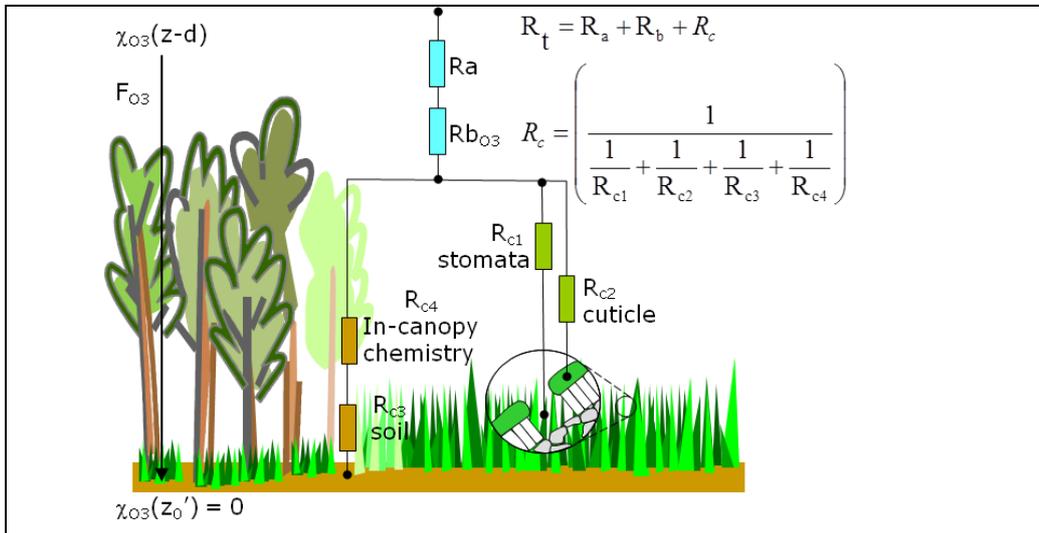


Figure 45 - The resistance analogy for ozone deposition to vegetation and soil- (for details of nomenclature see text).

1314
 1315 Ozone is a reactive gas and deposits readily on most surfaces simply by oxidative reactions (Grontoft,
 1316 2004)(Grontoft, 2004). Many studies have concluded that stomatal uptake is the main factor
 1317 regulating surface fluxes, but as stomata are usually open only a fraction of the time during daylight
 1318 hours, the smaller rates of deposition to non-stomatal surfaces are often a dominant component of
 1319 the annual deposition to the surface (Fowler et al., 2009)(Fowler et al., 2009).

1320 Considering the main sinks for ozone deposition at the surface in turn:

1321 a) Stomatal Uptake

1322 Plants open their stomata to take up CO₂ for photosynthesis and regulate stomatal opening to
 1323 minimise water loss by transpiration (Farquhar et al., 1980)(Farquhar et al., 1980). This also allows
 1324 the uptake of other gases that may not benefit the plant, including ozone, which as a powerful
 1325 oxidant, generates free radicals within the apoplast and cell fluids and is responsible for damage to
 1326 cell metabolism (Mills et al., 2011)(Mills et al., 2011a). Stomatal opening is controlled by water
 1327 pressure in the guard cells which the plant regulates depending on: light, in general stomata open
 1328 during the day and close at night; carbon dioxide concentration; plant water, with increasing CO₂
 1329 above current ambient inducing partial closure; soil water content, with stomata closing in drier soils
 1330 to control/reduce water loss; temperature, in most conditions with an increase in temperature
 1331 increases increasing stomatal opening. up to a species-specific maxima above which stomatal closing
 1332 is induced; humidity, with greatest stomatal opening under low vapour pressure deficits and
 1333 stomatal closing occurring at high vapour pressure deficits to conserve plant water; and phenology

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1334 ~~with stomatal conductance reducing as plants age. An example of the range of stomatal responses~~
 1335 ~~to these environmental and plant controls can be found for wheat in Grünhage et al. (Grünhage et~~
 1336 ~~al., 2012).~~ Other factors can affect stomatal opening although not as directly as those listed above:
 1337 ~~the age of a plant has an influence as older cells become damaged and respond more slowly to~~
 1338 ~~stimuli; pollutant gases or, for example,~~ aerosols can damage the guard cells or block stomata; ~~and~~
 1339 surface water ~~blockscan also block~~ stomata.

1341 The diffusion pathway through stomata as with atmospheric fluxes, can be quantified using a
 1342 resistance analogy as illustrated in ~~FigureFig.~~ 4. The inverse of stomatal resistance is stomatal
 1343 conductance (g_s) and is widely used by plant scientists to quantify stomatal exchanges of CO_2 and
 1344 H_2O . ~~The inverse of the total resistance between a reference height in the atmosphere and the~~
 1345 ~~absorbing surfaces is referred to as a deposition velocity (V_d) and is widely used to quantify dry~~
 1346 ~~deposition of pollutant gases in transport and deposition models,~~ and has also been used to develop
 1347 ~~stomatal flux-based risk assessments for ozone (Emberson et al., 2000;Mills et al., 2011b).~~ The
 1348 processes responsible for controlling stomatal function have been extensively studied in the field and
 1349 laboratory leading to a variety of instruments and models to quantify stomatal resistance, R_{c1} , or
 1350 conductance, g_s .

1351 **b) Non-Stomatal Uptake**

1353 The reaction of ozone with external surfaces of vegetation is referred to as non-stomatal uptake. In
 1354 controlled conditions in the laboratory, the flux may be measured quite straightforwardly by treating
 1355 the vegetation in order to fully close the stomata. In the field, there is the complication that multiple
 1356 sinks are present (leaf surfaces, senescent vegetation, soil and water) ~~as well as stomatal opening.~~
 1357 The usual approach to quantify non-stomatal deposition is by the difference between the total
 1358 deposition flux and the stomatal flux, choosing surfaces to minimise the soil and senescent
 1359 vegetation components. With reference to Figure 45, once R_{c1} has been determined from
 1360 measurements or by modelling the sum of R_{c2} , R_{c3} and R_{c4} (non-stomatal, R_{ns}) can be calculated as the
 1361 residual term:

1362
$$R_{ns} = \left(\frac{1}{R_c} - \frac{1}{R_{c1}} \right)^{-1}$$

 1363 ~~(101)~~

1364 Whether it is possible to separate the components of R_{ns} (R_{c2} – external surfaces, R_{c3} – soil, R_{c4} – in-
 1365 canopy chemistry) depends on the nature of the measurement site and canopy and there are some
 1366 models available based on measurements over bare soil, senescent vegetation and in-canopy
 1367 chemistry ~~(Launiainen et al., 2013;Fares et al., 2013;Fares et al., 2012;Bueker et al., 2012;Tuzet et al.,~~
 1368 ~~2011;Stella et al., 2011)(Launiainen et al., 2013;Fares et al., 2013b;Fares et al., 2012;Bueker et al.,~~
 1369 ~~2012;Tuzet et al., 2011;Stella et al., 2011).~~ Many research groups have taken this approach and Table
 1370 1 provides a summary of some of the different estimates of the values for R_{ns} in the literature.

Table 1 - Selected estimates of the non-stomatal resistance for ozone to snow, water, soil and plant surfaces from measurements.

Reference	Type	Surface	R_{ns} , s m^{-1}
(Chang et al., 2002)(Chang et al., 2002)	chamber Chamber	Agricultural soil (no data on moisture content)	ca 625 ca 475
(Wesely et al., 2002)	Field μ met	Wet bare soil	1000 \pm 100

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{1981}(Wesely et al., 1981)		Snow	-11°C	(370 – 2100)
			-5°C	2000 ± 200
			-1 to 2 °C	3500 ± 200
		Lake water		3300 ± 300
				9000 ± 300
{Sanchez et al., 1997}(Sanchez et al., 1997)	Field µmet	Semi-arid steppe (wet and dry)		Dry 275 Wet 437
{Rondon et al., 1993}(Rondon et al., 1993)	Field chambers	Coniferous trees		200 – 330
{Granat and Richter, 1995}(Granat and Richter, 1995)	Field chambers	Pine		500 – 2500
{Coe et al., 1995}(Coe et al., 1995)	Field µmet	Sitka spruce		ca 133
{Fowler et al., 2001}(Fowler et al., 2001)	Field µmet	Dry moorland		200 – 400
{Grantz et al., 1995}(Grantz et al., 1995)	Field µmet	Wet and dry grape		Dry 1020 Wet 292 ^a
{Grantz et al., 1997}(Grantz et al., 1997)	Field µmet	Wet and dry cotton		Dry 770 Wet 3030
{Zhang et al., 2002}(Zhang et al., 2002)^b	Field µmet	Wet and dry:		
		Mixed forest		244 – 970
		Deciduous forest		397 – 1831
		Corn		308 – 1332
		Soybean		137 – 735
		Pasture		571 – 879
{Gerosa et al., 2005}(Gerosa et al., 2005)	Field µmet	Mediterranean Oak (Quercus ilex)		Dry 249 Wet 177
{Gerosa et al., 2009}(Gerosa et al., 2009)				Avg 67 – 204
{McKay et al., 1992}(McKay et al., 1992) and references therein	Laboratory chambers and field	Sea water & saline solutions		650 – 6600
{Gallagher et al., 2004}(Gallagher et al., 2004)	Field µmet	Coastal waters		950 ± 70
{Coyle et al., 2009}(Coyle et al., 2009)	Field µmet	Potatoes		276
		Wet canopy		
		Dry canopy		
{Neiryck et al., 2012}(Neiryck et al., 2012)	Field µmet	Temperate Forest		136

a from data reported in ~~{Grantz et al., 1997}(Grantz et al., 1997)~~

b data derived from references therein

1371

1372 The literature reviewed in Table 1 reports estimates of R_{ns} , although few examine the controlling
1373 factors. In some of the studies, surface factors affecting ozone deposition, other than stomatal
1374 uptake, have been considered. ~~The main focus of work has been on the effects of~~For example,
1375 surface water ~~and researchers have been~~ found ~~to~~ both ~~positive~~enhance and ~~negative effects~~
1376 ~~on~~reduce deposition rates. The remainder of studies have considered solar radiation or surface
1377 temperature and found deposition rates increased with both variables, as well as in-canopy
1378 chemistry.

1379 c) Solar Radiation and Surface Temperature

1380 Given its reactive nature it is not unreasonable to find effects of surface temperature on R_{ns} and
1381 indications of such an effect were first reported by Rondon et al ([Rondon et al., 1993](#))([Rondon et al.,](#)
1382 [1993](#)). In their measurements of ozone deposition to a coniferous forest they found that the surface
1383 resistance to ozone was much lower than that predicted by stomatal conductance alone and the
1384 residual term (R_{ns}) varied with air temperature, radiation and stomatal conductance. It was proposed
1385 that this effect was due to temperature increasing the reaction rates of ozone with organic
1386 compounds on the canopy surface. Coyle et al ([Coyle et al., 2009](#))([Coyle et al., 2009](#)) found similar
1387 results in measurements for ozone uptake by potatoes, while also showing that surface wetness
1388 could enhance or reduce non-stomatal uptake depending on the initial status of the cuticles. While
1389 ([Fowler et al., 2001](#))([Fowler et al., 2001](#)) showed a clear reduction in R_{ns} with increasing total solar
1390 radiation and temperature. Cape et al ([Cape et al., 2009](#))([Cape et al., 2009](#)) examined deposition to
1391 metals and artificial leaf surfaces and found a strong temperature response, which yielded activation
1392 energies for the reaction of ~ 30 kJ mol⁻¹, similar to that found by Fowler *et al* ([Fowler et al.,](#)
1393 [2001](#))([Fowler et al., 2001](#)) for moorland of 36 kJ mol⁻¹ and from other unpublished studies ([Coyle et](#)
1394 [al., 2009](#))([Coyle et al., 2009](#)). The simplest explanation of non-stomatal ozone deposition is that it
1395 represents the thermal decomposition of ozone at the surface, which increases with surface
1396 temperature. Other, more complex processes have been suggested including rapid reactions with
1397 very reactive hydrocarbon compounds ([Hogg et al., 2007](#))([Hogg et al., 2007](#)).

1398 d) In-canopy chemistry

1399 The reaction of ozone with nitric oxide (NO) is an important cause of reduced ozone concentrations
1400 in urban areas. The same reaction also reduces ozone concentrations close to the surface where soil
1401 emissions of NO are large. Some types of vegetation such as coniferous forests also emit highly
1402 reactive volatile organic compounds ([Di Carlo et al., 2004](#))([Di Carlo et al., 2004](#)) which can rapidly
1403 deplete ozone concentrations immediately above the canopy ([Neiryck et al., 2012](#);[Stjernberg et al.,](#)
1404 [2012](#))([Neiryck et al., 2012](#);[Stjernberg et al., 2012](#);[Kurpius and Goldstein, 2003](#)). Over some forest
1405 canopies the major non-stomatal sink has been attributed to these reactions although the
1406 compound(s) responsible have not been identified. Other transient events such as grass cutting,
1407 which releases very reactive hydrocarbons ([Davison et al., 2008](#))([Davison et al., 2008](#)) can also lead to
1408 enhanced ozone deposition as a result ([Coyle, 2005](#))([Coyle, 2005](#)).

1409 e) Deposition to Water

1410 It has often been assumed that ozone deposition rates to water will be small and relatively constant
1411 as ozone has a low solubility in water. The resistance of a water layer to the uptake of ozone by
1412 dissolving and diffusing the gas is very large and values of 3×10^5 to 8×10^5 s m⁻¹ have been suggested
1413 ([Wesely et al., 1981](#))([Wesely et al., 1981](#)). They concluded that the far lower values found in their
1414 measurements (Table 1) resulted from surface chemical reactions, which has been supported by
1415 further work since then. However measurements have shown that over vegetated surfaces
1416 deposition can be either enhanced or suppressed by the presence of water films (([Fuentes et al.,](#)
1417 [1992](#))([Fuentes et al., 1992](#))), ([Padro, 1994](#))([Padro, 1994](#))), ([Grantz et al., 1995](#);[Grantz et al.,](#)
1418 [1997](#))([Grantz et al., 1995](#);[Grantz et al., 1997](#)), ([Pleijel et al., 1995](#))([Pleijel et al., 1995](#))), ([Coyle et al.,](#)
1419 [2009](#))([Coyle et al., 2009](#))) while over open water deposition varies with water chemistry, turbidity
1420 and agitation. The deposition velocity of ozone to open water is generally small, ~ 0.001 to 0.04 cm s⁻¹

1421 | ~~(Wesely et al., 1981;Chang et al., 2004)~~(Wesely et al., 1981;Chang et al., 2004) and increases in
1422 | deposition velocities to sea water have been observed with disturbance to the surface. McKay et al
1423 | ~~(McKay et al., 1992)~~(McKay et al., 1992) showed that increasing concentrations of chemical
1424 | surfactants in the seawater also enhanced deposition (see Table 1 for typical resistance values).
1425 | These results show that although deposition rates to water are generally small they can be significant
1426 | if other reactive compounds are present in the water, a conclusion also supported by measurements
1427 | to wet vegetated canopies. Research into ozone reactivity and solubility for the ozone water
1428 | treatment industry has highlighted the role of pH. Ganzeveld et al have explored the impact of dry
1429 | deposition of ozone over oceans ~~(Ganzeveld et al., 2009)~~(Ganzeveld et al., 2009). On a global scale,
1430 | model studies indicate that deposition to the ocean constitutes almost 40% of the total annual
1431 | surface dry deposition of ozone, far larger than that from any other land cover class ~~(Hardacre et al.,~~
1432 | ~~2014)~~(Hardacre et al., 2014).

1433
1434
1435
1436

1437 | 2.1.3 Transport and mixing processes

1438

1439 | a) 2.1.3.1 Long-range transport

1440

1441 | Research over the past two decades has clearly shown that ozone and ozone precursors are regularly
1442 | exported from their emission source, or point of production, to receptor regions far downwind on
1443 | the regional, intercontinental and even hemispheric scale (see reviews by Stohl and Eckhardt ~~(Stohl~~
1444 | ~~and Eckhardt, 2004)~~(Stohl and Eckhardt, 2004), Monks et al. ~~(Monks et al., 2009)~~(Monks et al., 2009),
1445 | National Research Council ~~(Council, 2009b)~~(Council, 2009b) and ~~(HTAP, 2010)~~(HTAP, 2010)). These
1446 | transport pathways are predominantly from west to east at mid-latitudes with in situ and satellite-
1447 | based observations showing that pollution plumes (anthropogenic and biomass burning) not only
1448 | travel from North America to Europe, or from East Asia to North America, but can also circle the
1449 | globe ~~(Jacob et al., 1999;Jaffe and et al., 1999;Lewis et al., 2007;Wild et al., 2004)~~(Jacob et al.,
1450 | ~~1999;Jaffe and et al., 1999;Lewis et al., 2007;Wild et al., 2004)~~.

1451

1452 | The scientific community has a very good understanding of the meteorological mechanisms that
1453 | export pollution from the boundary layer of a source region (warm conveyor belts, deep convection,
1454 | and horizontal advection) and then transport these plumes through the free troposphere towards
1455 | the downwind receptor region. The most recent development is the conceptualization of
1456 | intercontinental transport within an isentropic framework that illustrates how transport of pollution
1457 | plumes from East Asia are constrained by the thermal structure of the atmosphere, preferentially
1458 | impacting the high elevation terrain of the western USA rather than the relatively low-lying eastern
1459 | USA ~~(Brown-Steiner and Hess, 2011)~~(Brown-Steiner and Hess, 2011). The major uncertainty
1460 | concerning long-range transport is estimating the contribution of the imported pollution to the total
1461 | quantity of pollution at the surface of the receptor region. Some unique chemical tracers of upwind
1462 | pollution sources do exist, for example, stable lead isotopes can indicate events when Asian

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1463 | particulate matter reaches the surface of California (~~Ewing et al., 2010~~)(Ewing et al., 2010). However,
1464 | at present there are no routine and widespread monitoring methods that can distinguish ozone and
1465 | ozone precursors produced in a receptor region, such as California, from the same species produced
1466 | in an upwind region, such as East Asia.

1467 |
1468 | The only feasible method for quantifying the impact of imported pollution at the surface of a
1469 | receptor region is to use chemical transport models or chemistry-climate models. Such models have
1470 | been used for this purpose since the 1990s but until recently their effectiveness has been limited by
1471 | their coarse horizontal resolution, typically 2x2 degrees (~~HTAP, 2010~~)(HTAP, 2010). Such large grid
1472 | cells artificially dilute the imported pollution plumes as well as local emissions of ozone precursors in
1473 | the receptor region, and also smooth the topography of the receptor region, removing the influence
1474 | of terrain-driven transport mechanisms that affect the mixing of imported pollution down to the
1475 | surface. However, recent model studies have used finer scale grids to avoid the problems associated
1476 | with coarse resolution models (~~Lin et al., 2012b;Huang et al., 2013;Zhang et al., 2014;Choi et al.,~~
1477 | ~~2014~~)(Lin et al., 2012b;Huang et al., 2013;Zhang et al., 2014;Choi et al., 2014). Because these finer
1478 | resolution models provide more realistic simulations of observed pollution plumes scientists have
1479 | greater confidence in their quantification of the contribution of imported pollution at the surface of a
1480 | receptor region. Despite the improvements afforded by finer resolution, a recent study of the
1481 | quantity of background ozone advected into western North America shows monthly mean
1482 | differences as great as 10 ppbv between models, with the discrepancy attributed to differences in
1483 | the way that the models handle lightning NO_x, biogenic isoprene emissions and chemistry, wildfires,
1484 | and stratosphere-to-troposphere transport (~~Fiore et al., 2014~~)(Fiore et al., 2014).

1485 |
1486 | Future climate change may affect the contribution of long-range transport downwind effects
1487 | (~~Glotfelty et al., 2014;Doherty et al., 2013~~)(Glotfelty et al., 2014;Doherty et al., 2013). HTAP (2010)
1488 | suggests “changes in climate will affect meteorological transport processes as well as the chemical
1489 | environment and lifetime of the transported pollutants and hence the concentrations of pollutants
1490 | arriving at downwind continents.” Glotfelty et al. (2014) suggest a larger impact on the US from East
1491 | Asian Emissions and Doherty et al. (2013) showed stronger chemistry than transport positive climate
1492 | feedbacks on increased ozone.

1493 | 1494 | 2.1.3.2 Seasonal transport patterns:

1495 | b) Stratosphere-troposphere exchange and summer monsoons

1496 |
1497 | The tropospheric ozone burden varies seasonally as measured by the remotely sensed AURA
1498 | OMI/MLS tropospheric column product (~~Ziemke et al., 2006~~)(Ziemke et al., 2006). The region of the
1499 | Earth with the strongest seasonal ozone variation is the northern mid-latitudes where the ozone
1500 | burden is at a minimum in October and November and then increases by 30% through winter and
1501 | spring until it reaches a maximum in June (~~Cooper et al., 2014~~)(Cooper et al., 2014). Much of this
1502 | seasonal variability can be explained by the flux of ozone from the stratosphere to the troposphere
1503 | with a Northern Hemisphere peak flux in May and a minimum flux in November occurring
1504 | predominantly in mid-latitudes (Hsu and Prather, 2009). Until recently ozone transport from the
1505 | stratosphere to the troposphere was believed to be dominated by processes such as tropopause
1506 | folds and gravity wave breaking, with little attribution given to deep convection (~~Stohl et al.,~~

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1507 | [2003](#)([Stohl et al., 2003](#)). However, a new model-based estimate that accounts for deep convection
1508 | penetrating the lowermost stratosphere increases the Northern Hemisphere peak stratosphere-to-
1509 | troposphere ozone flux by 19% and shifts the peak month from May to June ([Tang et al., 2011](#))([Tang
1510 | et al., 2011](#)). Sudden STE events are well known to influence ground-based ozone measurements
1511 | over a short period (see e.g. ([Dempsey, 2014;Zanis et al., 2003](#))([Dempsey, 2014;Zanis et al., 2003](#))).

1512 |
1513 | While the tropospheric ozone burden responds seasonally to the flux from the stratosphere the
1514 | response is not uniform with altitude or latitude. Terao et al. ([Terao et al., 2008](#))([Terao et al., 2008](#))
1515 | using a chemical transport model calculated the stratospheric contribution to monthly mean ozone
1516 | above four ozonesonde sites between 75° N and 36° N. At 500 hPa the stratospheric contribution to
1517 | ozone is ~40% during spring and 25% in autumn at high latitudes, and 35-40% during spring and 10-
1518 | 15% during autumn at mid latitudes. At 800 hPa these values are slightly less on a percent scale but
1519 | much less on an absolute scale, being ~20 ppbv during spring and 5-7 ppbv during autumn with
1520 | relatively little latitudinal gradient. Model estimates of the stratospheric contribution to surface
1521 | ozone are smaller than those for 800 hPa but the estimates vary greatly depending on the model
1522 | used and the region under consideration. For example, Lamarque et al. ([Lamarque et al.,
1523 | 2005](#))([Lamarque et al., 2005](#)) estimated that the monthly mean stratospheric contribution to a range
1524 | of sites in the mid-latitudes and tropics of both hemispheres is always less than 5 ppbv, regardless of
1525 | season. In contrast, a study using a different model estimated the stratospheric contribution to US
1526 | surface ozone during late spring 2010 as being < 5 ppbv along the Gulf coast, 8-13 ppbv across the
1527 | eastern US, and 10-20 ppbv across the high elevations of the western USA ([Lin et al., 2012a](#))([Lin et
1528 | al., 2012a](#)). More recently Hess et al have claimed that that a large portion of the measured ozone
1529 | change in the sub-tropical NH are not due to changes in emissions, but can be traced to changes in
1530 | large-scale modes of ozone variability ([Hess et al., 2014](#))([Hess et al., 2014](#)).this

1531 | [c\) Seasonal transport patterns e.g. summer monsoons](#)

1532 |
1533 | In addition to the seasonal influence of stratosphere-troposphere exchange, ozone varies seasonally
1534 | at most sites around the world ([Oltmans et al., 2013](#))([Oltmans et al., 2013](#)) simply due to shifting
1535 | wind patterns associated with migratory mid-latitude cyclone tracks or the transition between winter
1536 | and summer monsoons. For example, Mauna Loa Observatory, Hawaii in the northern tropics is
1537 | influenced by westerly air masses from Asia in the spring when it is located beneath the subtropical
1538 | jet stream. But in autumn the subtropical jet stream is far to the north leaving the site well within
1539 | the tropical belt which reduces transport from Asia resulting in 25% decrease in monthly median
1540 | ozone ([Lin et al., 2014b](#))([Lin et al., 2014b](#)). Similarly, Bermuda in the subtropical North Atlantic
1541 | Ocean receives westerly transport from North America during winter and spring, but during summer
1542 | the westward and northward expansion of the Azores-Bermuda High advects tropical air masses to
1543 | Bermuda reducing ozone mixing ratios by 50% ([Moody et al., 1995](#))([Moody et al., 1995](#)). The North
1544 | American Summer Monsoon becomes established above central North America during July and
1545 | August producing a stationary upper tropospheric anticyclone that traps ozone precursors lofted
1546 | from the surface as well as large quantities of lightning NO_x ([Cooper et al., 2009](#))([Cooper et al., 2009](#)).
1547 | Over several days the ozone precursors enhance tropospheric ozone within the anticyclone by as
1548 | much as 30-40 ppbv compared to sites upwind of the anticyclone ([Cooper et al., 2007](#))([Cooper et al.,
1549 | 2007](#)). Ozonesondes launched from Ahmedabad, western India indicate that an upper tropospheric
1550 | ozone enhancement does not occur during the Asian Summer Monsoon despite the presence of an

1551 anticyclonic recirculation similar to that found above North America during its summer monsoon.
1552 Instead, an abrupt decrease in tropospheric column ozone is observed above Ahmedabad with the
1553 onset of the Asian Summer Monsoon, with decreases occurring at all levels of the troposphere (Ojha
1554 et al., 2014)(Ojha et al., 2014).
1555

1556 d) 2.1.3.3 Climate variability

1557 While mean climatic conditions are typically established over a 30-year period, climate variability
1558 occurs on much shorter time scales of one to several years. Short-term climate variability modifies
1559 the long-range transport pathways that channel ozone plumes on regional and intercontinental
1560 scales, and also impacts regional-scale ozone photochemistry by modifying cloud cover. The impact
1561 of climate variability on ozone transport and chemistry is a relatively new field of study that has
1562 explored the effects of such phenomena as El Niño/Southern Oscillation (ENSO), the Pacific–North
1563 American (PNA) pattern and the North Atlantic Oscillation (NAO). "The influence on ozone of low
1564 frequency climate variability on time scales longer than 30 years has not yet been assessed due to
1565 continuous ozone data sets being limited to durations shorter than 30-40 years."
1566

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1567 ENSO has been shown to modulate the tropospheric ozone burden across the tropical Pacific Ocean
1568 with lower tropospheric column ozone corresponding to the cloudiest regions (Ziemke and Chandra,
1569 2003; Sekiya and Sudo, 2014)(Ziemke and Chandra, 2003; Sekiya and Sudo, 2014). Zeng and Pyle
1570 (Zeng and Pyle, 2005)(Zeng and Pyle, 2005) calculated an anomalously large increase of stratosphere-
1571 troposphere exchange following a typical El Niño year, increasing the global tropospheric ozone
1572 burden. In addition, changes in emissions, especially biomass burning, associated with ENSO affect
1573 the ozone burden across the tropics (Doherty et al., 2006)(Doherty et al., 2006). ENSO has also been
1574 linked to column ozone changes above Europe with positive anomalies of tropospheric ozone column
1575 and surface ozone occurring in the spring following an El Niño year, for a variety of reasons including
1576 changes in STE, biomass burning emissions and pollution transport pathways from Asia and North
1577 America (Koumoutsaris et al., 2008)(Koumoutsaris et al., 2008). Voulgarakis et al. (Voulgarakis et al.,
1578 2010)(Voulgarakis et al., 2010) used a global chemical transport model to determine the interannual
1579 variability of the global tropospheric ozone burden for the period 1996-2000 which included the
1580 strong El Niño event of 1997-1998. They found that 79% of the interannual variability was due to
1581 changes in meteorology (mostly stratosphere-troposphere exchange) and only 11% was due to
1582 changes in emissions. Changes in cloudiness contributed a small but non-negligible amount (6%) to
1583 the interannual variability. ENSO and the Pacific –North American (PNA) pattern also control the
1584 transport of ozone from East Asia to Mauna Loa, Hawaii. A springtime decrease in transport from
1585 Asia to Mauna Loa over the period 1980-2011 has resulted in no significant ozone trend, while an
1586 increase in transport during autumn has produced a very strong ozone trend of 3.5 ± 1.4 ppb decade⁻¹
1587 (Lin et al., 2014b)(Lin et al., 2014b).
1588

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1589 The transport of ozone and other pollutants across the North Atlantic Ocean and the Arctic is heavily
1590 influenced by the NAO. During the high phase of the NAO, wintertime pollutant transport into the
1591 Arctic is enhanced by 70% compared to times dominated by the low phase, with the major source
1592 region being Europe, followed by North America (Eckhardt et al., 2003)(Eckhardt et al., 2003). The
1593 NAO has also been shown to modulate ozone at the high elevation site of Izaña in the subtropical
1594

1595 | North Atlantic Ocean (~~Cuevas et al., 2013~~)(Cuevas et al., 2013), with a relaxation of the
1596 | predominantly high positive phase of the NAO since the mid-1990s allowing increased ozone
1597 | transport from North America and the lower stratosphere to Izaña in more recent years. Across
1598 | Europe surface ozone observations tend to correlate positively with the NAO (~~Pausata et al.,~~
1599 | ~~2012~~)(Pausata et al., 2012).

1600 |
1601 | While transport of ozone from the stratosphere to the troposphere has a strong seasonal cycle,
1602 | recent work has also explored changes in the stratosphere-to-troposphere ozone flux and its impact
1603 | on the tropospheric ozone budget. Several modelling studies have given evidence for such a link
1604 | (~~Young et al., 2013; Kawase et al., 2011; Hess and Zbinden, 2013; Voulgarakis et al., 2011; Oman et al.,~~
1605 | ~~2013~~)(Young et al., 2013; Kawase et al., 2011; Hess and Zbinden, 2013; Voulgarakis et al., 2011; Oman
1606 | et al., 2013), even demonstrating that the Mount Pinatubo eruption of 1991 reduced the flux of
1607 | ozone from the stratosphere to the troposphere until 1995 (~~Tang et al., 2013~~)(Tang et al., 2013).
1608 | According to a model-based analysis, the interannual variability of the ozone mass flux from the
1609 | stratosphere to the troposphere is on the order of 15% for the Northern Hemisphere and 6% for the
1610 | Southern Hemisphere (~~Olsen et al., 2013~~)(Olsen et al., 2013). Correlations between remotely sensed
1611 | ozone in the lowermost stratosphere and mid-troposphere indicate that 16% of the northern
1612 | hemisphere mid-latitude tropospheric interannual ozone variability is controlled by the flux from the
1613 | stratosphere (~~Neu et al., 2014~~)(Neu et al., 2014). Ordoñez et al. (~~Ordonez et al., 2007~~)(Ordonez et
1614 | al., 2007) showed that the positive ozone trends and anomalies in the lower free troposphere over
1615 | Europe during the 1990s were probably due to enhanced stratospheric ozone contributions
1616 | (dominated by changes in lower stratospheric ozone concentrations rather than by variations of
1617 | cross-tropopause air mass transport), particularly in winter–spring. Similarly, Tarasick et al. (~~Tarasick~~
1618 | ~~et al., 2005~~)(Tarasick et al., 2005) using the ozonesonde network over Canada, and Terao et al. (~~Terao~~
1619 | ~~et al., 2008~~)(Terao et al., 2008) using a global model for the northern extratropics found positive
1620 | correlations between ozone in the troposphere and the lowermost stratosphere. Using the same
1621 | data as Ordoñez et al. (2007) for the lowermost stratosphere, Logan et al. (~~Logan et al., 2012~~)(Logan
1622 | et al., 2012) found no trends in ozone for the decades 1978-1989, 1990-1999, 2000-2009, except for
1623 | a marginally significant decrease in winter for 1978-1989, driven by very low values in early 1989 and
1624 | an increase in winter and spring for 1990-1999. They found no evidence in the time series for the
1625 | lowermost stratosphere from either ozonesondes over North America (~~Tarasick et al., 2005~~)(Tarasick
1626 | et al., 2005) or from satellite data (~~McPeters et al., 2007~~)(McPeters et al., 2007) to suggest that
1627 | changes in stratospheric input can explain the increase in ozone over Europe in the 1980s. However,
1628 | increases in stratospheric ozone after 1993 may have contributed to the increase in tropospheric
1629 | ozone in the 1990s.

1630 |

1631 | 2.2 Changing ozone – a brief history

1632 |

1633 | The German chemist C.F. Schönbein is credited with the discovery of ozone in 1839 (~~Schönbein,~~
1634 | ~~1843~~)(Schönbein, 1843) and its presence in the atmosphere was one of his earliest concerns
1635 | (~~Braslavsky and Rubin, 2011~~)(Braslavsky and Rubin, 2011). Early work by Hartley identified its UV
1636 | spectrum and its role as a UV filter in the atmosphere (~~Hartley, 1881~~)(Hartley, 1881).

1637 |

1638 Modelling studies coupled to limited surface measurements from the late 1800s and early 1900s
 1639 indicate that ozone concentrations have changed across all scales of the troposphere during the
 1640 modern era due to enhanced emissions of precursors from industrialisation. A recent assessment
 1641 (Cooper et al., 2014) of the most reliable historical ozone records (reliable due to the quantitative
 1642 measurement methods employed), indicates that surface tropospheric ozone levels in western
 1643 Europe increased by a factor of 3-5 between the late 1800s and late 1900s, and by a factor of 2
 1644 between the 1950s and 1990s (Wilson et al., 2012; Parrish et al., 2012; Marenco et al., 1994; Staehelin
 1645 et al., 1994; Volz and Kley, 1988)(Wilson et al., 2012; Parrish et al., 2012; Marenco et al.,
 1646 1994; Staehelin et al., 1994; Volz and Kley, 1988). Many locations around the world monitored ozone
 1647 in the late 1800s and early 1900s using the semi-quantitative Schönbein ozonoscope (Marenco et al.,
 1648 1994; Bojkov, 1986)(Marenco et al., 1994; Bojkov, 1986). These estimates indicate that surface ozone
 1649 was much lower in those days compared to modern times, but the uncertainty of the measurements
 1650 is so great that no accurate estimate can be made of the absolute increase in ozone (see the review
 1651 by (Cooper et al., 2014)(Cooper et al., 2014)).

1652
 1653 Recent modelling studies (Young et al., 2013; Lamarque et al., 2005)(Young et al., 2013; Lamarque et
 1654 al., 2005) suggest that the tropospheric ozone burden in 1850 was 30% lower than the present day
 1655 (see Figure 56), with the largest contribution to the change coming from the northern hemisphere
 1656 extratropics. Most current global models are still unable to reproduce the low surface ozone
 1657 concentrations reliably observed at Montsouris near Paris at the end of the 19th century (Young et
 1658 al., 2013; Hauglustaine and Brasseur, 2001)(Young et al., 2013; Hauglustaine and Brasseur, 2001),
 1659 despite the models having more detailed chemistry schemes, improved emissions estimates and
 1660 finer spatial resolution (e.g. (Lamarque et al., 2010)(Lamarque et al., 2010)). This suggests either
 1661 problems with interpretation of the original observations and their context, or weaknesses in our
 1662 assessment of precursor emissions or in our current understanding of atmospheric processes
 1663 (Mickley et al., 2001)(Mickley et al., 2001). However, the inclusion of bromine chemistry can help
 1664 models to approach the low ozone values measured at Montsouris in the late 1800s (Parrella et al.,
 1665 2012)(Parrella et al., 2012).

1666

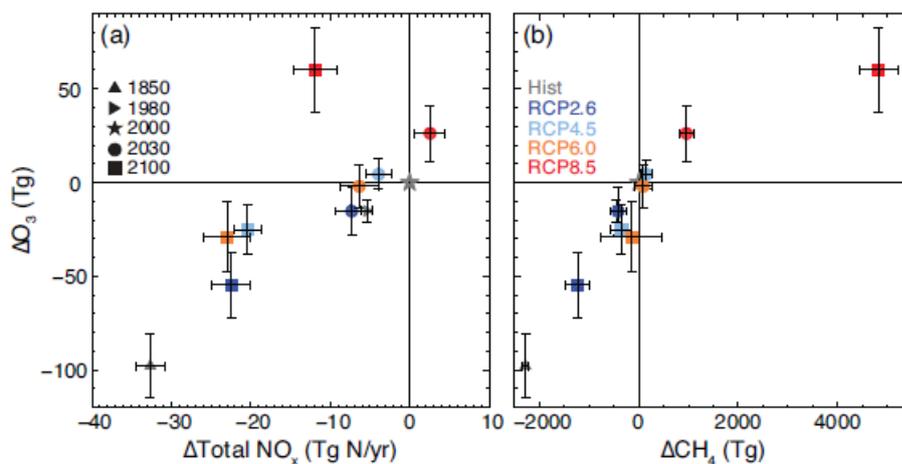


Figure 56 - Ensemble mean change in the tropospheric ozone burden compared to the year 2000 simulation as a function of (a) changes in total NO_x emissions and (b) changes in the tropospheric

methane burden. Error bars indicate ± 1 std. dev. of the changes in ozone, NO_x emissions and methane burdens, calculated from the spread of the models. Different colours represent the different scenarios, whereas different symbols represent the different time slices. (Young et al., 2013)(Young et al., 2013)

1667

1668 Establishing quantitative ozone trends from observations is important for testing our understanding
1669 of the processes affecting ozone in the troposphere and for the attribution of these ozone changes to
1670 changes in precursor emissions, removal processes and meteorology. It is also important from an air
1671 quality perspective for determining the effect of emission controls and for identifying the extent to
1672 which surface ozone is locally or regionally controllable. Observed ozone trends are a challenge to
1673 interpret as there are a number of factors that may be responsible (Cape, 2008)(Cape, 2008)
1674 including (a) changes in anthropogenic emissions of precursors (local, regional and global), (b) effects
1675 of biomass burning (both regional and global), (c) changes in Stratosphere-Troposphere exchange, (d)
1676 changes in geographical emission patterns and, (e) changes in meteorological land cover and (f)
1677 changes in meteorology e.g. transport patterns, rain, radiation, temperature etc. Long time series of
1678 high quality measurement data are required in order to detect the trend above the large inter-annual
1679 variation (Staelin, 2003; Weatherhead et al., 1998; Weatherhead et al., 2002)(Staelin,
1680 2003; Weatherhead et al., 1998; Weatherhead et al., 2002) and the sensitivity of the extracted trends
1681 to location necessitates that this is done at a range of sites to ensure representativeness and permit
1682 attribution of the observed changes. Jonson et al. (Jonson et al., 2006)(Jonson et al., 2006) have
1683 discussed some of these effects in relation to ozone trends.

1684

1685 Global tropospheric ozone trends were recently assessed by the Intergovernmental Panel on Climate
1686 Change (IPCC, 2013)(IPCC, 2013), and by Cooper et al. (2014) who provided an expanded discussion
1687 and update to the IPCC assessment. Following is a summary of the findings from Cooper et al.
1688 (2014). Prior to the 1970s, the only rural or remote quantitative ozone measurements outside of
1689 western Europe were some short-term observations from Antarctica (Wexler et al., 1960)(Wexler et
1690 al., 1960) and Mauna Loa, Hawaii (Junge, 1962)(Junge, 1962) during the late 1950s. By the early and
1691 mid-1970s quantitative and continuous ozone measurements were made at several rural and remote
1692 locations around the world for the purposes of detecting long-term changes in the global
1693 composition of the atmosphere. Continuous records in southern Germany began at the rural hilltop
1694 site of Hohenpeissenberg in 1971 and the mountaintop site of Zugspitze (2670 m) in 1978, and
1695 measurements began at the summit of Whiteface Mountain in upstate New York in 1973. Ozone
1696 measurements at remote sites were established by the U.S. National Oceanic and Atmospheric
1697 Administration at its baseline observatories of Mauna Loa, Hawaii (1973), Barrow, Alaska (1973), the
1698 South Pole (1975), and American Samoa (1976) (Oltmans et al., 2013)(Oltmans et al., 2013). Routine
1699 ozonesonde profiles became available in Germany, the US, Japan and Antarctica in the early 1970s
1700 (Oltmans et al., 2013)(Oltmans et al., 2013) and ship-borne monitoring of the marine boundary layer
1701 of the North and South Atlantic Oceans began in the late 1970s (Lelieveld et al., 2004)(Lelieveld et al.,
1702 2004). The number of urban rural and remote monitoring sites has continued to grow from the
1703 1970s to the present day.

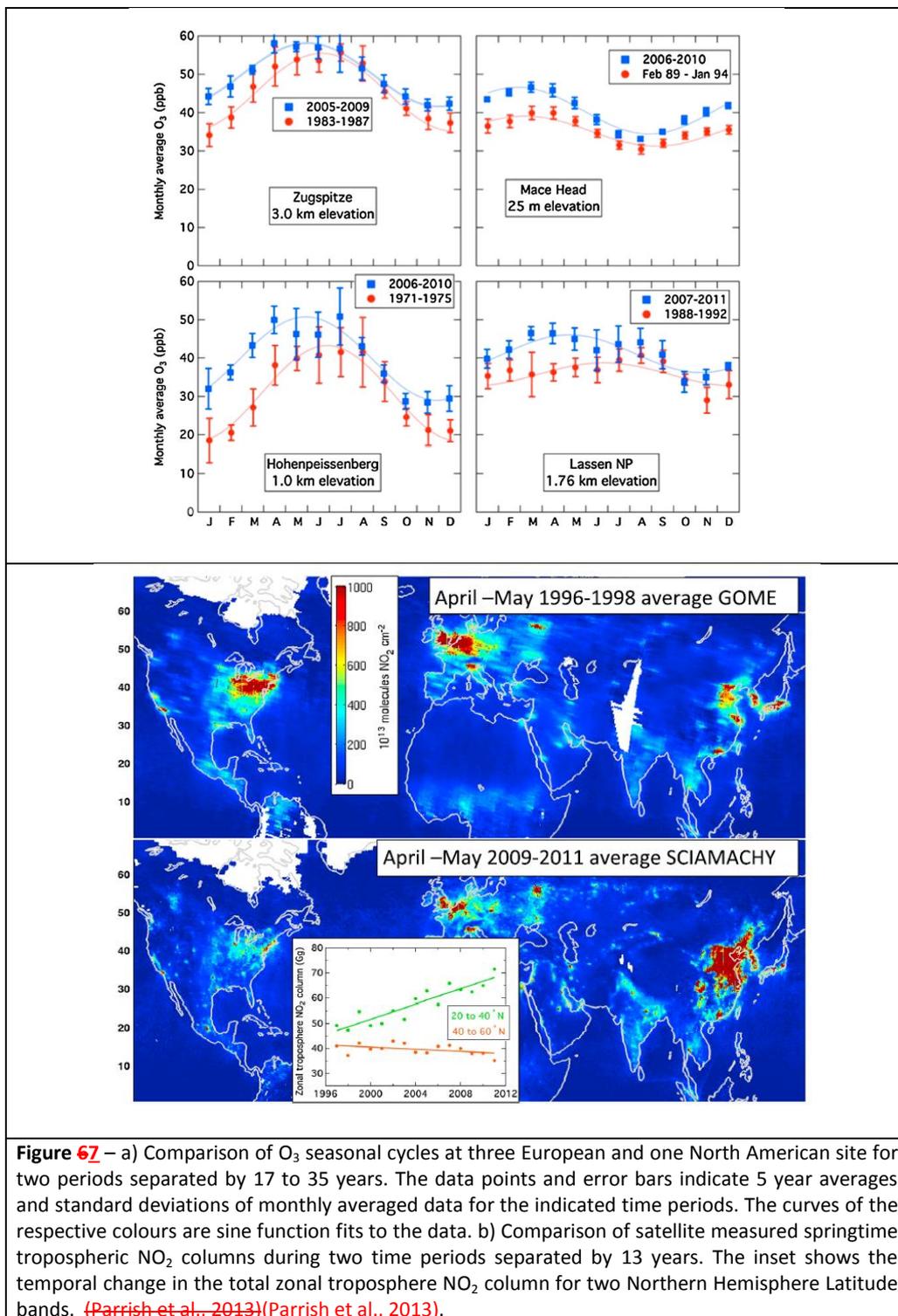
1704

1705 All available northern hemisphere surface monitoring sites indicate increasing ozone from 1950-1979
1706 until 2000-2010, with 11 of 13 sites having statistically significant trends of 1-5 ppbv decade⁻¹,
1707 corresponding to >100% ozone increases since the 1950s, and 9-55% ozone increases since the

1708 1970s. In the southern hemisphere only 6 sites are available since the 1970s, all indicating increasing
1709 ozone with 3 having statistically significant trends of 2 ppbv decade⁻¹. Ozone monitoring in the free
1710 troposphere since the 1970s is even more limited than at the surface. Significant positive trends
1711 since 1971 have been observed using ozonesondes above Western Europe, Japan and coastal
1712 Antarctica (rates of increase range from 1-3 ppbv decade⁻¹), but not at all levels. In addition, aircraft
1713 have measured significant upper tropospheric trends in one or more seasons above the north-
1714 eastern USA, the North Atlantic Ocean, Europe, the Middle East, northern India, southern China and
1715 Japan. Notably, no site or region has shown a significant negative ozone trend in the free
1716 troposphere since the 1970s. From 1990 until 2010, surface ozone trends have varied by region.
1717 Western Europe showed increasing ozone in the 1990s followed by a levelling off, or decrease since
1718 2000. In the eastern US rural surface ozone has decreased strongly in summer, is largely unchanged
1719 in spring, and has increased in winter. In contrast, few rural sites in the western US indicate
1720 decreasing ozone, with half of all rural sites showing a significant increase in spring. The decrease in
1721 ozone in Europe and the eastern USA is consistent with the decrease in ozone precursors in those
1722 regions. Meanwhile in East Asia surface ozone is generally increasing where ozone precursor
1723 emissions are growing.

1724
1725 Parrish et al. (~~Parrish et al., 2013~~)(Parrish et al., 2013) demonstrate that another
1726 manifestation of changes in tropospheric O₃ is a shift of the seasonal cycle at northern mid-latitudes
1727 so that the observed peak concentrations now appear earlier in the year than they did in previous
1728 decades (see Figure 67). The rate of this shift has been about 3 to 6 days per decade since the 1970s.
1729 Parrish et al. (2013) put forward an untested hypothesis that this shift may be due to changes in
1730 atmospheric transport patterns combined with spatial and temporal changes in emissions. This study
1731 relied on three long term (> 20 years) rural datasets in western Europe and one in the western USA.
1732 Cooper et al. (~~Cooper et al., 2014~~)(Cooper et al., 2014) also explored this topic using 21-year data
1733 sets from one additional site in western Europe and 5 additional sites in the USA, all in rural locations
1734 but more heavily influenced by regional ozone precursor emissions than those examined by Parrish
1735 et al. (2013). Of these 6 sites only three showed an earlier seasonal ozone peak, therefore the
1736 seasonal shift in the ozone cycle is not universal for time periods of 21 years or less. Similar shifts in
1737 the ozone seasonal cycle have been observed in the continental USA (Clifton et al., 2014;Simon et al.,
1738 2015;Bloomer et al., 2010).

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1741
1742 Episodic peak ozone levels in rural areas of Europe have been declining during the last three
1743 decades due to regional pollution emission controls applied to the VOC and NO_x emissions from fossil
1744 fuel combustion associated with motor vehicles and power plants (~~Jonson et al., 2006; Jenkin,~~
1745 ~~2008; Derwent et al., 2010~~)(~~Jonson et al., 2006; Jenkin, 2008; Derwent et al., 2010~~). Long-term
1746 downward trends have been observed at many long-running rural monitoring stations in the EMEP
1747 ozone monitoring network, and appear to be more pronounced at those stations where initial
1748 episodic peak levels were highest and insignificant at those stations where initial levels were lowest
1749 (~~Derwent and Hjellbrekke, 2013~~)(~~Derwent and Hjellbrekke, 2013~~). ~~The decreasing trend of highest~~
1750 ~~percentiles is compatible with the known emissions reductions. This behaviour has been interpreted~~
1751 ~~as resulting from the combined effect of regional pollution controls~~ since 1990 (~~Vautard et al.,~~
1752 ~~2006~~)(~~Vautard et al., 2006~~), ~~and this behaviour has also been interpreted as resulting from the~~
1753 ~~combined effect of regional pollution controls~~ and increasing hemispheric ozone levels (~~Derwent and~~
1754 ~~Hjellbrekke, 2013~~)(~~Derwent and Hjellbrekke, 2013~~). In contrast, episodic ozone levels in the high NO_x
1755 environments of major European towns and cities are now rising towards the levels found in the
1756 rural areas surrounding them, as exhaust gas catalysts fitted to petrol and diesel motor vehicles
1757 reduce the scavenging of ozone by chemical reaction with emitted nitric oxide. (~~Wilson et al.,~~
1758 ~~2012; Colette et al., 2011~~).

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1759
1760 Whereas the vast majority of the scientific literature on ozone trends relies on rural
1761 supersites dedicated to the assessment of long term atmospheric change, the records of regulatory
1762 monitoring networks are becoming progressively long enough to complete such assessments. In the
1763 1990s European air quality legislation began to include some targets with regards to the ozone
1764 monitoring network, so that the network now includes several thousand sites, a few hundred of
1765 which have records longer than 10 years. The size and diversity of the data gathered in the Airbase
1766 repository (maintained by EEA) offers an unprecedented view into air quality trends in Europe,
1767 provided that (1) 10-yr records can be considered long-enough to assess trends, and (2) the station
1768 representativeness of thousands of urban and rural background sites can be compared to the
1769 handful of supersites used elsewhere. Colette et al (~~Colette et al., 2011~~)(~~Colette et al., 2011~~) and
1770 Wilson et al. (2012) (Wilson et al., 2012) propose such assessments and conclude that the
1771 dominating picture is towards an increase of ozone in urban areas or non-significance of the rural
1772 background trends. They both report that the fraction of sites with an increasing trend is smaller
1773 when looking at peaks compared to daily means, but their findings suggest that the earlier reports of
1774 opposite signs of trends (~~Vautard et al., 2006~~)(~~Vautard et al., 2006~~) does not hold for this more
1775 recent period using this alternate monitoring dataset. By including urban stations in their analysis,
1776 Colette et al. (2011) also find an enhanced contribution of upwards trends, hence consolidating the
1777 results of Derwent and Hjellbrekke (~~Derwent and Hjellbrekke, 2013~~)(~~Derwent and Hjellbrekke, 2013~~)
1778 regarding the fact that urban areas are gradually becoming closer to rural levels.

1779
1780 The anthropogenic contributions to the observed trends are difficult to extract from the
1781 measurements, and reflect changes in both the magnitude and distribution of precursor emissions.
1782 While global average ozone responds to changes in global precursor emissions, trends at a given
1783 location are influenced by local, regional and global emission changes that may offset each other.
1784 Multi-model ensemble studies of the contribution of regional anthropogenic emission changes to

1785 regional and global surface ozone conducted under the LRTAP task force on Hemispheric Transport
1786 of Air Pollution, HTAP, (Fiore et al., 2009;HTAP, 2010)(Fiore et al., 2009;HTAP, 2010) have been used
1787 to assess the relative contributions of long range transport and regional emission changes to regional
1788 ozone (Wild et al., 2012)(Wild et al., 2012). These show annual mean surface ozone increases of
1789 nearly 0.3 ppb/yr at northern mid-latitudes between 1960-1980, similar to observed changes (e.g.,
1790 (Parrish et al., 2012)(Parrish et al., 2012)), but small net decreases in surface ozone over Europe and
1791 North America since 1990 when anthropogenic emissions over these regions are believed to have
1792 peaked. A recent comparison between ozone observations at northern mid-latitudes and output
1793 from three current chemistry-climate models shows that the models (1) overestimate ozone mixing
1794 ratios, on average by ~5 to 17 ppbv in the year 2000, (2) capture only ~50% of ozone changes
1795 observed over the past five to six decades, and little of observed seasonal differences, and (3)
1796 capture ~25 to 45% of the rate of change of the long-term changes (Parrish et al., 2014)(Parrish et al.,
1797 2014). These differences may reflect poor representation of emission changes or ozone changes due
1798 to natural sources, or they may reflect more fundamental weaknesses in resolving key chemical-~~or~~
1799 dynamical processes or climate variability over continental regions. Further detailed analysis of the
1800 spatial and seasonal variability of local and regional ozone trends should provide a more critical test
1801 of our current understanding of the processes affecting ozone as represented in current models.

1802
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1804 2.3 Ozone precursors

1805

1806 The spatial and temporal distribution of ozone and its precursors are in large part driven by the
1807 distribution of their emissions. An accurate knowledge of the surface emissions and of their
1808 evolution with time is therefore essential to support the analysis and modelling of air quality and
1809 climate change interactions. Up-to-date and consistent emissions are moreover required for the
1810 forecasting of the atmospheric composition (Frost et al., 2012)(Frost et al., 2012).

1811 Gridded global, regional, and national emission estimates exist for many of the pollutants that are
1812 important for understanding and analysing the distribution of ozone and its precursors, i.e. NO_x, CO,
1813 CH₄ and volatile organic compounds (VOCs). Some of these inventories are publicly available,
1814 whereas others are developed by individual research groups or government agencies to study
1815 specific aspects of emissions or atmospheric processes and are not always easy to access. In this
1816 paper, we will focus only on publicly available datasets.

1817 Some inventories provide global coverage with relatively coarse spatial resolution, while others focus
1818 only on a specific region for selected species, and provide information at a very high spatial
1819 resolution; other inventories consider only specific sectors (such as traffic, shipping, etc.)

1820 Over the past few years, several inventories providing the distribution of surface emissions of
1821 gaseous and particulate species were developed, at both the global and regional scales. This section
1822 will review the main advances during the past few years concerning surface emissions having the
1823 potential to impact ozone distributions and trends. The section will also discuss the current
1824 information on emissions uncertainties and we will briefly review the most recent studies using

1825 inverse modelling techniques to evaluate and optimize emission inventories.

1826 2.3.1 Quantification of global emissions of ozone precursors during the past decades

1827 Most anthropogenic emissions estimates are developed using the same methodology, based on the
1828 product of estimates for activity data for different sectors and emission factors (mass emitted per
1829 activity unit). Activity data are obtained from records provided by different countries or from
1830 international organizations such as the International Energy Agency or the United Nations. This leads
1831 to a country-based quantification of emissions, which are then gridded using different types of
1832 proxies such as population, traffic, ship routes or location of power plants/factories. The quality of
1833 the gridding depends on the quality and availability of the proxies, and therefore varies strongly
1834 among the different regions of the world.

1835 The data used for the quantification of emissions are not always consistent and many inventory
1836 developers adjust these data based on expert judgment. *A priori* emission factors are not always
1837 available or reported for all countries and for the differing emission sectors. They depend on
1838 different factors such as the level of economic development and the type of technology used in each
1839 region.

1840 The emissions inventories developed in the past years cover both global and regional scales. Some of
1841 these inventories provide emissions at the country level; some provide emissions for a few species or
1842 for a larger number of species, and some provide emissions for one year, a few years or a few
1843 decades. The main inventories providing emissions of gaseous and particulate compounds currently
1844 used in global modelling or analysis of the atmospheric composition at the global scale and its
1845 evolution are: ACCMIP (1850-2000 on a decadal time scale; ~~(Lamarque et al., 2010)~~(Lamarque et al.,
1846 2010)) and its extension MACCity (monthly averages for the 1980-2010 period; ~~(Granier et al.,~~
1847 ~~2011)~~(Granier et al., 2011)); EDGARv4.2 (1970-2008; ~~(Janssens-Maenhout et al., 2011)~~(Janssens-
1848 ~~Maenhout et al., 2011)~~) and HTAPv2 (2008 and 2010; ~~(Janssens-Maenhout et al., 2012)~~(Janssens-
1849 ~~Maenhout et al., 2012)~~); ECLIPSE (2005-2050; ~~(Klimont et al., 2013c)~~(Klimont et al., 2013b)); RETRO
1850 (1960-2000; ~~(Schultz et al., 2007a)~~(Schultz et al., 2007a)); HYDE (1890-1990; ~~(van Aardenne et al.,~~
1851 ~~2001)~~(van Aardenne et al., 2001)); POET (1990 and 2000; ~~(Granier et al., 2005)~~(Granier et al., 2005));
1852 EDGAR3.2 (year 2000; ~~(Olivier et al., 2005)~~(Olivier et al., 2005)).

1853 Several inventories also provide emissions at the regional scale for different regions of the world.
1854 Among these inventories, the TNO-MACC ~~(Kuenen et al., 2011)~~(Kuenen et al., 2011) and TNO-MACCII
1855 ~~(Kuenen et al., 2014)~~(Kuenen et al., 2014) provide emissions for Europe at a high spatial resolution
1856 (1/8th x 1/6th degree), based on the EMEP ~~(Droge et al., 2010)~~(Droge et al., 2010) emissions reported
1857 by the European countries. In North America, the US Environmental Protection Agency (EPA)
1858 provides emissions since 1980 for the USA (available at <http://www.epa.gov/ttnchie1/>), and
1859 Environment Canada provides non-gridded emissions for Canada since 1985 (available at:
1860 <http://www.ec.gc.ca>). Inventories are provided for Asia by the REAS-v1 (0.5x0.5 degree spatial
1861 resolution; ~~(Ohara et al., 2007)~~(Ohara et al., 2007)) and REAS v2 (0.25x0.25 degree spatial resolution;
1862 ~~(Kurokawa et al., 2013a)~~). The MEIC dataset (available at meicmodel.org) also provides emissions for
1863 China, at a 0.25x0.25 degree resolution, and Sahu et al. ~~(Sahu et al., 2012)~~(Sahu et al.,
1864 2012) provide emissions for China, at a 0.25x0.25 degree resolution, and Sahu et al. ~~(Sahu et al., 2012)~~(Sahu et al.,
1865 2012) provide emissions for India, as well as Garg et al. ~~(Garg et al.,~~
1866 ~~2012)~~(Garg et al., 2012).

1867 2006)(Garg et al., 2006) not gridded emissions). Recently, an inventory was also developed for Africa
 1868 by Liousse et al. (Liousse et al., 2014)(Liousse et al., 2014) at a 0.25x0.25 degree spatial resolution.

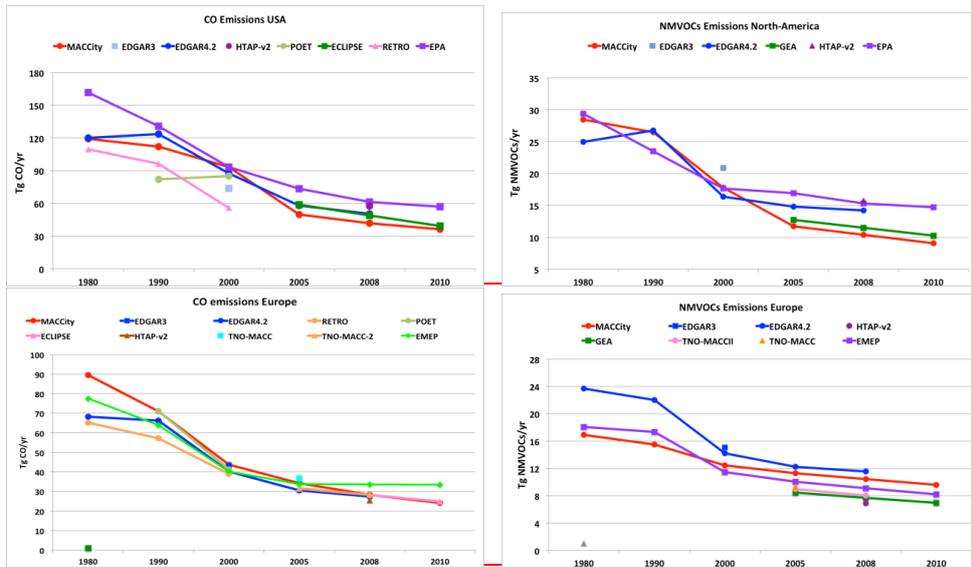
1869 2.3.3 Evolution of emissions in different regions since the 1980s

1870 During the past three decades and the past few years, surface anthropogenic emissions have shown
 1871 large changes in a few regions of the world. This section will focus on changes in emissions in Europe,
 1872 North America and Asia. There is still little information on emissions changes in other areas of the
 1873 world, and no detailed evaluation of the trends in emissions in these other regions can be made.

1874 a. Emissions in Europe and North America

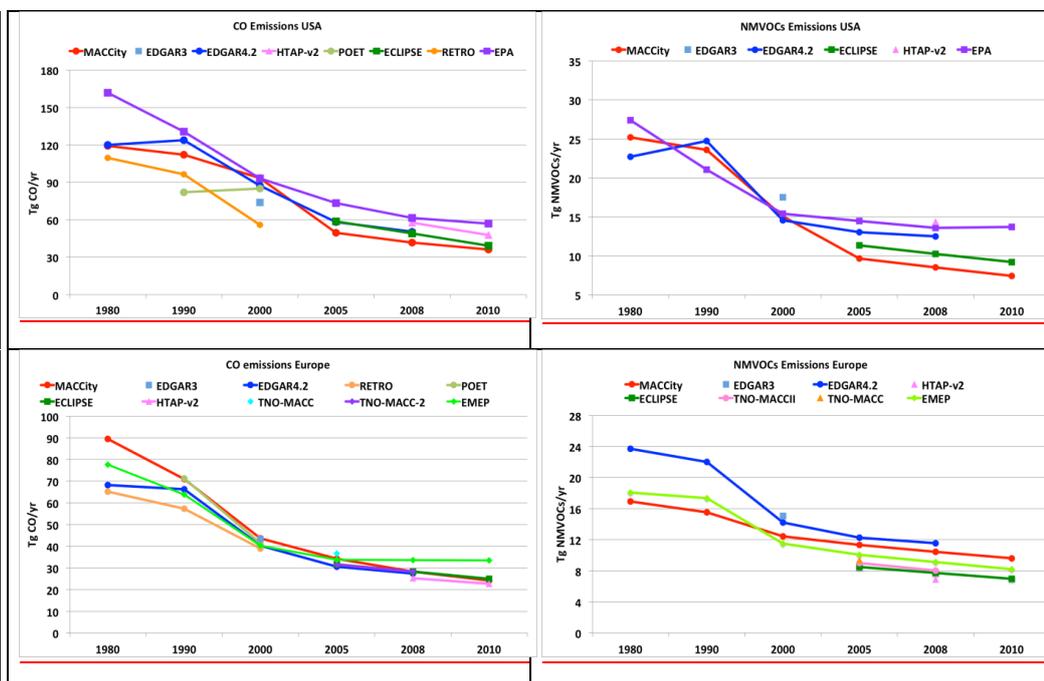
1875 The implementation of emissions policies reductions in Europe and in North America over the past
 1876 few decades has led to significant decreases in the emissions of ozone precursors. The agreement
 1877 between available global and regional inventories is rather good among ~~the global and regional~~ these
 1878 inventories providing emissions of CO and NOx for Europe and of NOx for North America, with
 1879 differences of 20-30% between the lowest and ~~largest~~ highest proposed values. However, larger
 1880 differences are obtained between inventories providing CO in the USA and NMVOCs in all regions, as
 1881 shown in Figure 78.

1882
 1883



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1885



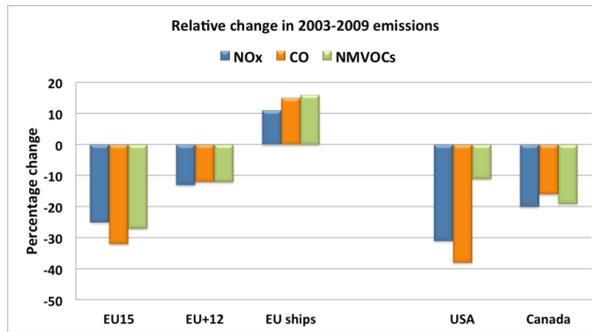
1886 **Figure 78** Emissions of CO and NMVOCs in the USA and of NMVOCs in Europe (Western and Central
 1887 Europe) from various emission inventories.

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1888 A detailed analysis of the changes in emissions of ozone precursors in Europe between 2003 and
 1889 2009 has been performed by Kuenen et al (Kuenen et al., 2014)(Kuenen et al., 2014). This analysis
 1890 has been extended in Figure 89 by adding emissions for USA and in Canada for the same period.
 1891 These data highlight the significant decrease in the emissions of NO_x, CO and NMVOC over the
 1892 European and North American continental regions. The changes in Figure 89 include shipping
 1893 activities around Europe and the European seas. In contrast to industrial activities and to
 1894 transportation in continental areas of Europe and North America, policies leading to the reduction of
 1895 emissions from shipping have not yet been defined or implemented, resulting in significant increases
 1896 in these emissions.

1897 It is worth noting that nearly 70% of ship emissions occur within 400 km of coastlines, causing air
 1898 quality problems through the formation of ground level ozone, sulphur emissions and particulate
 1899 matter in coastal areas and harbours with heavy traffic (Eyring et al., 2010). Offshore shipping
 1900 sources of NO_x may become significant as land based emissions decline (Eyring et al., 2010; Jonson et
 1901 al., 2009; Dalsøren et al., 2010). The impact of ship emissions on tropospheric oxidants is mainly
 1902 caused by the relatively large fraction of NO_x in ship exhaust. Dalsøren et al (Dalsøren et al., 2010)
 1903 have shown that typical increases in yearly average surface ozone concentrations in the most
 1904 impacted areas from shipping emissions are 0.5–2.5 ppbV. Transport emissions are predicted to be a
 1905 significant contributor to US and European ozone by 2050 (Hauglustaine and Koffi, 2012).

1906



1907

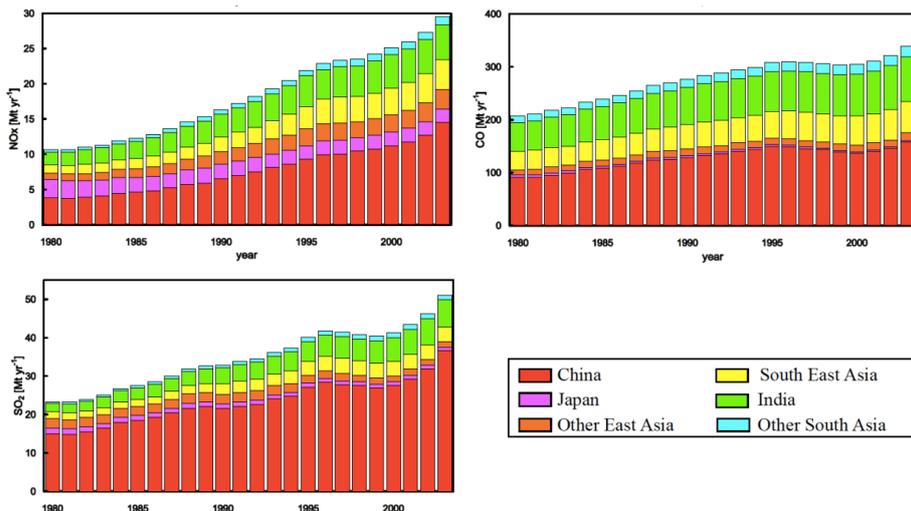
1908 | **Figure 89** - Relative change in NO_x, CO and NMVOCs emissions from 2003 to 2009 (from Kuenen et al., 2014), per country group: EU15 includes the EU Member States in 1995 as well as Norway and Switzerland, EU+12 includes the 12 New Member States, EU ships refers to all European sea regions combined. USA emissions are from the USA Environmental Protection Agency (EPA) and data from for Canada are from Environment Canada.

1913

1914 b. Emissions in Asia

1915 Emissions in Asia have shown dramatic increases over the past few years, which are now well documented, more particularly for China. Figure 910 shows the changes in the emissions of NO_x, CO and SO₂ for different Asian regions since 1980: the data in Figure 910 highlight the very large increase in NO_x emissions in China over the past few years. The main reason for these emissions changes are increases in coal use for energy generation and industrial activities, as well as a large increase in the number of vehicles (Kurokawa et al., 2013b). Zhao et al. (Kurokawa et al., 2013a). Zhao et al. (Zhao et al., 2013)(Zhao et al., 2013) have examined the impact of anthropogenic emissions control in China and have shown that these control measures have led to a decrease in the emissions factors and emissions for most compounds, except for nitrogen oxides.

1924



1925

1926 | **Figure 910** - Evolution of surface emissions of CO, NO_x and SO₂ for each Asian region (from Kurokawa et al., 2013).

1927

1928

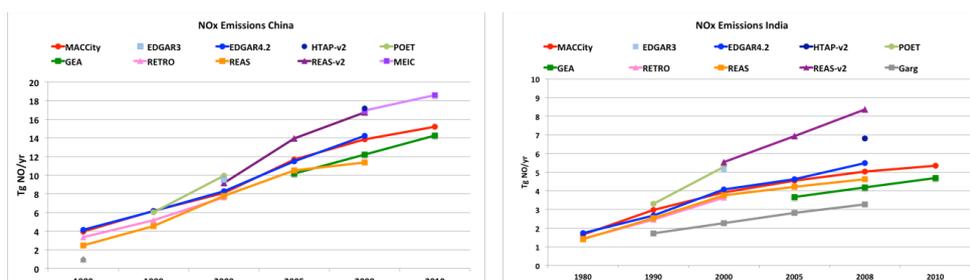
1929

1930 Comparisons between the regional and global inventories show significant differences, as shown in
1931 Figure 1011 (Granier et al., in preparation, 2014): these figures show the total anthropogenic
1932 emissions of NO_x in China and India from 1980 to 2010, based on the data provided by the
1933 inventories mentioned in Section 2.3.1. All the inventories show an increase in the emissions over the
1934 years of the NO_x emissions, but the magnitude of the emissions and the rate of increase differ
1935 significantly between the datasets. There is a difference of about 50% between the lowest and
1936 highest total emissions in China. For India, the range of values proposed by the different groups are
1937 even larger, with a factor of about 2.5 between the lowest and highest emissions. Since NO_x
1938 concentration levels are a key factor in the production of ozone (see Section 2.1), such large
1939 differences in the emissions could make the analysis of ozone concentrations and their trends rather
1940 difficult to achieve.

1941 It should be noted that Figure 1011 does not display any evaluation of the
1942 uncertainties on the inventories: since the data used for developing the inventories (activity data,
1943 emission factors) are not provided with estimates of the uncertainties, no estimation of the
1944 uncertainties on the emissions can be made.

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1946

1947 **Figure 1011** - Evolution of NO_x emissions in China and India from 1980 to 2010 provided by different
1948 global and regional inventories (Units are Tg NO/year)

1949 c) Shipping

1950 It is worth noting that nearly 70% of ship emissions occur within 400 km of coastlines, causing air
1951 quality problems through the formation of ground-level ozone, sulphur emissions and particulate
1952 matter in coastal areas and harbours with heavy traffic (Eyring et al., 2010). Offshore shipping
1953 sources of NO_x may become significant as land based-emissions decline (Eyring et al., 2010; Jonson et
1954 al., 2009; Dalsøren et al., 2010). The impact of ship emissions on tropospheric oxidants is mainly
1955 caused by the relatively large fraction of NO_x in ship exhaust. Dalsøren et al (Dalsøren et al., 2010)
1956 have shown that typical increases in yearly average surface ozone concentrations in the most
1957 impacted areas from shipping emissions are 0.5–2.5 ppbV. Transport emissions are predicted to be a
1958 significant contributor to US and European ozone by 2050 (Hauglustaine and Koffi, 2012).

1959

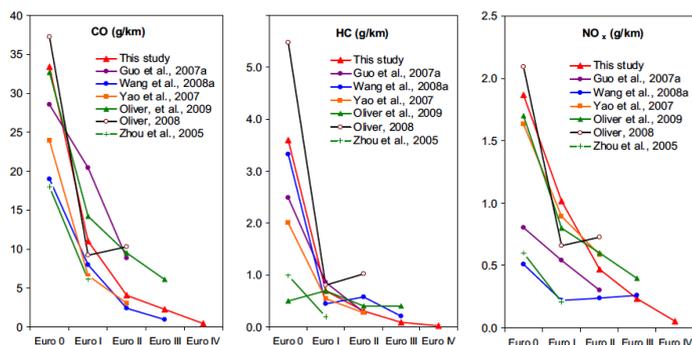
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1960 2.3.4 Uncertainties in anthropogenic emissions in different regions

1961 As indicated in the previous section, most emission datasets are provided without any information
1962 on uncertainties on the data used for quantifying the emissions. Several sources of uncertainties

1963 have been identified, which will be summarized in this section.

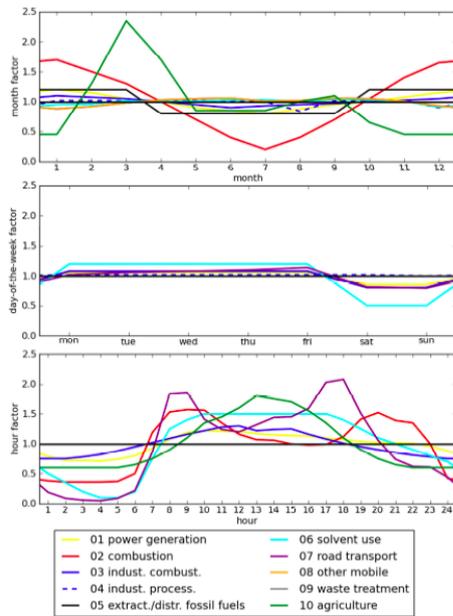
1964 Activity data and emission factors are generally provided without any information on their
1965 uncertainties. Emission factors are empirical functional relations between pollutant emissions and
1966 the activity that causes them: a comparison of emission factors is shown on Figure 1412, which
1967 displays measurements of emission factors in Chinese cities from different studies (Huo et al.,
1968 2012)(Huo et al., 2012). This study has shown that the differences between emission factors are
1969 larger for older vehicles than for newer vehicles, which could be an indication of varying
1970 deterioration of emissions whereas vehicles get older.



1971 **Figure 1412** - Comparison of emission factors of the LDGVs (light-duty goods vehicles) under real
1972 world driving conditions from different studies showing the greater variation for older vehicles. Note
1973 that the emission measurement technologies are different among these studies (from (Huo et al.,
1974 2012)(Huo et al., 2012)).
1975

1976 The seasonal, weekly and daily variations of the emissions are generally not provided with
1977 inventories, and are not well characterized. Simple temporal profiles have been developed to
1978 describe the changes in emissions over a day, a week and a season, as shown in Figure 1213 for the
1979 European LOTOS-EUROS model (Kuenen et al., 2011)(Kuenen et al., 2011). These profiles depend on
1980 the emission sectors. The same diurnal profiles are applied for all days of the week and to every
1981 country in the model domain for all periods, without taking into account differences in the way of
1982 living in different countries and its change with time. Similar profiles are not available for most the
1983 countries of the world, leading to large uncertainties in the temporal profiles of the emissions.

1984

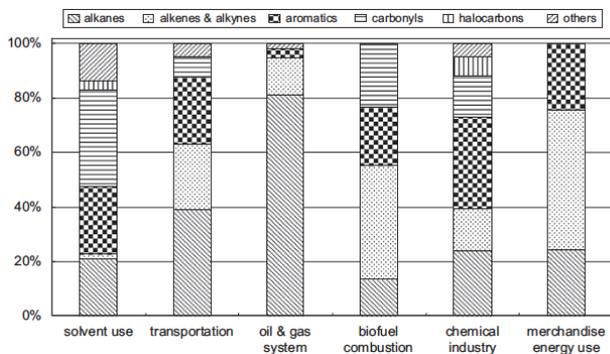


1985

1986 | **Figure 1213** - Monthly (top), weekly (middle) and hourly (bottom) time profiles applied to
 1987 anthropogenic emissions in Europe for different sectors (from Kuenen et al., 2011).

1988

1989 Good speciation of the NMVOCs lumped species is important for inventories. The translation of
 1990 speciation into global and sometimes regional models chemical schemes is generally poor: a gridded
 1991 speciated dataset has been developed as part of the RETRO inventory (Schultz et al., 2007a)(Schultz
 1992 et al., 2007a), and has not been revised or updated since. This speciation is currently applied to most
 1993 global inventories, for all the past and future decades. In different regions, VOC speciated inventories
 1994 have been developed for specific emission sectors often based on a few individual measurements
 1995 that are extrapolated with dubious reliability to related unmeasured emission sources. An example of
 1996 such a speciation is shown in Figure 1314 for China (Wei et al., 2008)(Wei et al., 2008). Li et al. (Li et
 1997 al., 2014)(Li et al., 2014a) have developed a speciation for Asia, which provides emissions of a large
 1998 set of VOCs for different model chemical schemes, for individual VOCs and lumped species.



1999

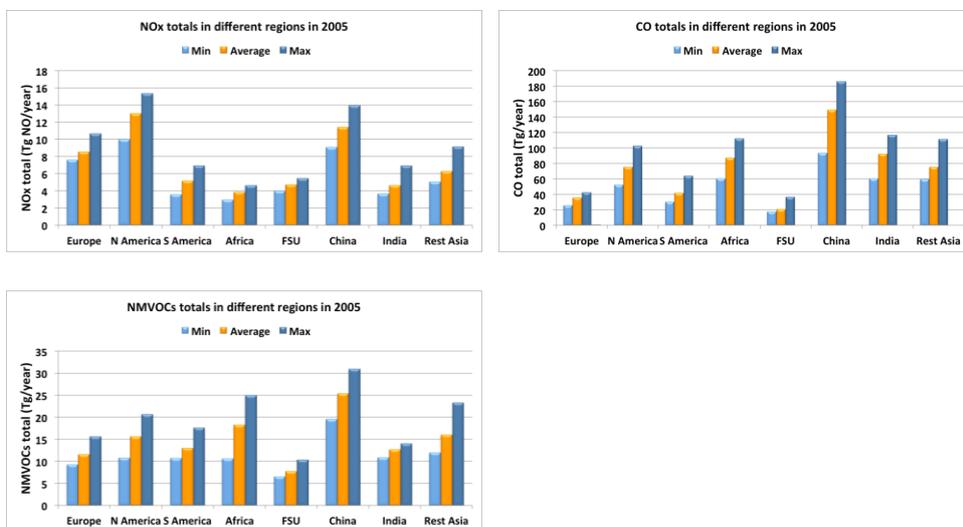
2000 | **Figure 1414** - Chemical speciation for various emission sectors in China (Wei et al., 2008)(Wei et al.,
 2001 | 2008)- showing the wide variation of VOCs with source that need to be represented in emission
 2002 | inventories.

2003 | Detailed information on the proxies used for the determination of the distribution in space of the
 2004 | emissions, i.e. on the proxies used for gridding emissions are also lacking. Other large uncertainties
 2005 | on the emissions are linked with resources (oil and gas) extraction and flaring, and to inland and
 2006 | international shipping. Emissions from agricultural practices (cattle, fertilizers, agriculture waste
 2007 | burning) are also very uncertain.

2008 | In order to summarize the uncertainties on anthropogenic emissions, we have calculated, for all the
 2009 | inventories mentioned in Section 2.3.1 the minimum, average and maximum values of the emissions
 2010 | for different world regions for NOx, CO and NMVOCs emissions in 2005. The ranges shown in Figure
 2011 | 1415 cannot be considered as a quantification of uncertainties, but they provide information on the
 2012 | differences between currently available datasets. Detailed evaluations of the uncertainties on
 2013 | anthropogenic emissions have started: for example, a methodology applying Monte Carlo
 2014 | simulations to quantify the uncertainties, represented as probability distributions, for the emissions
 2015 | of several compounds in China was proposed by Zhao et al. (Zhao et al., 2011)(Zhao et al., 2011).

2016 | Figure 1415 also highlights the growing of emissions in Africa and the rest of Asia (all Asian regions
 2017 | except China and India), regions where more detailed information is required for a better analysis
 2018 | and modelling of the global atmospheric composition.

2019



2020
 2021

2023 | **Figure 1415** - Minimum, average and maximum emissions taken from various inventories of NOx, CO
 2024 | and NMVOCs for different regions of the world in 2005. The emissions of NOx are reported in Tg
 2025 | NO/year.

2026 | [2.3.5 Emissions from fires](#)

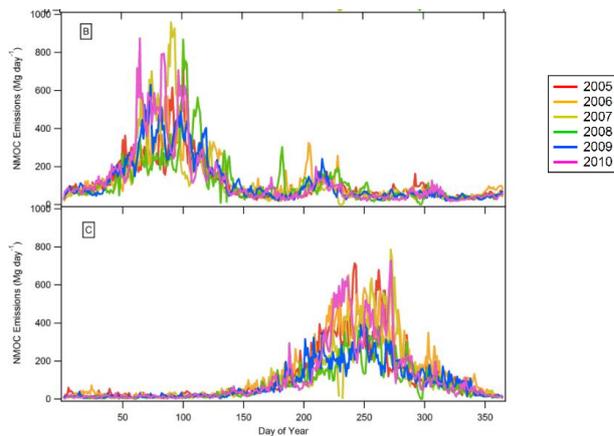
2027 | ~~In the last few decades biomass burning has been recognized as an important source of ozone~~

2028 ~~precursors (Hao and Liu, 1994;Schultz et al., 2008). Since wildfires are strongly dependent on~~
2029 ~~meteorological conditions, drought episodes and human behaviour, they have a large interannual~~
2030 ~~variability. In the last few decades biomass burning has been recognized as an important source of~~
2031 ~~ozone precursors (Hao and Liu, 1994;Schultz et al., 2008;Turquety et al., 2007). Since wildfires are~~
2032 ~~strongly dependent on meteorological conditions, drought episodes and human behaviour, they have~~
2033 ~~a large interannual variability (Duncan et al., 2003)(Duncan et al., 2003);~~ and the resulting emissions
2034 are very variable in time and space. It is therefore important to accurately characterize fire
2035 emissions, since they will partly drive the short-term variability of ozone precursors and ozone
2036 ~~(Parrington et al., 2012)(Parrington et al., 2012)~~. A review of the impact of biomass burning emissions
2037 on ozone was published by Jaffe and Wigder ~~(Jaffe and Wigder, 2012)(Jaffe and Wigder, 2012)~~.

2038 The first inventories relied on data reported by forest services, observations from the ground and
2039 from aircraft. Since the late 1990s, several inventories providing emissions from fires have been
2040 developed based on observation of active fires and burnt areas by satellites, and more recently of
2041 fire radiative energy. Other inventories have also been developed, through the representation of
2042 fires in biogeochemical models. The most recent global datasets providing a spatial distribution of
2043 the emissions of ozone precursors from fires are: MACCity (monthly, 1960-2008, 0.5x0.5 degree
2044 resolution; ~~(Granier et al., 2011)(Granier et al., 2011)~~), GFAS (daily, 2003 to present, 0.5x0.5 degree
2045 resolution; ~~(Kaiser et al., 2012)(Kaiser et al., 2012)~~), GFEDv2 (monthly, 1997-2010, 0.5x0.5 degree
2046 resolution)and GFEDv3 (monthly, 1997-2010, 0.5x0.5 degree resolution) ~~(van der Werf et al.,~~
2047 ~~2006;van der Werf et al., 2010)(van der Werf et al., 2006;van der Werf et al., 2010)~~, IS4FIRES (daily,
2048 2000-2013, 0.1x0.1 degree resolution, ~~(Sofiev et al., 2009)(Sofiev et al., 2009)~~), FINN (daily, 2002-
2049 current, 1x1 km resolution, ~~(Wiedinmyer et al., 2011)(Wiedinmyer et al., 2011)~~), GUESS-ES (monthly,
2050 1970-2009, 1x1 degree resolution, ~~(Knorr et al., 2012)(Knorr et al., 2012)~~), GICC (monthly, 1900-2005,
2051 0.5x0.5 degree resolution, ~~(Mieville et al., 2010)(Mieville et al., 2010)~~), Kloster (monthly, 1900-2004,
2052 1.9x2.5 degree resolution, ~~(Kloster et al., 2010)(Kloster et al., 2010)~~), RETRO (monthly, 1980-2000;
2053 0.5x0.5 degree resolution, ~~(Schultz et al., 2008)(Schultz et al., 2008)~~).

2054 The inventories providing daily emissions have highlighted the very large variability of emissions from
2055 fires in the different regions of the world. Only datasets providing emissions at a high temporal
2056 resolution are able to provide information on short and intense fire episodes, which happen often in
2057 different regions of the world, as a result of specific weather conditions or human-ignited fires. An
2058 example of this variability is shown on Figure ~~15~~16, from the FINN datasets ~~(Wiedinmyer et al.,~~
2059 ~~2011)(Wiedinmyer et al., 2011)~~, which also highlights the ~~constant~~large number of high fire episodes
2060 in each hemisphere.

2061

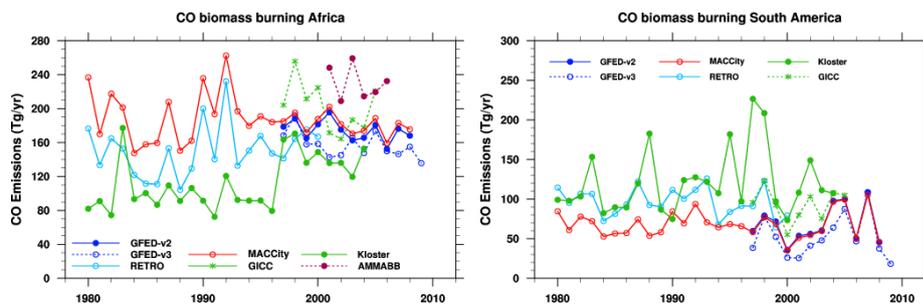


2062

2063 **Figure 1516** - Daily emissions of NMVOC from fires for 2005 through 2010 for the Northern
 2064 Hemisphere (top) and the Southern Hemisphere (bottom) as represented in the FINN dataset
 2065 ([Wiedinmyer et al., 2011](#))([Wiedinmyer et al., 2011](#)).

2066 Several studies have evaluated the differences between estimates of fire inventories, and have
 2067 highlighted the large differences remaining between available datasets in different regions. Figure
 2068 **1617** displays a comparison of several of the datasets previously detailed, for carbon monoxide in
 2069 Africa and South America (from ([Granier et al., 2011](#))([Granier et al., 2011](#))). Differences in the
 2070 emissions for each year, as well as in the interannual variability of the emissions are very large. It
 2071 should be noted that the AMMA inventory for Africa, which includes the use of satellite observations
 2072 as well as local information on the fires provides significantly larger emissions than the other
 2073 inventories, which are based either on satellite data only, or on fires modelling.

2074

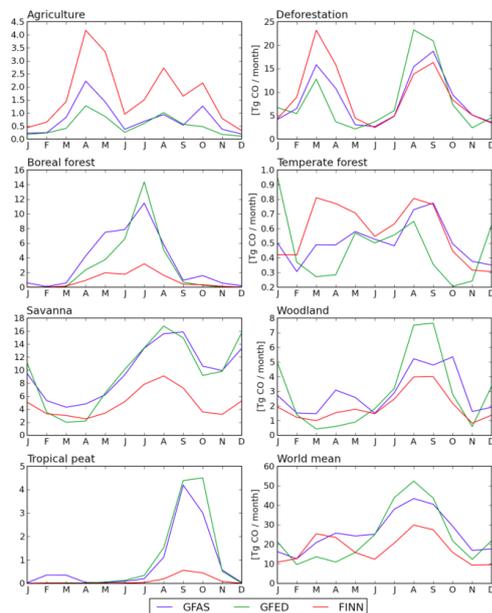


2075

2076 **Figure 1617** - Emissions of CO in Africa (left column) and South America (right column) from 1980 to
 2077 2009; from ([Granier et al., 2011](#))([Granier et al., 2011](#)).

2078 Another view of the differences between datasets is provided in Figure **1718**, where CO emissions
 2079 from different estimates are reported for different biomes (from ([Kaiser et al., 2013](#))([Kaiser et al.,](#)
 2080 [2013](#))). The largest differences are reports for peat and savannah fires. The seasonal variation shown
 2081 in these datasets is relatively consistent, except for temperate forests, where the consistency is very
 2082 low.

2083



2084

2085 **Figure 17-18** - Mean seasonal CO emissions for 7 biomes associated with dominant fire type in GFED
 2086 (see Figure 17-1b) and the world for 2003 until 2011. Emissions are shown for three different fire
 2087 emission inventories: GFAS, GFED and FINN (Kaiser et al., 2013)(Kaiser et al., 2013).

2088 The differences shown in Figure 17-18 demonstrate a limit to the accuracy of emissions resulting from
 2089 biomass burning. In turn, these impact on the distribution of ozone precursors and ozone through
 2090 uncertainties on burned area, fuel load, emission factors and injection heights.

2091 Data on burned areas from long-term monitoring have become available only recently, i.e. after
 2092 1995, and these data have been evaluated only for a few regions. The satellite data provide good
 2093 information on spatial and temporal patterns of fire occurrence, but so far they have not provided
 2094 quantitative information with high accuracy (e.g., (Giglio and Kendall, 2004)(Giglio and Kendall,
 2095 2004)). These data complement data on fires in the mid-latitude regions, which are well monitored in
 2096 many countries. However, monitoring by tropical countries is still scattered and scarce, owing to
 2097 limited resources at the local level for records and accessibility. The effect of temporal resolution of
 2098 fire emissions on models has been recently explored (Marlier et al., 2014)(Marlier et al., 2014).

2099 The amount of fuel load and the combustion factors are calculated either from ecosystem models or
 2100 through empirical formulas: these quantities depend on the fire severity, the fuel characteristics
 2101 (foliage and biomass density, moisture, vegetation type, organic content and moisture of the soil)
 2102 and the rate of spreading. These factors are highly variable and different studies have shown that
 2103 large uncertainties still remain on the spatial and temporal variation of this quantity. A study by
 2104 Knorr et al. (Knorr et al., 2012)(Knorr et al., 2012) has shown that the choice of the burned area
 2105 dataset has by far the largest impact on interannual variability of simulated emissions, and that for
 2106 the quantification of the total global emissions, burned area and combustion completeness have the
 2107 largest impact on emissions for most species.

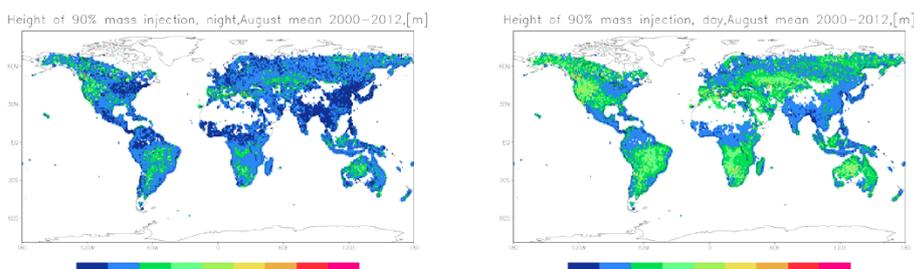
2108 Emissions factors for the different species are generally taken as constant for each type of ecosystem

2109 in different countries. However, emissions from a fire depend on the fuel type and fire
2110 characteristics, and they are often poorly determined. Most inventories currently use the emission
2111 factors compiled by Akagi et al. (Akagi et al., 2011) or the Andreae and Merlet
2112 (Andreae and Merlet, 2001) compilation and following updates. Results
2113 from different studies nevertheless stress that more complete combustion, as in flaming fires, would
2114 lead to a larger fraction of highly oxidized species (e.g., CO₂, NO_x), while smouldering fires release
2115 more material in reduced form (e.g., CO, NH₃ and NMVOC species), which indicates that emission
2116 factors may vary with season, and that fire characteristics can be very different from one fire to
2117 another even within the same geographical location. Emissions factors from peat fires are not well
2118 quantified with different studies reporting a wide range of values (Akagi et al., 2011).
2119

2120 The lifetime of the species released from fires depends on chemical processes in fire plumes
2121 and therefore on the vertical distribution of the plumes. The plume injection height can be critical in
2122 the impact of biomass burning (Leung et al., 2007). Several studies have proposed plume models or
2123 parameterization of the top height of the plumes, based on empirical methods or on the use of
2124 satellite observations (Tosca et al., 2011; Martin et al., 2010). A
2125 determination of the vertical profiles of fires emissions at the global scale was proposed by Sofiev et
2126 al. (Sofiev et al., 2013), based on satellite observations and semi-empirical
2127 formulas. An example is shown in Figure 1819, which highlights the importance of meteorology in the
2128 day-night variation of the height of fire plumes. It is worth noting that the diurnal variation of the
2129 injection height is significant such that one can practically consider two independent datasets, one
2130 for daytime and one for nighttime, with transition during morning and evening (Sofiev et al., 2013).
2131 The influence on ozone can be a combination of injection height and frequency (Williams et al.,
2132 2012).

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2133



2134

2135 **Figure 1819** - Injection height (in m) for 90% of the mass injection for night (left) and day (right) for
2136 August biomass burning from (Sofiev et al., 2013).

2137 2.3.7 Natural emissions

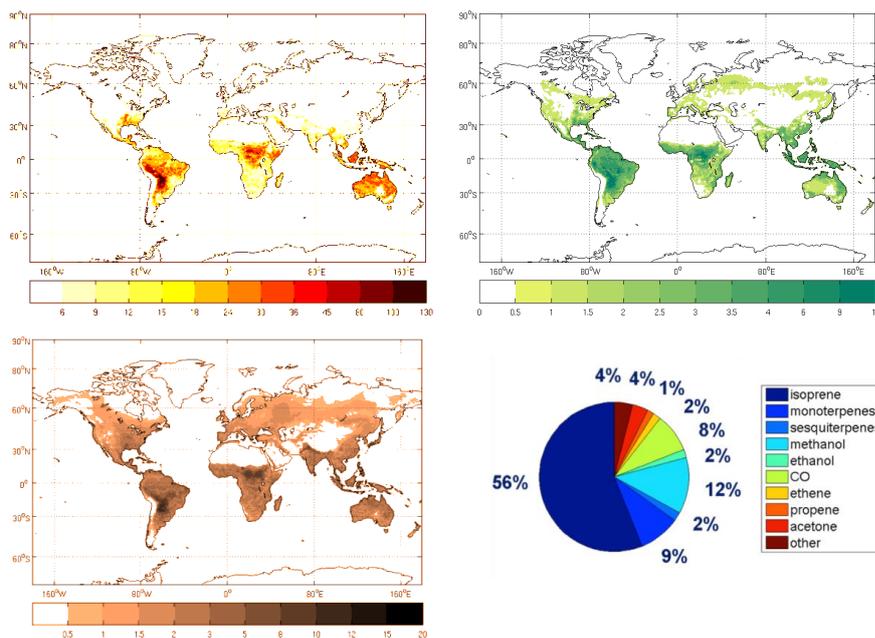
2138 Natural processes in the atmosphere, vegetation and oceans lead to the emissions of a wide range of
2139 compounds (Granier et al., 2004). Emissions resulting from lightning are
2140 discussed in Section 4.7. This section only focuses on emissions of hydrocarbons from vegetation,
2141 since they can have a large impact on the distribution of ozone and its precursors and because
2142 terrestrial vegetation is the dominant source of atmospheric VOCs (Guenther et al., 1995).
2143 The emissions of isoprene and monoterpenes are the largest, but many other

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2144 compounds are also emitted. Many studies performed during the past few years have used MEGAN
 2145 (Model of Emissions of Gases and Aerosols from Nature) to quantify the emissions of biogenic VOCs
 2146 to the atmosphere. MEGAN is a model system calculating temporal and spatial rates of emission of
 2147 chemical compounds from terrestrial ecosystems to the atmosphere under varying environmental
 2148 conditions. It was first introduced by Guenther et al. (Guenther et al., 1995; Guenther et al.,
 2149 2006; Guenther et al., 2003; Guenther et al., 1993)(Guenther et al., 1995; Guenther et al.,
 2150 2006; Guenther et al., 2003; Guenther et al., 1993). The most recent version of the model, MEGAN
 2151 v2.1 was introduced in Guenther et al. (Guenther et al., 2012)(Guenther et al., 2012) and Sindelarova
 2152 et al. (Sindelarova et al., 2014). The current algorithm takes into account the impact of past and
 2153 present temperature, light (sun-lit and sun-shaded leaves), leaf age, CO₂ inhibition of isoprene
 2154 emissions and losses of primary emissions in the canopy and soil moisture.

2155
 2156 Examples of emissions of isoprene, monoterpenes and methanol in 2003 are shown in Figure 1920,
 2157 together with the relative contribution of the different biogenic VOCs to the total amount emitted.
 2158 Isoprene emissions account for 56% of the total, which indicates that the emissions of the other
 2159 BVOCs and their chemical oxidation schemes (see section 4.3 and 4.10) also need to be taken into
 2160 account when simulating the atmospheric composition of the atmosphere.

2161



2162

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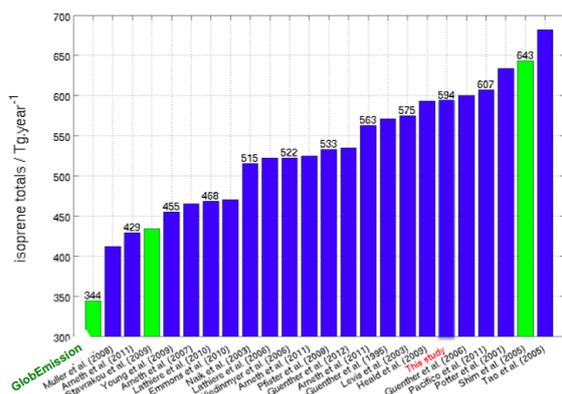
2164 **Figure 1920** - Mean annual emissions (in $\text{mg}\cdot\text{m}^{-2}\cdot\text{day}^{-1}$) of isoprene (top left), monoterpenes (top
 2165 right), methanol (bottom left), and contribution of each BVOCs to the annual global total average
 2166 (bottom right) (Sindelarova et al., 2014)(Sindelarova et al., 2014).

2167 Previous studies show significant differences between the total amount of BVOCs emitted by
 2168 vegetation (Sindelarova et al., 2014)(Sindelarova et al., 2014). These differences are related to the
 2169 large uncertainties of different factors driving BVOCs emissions, such as radiation amount, leaf

2170 temperature, and parameterization of the soil moisture factor or the representation of canopy
 2171 processes. There are also large differences in the vegetation types and in the distribution of plant
 2172 functional types used in different models and/or their assignment with BVOCs emission activity.

2173 Other methods have been used to calculate the emissions of BVOCs by the vegetation. Arneth et al.
 2174 (2007) (~~Arneth et al., 2007~~)(Arneth et al., 2007) have for example calculated the emissions using a
 2175 dynamic global vegetation model (LPJ-GUESS, (~~Sitch et al., 2003~~)(Sitch et al., 2003)) and a
 2176 physiological BVOCs emission algorithm. Studies conducted using the LPJ-GUESS model (~~Arneth et al.,~~
 2177 ~~2011~~)(Arneth et al., 2011) have shown that variations of driving input data such as meteorology and
 2178 vegetation description among different models might lead to substantially different emission
 2179 estimates.

2180 Figure 2021 shows the differences between previous estimates of global total isoprene emissions:
 2181 the difference between the lowest and ~~largest~~highest estimate is 65%. In ~~figure 20~~Figure 21, the
 2182 three values indicated by green bars correspond to an optimization of isoprene emissions using
 2183 inverse modeling techniques and formaldehyde satellite observations (see next section). The
 2184 difference between the highest and lowest value is about a factor of two, which highlights the large
 2185 uncertainties on the determination of isoprene emissions.



2186
 2187 **Figure 2021** - Isoprene global total estimated from different studies. Studies highlighted in green
 2188 used formaldehyde satellite data and an inversion modeling technique to constrain isoprene
 2189 emissions (~~Sindelarova et al., 2014~~)(Sindelarova et al., 2014).

2190
 2191 **2.3.8 Evaluation and optimization of emissions using inverse modelling techniques**

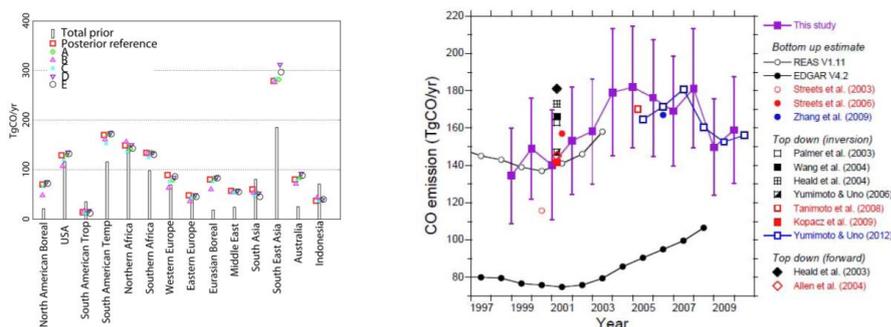
2192 Inverse modelling techniques have been widely used during the past few years to optimize surface
 2193 emissions so that simulated atmospheric concentrations can reproduce observed concentrations.
 2194 Most recent studies either minimize the weighted distance between model and observations
 2195 (variational approach) or minimize the error variance of the estimated parameters (Kalman Filter
 2196 techniques) (~~Saide et al., 2011~~)(Saide et al., 2011). Differences between the distributions provided by
 2197 the models and the observations are not only due to inaccuracies on the quantification of emissions,
 2198 errors in the model data (meteorological fields for example), but also in the representation of

2199 physical and chemical processes in the models: all these factors need to be taken into account in
 2200 inverse systems (Elbern et al., 2007)(Elbern et al., 2007). Estimates of emissions using inverse
 2201 techniques also depend on the *a-priori* emission dataset used, on the meteorological fields used to
 2202 drive the models used that infer emissions (Arellano and Hess, 2006)(Arellano and Hess, 2006), and
 2203 on the data used in the optimization of the emissions (Hooghiemstra et al., 2012)(Hooghiemstra et
 2204 al., 2012). A review of the current capabilities of inverse techniques to better quantify surface
 2205 emissions in North America using satellite observations was published by Streets et al., 2013 (Streets
 2206 et al., 2013)(Streets et al., 2013).

2207 These techniques have been mostly applied to the species for which observations from satellite are
 2208 available or for the optimization of local/regional emissions when comprehensive observation
 2209 datasets exist. Therefore, most studies have discussed the optimization of greenhouse gases (CO₂
 2210 and CH₄), of ozone precursors CO and NO₂, of aerosols and their precursors (see (Fuzzi et al.,
 2211 2014)(Fuzzi et al., 2015) companion paper), and of biogenic emissions using satellite observations of
 2212 formaldehyde.

2213
 2214 Several studies have applied inverse modelling techniques to the optimization of CO emissions using
 2215 observations from surface networks (Petron et al., 2004)(Petron et al., 2004) as well as observations
 2216 from the MOPITT and IASI satellite instruments. Most of these studies using satellite data have
 2217 concluded that the *a-priori* anthropogenic emissions of CO might be underestimated as indicated in
 2218 Figure 21a22a (Fortems-Cheiney et al., 2011)(Fortems-Cheiney et al., 2011) and 22b (Tohjima et al.,
 2219 2014)(Tohjima et al., 2014), more particularly at mid-latitudes of the northern hemisphere in winter
 2220 (Kopacz et al., 2010;Stein et al., 2014)(Kopacz et al., 2010;Stein et al., 2014). However, an analysis of
 2221 satellite and aircraft observations has also shown that emissions inventories in North America might
 2222 provide overestimated values (Miller et al., 2008)(Miller et al., 2008). Emissions resulting from fires
 2223 have also been optimized, and showed that current surface inventories might underestimate fires in
 2224 boreal regions (Pfister et al., 2005)(Pfister et al., 2005). Inverse techniques have also provided
 2225 information on the seasonal cycles of emissions from fires, for example in Africa as shown by
 2226 Chevallier et al. (Chevallier et al., 2009)(Chevallier et al., 2009).

2227
 2228



2229
 2230 **Figure 2122** - (a, left): Annual total posterior CO emissions per region for year 2004 compared to the
 2231 a priori reference : the different cases correspond to tests on different errors in the model (Fortems-
 2232 Cheiney et al., 2011)(Fortems-Cheiney et al., 2011). (b, right) : Comparison of estimated CO annual

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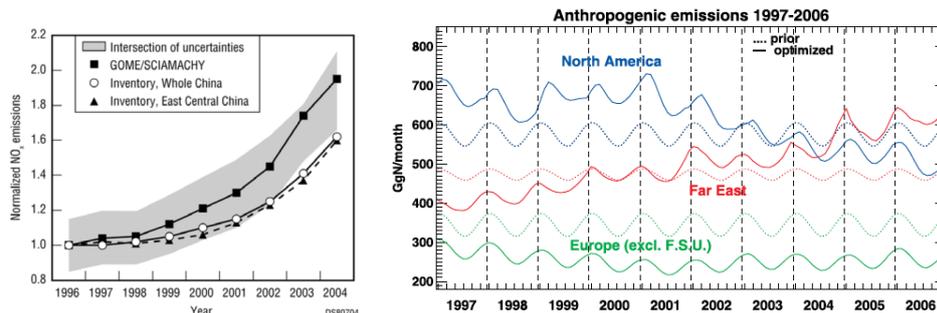
2233 emissions in China from inventories and inverse studies; the “This Study” refers to a top down
2234 estimate (Tohjima et al., 2014)(Tohjima et al., 2014).

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2235 Several studies have used satellite NO₂ column observations to constrain nitrogen oxide emission
2236 inventories through inverse or forward modeling (Martin et al., 2004;Kim et al., 2009;Lamsal et al.,
2237 2011;Wang et al., 2012)(Martin et al., 2004;Kim et al., 2009;Lamsal et al., 2011;Wang et al., 2012).
2238 These studies exploited the relatively short lifetime of tropospheric nitrogen oxides, which ranges
2239 from several hours to a few days. As a result, NO₂ has its largest concentrations in the boundary layer
2240 close to emission sources, making measurements of NO₂ columns well suited to improving our
2241 knowledge of the emissions of NO_x. Two examples of optimizations of NO_x emissions using satellite
2242 observations are given in Figure 2223. Figure 22a23a shows a quantification of emissions trends in
2243 China as provided by an inventory and optimized using the SAIAMACHY/SCIAMACHY/GOME
2244 observation for the 1996-2004 period is shown in Figure 2223 (Zhang et al., 2007)(Zhang et al., 2007).
2245 From 1996 to 2004, NO_x emissions over East Central China region increased by 61% according to the
2246 inventory, while a 95% increase in the NO₂ columns was obtained from the satellite observations
2247 during the same period. Stavrou et al. (Stavrou et al., 2008)(Stavrou et al., 2008) have
2248 evaluated the changes in the emissions in different regions of the world from 1997 to 2006 using an
2249 inverse method and found larger increases in the optimized emissions when compared to prior
2250 emissions in the Far East region, and emissions decreasing faster than in the inventories in North
2251 America.

2252



2253

2254 **Figure 2223** - Temporal evolution of NO_x emissions over China from the Zhang et al. (2007) inventory
2255 and inverse method using satellite observations. All data are normalized to the year 1996 (Zhang et
2256 al., 2007)(Zhang et al., 2007).

2257

2258 Optimization of the emissions of several other compounds have also been evaluated through inverse
2259 methods constrained by satellite observations, such as isoprene, methanol or formaldehyde
2260 (Stavrou et al., 2011;Stavrou et al., 2009;Palmer et al., 2003;Wells et al., 2014;Barkley et al.,
2261 2013)(Stavrou et al., 2011;Stavrou et al., 2009;Palmer et al., 2003;Wells et al., 2014;Barkley et
2262 al., 2013) and other studies have focused on the optimizations of the emissions from different
2263 sectors, such as ships ((de Ruyter de Wildt et al., 2012;Vinken et al., 2014)(de Ruyter de Wildt et al.,
2264 2012;Vinken et al., 2014).

2265

2266 2.4 A climatological view of ozone/linkages across the scales

2267

2268 The available observations show that tropospheric ozone is highly variable both in space and time, on
2269 long and short scales. Over the remote oceans, observations show low ozone amounts within the
2270 tropical Hadley circulation with little or no vertical gradient in mole fraction, then higher
2271 concentrations at mid latitudes, with an increase with height, corresponding to the additional source
2272 of tropospheric ozone from STE as well as in-situ chemistry (See [figures 23](#)[Figures 24](#) and [2425](#)). Over
2273 the continents and nearby oceans in the lower troposphere different behaviour is observed
2274 depending on the level of the ozone precursors. Analysis of the seasonal-diurnal variations allows
2275 separating the following types of ozone variations in the extra-tropics ([Tarasova et al.,
2276 2007](#))([Tarasova et al., 2007](#)): clean background, rural, semi-polluted non-elevated, semi-polluted
2277 semi-elevated, elevated and ~~polar~~Polar/remote marine. For the “clean background” regime in the
2278 Northern hemisphere the seasonal maximum is observed in March-April, both for night and day. For
2279 those sites with a double maximum or a wide spring-summer maximum, the spring maximum
2280 appears both for day and night, while the summer maximum is more pronounced for daytime and
2281 hence can be attributed to photochemical processes. There is much debate as to the origin on the
2282 spring maximum likely a combination of dynamical/transport processes and photochemistry ([Monks,
2283 2000](#))([Monks, 2000](#)).

2284 In July, northern summer, major regions of elevated lower troposphere ozone are observed over
2285 USA, Europe and East Asia from urban/industrial activities and regions of elevated ozone over the
2286 tropics due to both biomass burning and other human activities. Satellite observations of
2287 tropospheric ozone have revealed much detail about the spatial distribution including the discovery
2288 of the regional tropospheric ozone maximum off southwest Africa over the Atlantic Ocean (Figure
2289 [2324](#)) and the oscillation of tropospheric ozone over the Indonesian Archipelago in synchronicity with
2290 El Niño ([Ziemke et al., 2010](#))([Ziemke et al., 2010](#)). In highly dynamic regions such as the
2291 Mediterranean basin ([Lelieveld et al., 2002](#);[Lelieveld et al., 2009](#))([Lelieveld et al., 2002](#);[Lelieveld et
2292 al., 2009](#)), the strength of the meteorological systems (Azores anticyclone and Middle Eastern
2293 depression) are key factors in explaining both the variability and the anomalies of ozone in the lower
2294 troposphere in this region ([Doche et al., 2014](#))([Doche et al., 2014](#)). Recent observations have
2295 suggested there is a summertime free tropospheric ozone pool over the Mediterranean ([Zanis et al.,
2296 2014](#);[Kalabokas et al., 2013](#))([Zanis et al., 2014](#);[Kalabokas et al., 2013](#)).

2297

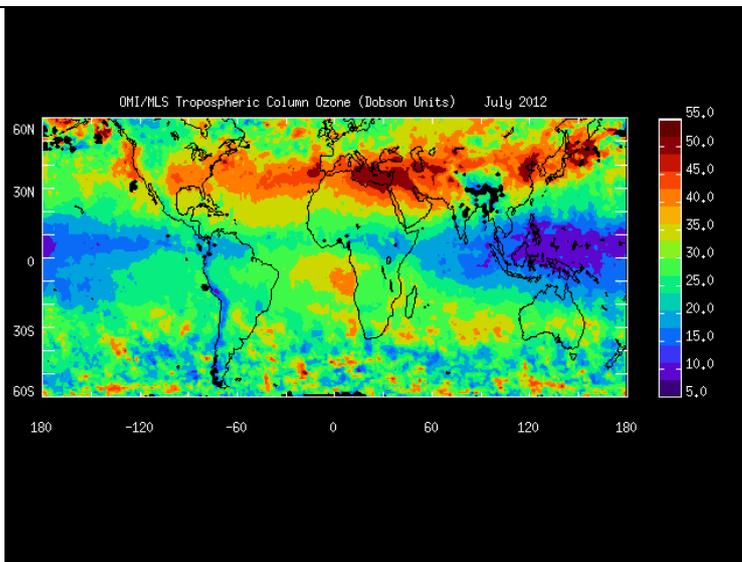


Figure 2324 - Tropospheric ozone columns determined by residual from OMI observations of total column O₃ and MLS observations of stratospheric O₃ (Ziemke et al., 2006)(Ziemke et al., 2006). Plots retrieved from http://acd-ext.gsfc.nasa.gov/Data_services/cloud_slice/index.html#nd

2298

2299 The data in Figure 2324 clearly highlights the benefits offered by satellite data for investigating the
 2300 global distribution of tropospheric ozone. However, it should be kept in mind that such data give an
 2301 incomplete picture (Monks and Bierle, 2011)(Monks and Bierle, 2011) which requires complementary
 2302 observing systems (Laj et al., 2009)(Laj et al., 2009). For example, the AMMA campaign (Redelsperger
 2303 et al., 2006) and associated analysis performed with the in situ MOZAIC (Sauvage et al., 2005) and
 2304 SHADOZ data (Thompson et al., 2003a;Thompson et al., 2003b) have revealed a somewhat different
 2305 feature of ozone distribution (both in terms of amount of the tropospheric columns and in the
 2306 location of the maxima) over Western and Southern Africa including the adjacent Atlantic Ocean. In
 2307 July, when biomass burning mainly occurs over the southern tropical part of Africa, the tropospheric
 2308 ozone column is about 45 DU over the continent (Thompson et al., 2014;Sauvage et al.,
 2309 2005;Sauvage et al., 2006). Export of polluted air masses is observed throughout the Gulf of Guinea
 2310 up to the coast of West Africa in the northern hemisphere. Reeves et al.. It is worth noting that
 2311 there are direct tropospheric ozone retrievals from satellite data (Liu et al., 2006). The AMMA
 2312 campaign (Redelsperger et al., 2006) and associated analysis performed with the in-situ MOZAIC
 2313 (Sauvage et al., 2005) and SHADOZ data (Thompson et al., 2003a;Thompson et al., 2003b) have
 2314 revealed a somewhat different feature of ozone distribution (both in terms of amount of the
 2315 tropospheric columns and in the location of the maxima) over Western and Southern Africa
 2316 including the adjacent Atlantic Ocean. In July, when biomass burning mainly occurs over the
 2317 southern tropical part of Africa, the tropospheric ozone column is about 45 DU over the continent
 2318 (Reeves et al., 2010)(Thompson et al., 2014;Sauvage et al., 2005;Sauvage et al., 2006) provide a clear
 2319 characterisation. Export of the ozone distribution polluted air masses is observed throughout the
 2320 troposphere over Gulf of Guinea up to the coast of West Africa during in the monsoon period. Thanks
 2321 to previous analysis based on MOZAIC data northern hemisphere. Reeves et al., (Sauvage et al.,

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2322 [2007a;Sauvage et al., 2005](#))([Reeves et al., 2010](#)) and provide a clear characterisation of the regular
2323 [ozone soundings distribution throughout the troposphere over Cotonou West Africa during 2-year the](#)
2324 [monsoon period. Owing to previous analysis based on MOZAIC data \(Thouret et al., 2009\)\(Sauvage et](#)
2325 [al., 2007a;Sauvage et al., 2005\)](#), it has been shown that the biomass burning seasons in both
2326 hemispheres impact the tropospheric ozone profiles in both hemisphere as well. As a consequence,
2327 a picture like the so-called “Ozone paradox” (Thompson and Hudson, 1999) was shown to be no
2328 longer valid (Sauvage et al., 2006) owing to the insight provided by additional in-situ observations.
2329 Recent work has shown evidence for the removal of ozone in Saharan dust plumes (Andrey et al.,
2330 2014) which is correlated with both dust loading and total amount of water present.

2331 [and the regular ozone soundings over Cotonou over 2 years \(Thouret et al., 2009\)](#), it has been shown
2332 [that the biomass burning seasons in both hemispheres impact the tropospheric ozone profiles in](#)
2333 [both hemispheres as well. As a consequence, a picture like the so-called “Ozone paradox”](#)
2334 [\(Thompson and Hudson, 1999\) was shown to be no longer valid \(Sauvage et al., 2006\) owing to the](#)
2335 [insight provided by additional in-situ observations. Recent work has shown evidence for the removal](#)
2336 [of ozone in Saharan dust plumes \(Andrey et al., 2014\) which is correlated with both dust loading and](#)
2337 [total amount of water present.](#)

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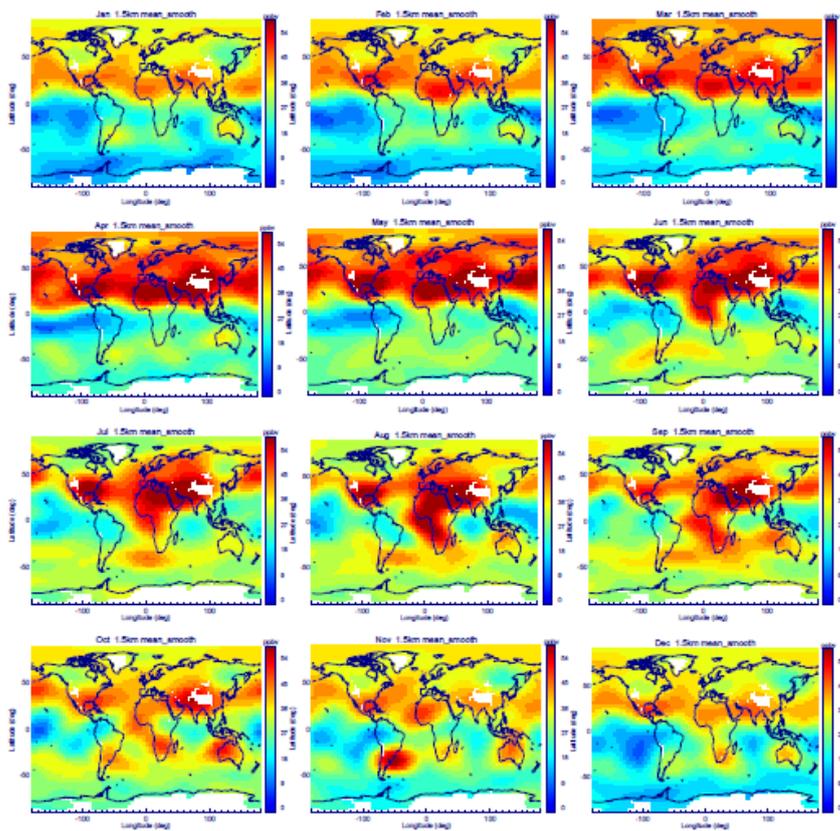


Figure 2425 – Monthly global ozone distributions at 1.5 km a.s.l from trajectory mapped ozone soundings ([Liu et al., 2013a](#))([Liu et al., 2013a](#)).

2338

2339 At coastal or island sites in the tropics diurnal variation in ozone generally show a minimum during
 2340 the daytime. Occasional air transport events from the mid-latitudes bring in higher ozone
 2341 concentrations (Lawrence and Lelieveld, 2010)(Lawrence and Lelieveld, 2010). The mole fractions
 2342 are low, typically 5 – 20 ppb, and the annual maximum occurs in June to September, southern
 2343 winter, a characteristic of ozone in the southern hemisphere troposphere. For more continental
 2344 tropical regional sources of ozone due to precursor emissions from such activities as biomass burning
 2345 (e.g. Ojha et al., 2014; Cristofanelli et al., 2010)(Ojha et al., 2014; Cristofanelli et al., 2010)), land
 2346 clearing, agriculture and human settlements may cause occasional elevated ozone concentrations.
 2347 (Jacob et al., 1996). The seasonal ozone variation may be affected by these activities which can be
 2348 influenced by seasonal meteorological conditions such as the occurrence of a wet/dry seasons (Valks
 2349 et al., 2014; Rama Gopal et al., 2014; Cristofanelli et al., 2010)(Valks et al., 2014; Rama Gopal et al.,
 2350 2014; Cristofanelli et al., 2010; Sinha et al., 2014). Under stable conditions, ozone depletion due to
 2351 ozone deposition and NO titration will occur. The diurnal variation of ozone will generally be
 2352 characterized by lower concentrations at night and higher concentrations during the daytime (see
 2353 e.g. (Rama Gopal et al., 2014)(Rama Gopal et al., 2014)). Vegetation cover plays an important role
 2354 together with local meteorology in (natural) trace gas emissions. For example, tropical forests and
 2355 plantations are among the greatest emitters of isoprene, a reactive hydrocarbon species which is
 2356 involved in the photochemical cycle of tropospheric ozone (see section 4.3). Replacement of tropical
 2357 forest by agriculture (e.g. palm trees) may cause potential future elevated ozone levels (Hewitt et al.,
 2358 2009; MacKenzie et al., 2011)(Hewitt et al., 2009; MacKenzie et al., 2011) (see Figure 2526).

2359

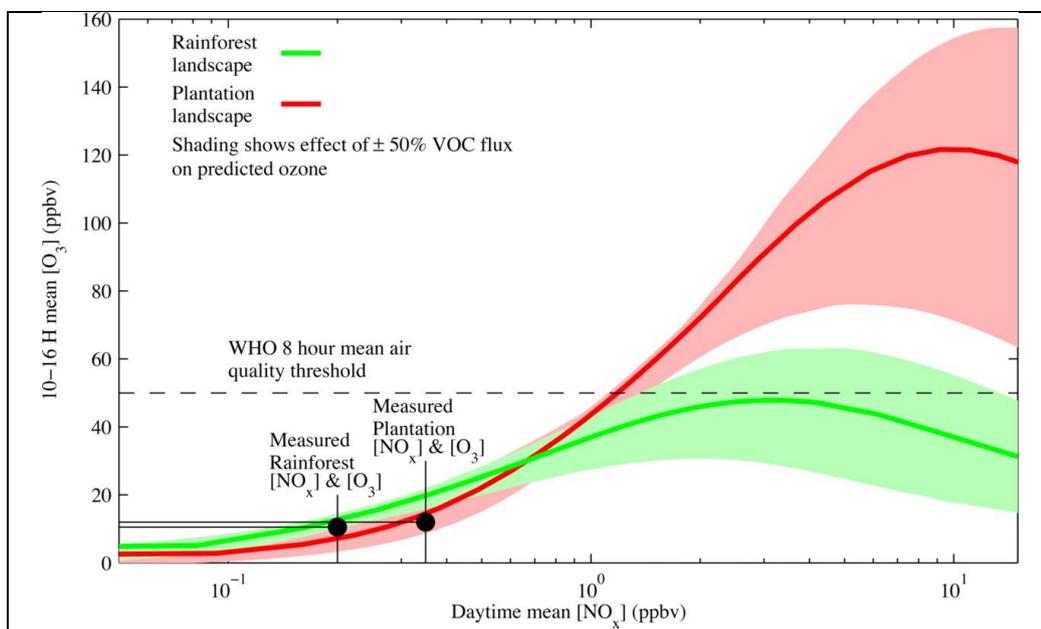


Figure 2526 – Sensitivity of daytime (10.00-16.00 h) average ozone concentrations to [NO_x] in the boundary layer for isoprene and monoterpene emission rates measured in a rainforest and palm oil landscape (Hewitt et al., 2009)(Hewitt et al., 2009).

2360

2361 | High latitude sites may be influenced by the near continuous presence of snow or ice cover ([Helmig et al., 2007](#))([Helmig et al., 2007](#)). Polar sites may have wind directions that remain relatively constant
2362 | for months. Coastal sites may have ozone depletion events, which are related to halogen chemistry
2363 | ([Simpson et al., 2007](#))([Simpson et al., 2007](#)), and which is evident for example at Barrow during
2364 | northern spring, March to May ([Oltmans et al., 2012](#))([Oltmans et al., 2012](#)). The high reflectivity of
2365 | snow and ice influences the photochemical reaction rates, because more UV radiation is available
2366 | than ~~what~~ would be expected for these latitudes if one assumes a global average value for the
2367 | surface UV albedo. In stable conditions with bright sunlight, local photochemical production has been
2368 | observed due to NO_x emission from the snow ([Helmig et al., 2008a](#);[Helmig et al., 2008b](#))([Helmig et al., 2008a](#);[Helmig et al., 2008b](#)) and this is evident at the South Pole during November to February,
2369 | centred on southern summer. The ozone concentrations in background air at the South Pole peak in
2370 | winter/spring and fall to a minimum in summer/autumn as with other southern hemisphere sites.
2371 |
2372 |

2373 | Moving to the urban scale the local coupling of NO_x and O₃ is important in that reductions in NO can
2374 | contribute to increases in ozone ([Kley et al., 1994](#))([Kley et al., 1994](#)). In areas of high NO_x, as is often
2375 | observed in urban areas, ozone levels can be suppressed through the following reaction:



2378 | This phenomenon, sometimes dubbed ‘NO_x titration’, thereby leads to the counter-intuitive effect
2379 | that reducing NO_x (NO) reduces the amount of ozone being suppressed and actually increases ozone
2380 | ([Heal et al., 2013b](#);[Sicard et al., 2013](#))([Heal et al., 2013a](#);[Sicard et al., 2013](#)). The spatial variability of
2381 | ozone in larger urban areas can be very much driven by “titration” ([Escudero et al., 2014](#))([Escudero et al., 2014](#)).
2382 | Figure [2627](#) shows the gradual increase in urban ozone, moving towards background
2383 | levels, driven by reducing NO_x emissions over the last couple of decades in the UK, decreasing the
2384 | suppression of ozone. Figure [2728](#) shows the “titration” in urban areas across Europe from
2385 | modelling studies, showing the widespread nature of this effect and the need to couple the regional
2386 | and hemispheric picture to the urban. In a sense there is an urban decrement driven by this process.
2387 | Conversely, there has been some work looking at megacity impacts on regional ozone ([Tie et al., 2013](#);[Beirle et al., 2011](#);[Guttikunda et al., 2005](#))([Tie et al., 2013](#);[Beirle et al., 2011](#);[Guttikunda et al., 2005](#)) (see also Section 4.1).
2388 |
2389 |

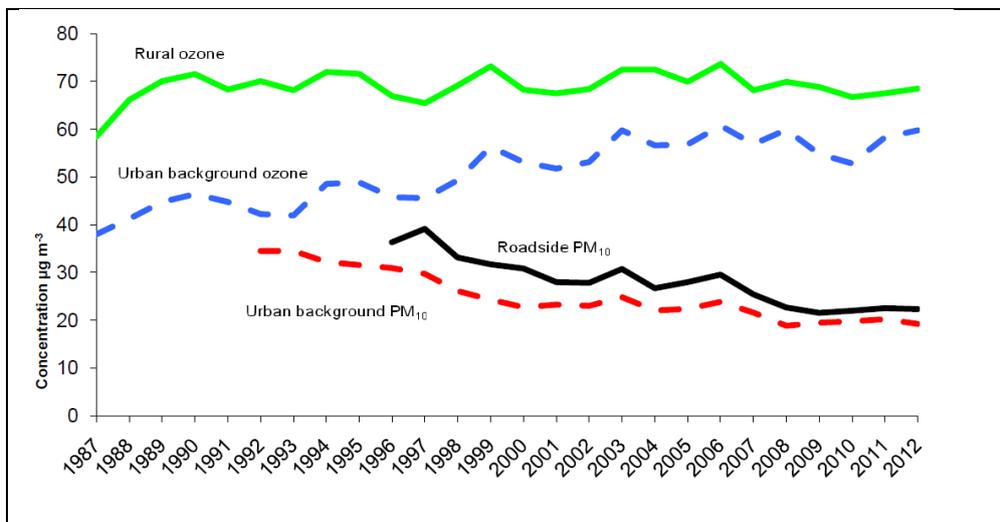


Figure 2627: Annual levels of PM_{10} and ozone (and PM_{10}) in the UK 1987-2012 (Defra, 2013) (Defra, 2013), showing the rise of urban ozone concentrations towards the rural ones and the relatively flat rural concentrations over the last decade.

2390

2391 Given the strong NO_x-O_3 (sometimes referred to as O_x (Zlatev et al., 1992; Yu et al., 2008) (Zlatev et al.,
 2392 1992; Yu et al., 2008), see also Figure 2728) couple at the urban scale there are a number of issues
 2393 around the role of an increase in the $NO_2:NO$ emission ratio caused by an increase in the share of
 2394 diesel vehicles in Europe and the role that may play in ozone formation (Weiss et al., 2012; Carslaw et
 2395 al., 2011; Carslaw, 2005) (Weiss et al., 2012; Carslaw et al., 2011; Carslaw, 2005). Further there are
 2396 questions as to what extent spatially (along roadsides, across urban areas, regionally) primary NO_2
 2397 emissions affect ozone concentrations (see e.g. (Yu et al., 2014) (Yu et al., 2014)). Meteorology also
 2398 plays a large role in local scale ozone concentration affecting deposition and transport (e.g. (Vieno et
 2399 al., 2010; Francis et al., 2011) (Vieno et al., 2010; Francis et al., 2011)) alongside transport and
 2400 dispersion in urban landscapes such as street canyons etc. (see e.g. (Baker et al., 2004; Zhong et al.,
 2401 2014; Kwak and Baik, 2014) (Baker et al., 2004; Zhong et al., 2014; Kwak and Baik, 2014)). Urban areas
 2402 are dynamic regions in terms of ~~emmission~~emmission of ozone precursors that have seen strong
 2403 changes owing to emmission reduction measures (see e.g. (vonSchneidemesser et al.,
 2404 2011; vonSchneidemesser et al., 2010; McMeeking et al., 2012; Warneke et al., 2012; Vijayaraghavan et
 2405 al., 2014) (vonSchneidemesser et al., 2011; vonSchneidemesser et al., 2010; McMeeking et al.,
 2406 2012; Warneke et al., 2012; Vijayaraghavan et al., 2014)).

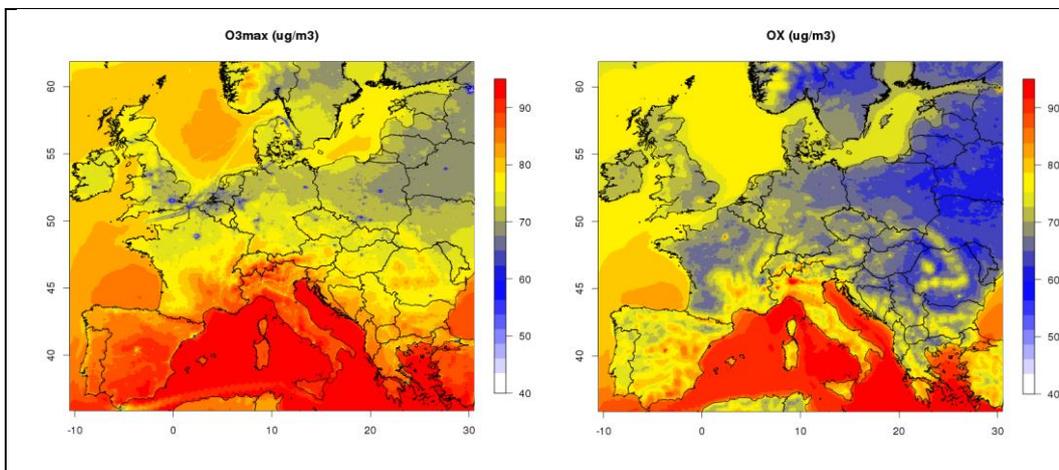


Figure 2728 - In a continental chemistry transport simulation with the CHIMERE model at a resolution of 8 km, major urban centres in Northern and Central Europe highlight the NO_x titration process (left: summertime average of daily maximum ozone) while the total oxidant level (right: O_x as NO₂ + O₃, annual mean) is high in most European cities (adapted from [\(Terrenoire et al., 2013\)](#)[\(Terrenoire et al., 2013\)](#), EC4MACS project).

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2407

2408 3 Impacts

2409

2410 Surface level ozone has multiple impacts. As an oxidant it can induce respiratory problems and has
 2411 been associated with premature human mortality [\(Bell et al., 2006\)](#)[\(Gryparis et al., 2004\)](#)[\(Bell et al.,](#)
 2412 [2006\)](#)[\(Gryparis et al., 2004\)](#). Further it can cause tree/vegetation damage [\(Krupa and Manning,](#)
 2413 [1988\)](#)[\(Krupa and Manning, 1988\)](#), reduce photosynthesis and growth [\(Reich and Amundson,](#)
 2414 [1985\)](#)[\(Reich and Amundson, 1985\)](#) and therefore crop yields [\(Fiscus et al., 2005\)](#)[\(Fiscus et al., 2005\)](#)
 2415 (see also [\(Felzer et al., 2007\)](#)[\(Ashmore, 2005\)](#)[\(Felzer et al., 2007\)](#)[\(Ashmore, 2005\)](#) for recent reviews).
 2416 Ozone is also an important greenhouse gas [\(IPCC, 2013\)](#)[\(IPCC, 2013\)](#).

2417 3.1 Health

2418

2419 Ozone has long been known to cause adverse effects on health [\(Bates, 2005\)](#)[\(Bates, 2005\)](#). The gas is
 2420 a powerful oxidant and short-term inhalation induces inflammation of the entire respiratory tract,
 2421 although the nasal cavity and the zone between the conducting and gas-exchange airways are
 2422 potentially more susceptible. Significant inflammation can be detected at ambient levels of ozone in
 2423 controlled human exposure studies, and although there is some degree of adaptation after repeated
 2424 exposures, pulmonary inflammation may persist, mainly in the terminal bronchiolar units. As well as
 2425 elucidating these effects in exposure studies, which naturally exclude the most sensitive or
 2426 susceptible parts of the population, associations between ozone and mortality and morbidity have
 2427 also been shown in epidemiological studies, covering the population at large.

2428 Early work on ozone health effects involved mainly exposure studies in chambers. On the basis of this
 2429 work ozone was included in the first tranche of National Air Quality Standards in the US, with a
 2430 standard of 80ppb as a 1 hour average, promulgated in 1971 as part of the US Clean Air Act. In

2431 Europe, the first edition of the WHO 'Air quality guidelines for Europe' in 1987 set a 1-hour guideline
2432 value of 75-100 ppb. This guideline was also based primarily on chamber studies. Since then much
2433 more evidence has accumulated regarding ozone effects, not least from epidemiology studies, and in
2434 the 2005 Global Update (~~WHO, 2005~~)(WHO, 2005) the most recent WHO Guideline is now 50ppb
2435 expressed as the maximum 8-hour mean in a day. The same document also sets an interim target of
2436 80 ppb as a daily maximum 8-hour mean, and a 'High Level' of 120 ppb daily maximum 8-hour mean
2437 where it notes that "significant health effects could occur" (interim) and that a "substantial
2438 proportion of vulnerable population affected' (sicaffected" (high).

2439 Earlier WHO guidelines were based on these chamber studies on humans and animals, but the more
2440 recent 2005 Global Update from WHO mentioned above additionally used time series
2441 epidemiological studies (~~WHO, 2005~~)(WHO, 2005). However, all guidelines up to and including 2005
2442 referred to short-term exposures and health effects, but the 2005 report noted that at that time
2443 there was some evidence that long-term exposure to ozone may have chronic effects but the review
2444 concluded that the evidence was insufficient to recommend a guideline. The policy implications of
2445 these purely health-based guidelines are discussed in Section 5.

2446 As part of their 'Year of the Air' in 2013 the European Commission requested WHO to review the
2447 health effect evidence on the pollutants covered in EU air quality legislation, including ozone. The
2448 review was in two parts, firstly a review (REVIHAAP) of the health effect evidence itself, in the form
2449 of answers to policy-relevant questions prepared by the Commission, in consultation with the WHO.
2450 The final REVIHAP report is available at
2451 [http://www.euro.who.int/_data/assets/pdf_file/0004/193108/REVIHAAP-Final-technical-report-](http://www.euro.who.int/_data/assets/pdf_file/0004/193108/REVIHAAP-Final-technical-report-final-version.pdf)
2452 [final-version.pdf](http://www.euro.who.int/_data/assets/pdf_file/0004/193108/REVIHAAP-Final-technical-report-final-version.pdf) . The second part of the process involved a shorter project (HRAPIE) which
2453 recommended concentration-response functions for use in health impact assessments and other
2454 quantification, for example in integrated assessment modelling to underpin the EU policy process.
2455 The HRAPIE report is available at
2456 [http://www.euro.who.int/_data/assets/pdf_file/0006/238956/Health-risks-of-air-pollution-in-](http://www.euro.who.int/_data/assets/pdf_file/0006/238956/Health-risks-of-air-pollution-in-Europe-HRAPIE-project-Recommendations-for-concentrationresponse-functions-for-costbenefit-analysis-of-particulate-matter,-ozone-and-nitrogen-dioxide.pdf)
2457 [Europe-HRAPIE-project-Recommendations-for-concentrationresponse-functions-for-costbenefit-](http://www.euro.who.int/_data/assets/pdf_file/0006/238956/Health-risks-of-air-pollution-in-Europe-HRAPIE-project-Recommendations-for-concentrationresponse-functions-for-costbenefit-analysis-of-particulate-matter,-ozone-and-nitrogen-dioxide.pdf)
2458 [analysis-of-particulate-matter,-ozone-and-nitrogen-dioxide.pdf](http://www.euro.who.int/_data/assets/pdf_file/0006/238956/Health-risks-of-air-pollution-in-Europe-HRAPIE-project-Recommendations-for-concentrationresponse-functions-for-costbenefit-analysis-of-particulate-matter,-ozone-and-nitrogen-dioxide.pdf) .

2459 The REVIHAAP report addressed three questions relating to ozone where WHO and the European
2460 Commission felt a more recent assessment was warranted. The questions posed were

- 2461 (i) What new evidence on health effects has emerged since the review work done for
2462 the 2005 global update of the WHO air quality guidelines, particularly with regard
2463 to the strength of the evidence of the health impacts associated with short-term
2464 and long-term exposure to ozone?
- 2465 (ii) What new health evidence has been published in relation to the evidence or
2466 likelihood(sic) of a threshold below which impacts are not expected? and
- 2467 (iii) Based on currently available health evidence, what ozone metrics , health
2468 outcomes and concentration-response functions can be used for health impact
2469 assessment

2470 The answer to the first question noted that the earlier 2005 global update found evidence only for
2471 short-term effects on mortality and morbidity. However an important advance is that several cohort
2472 studies have now emerged showing associations between long-term exposures and mortality. Of

2473 particular importance are the studies using the large American Cancer Society cohort. A study
2474 ~~(Krewski et al., 2009)~~(Krewski et al., 2009) using single pollutant models found an association
2475 between summertime average ozone and all-cause mortality as well as cardiopulmonary mortality,
2476 although the high correlation between PM_{2.5} and ozone in the summer months made it difficult to
2477 separate the effects of the two pollutants. A further study using the same cohort ~~(Jerrett et al.,~~
2478 ~~2009)~~(Jerrett et al., 2009) found unstable risk estimates for ozone and all-cause, cardiopulmonary
2479 and cardiovascular mortality with only respiratory mortality being associated with ozone after
2480 adjustment for PM_{2.5}. For a more detailed discussion of these and other recent cohort studies ~~(Smith~~
2481 ~~et al., 2009;Zanobetti and Schwartz, 2011;Lipsett et al., 2011)~~(Smith et al., 2009;Zanobetti and
2482 ~~Schwartz, 2011;Lipsett et al., 2011)~~ than space here permits, the reader is referred to the REVIHAAP
2483 report.

2484 In answering the second question regarding evidence for a threshold, the REVIHAAP report noted
2485 that epidemiological studies reporting an effect of long-term exposures to ozone on mortality do not,
2486 in general permit the firm identification of a threshold. However the report did draw some
2487 conclusions which are helpful to health impact studies.

2488 Earlier chamber exposure studies referred to above have been supplemented by more recent studies
2489 showing effects at lower levels. Studies since the 2005 global update have shown that healthy
2490 exercising human subjects showed impaired lung function at concentrations of 60ppb for 6.6 hours,
2491 relative to clean air controls ~~(Kim et al., 2011)~~(Kim et al., 2011). The report noted that the subjects
2492 were healthy adults and thus not representative of the general population, or of real-world
2493 combinations of susceptibility and exposure. The REVIHAAP report discusses a number of other
2494 studies which report adverse effects at concentrations lower than those in the above study and the
2495 reader is referred to that report for a more detailed discussion. The REVIHAAP report concluded that
2496 the evidence from epidemiological studies for a threshold for short-term exposures is inconsistent
2497 with some large multi-city studies which reported little evidence for a threshold down to near
2498 background ozone concentrations, whereas other short-term studies suggest a threshold between
2499 10ppb and 45 ppb daily maximum 1-hour ozone. The report concluded that in summary the evidence
2500 for a threshold for short-term exposure is inconsistent but where a threshold is observed it is likely to
2501 lie below 45 ppb maximum 1 hour ozone.

2502 The third question, relating to quantification of the health impacts of ozone overlaps with the HRAPIE
2503 project, the second part of the WHO review for the European Commission. The REVIHAAP report
2504 concluded that a range of health outcomes could be used where baseline rates are known, including
2505 all-age, all-cause, cardiovascular and respiratory mortality, and for ages 65 and over respiratory and
2506 cardiovascular hospital admissions. The report concluded that the evidence supports the use of all-
2507 year coefficients for daily maximum 8-hour average ozone concentrations. Bearing in mind the
2508 discussions on a threshold REVIHAAP recommended that cut-off points be used at 35ppb for daily
2509 maximum 8-hour ozone 'for consistency with previous work using SOMO35...' (for definition of
2510 SOMO see §5 or e.g. ~~(AQEG, 2009)~~(AQEG, 2009)). In an important further step, the report also
2511 recommended using a cut-off at 10ppb, i.e. SOMO10.

2512 This latter step is significant in that use of SOMO10 in assessing the effects of most future control
2513 strategies already in place or proposed in Europe (and probably elsewhere) are likely to lead to
2514 increases in health impacts from ozone exposures. The conclusions of the HRAPIE report essentially

2515 endorsed the findings of the REVIHAAP report, suggesting the use of SOMO10 and SOMO35, and an
2516 assessment of long-term exposure impacts as a sensitivity study.

2517 Interestingly, the Impact Assessment for the EU Clean Air package published on 18 December 2013
2518 (http://ec.europa.eu/environment/air/clean_air_policy.htm) only used SOMO35 to assess the health
2519 impact of proposed policies. This was justified on the grounds that below this level there are
2520 uncertainties in ozone modelling. However, it is convenient that using SOMO35 minimises the impact
2521 of increased ozone health impacts which are likely to result if SOMO10 is used. The policy process
2522 has yet to come to terms with the possibility that existing and planned control measures are likely to
2523 result in increased health impacts from ozone if a low threshold, or a long-term (annual or summer)
2524 average concentration is used in impact assessments. A recent modelling study (~~Heal et al.,~~
2525 ~~2013a~~)(~~Heal et al., 2013b~~) which looked at the burden of health impacts from ozone in 12 regions in
2526 the UK incorporated the uncertainty over thresholds using a population weighted daily maximum 8-
2527 hour mean ozone metric with a cut-off at 35 ppb and also with no threshold. They also incorporated
2528 future temperature increases and changes in the tropospheric baseline of ozone, and showed that as
2529 expected the assumption of zero threshold gave much larger health impacts. Moreover they showed
2530 that under a no-threshold assumption health impacts (deaths brought forward and hospital
2531 admissions) in the 'current legislation' scenario *increase* due both to the no-threshold assumption
2532 but also with the assumption of an increasing tropospheric ozone baseline. This interplay between
2533 the threshold assumption and the future trend in the tropospheric baseline has important
2534 implications for policy as discussed in section 5 below.

2535 [A not dissimilar process in followed the US context as part of the periodic review process for NAAQS](#)
2536 [\(National Ambient Air Quality Standards\). In 2013 the US EPA produced its most recent and](#)
2537 [substantial integrated science assessment for ozone \(http://www.epa.gov/ncea/isa/\).](http://www.epa.gov/ncea/isa/)

2538 Finally, an interesting new development bridging the gap between atmospheric chemistry and
2539 epidemiology is a time-series study of the associations between O_x (defined as O₃ + NO₂) and health
2540 outcomes (~~Williams et al., 2014~~)(~~Williams et al., 2014~~). As well as having a sound basis in atmospheric
2541 chemistry given the rapid interchange between ozone and NO₂, this also has a toxicological rationale
2542 since it is probable that both ozone and NO₂ cause harmful effects *via* oxidative stress (although it is
2543 recognised that NO₂ could also act *via* other mechanisms). The study showed that the associations
2544 between O_x and mortality were stronger than for either of the two pollutants individually, but similar
2545 to the pollutants when used in a two-pollutant model. The preliminary conclusion is that in
2546 epidemiological studies of either ozone or NO₂, some form of interaction should be incorporated,
2547 either as O_x or in a two-pollutant model. Single pollutant models for ozone or NO₂ should not be
2548 used.

2549

2550 3.2 Ecosystems

2551

2552 Ozone is phytotoxic. The primary ecosystem impact of ozone is *via* the stomatal uptake of ozone
2553 followed by reaction with the internal plant tissues generating highly reactive oxidants that interfere
2554 with physiological processes (see also §2.1.2) (~~Fowler et al., 2009;Matyssek et al., 2008;Matyssek et~~
2555 ~~al., 2010~~)(~~Fowler et al., 2009;Matyssek et al., 2008;Matyssek et al., 2010~~). ~~Ozone may also react with~~

2556 ~~leaf cuticles and other external plant surfaces as well as the volatile compounds emitted by~~
2557 ~~vegetation. Ozone may also react with leaf cuticles and other external plant surfaces as well as the~~
2558 ~~volatile compounds emitted by vegetation. Typical ozone effects on plants include reduced growth,~~
2559 ~~less seed production, lower functional leaf area and earlier leaf senescence. Data compilation~~
2560 ~~studies have shown that many species of plants are sensitive to ozone, including: agricultural crops~~
2561 ~~such as wheat, tomato, soybean and rice and salad crops such as lettuce, spinach and onion (Mills et~~
2562 ~~al., 2007a); grassland species such as clover species, buttercup and harebell (Hayes et al., 2007;Mills~~
2563 ~~et al., 2007b); and tree species such as beech, birch and Holm oak (Karlsson et al., 2007). These~~
2564 ~~effects impact on the important ecosystem services provided by plants, including food security,~~
2565 ~~carbon sequestration, timber production, and protection against soil erosion, avalanches and~~
2566 ~~flooding.~~

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2567

2568 Ozone has been recognised as a pollutant causing damage to vegetation since the 1960s and a
2569 research programme to quantify crop loss in North America was established during the 1970s
2570 ~~(Karnosky et al., 2007;Adams et al., 1982)(Karnosky et al., 2007;Adams et al., 1982). These early~~
2571 ~~studies showed that reductions in exposure to ozone of 40% would reduce the annual costs of ozone~~
2572 ~~damage by \$3 billion (at 1980 prices). These early studies used metrics for ozone exposure which~~
2573 ~~were simply the product of ozone mixing ratio and time above a threshold value (40ppb) and had~~
2574 ~~units of ppb hours. Global modelling results show that China, India and the United States are~~
2575 ~~currently by far the most affected countries, bearing more than half (AOT 40, see also section 5).~~
2576 ~~Since then, analysis of all global losses and threatened areas data from over 700 studies of field-based~~
2577 ~~ozone crop exposure of crops has indicated that many of our most globally important food crops~~
2578 ~~such as wheat, soybean, maize and rice are sensitive or moderately sensitive to ozone (Teixeira et al.,~~
2579 ~~2011)(Mills et al., 2007a). Crops such as rice have shown sensitivity to ozone. Research in Europe has~~
2580 ~~improved the methods of assessment and estimates of crop loss (Emberson et al., 2009). In the US~~
2581 ~~context, recent work by Ye and Unger has shown that O₃ damage decreases GPP (Gross Primary~~
2582 ~~Productivity) by 4–8% on average in the eastern US and leads to significant decreases of 11–17% in~~
2583 ~~east coast hot spots (Yue and Unger, 2014). Crops such as rice have shown sensitivity to ozone~~
2584 ~~(Ainsworth, 2008). Work in China has looked at the ozone exposure for rice regions, finding that~~
2585 ~~regions along the Yangtze and south China had the highest risks in 2000, and these risks will double~~
2586 ~~or triple by 2020 with estimated global crop production losses owing to ozone totalling 79-121~~
2587 ~~million metric tons, worth \$11-18 billion annually (USD2000) (Avnery et al., 2011a). Global~~
2588 ~~modelling results show that China, India and the United States are currently by far the most affected~~
2589 ~~countries, bearing more than half of all global losses and threatened areas from ozone crop exposure~~
2590 ~~(Teixeira et al., 2011;Sinha et al., 2015). In the US, recent work by Yue and Unger has shown that O₃~~
2591 ~~damage decreases GPP (Gross Primary Productivity) by 4–8% on average in the eastern US and leads~~
2592 ~~to significant decreases of 11–17% in east coast hot spots (Yue and Unger, 2014). Research in China~~
2593 ~~has looked at the ozone exposure for rice regions, finding that regions along the Yangtze and south~~
2594 ~~China regions had the highest risks in 2000, and these risks will double or triple by 2020 (Tang et al.,~~
2595 ~~2014)(Tang et al., 2014). It has been recently estimated that the nationally aggregated yield loss~~
2596 ~~owing to ozone in India is sufficient to feed about 94 million people living below the poverty line~~
2597 ~~(Ghude et al., 2014). Future moves to biofuel crops could enhance the formation of ozone with~~
2598 ~~concomitant negative health impacts (Ashworth et al., 2013;Hardacre et al., 2013).~~

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2599

2600 ~~Ozone risks for crops and pastures depend on exposure, leaf uptake and the plant's defence capacity~~
2601 ~~(Fuhrer, 2009; Kangasjarvi et al., 1994). The extent of ozone damage has been shown to be directly~~
2602 ~~proportional to the stomatal ozone flux for a range of crop plants in excess of a threshold value,~~
2603 ~~recognising that vegetation has an innate capacity to detoxify oxidants. In India, The total economic~~
2604 ~~cost losses for ozone on crop yields in the Indian states of Punjab and Haryana amounted to USD6.5~~
2605 ~~billion in the fiscal year 2012–2013 and USD3.7 billion in the fiscal year 2013–2014 (Sinha et al.,~~
2606 ~~2015). It has been recently estimated that the nationally aggregated yield loss owing to ozone in~~
2607 ~~India is sufficient to feed about 94 million people living below the poverty line (Ghude et al., 2014). In~~
2608 ~~Europe, wheat yield losses were estimated at 14% due to ozone in the year 2000 in EU27 (plus~~
2609 ~~Switzerland and Norway), equivalent to 3.2 billion Euros falling to 9% and 2 billion Euro in 2020 with~~
2610 ~~implementation of current legislation (Mills and Harmens, 2011). It has been argued that plant~~
2611 ~~breeding that incorporates O₃ sensitivity into selection strategies will be increasingly necessary to~~
2612 ~~achieve sustainable production with changing atmospheric composition (Fowler et al.,~~
2613 ~~2009)(Wilkinson et al., 2012; Booker et al., 2009). Recent analyses have shown a crop loss of 14% due~~
2614 ~~to ozone in the year 2000, equivalent to 3.2 billion Euros. Future moves to biofuel crops could~~
2615 ~~enhance the formation of ozone with concomitant negative health impacts (Mills et al.,~~
2616 ~~2011)(Hardacre et al., 2013; Ashworth et al., 2013). The same authors show that semi-natural~~
2617 ~~vegetation has a similar range of sensitivity to ozone as crop plants and is a cause of decreased~~
2618 ~~productivity of European forests. It has been argued that plant breeding that incorporates O₃~~
2619 ~~sensitivity into selection strategies will be increasingly necessary to achieve sustainable production~~
2620 ~~with changing atmospheric composition.~~

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2621
2622 ~~Critical levels for effects of ozone on vegetation have been set and revised at a series of LRTAP~~
2623 ~~Convention workshops from 1988 to date. Initial critical levels were based on mean concentrations,~~
2624 ~~followed by AOT40-based values reflecting appreciation of the cumulative nature of effects and most~~
2625 ~~recently critical levels have been derived based on stomatal fluxes. The latter take into account the~~
2626 ~~modifying effects of climate (temperature, vapour pressure deficit, light), soil moisture, ozone and~~
2627 ~~plant growth stage (phenology) on the amount of ozone absorbed by leaves (discussed in Section~~
2628 ~~2.1.2a). Calculated using the DO₃SE model developed by Emberson et al. (Booker et al.,~~
2629 ~~2009)(Emberson et al., 2000).~~

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2630
2631 ~~The reduced carbon assimilation due, the stomatal ozone flux method thus allows the dynamics of~~
2632 ~~stomatal opening and therefore ozone uptake to be modelled throughout the growing season. For~~
2633 ~~example, using the DO₃SE model parameterised for a specific species, the substantially higher~~
2634 ~~stomatal uptake of 80 ppb of ozone under warm humid conditions with adequate soil moisture than~~
2635 ~~at higher temperatures with lower humidity and drier soils would be captured. The flux~~
2636 ~~methodology also incorporates the natural ozone detoxification capacity of plants by accumulating~~
2637 ~~the Phytotoxic Ozone Dose of ozone above an ozone threshold flux of $Y \text{ nmol m}^{-2} \text{ s}^{-1}$ (POD_Y, units~~
2638 ~~nmol m^{-2} projected leaf area). The value for "Y" varies between species, with the highest values~~
2639 ~~being for crops such as wheat and potato and the lowest for some tree species such as beech and~~
2640 ~~birch. The LRTAP Convention's Modelling and Mapping Manual now contains flux-based critical~~
2641 ~~levels for ozone effects on food security (wheat, potato, tomato yield), carbon sequestration and~~
2642 ~~timber production (Norway spruce, beech and birch) and biodiversity in conservation-grade~~
2643 ~~grasslands (Mills et al., 2011b; Mills, 2014).~~

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[The use of the flux-based methodology is supported by recent analyses indicating that field evidence of ozone effects in Europe fits more closely with areas of greatest risk when flux-based rather than AOT40-based maps are produced. Furthermore, Fares et al. \(Fares et al., 2013a\) showed a strong correlation between measured and modelled fluxes in a mixed pine and oak Mediterranean forest and epidemiological studies conducted in Switzerland provided supporting evidence for both critical levels for deciduous trees and the DO₃SE parameterisation.](#)

[Reduced carbon assimilation owing to ozone by forests globally has been estimated by Sitch et al \(Sitch et al., 2007\)\(Sitch et al., 2007\) to represent a substantial contribution to the indirect radiative forcing of climate by ozone \(see Figure 2829\). Through these effects on the productivity of ecosystems and the potential effects on biodiversity, ozone has become the most important pollutant threat to terrestrial ecosystems globally and is likely to remain so through most of the current century. Witting et al \(2009\) have shown that the carbon-sink strength of northern hemisphere forests is likely reduced by current ozone and will be further reduced in future if ozone rises therefore implying that a key carbon sink currently offsetting a significant portion of global fossil fuel CO₂ emissions could be diminished or lost in the future \(Wittig et al., 2009\)\(Wittig et al., 2009\).](#)

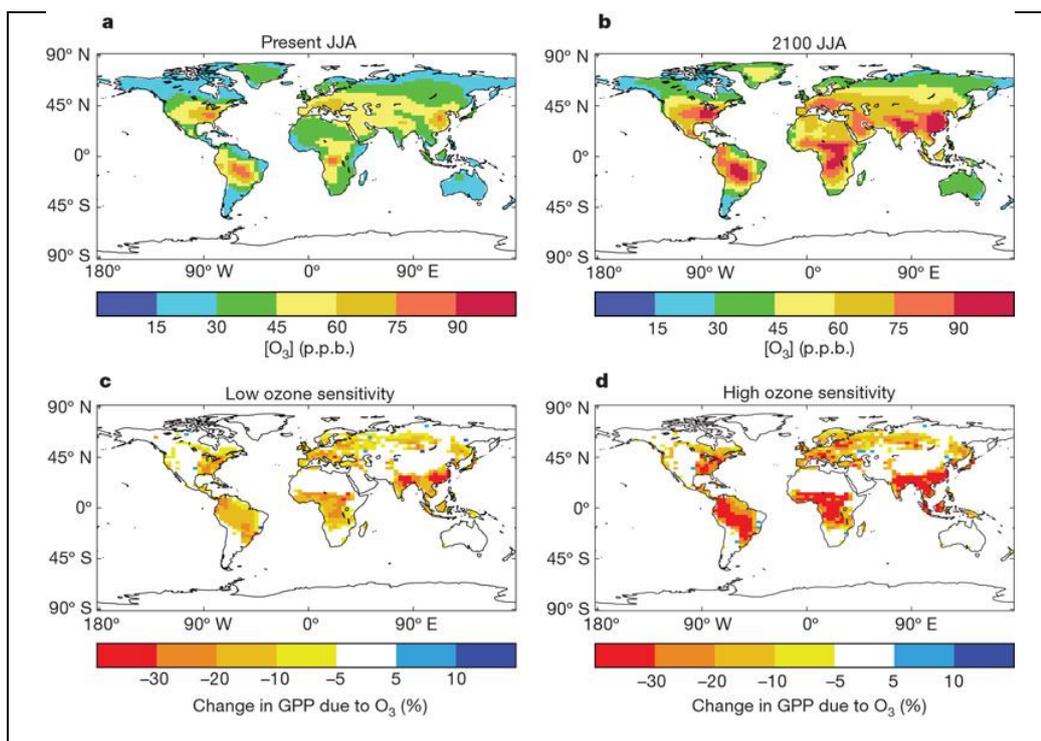


Figure 2829 - a,b - Modelled diurnal (24-h) mean surface [O₃] in pbb averaged over June, July and August (JJA) for the present day (a) and the year 2100 under the highly polluted SRES A2 emissions scenario (b). **c, d**, Simulated percentage change in gross primary productivity (GPP) between 1901 and 2100 due to O₃ effects at fixed pre-industrial atmospheric [CO₂] for 'low' (c) and 'high' (d) ozone

plant sensitivity (Sitch et al., 2007)(Sitch et al., 2007).

2662

2663 | In a recent review, Fuhrer looked at the ozone risk for crops and pastures in respect of future climate
2664 | and concluded that the facets of exposure, leaf uptake and the plant's defence capacity are likely to
2665 | change with a combination of changing anthropogenic ozone precursor emissions and climatic
2666 | conditions. For example, in areas where the frequency of hot/dry periods is projected to increase
2667 | ozone episodes may become more frequent and cumulative exposures will grow, but leaf uptake of
2668 | ozone will decline. In contrast, in more remote rural areas with rapid warming and less drying, ozone
2669 | risks are likely to grow because of increasing hemispheric transport of pollution leading to peak
2670 | ozone levels at times when plant sensitivity is high (Fuhrer, 2009)(Fuhrer, 2009). Tai et al (Tai et al.,
2671 | 2014)(Tai et al., 2014) have shown that warming reduces global crop production by >20% by 2050.
2672 | In the range of the scenarios used ozone changes either exacerbate or offset a substantial part of the
2673 | climate impacts. The authors note that given the competing sensitivities to heat or ozone that it is
2674 | possible to measure the relative benefits for climate adaptation versus air pollution regulation for
2675 | food security- (see also (Avnery et al., 2011b)). It is of further concern that new evidence suggests
2676 | that ozone can reduce the sensitivity of plants to drought by interfering with stomatal control
2677 | mechanisms (Wilkinson and Davies, 2010;Wagg et al., 2012), thereby exacerbating effects of extreme
2678 | weather events.

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3.3 Impact of Ozone on Climate

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2682 | Tropospheric ozone interacts with both solar (short-wave, SW) and terrestrial (long-wave, LW)
2683 | radiation – consequently changes in its distribution can generate radiative forcings (RF) and lead to
2684 | changes in climate. In their Fifth Assessment Report of climate change, IPCC found that changes in
2685 | tropospheric ozone between 1750 and 2010 had generated a global mean radiative forcing of +0.40
2686 | (90% confidence range: 0.20 to 0.60) W m⁻² (Myhre et al., 2013)(Myhre et al., 2013). This was largely
2687 | based on the ACCMIP assessment which considered changes in ozone (1850-2000) simulated by 17
2688 | different global models, and calculations from three different radiation schemes (Stevenson et al.,
2689 | 2013)(Stevenson et al., 2013). The ACCMIP results were augmented by simulations back to 1750 and
2690 | up to 2010 by the OsloCTM2 model (Skeie et al., 2011)(Skeie et al., 2011), and were broadly
2691 | consistent with earlier assessments from the literature.

(c) MMM 2000s-1850s O₃T RF (355) mWm⁻²

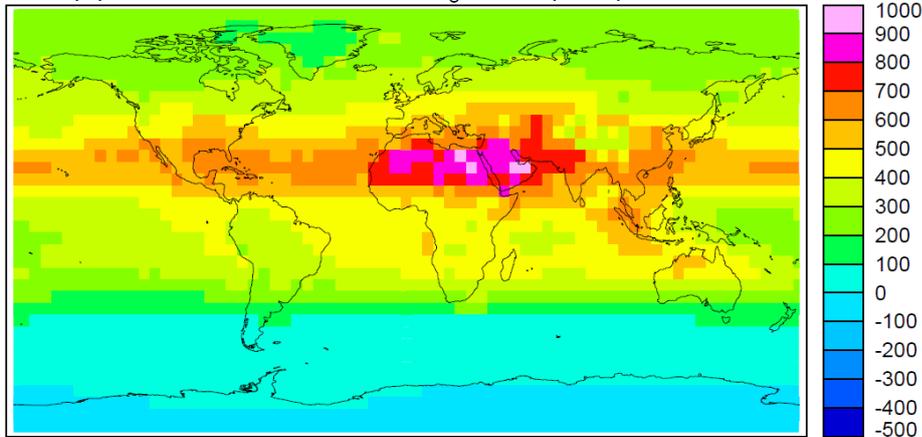


Figure 2930 - The global distribution of (annual mean) tropospheric ozone (O₃T) radiative forcing (1850-2000, net SW+LW, including stratospheric adjustment, in mW m⁻²), based on results from the multi-model mean (MMM) of the ACCMIP models, using the Edwards-Slingo radiation scheme (Stevenson et al., 2013)(Stevenson et al., 2013). (NB the value of 0.355 W m⁻² shown here compares to the recommended IPCC value for the 1750-2010 O₃ RF of 0.40 W m⁻²; the IPCC value includes the additional time periods 1750-1850 and 2000-2010, and also incorporates calculations with additional radiation schemes, and from other studies.)

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2693 An important feature of the O₃ RF is that, in comparison to the RFs from the well-mixed greenhouse
2694 gases, it is more spatially variable (see Figure 2930). The largest changes in ozone since 1750 have
2695 occurred over the industrial regions that are the main sources of ozone's precursors (NO_x, CO, and
2696 VOCs) (see also section 2.3). The industrial emission focus is tempered somewhat by ozone's other
2697 key precursor, CH₄, which is relatively well-mixed due to its decadal lifetime, and also by ozone's
2698 lifetime of a few weeks, which allows transport to reduce zonal heterogeneities. In addition, to the
2699 variations in ozone changes, the spatial distribution of the O₃ RF also depends on other factors. The
2700 LW RF is largest where the temperature difference between the surface and the tropopause reaches
2701 a maximum: that is over land in the tropics and sub-tropics. The SW RF is largest over more reflective
2702 surfaces e.g., snow/ice and desert. The presence of clouds reduces the LW RF, and also modulates
2703 the SW RF. This combination of contributing factors leads to the net (LW+SW) O₃ RF peaking over the
2704 southern margins of the northern mid-latitudes and sub-tropics over land, and in particular over N.
2705 Africa and the Middle East (Figure 2930).

2706 Shindell et al (Shindell et al., 2009;Shindell et al., 2005)(Shindell et al., 2009;Shindell et al., 2005) and
2707 Stevenson et al (2013) have attributed the O₃ RF to O₃ precursor emissions (CH₄, NO_x, CO and VOCs).
2708 Results from these studies are reported in the 'emissions-based' RF summary diagram of the IPCC-
2709 AR5-WGI Summary for Policymakers (IPCC, 2013)(IPCC, 2013). Stevenson et al (2013) found that
2710 increases in anthropogenic emissions of CH₄, NO_x, CO and NMVOCs are respectively responsible for
2711 44±12% (±1 standard deviation range, based on results from 6 different global models), 31±9%,

2712 15±3% and 9±2% of the 1850s-2000s ozone RF. All these O₃ precursors also affect the abundance of
2713 atmospheric OH, and hence the lifetime of longer-lived greenhouse gases such as CH₄.
2714 Consequently, emissions of ozone precursors generate further impacts on climate in addition to
2715 those related to changes in ozone; often these additional climate impacts occur over different
2716 timescales, and the net integrated impact on climate of emissions of a particular ozone precursor are
2717 complex to diagnose e.g. (Fuglestedt et al., 2010)(Fuglestedt et al., 2010). A further complication is
2718 that net impact on atmospheric composition, and hence climate, depends upon the location of ozone
2719 precursor emissions, both in the horizontal e.g., (Fry et al., 2012)(Fry et al., 2012) and vertical e.g.
2720 (Stevenson and Derwent, 2009)(Stevenson and Derwent, 2009).

2721 A major source of uncertainty in the O₃ RF is the pre-industrial distribution of ozone (see Section 2.2).
2722 The few measurements that do exist are mainly thought to be unreliable, but the two more rigorous
2723 datasets, from Montsouris in Paris and Pic du Midi, a mountain top site in central France (Marenco et
2724 al., 1994)(Marenco et al., 1994), suggest that current generation models significantly overestimate
2725 late 19th century ozone levels (Cooper et al., 2014)(Cooper et al., 2014). Cooper et al. (2014) highlight
2726 that global models significantly underestimate recent trends in surface ozone, suggesting that they
2727 overestimate past ozone levels. Consequently, there are suggestions that the ozone RF may be
2728 substantially underestimated (also see, e.g., (Mickley et al., 2001)(Mickley et al., 2001)). It is unclear
2729 how these apparent discrepancies in surface ozone translate into uncertainties in O₃ RF, which is
2730 more strongly related to changes in the upper troposphere. The error estimate quoted by IPCC
2731 makes only a cursory attempt to include this source of uncertainty. Other sources of uncertainty are
2732 estimated from the model spread in the 1850-2000 change in ozone, and differences between
2733 radiation schemes. Nevertheless, the uncertainties associated with O₃ RF are far smaller than those
2734 due to, e.g., aerosol, because the key source of uncertainty is the O₃ change rather than our
2735 understanding of the fundamental processes involved. For example, the radiative forcing from
2736 aerosols has a larger uncertainty because we are less sure of the changes in aerosol since the pre-
2737 industrial, both in terms of their magnitude and geographical distribution, but also because the
2738 aerosol forcing originates from changes in multiple different aerosol types, including mixtures of
2739 different aerosol species, with highly uncertain optical properties (Fuzzi et al., 2015).

2740 The spatial structure of the climate response to a particular radiative forcing is not directly related to
2741 the spatial distribution of the RF, but the climate response is typically spread out over the same
2742 latitudes e.g. (Shindell et al., 2010)(Shindell et al., 2010).

2743 The efficacy, or relative effectiveness of an O₃ RF relative to a CO₂ RF in generating a global mean
2744 surface temperature change, has been estimated by Hansen et al. (Hansen et al., 2005)(Hansen et al.,
2745 2005) and Stuber et al. (Stuber et al., 2005)(Stuber et al., 2005). For stratospherically adjusted RFs,
2746 Stuber et al (2005) found that the tropospheric O₃ efficacy has values in the range 0.7-1.7, with O₃
2747 changes in the lower troposphere showing higher efficacies than the upper troposphere, and O₃
2748 changes in the extra-tropical N. Hemisphere showing higher efficacies than globally uniform changes.
2749 The spatial dependence of the efficacy appears to relate to the interactions with climate feedback
2750 mechanisms e.g., (Shindell, 2014)(Shindell, 2014).

2751 It is quite arbitrary (and often scientifically dubious) to consider RFs owing to ozone changes
2752 in the troposphere and stratosphere separately, not least because the tropopause is often difficult to
2753 locate (e.g., (Prather et al., 2011)(Prather et al., 2011)), and because coherent ozone changes often

2754 | straddle the tropopause e.g. ~~(Gauss et al., 2006; Shindell et al., 2013)~~(Gauss et al., 2006; Shindell et al.,
2755 | [2013](#)). Changes in tropopause height and morphology associated with climate change (e.g., ~~(Wilcox~~
2756 | ~~et al., 2012)~~([Wilcox et al., 2012](#)) suggest that there may be very important chemistry climate
2757 | ~~feedbacks related to ozone, such as those related to changes in the Brewer-Dobson Circulation (e.g.~~
2758 | ~~(Hegglin and Shepherd, 2009))~~.

2759 |

2760 **4** ~~Topics~~
2761) suggest that there may be very important chemistry-climate feedbacks related to ozone, such as
2762 those related to changes in the Brewer-Dobson Circulation (e.g. (Hegglin and Shepherd,
2763 2009;Nowack et al., 2015)).
2764

2765 4 Recent Advances

2766

2767 The topics in this section have been selected to represent areas of active research highlighting recent
2768 advances associated with key sources or with particular chemical environments. The diversity of
2769 topics reflect the improved understanding of the richer and more diverse range of interactions
2770 between atmospheric ozone, other parts of the Earth system and human activity.

2771 4.1 Megacities

2772

2773 As of 2007 more than half of the world's population now lives in urban areas, and many of them in
2774 megacities- (Zhu et al., 2012). This statistic is often cited, but what defines an 'urban' area or a
2775 'megacity'? While these definitions evolve and differ often depending on the context, here we will
2776 consider a common definition of a megacity that is a city or urban agglomeration of greater than 10
2777 million people- (Molina and Molina, 2004). As per Parrish et al. (2011), megacities are dense centres
2778 of population, economic activity, and pollutant emissions, but also areas where effective pollution
2779 control strategies could realize maximum benefit (Parrish et al., 2011)(Parrish et al., 2011). Ground-
2780 level ozone is a serious air quality issue in many of the world's megacities. Monitoring and
2781 measurement campaigns have documented ozone levels exceeding air quality standards in many
2782 megacities. For example, based on the UNECE and WHO guidelines for protection of human health,
2783 daily 8-hr ozone should not exceed 60 ppb on more than 25 days per calendar year. In Delhi, India
2784 this threshold was exceeded approximately 45 days per year on average during the 7 year period
2785 (1997-2004), a significant concern for human health in the megacity (Ghude et al., 2008)(Ghude et
2786 al., 2008), especially since ozone concentrations there are still on the rise (Chelani, 2012)(Chelani,
2787 2012). While some cities have extensive monitoring of ozone (and ozone precursors, more often NOx
2788 than NOx and NMVOCs), others have limited to no measurements. Consider South America. Buenos
2789 Aires, Argentina for example, has very few ozone measurements, so that the overall situation with
2790 regard to ozone pollution cannot be thoroughly assessed for the city (Zhu et al., 2012)(Zhu et al.,
2791 2012). Sao Paulo, Brazil on the other hand has a more extensive monitoring network and increasing
2792 ozone concentrations are have been observed over the last decade, despite decreases in other
2793 pollutants such as NOx and CO, as well as regular violations of the national ozone air quality standard
2794 (Sanchez-Ccoyllo et al., 2006;Zhu et al., 2012)(Sanchez-Ccoyllo et al., 2006;Zhu et al., 2012). A variety
2795 of reported ozone concentrations from megacities are shown in Table 2.

2796 Sources of ozone precursors in megacities include, but are not limited to, traffic-related emissions,
2797 combustion of fossil fuels, solvent use, domestic biofuel usage and industrial activities such as power
2798 generation. The relative contribution of sources and specific activities leading to the local ozone
2799 precursor compound mix varies depending on location, including between cities. For example, in Sao
2800 Paulo, Brazil light-duty vehicles are run on ethanol or gasoline/ethanol mix fuels and ethanol from
2801 sugar cane accounts for 65% of all fuel consumed (de Fatima et al., 2012)(de Fatima et al., 2012);
2802 light-duty vehicles in North America predominantly run on gasoline which in some cases contains a
2803 small fraction of ethanol; in China gasoline-powered vehicles dominate the vehicle fleet, but there is
2804 also significant use of liquefied petroleum gas (LPG) for some types of vehicles in the fleet (Liu et al.,
2805 2008)(Liu et al., 2008). Overall, the emissions from vehicles/traffic dominate as a major (if not the
2806 main) source of ozone precursors in many megacities, especially because growing vehicle fleets

2807 | generally accompany megacity development (~~Parrish and Zhu, 2009~~)(Parrish and Zhu, 2009). For
2808 | example, Shanghai, the largest city in China with a population of roughly 23 million, has undergone
2809 | rapid development; between 1996 and 2008 industrial gross output increased by ≈ 4 times and the
2810 | number of cars increased from 0.47 to 2.61 million (~~Tie et al., 2013~~)(Tie et al., 2013). Other
2811 | megacities in which vehicles are cited as the top or among the top sources of ozone precursors are
2812 | Cairo, Delhi, Istanbul, Los Angeles, New York, Mexico City, Sao Paulo, and the Ruhr-Rhein
2813 | metropolitan region (~~Chelani, 2012; de Fatima et al., 2012; Parrish et al., 2011; Khoder, 2009; Im and~~
2814 | ~~Kanakidou, 2012; Melkonyan and Kuttler, 2012~~).(Chelani, 2012; de Fatima et al., 2012; Parrish et al.,
2815 | 2011; Khoder, 2009; Im and Kanakidou, 2012; Melkonyan and Kuttler, 2012; Bon et al., 2011). In cities
2816 | like Lagos in Nigeria it is a combination of traffic and open biomass burning driving large ozone
2817 | exceedances from emission of NMVOCs (~~Marais et al., 2014~~)(Marais et al., 2014).

2818 | Extreme events in terms of air quality have also been documented in a number of megacities. These
2819 | result from a combination of elevated emissions and meteorological conditions (e.g., stagnant, hot,
2820 | sunny) that lead to extremely high concentrations of ozone, typically for a short duration. During the
2821 | summer of 2010 in Moscow, blocking anticyclonic meteorological conditions with record-breaking
2822 | high temperatures led to larger than normal evaporative emissions from fuels, paints, etc, as well as
2823 | emissions from a large number of forest and turf fires, in addition to the 'normal' megacity
2824 | emissions- (~~Zvyagintsev et al., 2011; Elansky et al., 2011~~). These conditions which persisted from July
2825 | through mid-August resulted in maximum hourly mean concentrations of ozone that peaked at 134.2
2826 | ppbv (~~Elansky et al., 2011~~)(Elansky et al., 2011). An oft cited example of extreme ozone was during
2827 | the European heat wave during summer 2003, which adversely affect the residents of a number of
2828 | European megacities. For about 2 weeks at the beginning of August a high pressure system was
2829 | centred on Western Europe, resulting in record-setting temperatures exceeding 40° C, and
2830 | accompanied by a persistent photochemical episode. In France, the highest hourly ozone value
2831 | reached $417 \mu\text{g m}^{-3}$; 208.5 ppbV and concentrations regularly exceeded the EU threshold of $180 \mu\text{g m}^{-3}$
2832 | 90 ppbV (~~Lee et al., 2006; Vautard et al., 2005; Stedman, 2004; Tressol et al., 2008~~)(Lee et al.,
2833 | 2006; Vautard et al., 2005; Stedman, 2004; Tressol et al., 2008).

2834 | Because of their high population density, the adverse health effects of elevated ozone levels affect a
2835 | disproportionately large fraction of the population. A recent study investigated the health impacts of
2836 | air pollutants in Tehran, Iran where the annual 8 hr average ozone concentration in 2010 was 68.84
2837 | $\mu\text{g m}^{-3}$; 34 ppbV , with a maximum 8-hr h concentration of $187 \mu\text{g m}^{-3}$; 93.5 ppbV during the summer
2838 | period. The health impact owing to ozone was estimated to cause an excess total mortality of 819
2839 | out of 47,284 people in a year, relative to 2,194 cases for PM_{10} (~~Naddafi et al., 2012~~)(Naddafi et al.,
2840 | 2012). For comparison (from a study using the same method), they cited the excess total mortality
2841 | for PM_{10} was 677 out of 1,308,000 people for Milan, Italy, located in the megacity region of the Po
2842 | Valley (~~Naddafi et al., 2012~~)(Naddafi et al., 2012). Cifuentes, et al. (~~Cifuentes et al., 2001~~)(Cifuentes et
2843 | al., 2001) assessed the health benefits associated with reductions of PM and O_3 from climate change
2844 | mitigation strategies (2000-2020) in 4 megacities – Santiago, Sao Paulo, Mexico City, and New York.
2845 | Without going into detail, substantial premature deaths could be avoided from such policies; in all
2846 | cases the adverse health effects avoided from ozone were significantly less than those from PM
2847 | (~~Cifuentes et al., 2001~~)(Cifuentes et al., 2001). Generally, small but substantial associations between
2848 | ozone and total mortality are found in epidemiological studies (~~Bates, 2005~~)(Bates, 2005).

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2849 The emissions from megacities also have an effect on the surrounding region. In many cases the
2850 urban areas have been identified to be VOC-limited, where high NO_x concentrations in the cities
2851 suppress ozone concentrations (~~Im and Kanakidou, 2012;Tie et al., 2013~~)(~~Im and Kanakidou, 2012;Tie~~
2852 ~~et al., 2013~~). For example, a modeling study conducted in the context of the MILAGRO measurement
2853 campaign in Mexico City identified that reductions in VOC emissions led to decreases in maximum
2854 ozone concentrations while NO_x emission reductions led to increased maximum ozone
2855 concentrations, demonstrating that the urban core of the Mexico City metropolitan area was VOC-
2856 limited, a conclusion supported by the *in-situ* measurements. The surrounding mountain/rural areas
2857 were identified as mostly NO_x-limited, although the range of these areas was meteorologically
2858 dependent (Song et al., 2010). This suppression of ozone by high NO_x has been identified as
2859 generally more characteristic of extratropical megacities in the northern hemisphere, relative to
2860 tropical cities, because of differences in transport patterns. Tropical cities are more influenced by
2861 rapid convective transport, while the extratropical cities (~~despite the Mexico City example~~) allow for
2862 a greater build-up of local pollution (~~Butler and Lawrence, 2009~~)(~~Butler and Lawrence, 2009~~).
2863 Local/regional transport of these air masses into the surrounding areas can lead to greater
2864 concentrations outside of the megacities. For example, in Tokyo, modelling studies showed that sea
2865 breezes developed during the daytime transported emissions from the urban centre to the north,
2866 which caused enhanced ozone in downwind areas 50-100 km away (~~Kondo et al., 2010~~)(~~Kondo et al.,~~
2867 ~~2010~~). Furthermore, regional enhancements of ozone during summer were found in the area (200
2868 km x 200 km) surrounding the Tokyo metro area (~~Kondo et al., 2010~~)(~~Kondo et al., 2010~~). A modelling
2869 study looking at the impacts of Istanbul (and Athens) on air quality in the East Mediterranean also
2870 found much lower concentrations of ozone within the (mega)cities owing to significant NO_x
2871 emissions depressing O₃. The rural sites in the surrounding area had much higher ozone
2872 concentrations, 11-24 ppbv (9-14 ppbv) greater in summer (winter) than in the urban areas,
2873 emphasizing the importance of the megacity emissions on regional air quality (~~Im and Kanakidou,~~
2874 ~~2012~~)(~~Im and Kanakidou, 2012~~). Further sources of emissions in areas surrounding the megacity
2875 regions also play a role in ozone formation. For example, the photochemistry in plumes transported
2876 out of London or the Ruhr-Rheine region will be influenced significantly by further emissions sources,
2877 while this is much less the case for e.g., Cairo which has far fewer emissions sources in the
2878 surrounding area (~~Hodnebrog et al., 2011~~)(~~Hodnebrog et al., 2011~~). Biogenic emissions will also
2879 influence ozone formation downwind of cities. For the Shanghai region, results from a model study
2880 suggested that ozone formation was enhanced in the city plumes in the downwind region because of
2881 the strongly VOC-limited conditions in the plume which resulted in continuous ozone production
2882 during transport. Reasons for this were oxidation of OVOCs and CO in the plume, and biogenic
2883 isoprene emissions (~~Tie et al., 2013~~)(~~Tie et al., 2013~~). Finally, an estimate of the footprint of
2884 southeastern megacities in South America found contributions in excess of 30% to downwind levels
2885 of surface ozone under certain conditions (~~Zhu et al., 2012~~)(~~Zhu et al., 2012~~).

2886 Considering ozone concentrations as a relative contribution to the global scale, megacities contribute
2887 a ~~surprisingly~~ small amount to global ozone (~~Stock et al., 2013~~)(~~Stock et al., 2013~~). In a model
2888 'annihilation' experiment where the emissions from grid cells containing megacities were removed,
2889 emissions from megacities contributed only 0.84% to the global average tropospheric ozone column
2890 density, proportionally smaller than the 6% of global anthropogenic ozone precursor emissions from
2891 megacities (~~Butler et al., 2012~~)(~~Butler et al., 2012~~). This however does not represent human health
2892 exposure to ozone in megacities and is also a global average.

2893 Previous work has shown that individual megacities are not well represented by global emission
 2894 inventories compared to the detailed city-scale inventories (Butler et al., 2012)(Butler et al., 2012),
 2895 which can be an issue when trying to model ozone on a larger scale. Additional complexity is added
 2896 by the non-linearity of the atmospheric chemistry involved in ozone production processes.
 2897 Hodnebrog et al. investigated the importance of resolution for the representation of large scale
 2898 tropospheric ozone from megacity emissions and found that higher resolution was much more
 2899 important for local air pollution studies than for larger climate studies, as might be expected, due to
 2900 the integration of relatively small changes over a large volume (changes in the resolution of
 2901 meteorology were not considered) (Hodnebrog et al., 2011)(Hodnebrog et al., 2011). A key point was
 2902 whether or not the model resolution could capture the NOx saturated regime within the plume
 2903 common to many megacities (Hodnebrog et al., 2011)(Hodnebrog et al., 2011). That said, recent
 2904 work by Siour, et al. has shown that for chemistry transport models, using a 'zooming' technique
 2905 (introducing higher resolution 'zooms' to certain areas within a coarser overall resolution) allows for
 2906 better representation of scale interactions, including better representation of megacity areas, such
 2907 as the BeNeLux region (Siour et al., 2013)(Siour et al., 2013). With the growing interest in the study of
 2908 urbanization, more research and improved methods are still needed to develop a better
 2909 understanding of pollutants such as ozone in megacities and their effects on all scales.

2910 **Table 2** - Reported mean O₃ for megacities.

Location	Mean O ₃ (ppbv)*	Statistic/year	Ref
Beijing, China	31	6 site average, 2001-2006, JJAS	(Tang et al., 2009)(Tang et al., 2009)
Bangkok, Thailand	17	Annual average, 1-hr, 1995-2008	(Zhu et al., 2012)(Zhu et al., 2012)
Cairo, Egypt	46	Dec 2004-Nov 2005	(Khoder, 2009)(Khoder, 2009)
Delhi, India	24	1997-2004, annual avg	(Ghude et al., 2008)(Ghude et al., 2008)
London, UK	19 (38 µg m ⁻³)	Annual average hourly data; suburban site (Eltham) 2009	http://uk-air.defra.gov.uk
Los Angeles, CA, USA	122	2005-2007, national 8-hr ozone design values (daily 8-hr max from running avgs)	(Cox et al., 2009)(Cox et al., 2009) (accessed 8.8.13)
Mexico City, Mexico	93	City plume, avg from flight measurements, MIRAGE-MEX campaign	(Tie et al., 2009)(Tie et al., 2009)
Tehran, Iran	34 (68.84 µg m ⁻³)	2010, annual 8-hr avg	(Naddafi et al., 2012)(Naddafi et al., 2012)

2911 *concentrations originally reported in µg m⁻³ were converted to ppbv using the simple conversion of
 2912 1 ppb = 2.0 µg m⁻³, assuming 1 atm pressure and 25° C.

2913

4.2 Biomass burning

2914
2915

2916 Wildfires/Biomass burning are the second largest source of carbon dioxide emissions, after fossil fuel
2917 combustion with ca. 1-2 GtC/year (~~(IPCC, 2013)~~(IPCC, 2013). Tropical biomass burning dominates total
2918 emissions with boreal fires (50-70°N) accounting for around 9% of all fire carbon emissions. Burning
2919 is thus a major component of the carbon cycle having a direct global impact on climate (~~(van der Werf~~
2920 ~~et al., 2010)~~(van der Werf et al., 2010). Further, there is also an indirect impact because biomass
2921 burning is a significant source of ozone precursors (e.g. CO, NO_x, VOCs) and aerosol particles. Indeed,
2922 Jaffe and Wigder (~~(Jaffe and Wigder, 2012)~~(Jaffe and Wigder, 2012) estimated that global wildfires
2923 produce approximately 170 Tg of ozone per year, which is 3.5% of all tropospheric ozone production.
2924 However, as they pointed out, many areas of uncertainty remain in wildfire-related ozone
2925 production. These uncertainties centre around the interplay of fire emissions, chemical reactions
2926 within the plumes, aerosols effects on chemistry and radiation and atmospheric dynamics (local and
2927 downwind meteorological patterns). These factors, along with a strong interannual variability of the
2928 fire activity itself, make the assessment of global impact a challenge. The impact of boreal forest fires
2929 is less documented and quantified than tropical biomass burning.

2930

2931 Recently, the atmospheric chemistry challenge of biomass burning in ~~Bora~~Boreal regions provided
2932 the motivation for the Quantifying the impact of Boreal forest fires on Tropospheric oxidants over
2933 the Atlantic using Aircraft and Satellites (BORTAS) campaign, held in July-August 2010 and 2011 over
2934 Eastern Canada and Western Atlantic. The objective of BORTAS ~~where~~was to better understand the
2935 chemical evolution of plumes emitted from the burning of biomass, with a particular emphasis on the
2936 net production of tropospheric ozone and downwind impacts on air quality (~~(Palmer et al.,~~
2937 ~~2013)~~(Palmer et al., 2013) and references therein)(see Figure 31). Parrington et al (~~(Parrington et al.,~~
2938 ~~2013)~~(Parrington et al., 2013) have shown that ozone formation increases with plume age. They used
2939 NMHC ratios (extensively described for this BORTAS campaign in ~~(Lewis et al., 2013)~~(Lewis et al.,
2940 2013)) to estimate photochemical ages of the observed biomass burning plumes between 0 and 10
2941 days. Parrington et al., (~~(Parrington et al., 2013)~~(Parrington et al., 2013) found that Ozone Production
2942 Efficiency (OPE) increased with plume ages as calculated by NMHC ratios. They show $\Delta\text{O}_3/\Delta\text{CO}$
2943 enhancement ratios increasing from 0.020 ± 0.008 ppbv/ppbv in plumes with photochemical ages less
2944 than 2 days to 0.55 ± 0.29 ppbv/ ppbv in plumes with photochemical ages greater than 5 days.
2945 According to this study, it seems that the aerosol loading is one of the main ~~driver~~drivers of such
2946 OPE, as aged plumes were typically associated with low aerosol loading and relatively high OPE. In
2947 one of the BORTAS flights that made multiple interceptions of a plume with high aerosol loading,
2948 Parrington et al. (2013) report that the photolysis rate of NO₂ was reduced by a factor of
2949 approximately 2 within this plume, suggesting that increased optical depth of the plume can reduce
2950 the photolysis rate of NO₂ to form ozone.

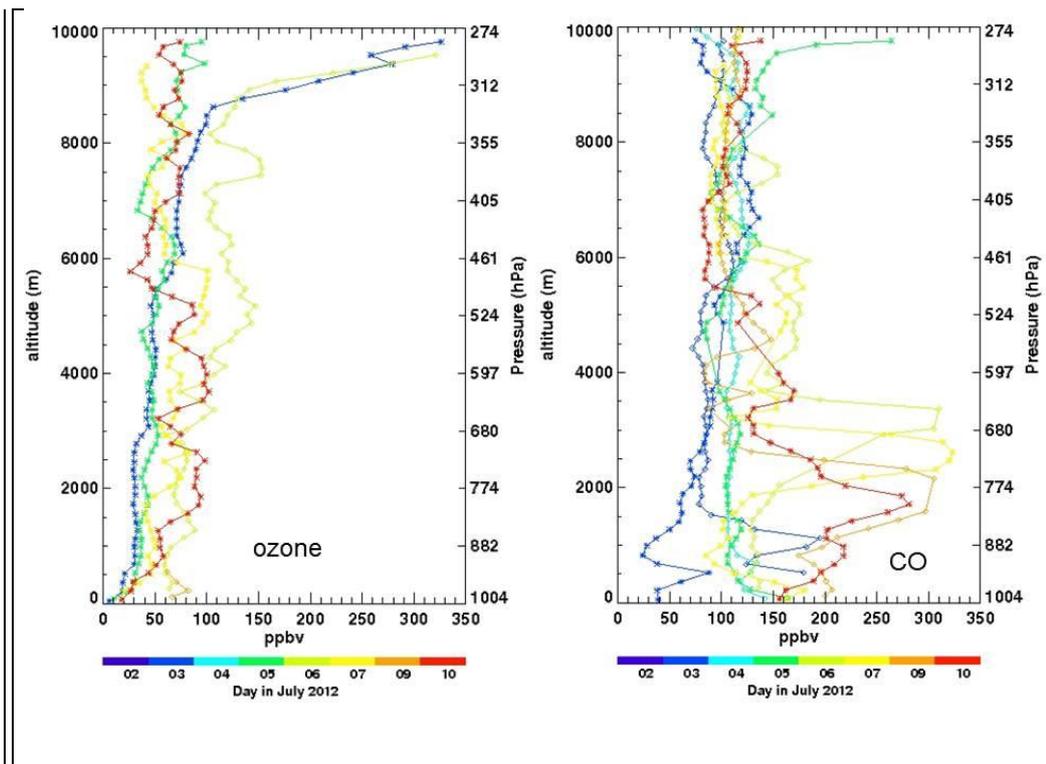
2951

2952 Still in the frame of BORTAS and WMO-GAW programme, results exploring NMVOC ozone precursors
2953 from measurements of biomass-burning (~~(Lewis et al., 2013)~~(Lewis et al., 2013) have suggested that
2954 biomass burning can be the largest fractional contributor to observed benzene, toluene, ethene and
2955 propene levels in many global locations. The extrapolated widespread biomass burning contribution
2956 to atmospheric benzene, a heavily regulated air pollutant, suggesting a pragmatic approach when
2957 setting air quality targets as tailpipe and solvent emissions decline in developed countries.

2958
2959 Jaffe and Widger (2012) also indicated that boreal wildfires are likely to produce less NO_x on average
2960 than fires in other biomes. This observation makes sense with a more obvious impact of biomass
2961 burning on ozone production in the tropics than at mid-high northern latitudes. However, boreal
2962 wildfires may have diverse impacts on the global ozone distribution as they are also often linked to
2963 convection and pyro-convection allowing thus the injection of ozone precursors and particulate
2964 matter up to the upper troposphere-lower stratosphere where long-range transport processes can
2965 widely distribute the emissions. Subsequent photochemical processes leading to ozone production
2966 and their impact on larger scale may be different than in the tropics. Several groups are actively
2967 developing ~~algorithm~~algorithms and models to ~~propose~~derive appropriate injections heights for
2968 biomass burning emissions (e.g. ~~(Kaiser et al., 2012; Freitas et al., 2007)~~(Kaiser et al., 2012; Freitas et
2969 al., 2007)). Depending on the fire radiative power (FRP), it is shown that boreal forest fires are likely
2970 able to emit products up to 10 km altitude with a maximum between 1 and 4 km altitude for fires
2971 with the highest radiative power, while tropical biomass burning is limited to the first km of the
2972 atmosphere. Depending on the fire radiative power (FRP) and size of the fire, Freitas et al. (Freitas
2973 et al., 2007) have shown that plumes from fires are likely able to reach 10 km altitude. Based on a
2974 statistical analysis of 5 years of satellite observation by MISR (Multi-angle Imaging Spectrometer)
2975 over North America, Val Martin et al. (Martin et al., 2010) have shown that the median altitude of
2976 plumes is found below 3 km altitude for boreal forest fires. A significant fraction (4-12%) of those
2977 plumes are injected above the boundary layer and are more spread-out vertically depending on the
2978 atmospheric stability conditions. In comparison, tropical biomass burning plumes are mostly limited
2979 to the first km of the atmosphere.

2980
2981 Numerous studies report high mixing ratios of biomass burning products, CO in particular, measured
2982 in the middle to upper troposphere and eventually up to the lower stratosphere, thousands of km
2983 from the fire sources (e.g. ~~(Nedelec et al., 2005; Cammas et al., 2009; Fromm et al., 2010; Elguindi et~~
2984 ~~al., 2010)~~(Nedelec et al., 2005; Cammas et al., 2009; Fromm et al., 2010; Elguindi et al., 2010)).
2985 Biomass burning signatures from Canadian forest fires have also been observed in the lower
2986 troposphere at the Pico Observatory on the opposite of the North Atlantic ~~(Val Martin et al.,~~
2987 ~~2006)~~(Val Martin et al., 2006). Implications for the ozone budget in Boreal regions are not as direct as
2988 in the Tropics. Photochemical impact remains uncertain. Clearly, long range transported biomass
2989 burning plumes can influence Europe ~~(Cook et al., 2007; Real et al., 2007)~~(Cook et al., 2007; Real et al.,
2990 2007) though the impact is variable ~~(Hudman et al., 2004)~~(Hudman et al., 2004). More recently,
2991 European fires in Portugal and Russia have been shown to contribute to air pollution ~~(Tressol et al.,~~
2992 ~~2008; Martins et al., 2012)~~(Tressol et al., 2008; Martins et al., 2012) and the frequency is expected to
2993 increase with climate change ~~(Carvalho et al., 2011)~~(Carvalho et al., 2011). Long range transport of
2994 biomass burning products from Siberia can influence the west coast of the North American continent
2995 (Seattle, Vancouver) and such haze may have strong impact on meeting their air quality standards.
2996 Figures 30 summarizes ozone and CO profiles recorded over Vancouver in the beginning of July 2012
2997 recorded in the frame of IAGOS. Further details and profiles may be found on the dedicated MACC
2998 web pages: <http://www.iagos.fr/macc> under the rubric "Profile of the day" in relation to the huge
2999 fires in summer 2012. Figure 30 also illustrates the difficulty in assessing the ozone production from
3000 such boreal wildfires based on *in-situ* measurements (of limited compounds). Differing profiles over
3001 Vancouver have different $\Delta O_3/\Delta CO$ (from almost 0 to 1) and given plume age is increasing everyday
3002 (same source, same receptor, same transport pathway apparently). Finally, it is noticeable that the

3003 ~~highest ozone concentrations in the lower troposphere are observed on the last day of this time-~~
3004 ~~series.~~ Besides their impact on air quality over populated mid-latitudes, boreal fires are also of
3005 particular importance for the Arctic region. For example, the ARCTAS mission held in April 2008 and
3006 June-July 2008 was designed to observe spring time fires from Siberia, and summertime fires plumes
3007 from Canada and Siberia along with their impact on Arctic pollution (Jacob et al., 2010) (see also
3008 section 4.8).
3009



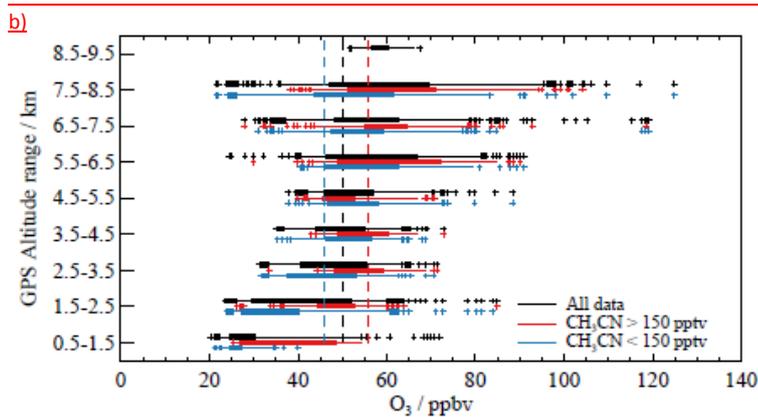
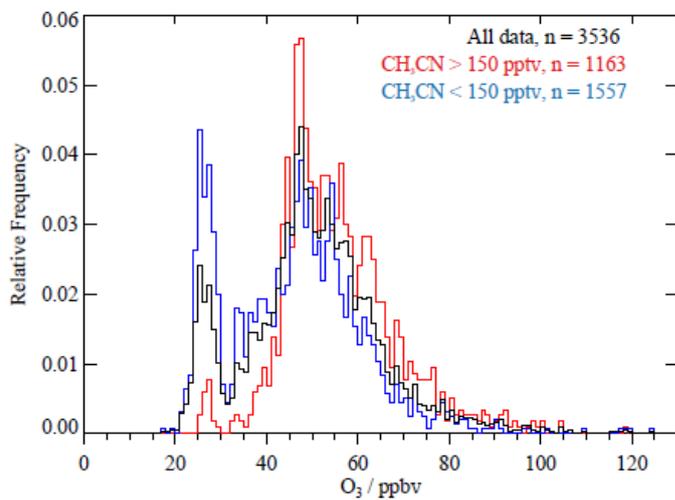


Figure 30 — Ozone and Carbon monoxide vertical profiles over Vancouver recorded by IAGOS* (<http://www.iagos.org/>) in July 2012 (Courtesy of Hannah Clark). **Figure 31** — a) Frequency distribution for ozone over the BORTAS (Palmer et al., 2013) flights, the black line are the measured mixing ratios, where the red line shows the distribution of measurements identified as biomass burning plumes using a threshold of CH_3CN mixing ratios greater than 150 pptv, and the blue line shows measurements made in air masses with CH_3CN mixing ratios less than 150 pptv. ; b) the same data plotted as a box and whisker (solid boxes show the range of the 25th to 75th percentiles and whiskers show the range of the 5th to 95th percentiles with outliers shown as plus symbols) (GPS) altitude profile (Parrington et al., 2013).

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3010

3011 Biomass burning in the tropics has long been recognized as the major source of tropospheric ozone
 3012 locally and regionally (Fishman et al., 1990;Thompson et al., 1996;Sauvage et al., 2006;Jonquieres et
 3013 al., 1998;Thompson et al., 2003e)(Fishman et al., 1990;Thompson et al., 1996;Sauvage et al.,
 3014 2006;Jonquieres et al., 1998;Thompson et al., 2003c). Tropospheric seasonal cycles of ozone are in
 3015 phase with the local biomass burning season (coinciding with the dry season) of each region. It is
 3016 worth noting one exception over Equatorial Africa (Gulf of Guinea and adjacent continent) which is
 3017 impacted by two biomass burning regions, one from each hemisphere. For example, over Lagos,

3018 Nigeria, ozone enhanced layers are observed in December-February when burning occurs regionally
3019 and also in July-August when biomass burning occurs over southern Equatorial Africa (~~Sauvage et al.,~~
3020 ~~2005;Sauvage et al., 2007b~~)(~~Sauvage et al., 2005;Sauvage et al., 2007b~~). Such processes have been
3021 further investigated in the frame of AMMA (~~Reeves et al., 2010;Thouret et al., 2009;Mari et al.,~~
3022 ~~2008~~)(~~Reeves et al., 2010;Thouret et al., 2009;Mari et al., 2008~~). Similarly, over Brazzaville, the
3023 seasonal cycle of ozone peaks in July-August but a secondary maximum appears in January-February
3024 due to transport of ozone and precursors from fires occurring in the northern hemisphere (~~Sauvage~~
3025 ~~et al., 2005;Sauvage et al., 2007b~~)(~~Sauvage et al., 2005;Sauvage et al., 2007b~~). Thus, the so-called
3026 zonal wave-one as described by Thompson et al., 1999 and Sauvage et al., 2006, which describe the
3027 ozone maximum over the South Atlantic and adjacent continents clearly designates biomass burning
3028 as the main contributor of the ozone burden in the tropical lower to middle troposphere at least.
3029 Biomass burning in the tropics may have a global impact too. The extreme El-Nino event in 1997 and
3030 the consequent long lasting fires in Indonesia (~~Siegert et al., 2001~~)(~~Siegert et al., 2001~~) have
3031 contributed to the increase of CO and other trace gases (e.g. CO₂) observed globally from late 1997
3032 through 1998.

3033

3034 4.3 Role of biogenics in the formation of ozone

3035

3036 There is much debate as to the influence of biogenic VOCs in urban (~~Calfapietra et al.,~~
3037 ~~2013~~)(~~Calfapietra et al., 2013~~), regional and global ozone budgets. Variation in natural plant
3038 emissions of ozone precursors can influence ozone concentrations. Goldstein and Galbally suggested
3039 we cannot account for the loss of approximately half the non-methane organic carbon entering the
3040 atmosphere (~~Goldstein and Galbally, 2007~~)(~~Goldstein and Galbally, 2007~~) much of biogenic origin.
3041

3042 Much of the focus on biogenic VOCs and ozone is on isoprene and the monoterpenes owing to their
3043 apparent dominance of the global VOC budgets (see Figure ~~2122~~). New insights from field
3044 observations, models and quantum chemistry into isoprene chemistry and its impacts have been
3045 recently reviewed by Whalley et al. (~~Whalley et al., 2014~~)(~~Whalley et al., 2014~~). Some of the new
3046 radical chemistry from isoprene oxidation is detailed in section 4.10 and the emissions in section 2.3.
3047 This section deals with the relationships between biogenics and formation of ozone.

3048 Work by Hewitt et al has suggested that the circadian rhythms of the isoprene emitters have an
3049 effect on the ozone budgets (~~Hewitt et al., 2011~~)(~~Hewitt et al., 2011~~) (see Figure ~~3132~~), but there has
3050 been some debate as to the nature of the circadian control (~~Keenan and Niinemets, 2012~~)(~~Keenan~~
3051 ~~and Niinemets, 2012~~).

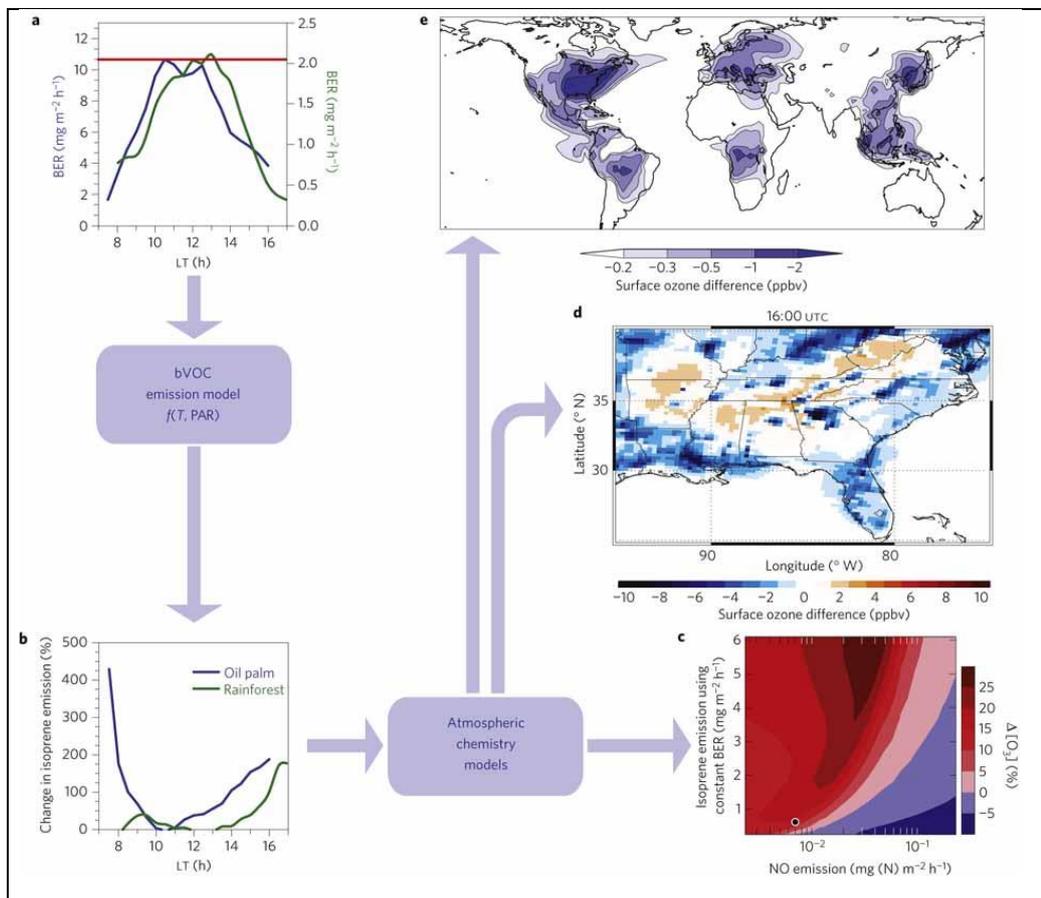


Figure 3132 - The effect of a circadian rhythm based biogenic emission rate (BER) on ozone; **a**) The oil palm plantation (blue) and rainforest (green) BERs of isoprene compared with a constant BER (red). **b**), The differences in isoprene emission rates between constant (red line in **a**) and circadian-controlled BERs (oil palm: blue; rainforest: green); **c**), Changes in ground-level ozone resulting from changing isoprene and nitrogen oxide emission rates. **d**), Changes in regional-scale modelled ground-level ozone for 11:00 LT using an 'oil palm' circadian-controlled BER compared with a constant BER. **e**), Changes in global-scale modelled ground-level ozone for July using the same scenario as **d** ([Hewitt et al., 2011](#))([Hewitt et al., 2011](#)).

3052

3053 Model calculations have indicated that the biogenic isoprene emissions represent a major
 3054 uncertainty in the achievement of AOT40 levels by a factor of at least two ([EEA, 2009](#))([EEA, 2009](#)).
 3055 The impact of North American (NA) isoprene on European ozone has been assessed and it has been
 3056 shown that future increases in NA isoprene emissions could offset decreases in EU surface O₃
 3057 resulting from controls on NA anthropogenic emissions ([Fiore et al., 2011](#))([Fiore et al., 2011](#)).
 3058 Archibald et al. have shown that there is a significant impact of mechanism uncertainties on the
 3059 global impact of isoprene chemistry on ozone ([Archibald et al., 2011](#))([Archibald et al., 2011](#)). Recent
 3060 estimates from Zare et al ([Zare et al., 2014](#))([Zare et al., 2014](#)) found BVOC to be the most significant
 3061 contributor to ozone formation over land areas in the NH enhancing the mixing ratio by about 11%.
 3062 In the Pearl River Delta in China ([Situ et al., 2013](#))([Situ et al., 2013](#)) ozone is sensitive to the BVOC

3063 levels particularly in urban areas. At a regional European scale, biogenic emissions increase O₃,
3064 locally, while reducing PM_{2.5} levels (~~Tagaris et al., 2014~~)(Tagaris et al., 2014). At the city scale,
3065 modelling has shown that O₃ concentration can be increased by 37 ppb in Seoul, Korea owing to
3066 biogenic isoprene emission and transport of ~~its~~ key oxidation products into the metropolitan area
3067 (~~Lee et al., 2014~~)(Lee et al., 2014).

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3068 The role of isoprene nitrates has been highlighted as a key uncertainty in ozone and NO_x chemistry
3069 (~~Horowitz et al., 2007~~)(Horowitz et al., 2007). In the tropics regional simulation of ozone is shown to
3070 be very sensitive to the removal and export of NO_x by isoprene nitrates (~~Paulot et al., 2012~~)(Paulot et
3071 al., 2012). For example, in South America and New Guinea, the high ratio of isoprene to-NO_x
3072 emissions makes isoprene nitrates chemistry the primary sink of NO_x. Paulot et al. (2012) suggest
3073 that the impact of isoprene photooxidation on tropical ozone is not limited to the regional scale but
3074 can have a long-range effect as a result of dynamic and photochemical processes such as efficient
3075 vertical mixing (through deep convection) and low boundary layer HO_x (as a result of high biogenics
3076 and low NO_x emissions). It is suggested that these physical and chemical conditions, promote the
3077 oxidation of isoprene outside of the boundary layer, where its contribution to ozone production is
3078 amplified (~~Paulot et al., 2012~~)(Paulot et al., 2012). Enhancing the isoprene oxidation mechanism for
3079 isoprene-nitrates in a global model and running it over the USA, Mao et al (~~Mao et al., 2013~~)(Mao et
3080 al., 2013) have shown that the dependence of surface ozone on isoprene emission is positive
3081 throughout the U.S., even if NO_x emissions are reduced by a factor of 4, as well as the chemistry
3082 leading to the export of nitrates to the background atmosphere. Browne et al (~~Browne et al.,
3083 2014~~)(Browne et al., 2014) have shown that the monoterpene nitrates have the potential to impact
3084 ozone levels over the remote continental boundary layer. The change in global O₃ burden due to an
3085 increase in BVOC emissions associated with temperature depends critically on the assumed
3086 treatment for the fraction of NO_x recycled from isoprene nitrates (~~Ito et al., 2009~~)(Ito et al., 2009).

3087 ~~There is significant debate about the role of a wide range of biogenics in ozone chemistry under
3088 future climate e.g. (Andersson and Engardt, 2010) with up to a factor five difference between
3089 different models for Europe (Langner et al., 2012b). A limiting role of increased CO₂ on the potential
3090 of ozone precursor emissions from the vegetation has been recently noted at the global scale
3091 (Lathiere et al., 2010). Earlier studies showed that the vegetation response to climate change is
3092 important in driving BVOC emissions and subsequent formation of ozone (Sanderson et al., 2003).~~

3093 A future uncertainty revolves around the impact of biofuels on ozone concentrations, via changes to
3094 vegetation (Ashworth et al., 2013) and BVOC emissions (Porter et al., 2012).

3095 There is significant debate about the role of a wide range of biogenics in ozone chemistry under
3096 future climate e.g. (Andersson and Engardt, 2010;Wiedinmyer et al., 2006) with up to a factor five
3097 difference between different models for Europe (Langner et al., 2012c). It has been recently shown
3098 that the choice of isoprene chemical schemes in climate models can be important (Squire et al.,
3099 2015). A limiting role of increased CO₂ on the potential of ozone precursor emissions from the
3100 vegetation has been recently noted at the global scale (Lathiere et al., 2010). Earlier studies showed
3101 that the vegetation response to climate change is important in driving BVOC emissions and
3102 subsequent formation of ozone (Sanderson et al., 2003).

3103 Several other studies have focused only on the regional impact of climate change (~~Langner et al.,
3104 2012a;Langner et al., 2012b;Andersson and Engardt, 2010;Manders et al., 2012;Hedegaard et al.,~~

3105 ~~2008;Hedegaard et al., 2013b;Katragkou et al., 2011)~~(Langner et al., 2012a;Langner et al.,
3106 ~~2012c;Andersson and Engardt, 2010;Manders et al., 2012;Hedegaard et al., 2008;Hedegaard et al.,~~
3107 ~~2013b;Katragkou et al., 2011)~~. They found a general increase of ozone concentrations owing to
3108 increasing biogenic emissions and favourable conditions for the build-up of ozone pollution as a
3109 result of increased temperature, more frequent summer blocking and heat waves and increased
3110 shortwave radiation. Whereas all these studies agreed on the main outcome of climate change on
3111 ozone in Europe, they also pointed out significant differences depending on the regional climate
3112 model used. The main factors influencing these differences are the projections of cloud cover and
3113 frequency of stagnation episodes that are highly variable across the ensembles of regional climate
3114 projections.

3115

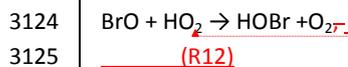
3116 4.4 Halogens

3117

3118 Halogens (chlorine, bromine, iodine) influence the concentrations of ozone in the troposphere either
3119 directly, by reacting with O₃ itself (e.g.,



3121 or indirectly, by affecting its sources and sinks. Indirect influence results in production or destruction
3122 of O₃, depending on the conditions. Halogens, especially chlorine, react with VOCs to form peroxy
3123 radicals, which convert NO into NO₂, and change the OH/HO₂ ratio (e.g., via



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3126 followed by HOBr photolysis to give OH). Reactions between halogen species and nitrogen oxides
3127 also affect the NO/NO₂ ratio, and form stable compounds, which can act as nitrogen reservoirs and
3128 allow long-range transport of O₃ precursors. A detailed model analysis by Saiz-Lopez et al (Saiz-Lopez
3129 et al., 2012a)(Saiz-Lopez et al., 2012a) estimated that halogens are responsible for up to 10% yearly
3130 depletion of the total tropospheric O₃ column (and up to 20% in the tropical troposphere), especially
3131 in the middle and upper troposphere. The model calculations by Parrella et al. (Parrella et al.,
3132 2012)(Parrella et al., 2012) and Long et al. (Long et al., 2014)(Long et al., 2014) generally agree with
3133 these estimates. The Long et al. (2014) model analysis suggest that models may overestimate the
3134 role of bromine (and the extent of ozone destruction), because they tend to overestimate marine
3135 aerosol, especially in the Southern Hemisphere.

3136 Our understanding of the role of halogens in ozone chemistry ultimately depends on our
3137 understanding of the physio-chemical processes that release, transform and destroy halogen species
3138 into the atmosphere. There are significant uncertainties in our knowledge of the source processes
3139 and geographical distribution of halogens and severe limitations in the databases of chemical kinetics
3140 parameters (Abbatt et al., 2014)(Abbatt et al., 2014). Part of the problem is related to the difficulty
3141 in measuring halogens species at the low concentrations found in the atmosphere (Finlayson-Pitts,
3142 2010)(Finlayson-Pitts, 2010): this area of research has been very active in recent years and many of
3143 the most recent advancements in our knowledge are related to developments in the field of
3144 analytical chemistry, particularly mass spectrometry.

3145 Very detailed reviews have been published in recent years both on halogen chemistry in general
3146 Saiz-Lopez & von Glasow (~~Saiz-Lopez and von Glasow, 2012~~)(Saiz-Lopez and von Glasow, 2012) and
3147 on specific aspects of halogen chemistry, such as bromine (~~Sander et al., 2003~~)(Sander et al., 2003),
3148 iodine (~~Saiz-Lopez et al., 2012b;Carpenter, 2003~~)(Saiz-Lopez et al., 2012b;Carpenter, 2003) and
3149 ~~polar~~Polar chemistry (~~Simpson et al., 2007~~)(Simpson et al., 2007). These reviews detail the broader
3150 picture of halogen chemistry and the focus hereafter will be on recent advances that directly affect
3151 tropospheric ozone.

3152

3153 Overall, observations of halogen species in the boundary layer show a consistent picture across the
3154 globe (see (~~Saiz-Lopez and von Glasow, 2012~~)(Saiz-Lopez and von Glasow, 2012)), with comparable
3155 levels of reactive halogens in different unpolluted/semi-polluted regions, and more variable levels of
3156 reactive halogens in continental/coastal environments, reflecting the larger variability in their
3157 sources and sinks under polluted conditions.

3158 The major source of chlorine and bromine in the boundary layer is known to be sea-salt: current
3159 model parameterizations are able to reproduce measured BrO levels in the unpolluted MBL
3160 reasonably well (~~Keene et al., 2009;Sommariva and von Glasow, 2012;Saiz-Lopez et al., 2012a~~)(Keene
3161 ~~et al., 2009;Sommariva and von Glasow, 2012;Saiz-Lopez et al., 2012a~~). However, models struggle to
3162 reproduce total bromine and particulate bromide concentrations and their diurnal cycles, suggesting
3163 that our understanding may be limited by the lack of speciation of bromine in current analytical
3164 techniques.

3165 Inorganic chlorine observations are better constrained, although concurrent measurements of Cl₂
3166 and HOCl, especially under semi-polluted conditions (~~Keene et al., 2009;Lawler et al., 2009;Lawler et
3167 al., 2011~~)(Keene et al., 2009;Lawler et al., 2009;Lawler et al., 2011), are difficult to reconcile and BrCl,
3168 which the models predict as one of the major components of total Br, has not been detected in
3169 marine environments. It is unclear whether the models-measurements disagreements are due to
3170 instrumental issues or to some fundamental problem in our understanding of the chemistry,
3171 particularly in the aqueous phase (~~Long et al., 2014;Sommariva and von Glasow, 2012~~)(Long et al.,
3172 ~~2014;Sommariva and von Glasow, 2012~~), but they make it difficult to accurately assess the effect of
3173 chlorine and bromine on ozone under unpolluted and semi-polluted conditions.

3174 In the past few years, several groups have reported observations of significant concentrations of
3175 ClNO₂ (from 80 ppt to over 2000 ppt) in a variety of environments and under different conditions
3176 (~~Osthoff et al., 2008;Kercher et al., 2009;Thornton et al., 2010;Mielke et al., 2011;Mielke et al.,
3177 2013;Phillips et al., 2012;Riedel et al., 2012;Riedel et al., 2013;Wagner et al., 2012~~)(Osthoff et al.,
3178 ~~2008;Kercher et al., 2009;Thornton et al., 2010;Mielke et al., 2011;Mielke et al., 2013;Phillips et al.,
3179 2012;Riedel et al., 2012;Riedel et al., 2013;Wagner et al., 2012~~). These studies indicate that nocturnal
3180 formation of ClNO₂, *via* reaction of N₂O₅ with aerosol chloride, followed by its photolysis at sunrise to
3181 form Cl + NO₂ is a strong and widespread source of reactive chlorine in the polluted troposphere. In
3182 addition, to kick starting VOC oxidation in the morning before the formation of OH, the ClNO₂
3183 mechanism also acts as a NO_x reservoir, preventing NO₂ loss overnight as HNO₃ and hence making it
3184 available in the morning to form O₃. Although these two effects are not yet fully decoupled and
3185 understood (see discussion in (~~Young et al., 2014~~)(Young et al., 2014)), it is clear that this chemistry is
3186 potentially very important for ozone formation in polluted environments. Osthoff et al. (~~Osthoff et
3187 al., 2008~~)(Osthoff et al., 2008), Thornton et al. (~~Thornton et al., 2010~~)(Thornton et al., 2010)

3188 estimated that up to 13 ppb additional ozone can be formed *via* this chemistry. It must be noted,
3189 however, that the actual impact on local ozone may be much less than that, because of the effect of
3190 transport and local circulation on ozone levels (~~Sarwar et al., 2012; Simon et al., 2010; Simon et al.,~~
3191 ~~2009~~)(~~Sarwar et al., 2012; Simon et al., 2010; Simon et al., 2009~~). Furthermore, model analysis
3192 suggests that recirculation of air masses from urban coastal areas over the ocean may lead to high
3193 Cl₂ levels during the night, thus increasing VOC oxidation and ozone formation at sunrise (~~Pechtl and~~
3194 ~~von Glasow, 2007~~)(~~Pechtl and von Glasow, 2007~~), a mechanism which has particular relevance for
3195 coastal megacities.

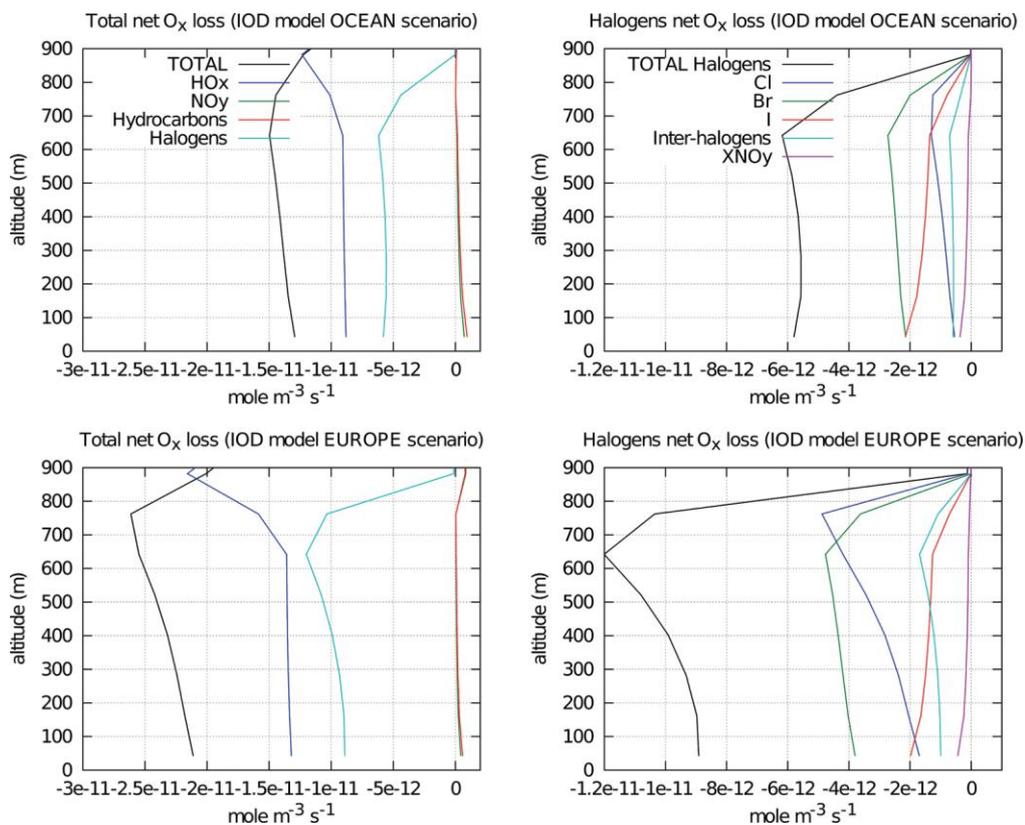
3196 There are several inconsistencies in the observations of reactive iodine, especially under conditions
3197 representative of the open ocean: for example, Carpenter et al. (~~Carpenter et al., 2010~~)(~~Carpenter et~~
3198 ~~al., 2010~~) highlighted the discrepancy between observations of IO made by different groups at the
3199 Cape Verde islands, while Mahajan et al. (~~Mahajan et al., 2010~~)(~~Mahajan et al., 2010~~), found that IO
3200 concentrations in the eastern Pacific did not agree with the observations of Chlorophyll-a and
3201 Dissolved Organic Matter, and were much lower than the SCIAMACHY satellite observations.
3202 Additionally, the measured emission rates of iodinated VOCs are unable to explain the observed
3203 levels of IO and I₂ (~~Carpenter et al., 2013; Jones et al., 2010b; Grossmann et al., 2013; Lawler et al.,~~
3204 ~~2014~~)(~~Carpenter et al., 2013; Jones et al., 2010b; Grossmann et al., 2013; Lawler et al., 2014~~). Several
3205 mechanisms have been introduced to explain the observations and are still under active discussion
3206 (e.g., (~~Lawler et al., 2014; Carpenter et al., 2013; MacDonald et al., 2014~~)(~~Lawler et al.,~~
3207 ~~2014; Carpenter et al., 2013; MacDonald et al., 2014~~)). Some of these discrepancies may be explained
3208 by assuming seasonal or yearly cycles of the iodine sources, but there are also several uncertainties
3209 in the iodine chemical mechanism (~~Sommariva et al., 2012; Saiz-Lopez et al., 2012b~~)(~~Sommariva et al.,~~
3210 ~~2012; Saiz-Lopez et al., 2012b~~). The many uncertainties in iodine sources and chemistry make it
3211 difficult to assess accurately the role of iodine as ozone sink in unpolluted/semi-polluted conditions,
3212 especially over the open ocean. Although the sources and chemistry of iodine species are still being
3213 debated, it is well established that iodine species do not react with VOC and, when NO_x levels are
3214 high, they form stable iodine nitrates (INO_x) which can be taken up on aerosol, leading to net loss of
3215 NO_x (and hence O₃). In addition, recycling of INO₃ in the gas-phase {



3217 may increase ozone depletion by up to 60% (~~Mahajan et al., 2009~~)(~~Mahajan et al., 2009~~).

3218 The interaction between iodine and ozone also leads to the formation of ultra-fine particles under
3219 certain conditions (e.g. coastal regions with strong emissions of iodine from macro-algae,
3220 (~~McFiggans, 2005~~)(~~McFiggans, 2005~~)), although this process, and its impact on local O₃
3221 concentrations, is not yet completely understood. Recent work has shown increased O₃ deposition
3222 velocities during low tides, owing to direct deposition to macro-algae and/or to a-O₃ involvement in
3223 iodine-mediated particle formation (~~McFiggans et al., 2010~~)(~~McFiggans et al., 2010~~).

3224



3225

3226

3227 | **Figure 3233** - Vertical profiles at 12:00midday of total and halogenshalogen induced net O_x loss in an
 3228 unpolluted (OCEAN scenario) and semi-polluted (EUROPE scenario) marine boundary layer
 3229 ([Sommariva and von Glasow, 2012](#))([Sommariva and von Glasow, 2012](#)).

3230

3231 All recent observational and modelling studies of halogen chemistry generally agree that reactive
 3232 halogen species are present in the unpolluted/semi-polluted marine boundary layer at levels that
 3233 result in significant depletion of tropospheric ozone and loss of O_x. Read et al. ([Read et al.,
 3234 2008](#))([Read et al., 2008](#)), Sommariva & von Glasow ([Sommariva and von Glasow, 2012](#))([Sommariva
 3235 and von Glasow, 2012](#)) and Saiz-Lopez et al. ([Saiz-Lopez and von Glasow, 2012](#))([Saiz-Lopez and von
 3236 Glasow, 2012](#)) calculated the contribution of halogens to ozone destruction using different models.
 3237 The results, varying from 3 to 14 ppb per day, indicate that halogens overall are the second most
 3238 important sink for O_x in unpolluted and semi-polluted conditions, accounting for up to a third of
 3239 total ozone loss in the tropical troposphere (Figure 3233). Bromine is ~~about~~ twice more important
 3240 than chlorine as ozone sink, and iodine is potentially more efficient than bromine although its
 3241 concentration shows a steeper vertical gradient, making it less important than bromine and chlorine
 3242 above 500-600 m (Sommariva & von Glasow, 2012). It must also be noted that the co-presence of
 3243 iodine and bromine makes the loss of ozone more efficient than if bromine alone were present, due
 3244 to the multiplying effect of inter-halogens reactions (e.g., IO + BrO, ([Read et al., 2008](#))([Read et al.,
 3245 2008](#))). In polluted regions, due to the higher concentrations of NO_x, the oxidation of VOC by Cl and,

3246 | to a lesser extent, Br, results in net ozone production. For example, Finley & Saltzmann (~~Finley and~~
3247 | ~~Saltzman, 2008~~)([Finley and Saltzman, 2008](#)) calculated that the levels of dihalogens (up to 26 ppt of
3248 | Cl₂ and up to 9 ppt of Br₂) observed in California could account for up to 10% of measured ozone.

3249 | The role of halogens in the boundary layer is likely to be impacted, and possibly become even more
3250 | relevant for O₃ photochemistry, by changes in the Earth System, which may alter the formation of
3251 | marine aerosol, as well as increased emissions of acids and acid precursors, which affect the halogen
3252 | activation processes (~~Long et al., 2014~~)([Long et al., 2014](#)).

3253 |

3254 | At ~~polar~~Polar latitudes (> 60°), periodic ozone depletion events (ODE) have been observed since the
3255 | mid-1980s and have been linked to bromine release from the snowpack (~~Simpson et al.,~~
3256 | ~~2007~~)([Simpson et al., 2007](#)) with observed BrO levels ranging from 5 to 41 ppt (~~Roscoe et al.,~~
3257 | ~~2014~~;[Saiz-Lopez and von Glasow, 2012](#))([Roscoe et al., 2014](#);[Saiz-Lopez and von Glasow, 2012](#)).

3258 | Recently, novel observations of BrO (~~Liao et al., 2011~~)([Liao et al., 2011](#)) have shown that “BrO
3259 | clouds” tend to be distributed homogeneously over scales of a few km during ODE, while Salawitch
3260 | et al. (~~Salawitch et al., 2010~~)([Salawitch et al., 2010](#)) found that BrO columns are correlated with the
3261 | height of the tropopause, although some “hotspots” are related to free tropospheric or stratospheric
3262 | intrusions.

3263 | There is much less information regarding iodine and chlorine in Polar Regions. Chlorine presence has
3264 | been inferred only by VOC ratios in the past; recently Pohler et al. (~~Pohler et al., 2010~~)([Pohler et al.,](#)
3265 | [2010](#)) and Liao et al. (~~Liao et al., 2014~~)([Liao et al., 2014](#)) have reported the first observations of OClO
3266 | (up to 24 ppt) and Cl₂ (up to 100 ppt), respectively, in the Arctic. These findings suggest that Cl may
3267 | enhance O₃ depletion in the Polar Regions, mostly *via* its close coupling with bromine chemistry (with
3268 | the ClO + BrO reaction contributing up to 73% to ozone depletion). Iodine species (IO) have been
3269 | detected in Antarctica at levels comparable to BrO (~20 ppt, (~~Saiz-Lopez et al., 2008~~)([Saiz-Lopez et](#)
3270 | [al., 2008](#))), which increase O₃ loss rate by up to 3 times. Only one study reported IO in the Arctic at
3271 | <3.4 ppt (~~Mahajan et al., 2010~~)([Mahajan et al., 2010](#)); whether this is due to differences between the
3272 | two ~~polar~~Polar regions (e.g., biological communities or sea ice characteristic) is at present unclear.

3273 | Many questions remain on the mechanism of release of chlorine/bromine species from the
3274 | snowpack (see (~~Abbatt et al., 2012~~)([Abbatt et al., 2012](#)) for a detailed review). An important issue is
3275 | whether it can explain observed HOx and NOx levels (~~Saiz-Lopez et al., 2008~~;~~Bloss et al., 2010~~)([Saiz-](#)
3276 | [Lopez et al., 2008](#);[Bloss et al., 2010](#)), as well as observed halogens levels. A modelling study by
3277 | Thomas et al. (~~Thomas et al., 2011~~)([Thomas et al., 2011](#)), Thomas et al. (~~Thomas et al., 2012~~)([Thomas](#)
3278 | [et al., 2012](#)) indicated that up to 10 ppt of tropospheric BrO can be explained by a mechanism
3279 | involving nitrate formation in the interstitial snow; if this is the case, the resulting formation of NOx
3280 | may compensate the depletion of O₃ due to reactive Br, possibly leading to net ozone formation.
3281 | Additionally, since bromine release is connected with first-year sea ice (~~Saiz-Lopez and von Glasow,~~
3282 | ~~2012~~)([Saiz-Lopez and von Glasow, 2012](#)), changes in sea ice formation due to climatic change are
3283 | likely to affect ozone photochemistry significantly.

3284 |

3285 | The importance of localized sources of halogens, such as salt lakes, salt beds, saline soils and
3286 | marshes, for O₃ photochemistry is at present unclear. Large concentrations of bromine and iodine
3287 | (up to 200 ppt of BrO (~~Tas et al., 2005~~)([Tas et al., 2005](#)) and up to 10 ppt of IO, (~~Zingler and Platt,~~

3288 | ~~2005~~(Zingler and Platt, 2005)) have been reported over the Dead Sea (Israel) causing O₃ as low as 2
3289 | ppb; however, observations in other locations have shown much lower values (e.g., 6 ppt of BrO and
3290 | 15 ppt of ClO over the Great Salt Lake, ~~(Stutz et al., 2002)~~(Stutz et al., 2002)). The database of
3291 | observations is very sparse and shows large variability between different locations, suggesting that
3292 | the local characteristics (e.g, latitude, pH, geology, ecosystem and local meteorology) of the salt
3293 | lakes are crucial in determining the strength of halogen emissions and therefore of the magnitude of
3294 | the ODE they trigger ~~(Smoydzin and von Glasow, 2009)~~(Smoydzin and von Glasow, 2009)

3295 | Chlorine and bromine species have also been detected in volcanic plumes at ppm and ppb levels,
3296 | respectively ~~(Saiz-Lopez and von Glasow, 2012)~~(Saiz-Lopez and von Glasow, 2012), leading to ozone
3297 | depletion inside the plume which can persist for at least 3 days and possibly longer (see ~~(von Glasow,~~
3298 | ~~2010)~~(von Glasow, 2010), for a detailed discussion). Similar chemistry may be expected from other
3299 | types of volcanic activity, such as volcanic fields, vents, fumaroles, etc... and may affect ozone levels
3300 | both in the boundary layer and in the free troposphere. The extent to which halogens from volcanic
3301 | activity affect ozone photochemistry on a larger (regional/global) scale depends on several variables,
3302 | including the type, frequency and extent of volcanic activity, the mixing with the background air and
3303 | the synoptic circulation.

3304 |

3305 | Besides the injection of halogen species by volcanic activity (see above), the main sources of
3306 | halogens in the free troposphere are believed to be the oxidation of halocarbons, the uplift of
3307 | aerosol and stratospheric intrusions ~~(Saiz-Lopez and von Glasow, 2012)~~(Saiz-Lopez and von Glasow,
3308 | ~~2012)~~, although direct observations are scarce. Pommier et al. ~~(Pommier et al., 2012)~~(Pommier et al.,
3309 | ~~2012)~~ found evidence of biomass burning from Siberian fires as a source of BrO (>5 ppt) in the Arctic
3310 | free troposphere, although the release mechanism is unclear; Roscoe et al., ~~(Roscoe et al.,~~
3311 | ~~2014)~~(Roscoe et al., 2014) compared ground based and remote sensing measurements in Antarctica
3312 | and concluded that they could only be reconciled by assuming large amounts of BrO at high
3313 | altitudes, in broad agreement with the findings by Koo et al. ~~(Koo et al., 2012)~~(Koo et al., 2012).

3314 | Jones et al. ~~(Jones et al., 2010a)~~(Jones et al., 2010a) found a strong association between high altitude
3315 | ODEs and atmospheric low pressure systems which drive the formation of large scale BrO clouds
3316 | over Antarctica. Free troposphere-boundary layer dynamic may also be important outside the Polar
3317 | Regions. For example, Long et al. (2014) suggested that the accumulation of insoluble bromine in
3318 | the free troposphere (*via* sulphur oxidation by HOBr and/or HBr condensation on aerosol) may
3319 | explain the observed Br enrichment of submicron aerosol after entrainment of these species back in
3320 | the boundary layer. These recent findings suggest the presence of large concentrations of inorganic
3321 | Br at high altitudes in the troposphere and model calculations show that they may account for 3-15%
3322 | reduction in tropospheric O₃ ~~(von Glasow and Crutzen, 2004)~~(von Glasow and Crutzen, 2004). Lary
3323 | ~~(Lary, 2005)~~(Lary, 2005) has examined the role of halogens in the free troposphere using chemical
3324 | data assimilation technique and concluded that chlorine and bromine can affect significantly the
3325 | levels of methane (10-50% of the total oxidation rate) and ozone in the upper troposphere,
3326 | especially at ~~polar~~Polar and tropical latitudes.

3327 | Recently, IO has also been observed in the tropical and sub-tropical free troposphere at
3328 | concentrations between 0.2 and 0.4 ppt ~~(Dix et al., 2013; Puente-dura et al., 2012)~~(Dix et al.,
3329 | ~~2013; Puente-dura et al., 2012)~~. Assuming 0.5 ppt of BrO and 0.6 ppt of IO, Dix et al. (2012) estimated

3330 that iodine accounts for 11% and 26% of total ozone loss in the free troposphere and in the
3331 boundary layer-free troposphere transition region, respectively.

3332

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3334 4.5 Tropospheric ozone and global perturbation of the nitrogen cycle

3335

3336 The drivers of enhanced tropospheric ozone are emissions of the precursors, NO_x and VOC, and
3337 viewed globally the hot spots for ozone production coincide with the areas of largest NO_x emissions
3338 (~~Sutton et al., 2006~~)([Stevenson et al., 2006](#)), most notably in Europe, North America and Asia.
3339 Thus the effects of elevated tropospheric ozone are coupled to anthropogenic emissions of NO_x, an
3340 important anthropogenic contribution to the current global cycling of fixed nitrogen (Nr), to
3341 distinguish it from the atmospheric reservoir of unreactive N₂. Recent assessments of the global
3342 nitrogen cycle have quantified the major natural and anthropogenic fluxes of Nr (~~Sutton et al.,~~
3343 ~~2011;Galloway et al., 2004~~)([Sutton et al., 2011;Galloway et al., 2004](#)). The scale of the human
3344 perturbation of the global nitrogen cycle is substantial, with approximately half of the 413 Tg-N
3345 annual fixation of atmospheric nitrogen into reactive oxidized NO_y and reduced, NH₃ forms (~~Fowler et~~
3346 ~~al., 2013b~~)([Fowler et al., 2013b](#)) contributed by anthropogenic activities (~~Lu et al., 2013~~)([Lu et al.,](#)
3347 [2013](#)). Within this total, most of the nitrogen fixed annually is by industrial production of ammonia in
3348 Haber-Bosch industrial plants and within soils and oceans by microbial processes. Only a small
3349 fraction of this Nr is emitted to the atmosphere (see Figure ~~3334~~). The emissions most important for
3350 tropospheric ozone production are of NO_x from combustion, in vehicles and industry, which totals
3351 approximately 40Tg-N annually, just 10% of total Nr production.

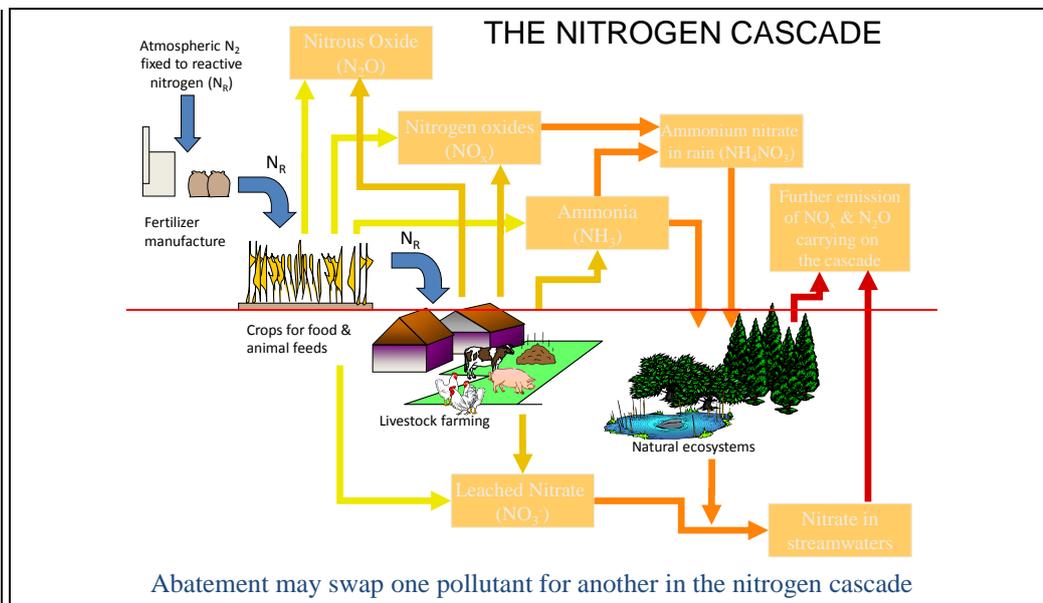
3352 While the focus of this review is tropospheric ozone, it is important to recognise the wider impacts of
3353 the human perturbation of the nitrogen cycle (~~Fowler et al., 2014~~)([Fowler et al., 2015](#)). These
3354 include eutrophication of terrestrial and marine ecosystems, reductions in biodiversity of terrestrial
3355 ecosystems over Europe and North America, widespread effects on human health from the aerosols
3356 containing nitrogen compounds and radiative effects on global climate and reductions in
3357 stratospheric ozone from elevated N₂O emission, as detailed by Erisman et al (~~Erisman et al.,~~
3358 ~~2013~~)([Erisman et al., 2013](#)). The negative effects of contamination of the environment by nitrogen
3359 compounds has been extensively documented for Europe (~~Sutton et al., 2011~~)([Sutton et al., 2011](#))
3360 and North America (~~Suddick et al., 2013~~)([Suddick et al., 2013](#)). However, evidence from Asia and
3361 especially China and India is increasing rapidly and the absolute values for deposition of Nr in these
3362 regions are among the largest globally and effects on ozone production and terrestrial eutrophication
3363 are widespread.

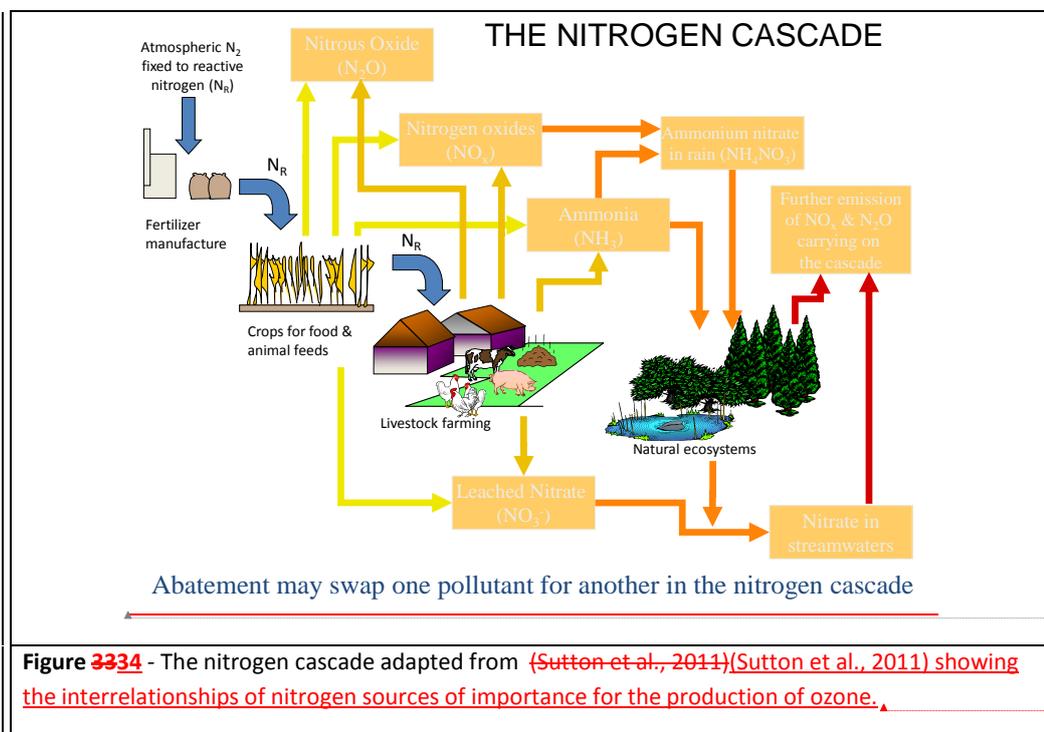
3364 Relatively little research on the effects of nitrogen emission has been reported ~~for in the~~ tropics, but
3365 new evidence from work in SE Asia by Hewitt et al (~~Hewitt et al., 2009~~)([Hewitt et al., 2009](#)) suggests
3366 that these regions are very sensitive to emissions of NO_x which are projected to increase
3367 substantially over coming decades as natural rainforest ecosystems are replaced by agriculture e.g.
3368 plantations of oil palm.

3369 One of the distinctive features of the nitrogen cycle is the rapid transformation of nitrogen
3370 compounds within, and transfers between the atmosphere, vegetation, soils, fresh and marine

3371 waters. Thus emissions of oxidized nitrogen to the atmosphere are rapidly oxidized and deposited
 3372 into terrestrial ecosystems, where it may be transformed into amino acids and subsequently
 3373 decomposed to ammonium following decay and returned to the atmosphere as ammonia. In the
 3374 processes within this short sequence, the NO_x in the atmosphere may have generated ozone,
 3375 reduced the lives of humans breathing NH_4NO_3 containing aerosols and contributed to species loss in
 3376 terrestrial ecosystems. The same emitted N atom may have contributed to a series of different
 3377 effects within the Earth system before it is returned back to the atmospheric reservoir as N_2
 3378 following denitrification in soils or within the ocean. This effect has been referred to as the nitrogen
 3379 cascade (Galloway et al., 2003)(Galloway et al., 2003) and shows a very large range of effects of
 3380 reactive nitrogen on climate, terrestrial and marine ecosystems and on human health. A full analysis
 3381 of the global effect of nitrogen has yet to be completed, but assessments have been recently
 3382 published for Europe (Sutton et al., 2011)(Sutton et al., 2011) and for North America (Suddick et al.,
 3383 2013)(Suddick et al., 2013).

3384





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3385

3386 One further aspect for consideration is the coupling between tropospheric ozone, carbon uptake in
 3387 the terrestrial biosphere and nitrogen (Simpson et al., 2014)(Simpson et al., 2014). It has been
 3388 recently shown that limitations of available nitrogen for sufficient plant growth reduce the negative
 3389 impact of tropospheric ozone on carbon uptake in plants, leading to a smaller indirect change in
 3390 radiative forcing than previously calculated (Kvalevåg and Myhre, 2013)(Kvalevåg and Myhre, 2013).

3391 4.6 Challenges in modelling ozone

3392

3393 Models are the integrator of the chemical and physical knowledge with respect to ozone in the
 3394 atmosphere. They are much relied on for analysis of observational data, hindcasting, policy support
 3395 and forecasting across a range of temporal and spatial scales. Models vary much in scale, resolution
 3396 and with physical/chemical complexity.

3397 With respect to current global ozone, Young et al. (Young et al., 2013)(Young et al., 2013) compared
 3398 modelled (ACCMIP models) and observed (ozone-sonde and satellite (OMI/MLS)) present-day spatial
 3399 distributions and seasonality of global tropospheric ozone. The models captured the seasonality well
 3400 at most locations. Based on the comparison to OMI/MLS data the models typically overestimated
 3401 annual mean tropospheric column ozone at 0-50°N by ≈0-30% and underestimate it at 0-50°S by ≈0-
 3402 25%. Similarly, re-analysis model data for ozone from MACC shows biases of -5% to +10% with
 3403 respect to ozonesondes and aircraft data in the extratropics, but with larger negative biases in the
 3404 tropics (Inness et al., 2013)(Inness et al., 2013). These results indicate that models don't fully
 3405 represent the processes controlling the present-day ozone distribution.

3406 Parrish et al. (~~Parrish et al., 2014~~)(Parrish et al., 2014) have shown that three of the ACCMIP models,
3407 driven by trends in emissions, underestimate observed trends in surface ozone over the period since
3408 ~1950, at NH mid-latitudes; similar results have been found earlier, e.g. (~~Schultz et al., 2007b~~)(Schultz
3409 et al., 2007b). Cooper et al. (~~Cooper et al., 2014~~)(Cooper et al., 2014) extended this analysis to show
3410 that models also underestimate trends in other regions. These observations suggest that modelled
3411 ozone appears to be rather too insensitive to emissions perturbations (or possibly that historical
3412 emissions changes are not well described, see section 2.3). Emissions remain a key uncertainty for
3413 global models (~~Frost et al., 2012~~)(Frost et al., 2012).

3414 Hess and Zbinden (~~Hess and Zbinden, 2013~~)(Hess and Zbinden, 2013) and Hess et al. (~~Hess et al.,~~
3415 ~~2014~~)(Hess et al., 2014) find that the evolution of stratospheric ozone, and how this ozone is
3416 transported into the troposphere, is a major determinant of historical inter-annual variability of NH
3417 mid-latitude ozone throughout the troposphere, and may have significantly contributed to long-term
3418 trends. This work indicates that global models need a detailed representation of stratospheric and
3419 UT/LS processes in order to simulate ozone trends and variability correctly.

3420 Further challenges to models include, the representation of different aspects of ozone chemistry
3421 (e.g., isoprene: (~~Archibald et al., 2010;Dunker et al., 2014~~)(Archibald et al., 2010;Dunker et al., 2014);
3422 halogens: (~~Yang et al., 2005;Saiz-Lopez et al., 2012a~~)(Yang et al., 2005;Saiz-Lopez et al., 2012a);
3423 chemical mechanism (~~Saylor and Stein, 2012~~)(Saylor and Stein, 2012)) and deposition (~~Val Martin et~~
3424 ~~al., 2014~~)(Val Martin et al., 2014). For models to represent many of these processes, adequate
3425 resolution is required (~~Colette et al., 2014~~)(Colette et al., 2014).

3426 A couple of studies (~~Wu et al., 2007;Wild, 2007~~)(Wu et al., 2007;Wild, 2007) have argued that much
3427 of the variance in ozone production across models can be explained by differences in NOx emissions,
3428 inclusion of nonmethane volatile organic compounds (NMVOCs, mostly biogenic isoprene) and ozone
3429 influx from stratosphere-troposphere exchange. Model tagging offers insights into separating
3430 processes such as transport and chemistry (see e.g. (~~Garny et al., 2011~~)(Garny et al., 2011)).

3431 The challenge for global modellers is prioritising and including all relevant processes in a model with
3432 sufficient resolution and while keeping it sufficiently computationally efficient so that it can be useful
3433 for a wide range of studies.

3434 A large range of models are used to address ground-level ozone at the regional scale (~~Kukkonen et~~
3435 ~~al., 2012~~)(Kukkonen et al., 2012). Taking into account only emission control policies for the present or
3436 near future (2010 or 2020), several multi-model exercises have been conducted in support to the
3437 Clean Air For Europe programme (CAFE). The CityDelta project (~~Cuvelier et al., 2007~~)(Cuvelier et al.,
3438 2007) aimed to predict the impact and uncertainty of emission reductions of several emission
3439 scenarios for 2010 in several European cities. The same exercise was carried out at the larger scale of
3440 the European continent (EuroDelta: (~~van Loon et al., 2007~~)(van Loon et al., 2007)). At the continental
3441 scale the efficiency of emission reductions was demonstrated but models provided a large spread of
3442 responses in city centres. The ability of an ensemble of six chemistry transport models to capture
3443 recent observed ozone trends was also discussed by Colette et al. (~~Colette et al., 2012~~)(Colette et al.,
3444 2012). They found that models efficiently captured the increase in NOx saturated areas, such as the
3445 Benelux region and the decrease in NOx limited areas (many rural regions in Europe). The
3446 quantitative skill of the model was however difficult to retrieve owing to the lack of significant trends
3447 throughout Europe over the period selected for the experiment. They also emphasized that modelled

3448 trends where highly sensitive to the trends in precursors prescribed in the inventory which exhibited
3449 some significant inconsistencies compared to observations of NO₂, in particular.

3450 A regional model comparison of ozone is shown in Figure 3435. The comparison found that no one
3451 model was the 'best' model on all days, indicating that no single air quality model could currently be
3452 relied upon to inform policymakers robustly in terms of NO_x- versus VOC-sensitivity. For this reason
3453 coupled to basic statistical arguments, it was argued that it is important to maintain diversity in
3454 model approaches (Derwent et al., 2014)(Derwent et al., 2014).

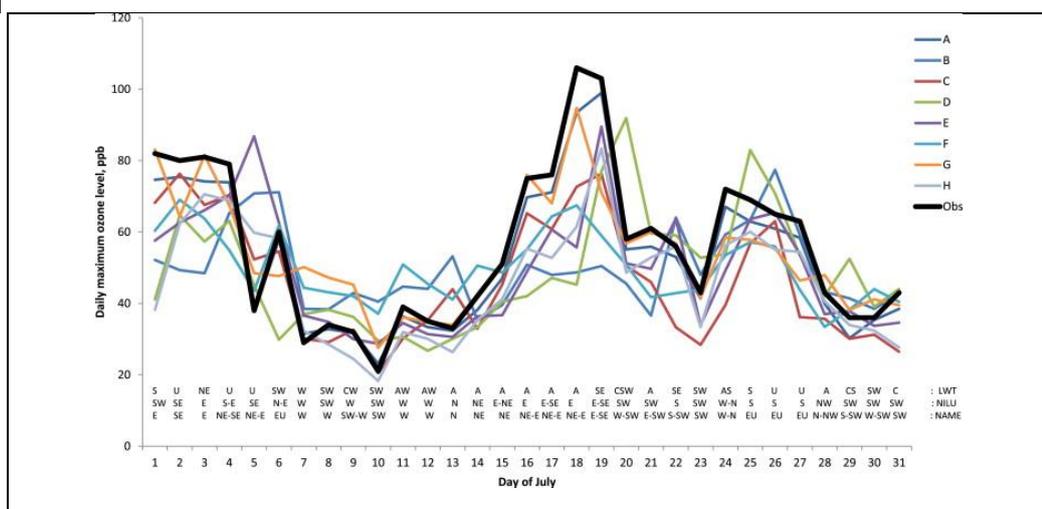


Figure 3435 - Daily maximum hourly ozone concentrations for eight models A-H in a comparison exercise against observations for July 2006 at Harwell, Oxfordshire, UK. Also shown are the daily advection regimes as Lamb Weather types (LWT), NILU FLEXTRA trajectories (NILU) and NAME air history maps (NAME), see (Derwent et al., 2014)(Derwent et al., 2014)

3455
3456 With respect to regional models Kukkonen et al. (Kukkonen et al., 2012)(Kukkonen et al., 2012) have
3457 highlighted the most prominent gaps of knowledge for chemical weather forecasting models; these
3458 include emission inventories, the integration of numerical weather prediction and atmospheric
3459 chemical transport models, boundary conditions and nesting of models, data assimilation of the
3460 various chemical species (see e.g. (Gaubert et al., 2014)(Gaubert et al., 2014)), improved
3461 understanding and parameterization of physical processes, better evaluation of models against data
3462 and the construction of model ensembles.

3463 It is clear that next generation models will push to greater resolution on the regional scale (Colette et
3464 al., 2014)(Colette et al., 2014).

3465

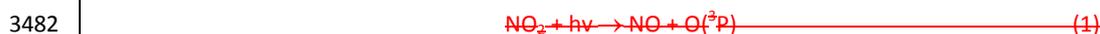
3466 4.7 Lightning

3467

3468 Globally, lightning flashes occur about 50 times per second, equal to 4.3 million times per day and
 3469 roughly 1.5 billion times per year. Lightning flashes dissociate N₂ molecules, leading to NO production
 3470 (~~Schumann and Huntrieser, 2007~~)(Schumann and Huntrieser, 2007), a key source of NOx in much of
 3471 the troposphere, especially the tropical upper troposphere (Schumann and Huntrieser, 2007;Grewe,
 3472 2008;Grewe et al., 2012a;Levy et al., 1996).(Schumann and Huntrieser, 2007;Grewe, 2008;Grewe et
 3473 al., 2012a;Levy et al., 1996). More than 80% of summertime upper tropospheric NOx above the
 3474 eastern United States is produced by lightning (Cooper et al., 2009)(Cooper et al., 2009) and can be
 3475 significant contributor to surface ozone (Hudman et al., 2009)(Hudman et al., 2009). Lightning NOx
 3476 (NO₂) has been detected from space (Beirle et al., 2010;Choi et al., 2005)(Beirle et al., 2010;Choi et
 3477 al., 2005). The lightning produced NO perturbs atmospheric composition in several ways relevant to
 3478 climate, as NO is an important determinant of OH/HO₂ ratios *via* the reaction:



3481 ~~The NO₂ photolyses to release an oxygen atom that can go on to form ozone:~~



3484 Given sufficient UV radiation and a supply of HO₂, NOx will efficiently generate O₃ (see section 2.1.1).
 3485 Figure ~~3536~~ shows modelled contribution of individual sectors in terms of NOx emissions, including
 3486 lightning, to the tropospheric ozone column (~~Grewe et al., 2012a~~)(Grewe et al., 2012a). Toumi et al
 3487 (~~Toumi et al., 1996~~)(Toumi et al., 1996) highlighted that O₃ production from lightning NOx (L_{NOx}) may
 3488 represent an important positive climate feedback, if a warmer world generates more lightning
 3489 (~~Reeve and Toumi, 1999~~)(Reeve and Toumi, 1999).

3490 However, in addition to leading to O₃ production, (~~see section 2.1.1~~), equation (~~3R3~~) enhances OH.
 3491 This increases the methane removal *via* the reaction:



3494 shortening the CH₄ lifetime, and hence reducing its atmospheric concentration, exerting reducing
 3495 radiative forcing (RF) (e.g. (~~Derwent et al., 2001~~)(Derwent et al., 2001);(~~Wild, 2007~~)(Wild et al., 2001);(~~Wild et al., 2001~~)).
 3496 (~~Wild, 2007~~)(Wild et al., 2001)).

3497 ~~Enhanced levels of OH will tend to increase production of all secondary aerosols (e.g., including~~
 3498 ~~sulphate), which would increase the magnitude of the negative radiative forcing owing to aerosols~~
 3499 ~~(e.g. (Stevenson et al., 2005)).~~

3500 ~~The catalytic cycle of O₃ production described by equations (1-3) is terminated when NOx is removed~~
 3501 ~~by the reaction:~~



3503 ~~This and other analogous reactions produce various forms of nitrates, including nitrate aerosol.~~
 3504 ~~Enhanced levels of nitrate aerosol, which typically scatters solar radiation, will exert a negative RF~~

3505 (e.g. (Shindell et al., 2009)), although specific studies of nitrate aerosol associated with L_{NOx} are
3506 apparently lacking.

3507 Any perturbations to NO_x (including lightning) will potentially lead to climate feedbacks *via* the
3508 biosphere, through deposition of NO₃ and O₃, and impacts on the carbon cycle (e.g. enhanced or
3509 reduced uptake of CO₂ by vegetation; (Sitch et al., 2007; Felzer et al., 2007)(Sitch et al., 2007; Felzer et
3510 al., 2007)). It is unclear if the net effect on CO₂ would result in a positive or negative RF.

3511 Most studies suggest more lightning (NO_x) in a warmer world (Schumann and Huntrieser,
3512 2007; Williams, 2005; Banerjee et al., 2014)(Schumann and Huntrieser, 2007; Williams, 2005; Banerjee
3513 et al., 2014), but these are typically based on the Price and Rind (Price and Rind, 1992)(Price and
3514 Rind, 1992) parametrization that links L_{NOx} emissions to cloud top height (raised to the power 4.9
3515 over land; to a lower power over ocean). Other parametrizations link L_{NOx} emissions to cloud ice (see
3516 e.g. (Finney et al., 2014; Tost et al., 2007)(Finney et al., 2014; Tost et al., 2007)), and as warming
3517 increases, ice declines, and so does LNO_x (Jacobson and Streets, 2009)(Jacobson and Streets, 2009).
3518 In addition, some schemes relate L_{NOx} to aerosols (Yuan et al., 2012; Venevsky, 2014)(Yuan et al.,
3519 2012; Venevsky, 2014), with potentially complex links between climate and L_{NOx} . With global
3520 warming, tropical stability increases (Held and Soden, 2006; Chou et al., 2013)(Held and Soden,
3521 2006; Chou et al., 2013) tending to reduce convection. However, the tropopause tends to rise,
3522 allowing convection to reach greater heights. In the Price and Rind (1992) scheme, the higher
3523 tropopause affect typically dominates, although there can be regional increases/decreases in some
3524 models (e.g., Stevenson et al., 2005).

3525 In summary, the literature suggests that L_{NOx} increases lead to higher levels of O₃, OH, nitrate, and
3526 secondary aerosols in general, but less CH₄, and have unknown impacts on CO₂. It remains unclear if
3527 the net impact of increases in L_{NOx} on climate is warming or cooling; it is also unclear if L_{NOx}
3528 represents a positive or negative climate feedback.

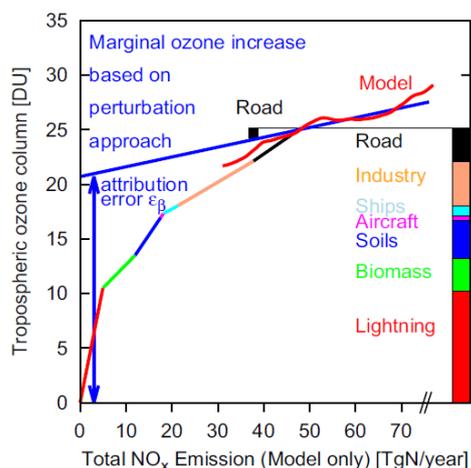


Figure 3536 - Contribution of individual sectors in terms of NO_x emissions to the tropospheric ozone column (Grewe et al., 2012b)(Grewe et al., 2012b).

3529 **4.8 Arctic**

3530 The Arctic is under-going rapid change as a result of global warming. This can, in part, be attributed
3531 to production of ozone from tropical and mid-latitude emissions of ozone precursors, especially
3532 methane. Climate change together with economic drivers, is also opening up the Arctic to new
3533 sources of pollution, such as shipping or oil/gas extraction which may lead to significant local or
3534 regional increases in surface ozone and associated impacts on Arctic air quality and deposition to
3535 fragile ecosystems (Law and Stohl, 2007)(Law and Stohl, 2007).

3536 Observed Arctic O₃ mixing ratios vary between 20 and 40 ppbv at the surface and increase with
3537 altitude up to the tropopause (8-10 km) (Hirdman et al., 2010)(Hirdman et al., 2010). Even though
3538 annual data on Arctic O₃ is rather limited, significant differences can be seen in the seasonal cycle at
3539 different locations, driven by both remote and local processes (see Figure 3637) (Hirdman et al.,
3540 2010)(Hirdman et al., 2010). Certain sites, such as Barrow (Alaska) or Alert (Canada), show evidence
3541 of halogen influenced depletion during the spring months. As discussed in section 4.4, halogen
3542 chemistry over sea-ice or snow covered regions can lead to very low or even near-zero O₃
3543 concentrations near the surface in the Arctic spring (e.g. (Barrie et al., 1988)(Barrie et al., 1988)).
3544 Other sites, such as Zeppelin, exhibit a spring maximum. Higher concentrations in the spring appear
3545 to be due to O₃ formation from precursors related to the breakdown of Arctic Haze at this time of
3546 year in the lower and middle troposphere (Emmons et al., 2003)(Emmons et al., 2003). Arctic Haze
3547 builds up during the winter each year as a result of transport of mid-latitude pollution into the
3548 polarPolar dome (e.g. (Barrie et al., 1981)(Barrie et al., 1981)) and contains elevated levels of O₃
3549 precursors (CO, VOCs, NO_x) as well as PAN, an important source of NO_x (e.g. (Hov et al., 1989)(Hov et
3550 al., 1989)).

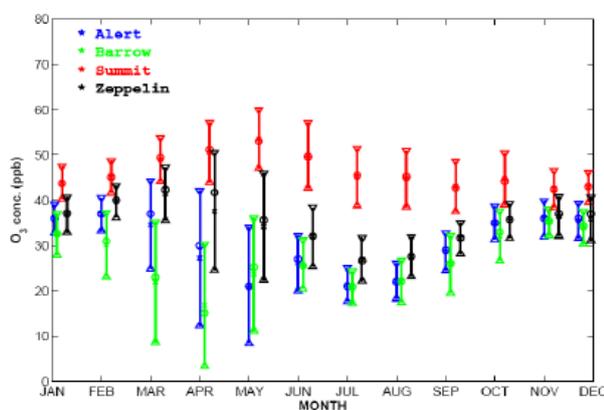


Figure 3637 - Seasonal cycle of O₃ at various surface sites in the Arctic. From (Hirdman et al., 2010)(Hirdman et al., 2010).

3551 Summit, which is at a higher elevation (3 km), on Greenland, has a late spring/ early summer
3552 maximum, likely owing to transport of polluted air masses, primarily from North America, but which
3553 could also include a contribution from snow NO_x emissions (e.g. (Grannas et al., 2007)(Grannas et al.,

3554 | [2007](#)) or from the stratosphere. There are indications that O₃ concentrations continue to increase in
3555 | the Arctic both at the surface and at higher altitudes in the troposphere. Hess and Zbinden ([Hess and](#)
3556 | [Zbinden, 2013](#))([Hess and Zbinden, 2013](#)) reported an increasing trend in the European Arctic middle
3557 | troposphere (500 hPa) of 0.36+/-0.23 ppb/yr from ozonesonde measurements over the period 1996-
3558 | 2010, and Oltmans et al. ([Oltmans et al., 2006](#);[Oltmans et al., 2013](#))([Oltmans et al., 2006](#);[Oltmans et](#)
3559 | [al., 2013](#)) reported a trend of 0.87+/-0.50 %/yr from surface measurements in 1992-2004 at Alert in
3560 | the Canadian Arctic.

3561 | Several studies have examined sources of pollution transported to the Arctic- ([Mauzerall et al., 1996](#)).
3562 | In a multi-model assessment, Shindell et al. ([Shindell et al., 2008](#))([Shindell et al., 2008](#)) examined
3563 | precursor emission sensitivities to emissions from different mid-latitude source regions on the Arctic
3564 | and found, for example, European CO emissions to be important in the winter. However, surface
3565 | Arctic O₃ in winter was most sensitive to European NO_x owing to the domination of strong titration
3566 | (O₃ removal) in air masses. This was also confirmed by Hirdman et al ([Hirdman et al., 2010](#))([Hirdman](#)
3567 | [et al., 2010](#)). Emissions from Asia and Europe have been found to be important sources of Arctic O₃
3568 | in spring in the free troposphere together with stratospheric injection although relative contributions
3569 | vary between studies and years ([Wespes et al., 2012](#);[Shindell et al., 2008](#))([Wespes et al.,](#)
3570 | [2012](#);[Shindell et al., 2008](#)). [Based on analysis of ARCTAS data \(Jacob et al., 2010\)](#), Wespes et al.
3571 | (2012) found that European anthropogenic emissions were important for lower tropospheric
3572 | summertime Arctic O₃ in contrast to a previous study ([Shindell et al., 2008](#))([Shindell et al., 2008](#)).
3573 | Production of O₃ from PAN decomposition in air masses transported from mid-latitudes has also
3574 | been identified as an important source of Arctic tropospheric ozone in the summer months ([Walker](#)
3575 | [et al., 2012](#))([Walker et al., 2012](#)). Indeed, aircraft observations collected during POLARCAT-IPY show
3576 | elevated PAN and CO concentrations in air masses transported from Asian and North American
3577 | anthropogenic emission regions in summer 2008 ([Law et al., 2014](#))([Law et al., 2014](#)) and references
3578 | therein). Boreal forest fires are also an important source of PAN and, due to their proximity to the
3579 | Arctic, plumes can be transported to high latitudes during the spring and summer months ([Brock et](#)
3580 | [al., 2011](#);[Singh et al., 2010](#))([Brock et al., 2011](#);[Singh et al., 2010](#)). Whilst little O₃ production appears
3581 | to occur close to boreal fires ([Alvarado et al., 2010](#);[Paris et al., 2010](#))([Alvarado et al., 2010](#);[Paris et al.,](#)
3582 | [2010](#)), several recent studies have shown O₃ production downwind from boreal fires in the Arctic
3583 | during the summer months ([Wespes et al., 2012](#);[Parrington et al., 2012](#);[Thomas et al., 2013](#))([Wespes](#)
3584 | [et al., 2012](#);[Parrington et al., 2012](#);[Thomas et al., 2013](#)). Nevertheless, O₃ production is higher in air
3585 | masses influenced by anthropogenic emissions.

3586 | However, global and regional models still struggle to capture vertical distributions of trace gases,
3587 | including ozone in the Arctic. CO concentrations are often underestimated even in multi-model
3588 | simulations carried out as part of the POLARCAT model inter-comparison project (POLMIP) using the
3589 | same emission datasets ([Monks et al., 2014](#);[Emmons et al., 2014](#))([Monks et al., 2014](#);[Emmons et al.,](#)
3590 | [2014](#)). Discrepancies appear to be related to either differences in oxidative capacity (OH) or vertical
3591 | transport of pollutants from mid-latitude source regions into the Arctic. Modelled concentrations of
3592 | NO_y species such as PAN or HNO₃ also show large variability and significant discrepancies compared
3593 | to measurements ([Arnold et al., 2014](#))([Arnold et al., 2014](#)) pointing to lack in our understanding
3594 | about chemical processing in polluted air masses transported to the Arctic (Law et al., 2014).

3595 | As a short-lived climate forcer, tropospheric O₃ contributes to Arctic warming. For example, Shindell
3596 | et al. ([Shindell et al., 2006](#))([Shindell et al., 2006](#)) estimated that anthropogenic emission increases

3597 since 1900 could be responsible for 25% of surface temperature changes in the Arctic. Shindell
3598 (~~Shindell, 2007~~)(~~Shindell, 2007~~), based on the results from one model, estimated that about 50% of
3599 the Arctic radiative forcing owing to ozone may be coming from O₃ produced at mid-latitudes
3600 (impacting poleward heat transport) and about 50% from ozone produced in the Arctic. New local
3601 sources of pollution may also impact Arctic O₃ in the future such as increased emissions from
3602 shipping (~~Granier et al., 2006~~)(~~Granier et al., 2006~~) or oil and gas production. Dalsøren et al.
3603 (~~Dalsoren et al., 2013~~)(~~Dalsoren et al., 2013~~) examined impacts of future shipping on radiative forcing
3604 in the Arctic and found, particularly in the high-end scenario (from (~~Corbett et al., 2010~~)(~~Corbett et~~
3605 ~~al., 2010~~)), that O₃ radiative forcing from shipping is important in the summer and transit season
3606 (May to October) when sea-ice is at a minimum.

3607 4.9 Unconventional oil and natural gas production: “fracking” and air quality

3608
3609 Fossil fuel energy production is rapidly transforming and expanding owing to unconventional oil and
3610 natural gas extraction techniques, with implications for regional-scale ozone production.
3611 Unconventional oil and natural gas is extracted from tight geological formations (such as sandstone,
3612 coal and shale) through the use of hydraulic fracturing (also known as fracking) and directional
3613 drilling (~~Field et al., 2014~~;~~Bickle, 2012~~)(~~Field et al., 2014~~;~~Bickle, 2012~~). These techniques allow wells
3614 to be drilled vertically into a shale formation and then horizontally through the formation, after
3615 which a mixture of water, sand and chemicals is pumped into the well at high pressure, fracturing the
3616 rock and allowing oil and gas to escape.

3617
3618 In the USA, the most extensive extractors of unconventional gas/oil, the shale plays (a name for the
3619 deposits) are located in many large basins across the country, primarily in the Rocky Mountains,
3620 Great Plains and the Appalachian Mountains. In terms of natural gas, the most productive shale
3621 plays are the Marcellus Shale (West Virginia, Pennsylvania, New York) (~~Kargbo et al., 2010~~)(~~Kargbo et~~
3622 ~~al., 2010~~) and the Haynesville-Bossier Shale (Texas and Louisiana). The greatest unconventional oil
3623 production is from the Bakken (North Dakota, Montana), Eagle Ford (Texas) and the Niobrara
3624 (Wyoming and Colorado) shale formations (~~Administration, 2014a~~);(~~Administration, 2014b~~).

3625
3626 In 2012 the USA produced 8.9 million barrels of oil per day (this includes crude oil, shale oil, oil sands
3627 and natural gas liquids), the third largest producer in the world, behind Russia and Saudi Arabia.
3628 However, the USA is the world leader in producing oil and natural gas from hydraulic fracturing with
3629 1.6 million barrels per day extracted from tight oil formations in 2012. Projections suggest that
3630 hydraulic fracturing will produce 4.5 million barrels per day by 2035, accounting for most of the US
3631 increase in oil production (~~BP, 2013~~)(~~BP, 2013~~). The US is poised to become the largest producer of
3632 liquid fuels in the world, primarily due to tight oil growth (~~BP, 2014b~~)(~~BP, 2014a~~).

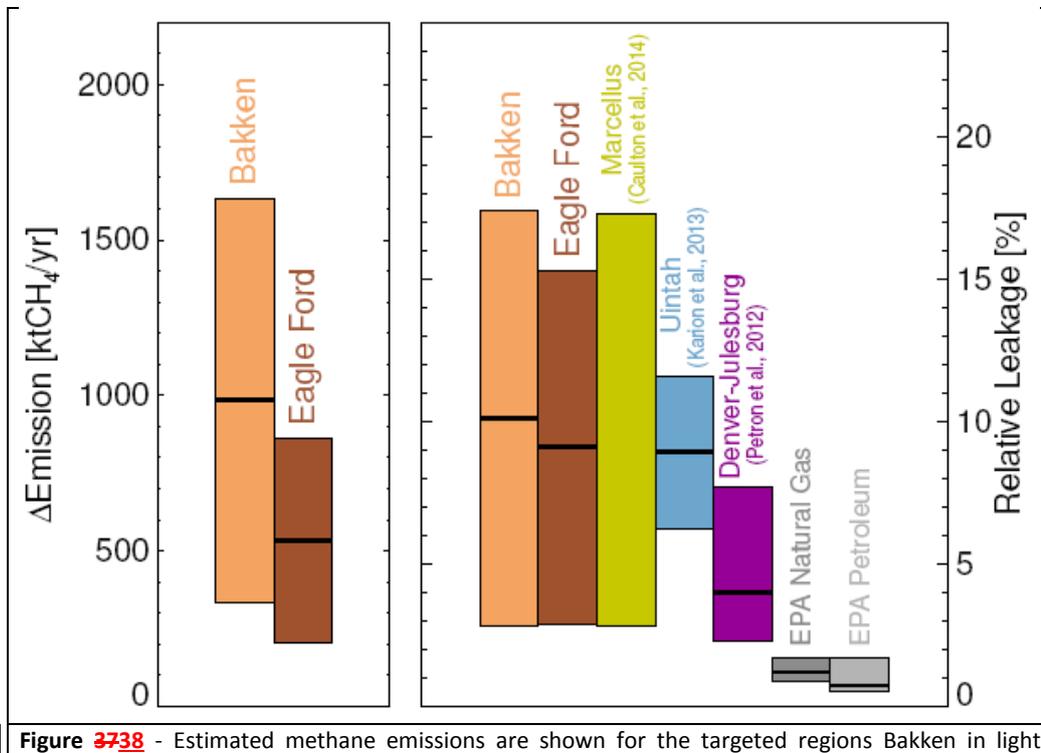
3633
3634 In 2012 the USA was the world’s largest producer of natural gas, producing 681 billion cubic meters
3635 (Bcm). US production of shale gas output is projected to rise from 255 Bcm/yr in 2012 (37% of total
3636 production) to 672 Bcm/yr in 2035 (~~BP, 2014a~~);(~~BP, 2014b~~). Shale gas was just 2% of US natural gas
3637 production in 2004 but by 2035 it will be 63%. However, these oil and natural gas projections are
3638 highly uncertain with the quantity of oil produced in 2040 differing by a factor of two under a
3639 scenario of high oil and gas resources vs a scenario of low oil and gas resources (~~Administration,~~
3640 ~~2014b~~)(~~Administration, 2014a~~).

3641
3642 With the increase in unconventional oil and natural gas has come an increase in fugitive emissions of
3643 hydrocarbons to the atmosphere in quantities great enough to influence local and regional-scale
3644 ozone production (~~Katzenstein et al., 2003;Kemball-Cook et al., 2010;Edwards et al.,~~
3645 ~~2014~~)(Katzenstein et al., 2003;Kemball-Cook et al., 2010;Edwards et al., 2014). This impact was
3646 revealed in an unexpected way during early 2008 when hourly average ozone mixing ratios exceeded
3647 140 ppbv during February in the rural Upper Green River Basin, of Wyoming (~~Schnell et al.,~~
3648 ~~2009~~)(Schnell et al., 2009). In situ measurements revealed that the high wintertime ozone levels
3649 were due to a strong temperature inversion that trapped NO_x and hydrocarbon emissions from the
3650 natural gas industry. The trapping of the emissions was facilitated by high mountains on three sides
3651 of the basin. A key contributor to the ozone formation was deep snow cover which reinforced the
3652 temperature inversion, and also reflected UV radiation back through the inversion layer, essentially
3653 doubling the amount of available UV radiation necessary for ozone production. This phenomenon
3654 has also been observed in the Uintah Basin, Utah, impacted by both oil and natural gas extraction
3655 (~~Edwards et al., 2013;Edwards et al., 2014~~)(Edwards et al., 2013;Edwards et al., 2014). Multiple years
3656 of data from these two regions demonstrate that without deep snow cover, the high ozone values do
3657 not occur (~~Oltmans et al., 2014~~)(Oltmans et al., 2014). Recent work in the UK context (~~Sommariva et~~
3658 ~~al., 2014~~)(Sommariva et al., 2014) has shown the range and difference in UK shale VOC profiles.
3659
3660 The impact of emissions from the oil and natural gas industry on ozone outside of these rural,
3661 mountain-ringed basins is less certain. For example, field studies have been carried out across the
3662 Denver-Julesburg Basin in north-eastern Colorado to quantify emissions from the oil and natural gas
3663 industry but accurate estimates for some gases are complicated by emissions from nearby urban
3664 areas (~~Pétron et al., 2012~~)(Pétron et al., 2012). However, relationships between particular VOCs can
3665 be used to clearly distinguish oil and natural gas emissions from urban emissions. VOC
3666 measurements from a site on the northern edge of the Denver metropolitan area were analysed to
3667 demonstrate that more than 50% of the VOC-OH reactivity was attributable to emissions from oil and
3668 natural gas operations, indicating that these emissions are a significant source of ozone precursors
3669 (~~Gilman et al., 2013~~)(Gilman et al., 2013).
3670
3671 Several recent studies using chemical transport models suggest that emissions from the oil and
3672 natural gas industry can produce local and regional scale ozone enhancements (~~Kemball-Cook et al.,~~
3673 ~~2010;Rodriguez et al., 2009;Carter and Seinfeld, 2012;Olague, 2012~~)(Kemball-Cook et al.,
3674 ~~2010;Rodriguez et al., 2009;Carter and Seinfeld, 2012;Olague, 2012~~). The reliability of these
3675 modelling studies is limited by the emissions inventories which are difficult to accurately produce
3676 owing to the heterogeneity of sources: type of gases emitted from a shale play; number of active
3677 well heads; integrity of infrastructure; emissions associated with well-completion vs. well operation;
3678 usage of venting or flaring (~~Field et al., 2014~~)(Field et al., 2014). The most recent U.S. EPA estimates
3679 indicate that NO_x emissions from US petroleum and related industries only amounted to 5% of total
3680 US anthropogenic NO_x emissions in 2013, but doubled between 2004 and 2013 (~~Agency,~~
3681 ~~2014~~)(Agency, 2014). Likewise VOC emissions increased by a factor of four over the same period,
3682 accounting for 14% of total US anthropogenic VOC emissions in 2013. However, these U.S. EPA
3683 estimates are highly uncertain. The U.S. EPA recently stated that it had not anticipated the
3684 tremendous nationwide growth in this sector and that it has limited directly-measured air emissions
3685 data from several important oil and gas production processes. The report recommends that the EPA

3686 produce a comprehensive strategy for improving air emissions estimates for oil and natural gas
 3687 production (Agency, 2013)(Agency, 2013). Until the emission inventories can be improved great
 3688 uncertainty will surround model estimates of the impact of the oil and natural gas industry on ozone
 3689 pollution.

3691 One final consideration of the potential for the oil and gas industry to affect tropospheric ozone is
 3692 through its impact on global methane levels. Methane leaks from the oil natural gas industry may be
 3693 a contributing factor to the renewed increase in global methane concentrations (Nisbet et al.,
 3694 2014)(Nisbet et al., 2014). For example, hydraulic fracturing in Utah may locally leak 6 to 12% of gas
 3695 production to the air (Karion et al., 2013)(Karion et al., 2013). Figure 37 shows data from a series of
 3696 studies including satellite remote sensed data that all show significant methane leakage beyond
 3697 official EPA estimates (Schneising et al., 2014;Kort et al., 2014). However, owing to the great
 3698 uncertainty of many aspects of global methane emissions, especially from US unconventional oil and
 3699 natural gas activities, much more research is required to understand the impact of unconventional oil
 3700 and natural gas activities on global methane concentrations (Brandt et al., 2014). Kang et al (Kang et
 3701 al., 2014) have highlighted the issue of abandoned wells and their significance. Figure 38 shows data
 3702 from a series of studies including satellite remote sensed data that all show significant methane
 3703 leakage beyond official EPA estimates (Schneising et al., 2014;Kort et al., 2014). However, owing to
 3704 the great uncertainty of many aspects of global methane emissions, especially from US
 3705 unconventional oil and natural gas activities (see e.g. (Allen et al., 2013)), much more research is
 3706 required to understand the impact of unconventional oil and natural gas activities on global methane
 3707 concentrations (Brandt et al., 2014).

3708



brown, and Eagle Ford in dark brown from satellite remote sensing measurements (Schneising et al., 2014)(Schneising et al., 2014). Shown are absolute emission ~~increase~~increases (2009–2011 relative to 2006–2008) in the left panel, and the leakage ~~raterates~~ relative to production in the right panel, with the 1 σ uncertainty ranges. For comparison, leakage estimates from previous studies in Marcellus (2012) (Caulton et al., 2014)(Caulton et al., 2014), Uintah (2012) (Karion et al., 2013)(Karion et al., 2013), Denver-Julesburg (2008) (Pétron et al., 2012)(Pétron et al., 2012) . EPA bottom-up inventory estimates for natural gas and petroleum systems (2011) are shown for comparison (EPA, 2014)(EPA, 2014).

3709

3710 4.10 Radical Chemistry, Radical changes

3711

3712 Radicals are central to the chemistry of the atmosphere; from the destruction of O₃ in the
3713 stratosphere, to the production and destruction of O₃ in the troposphere, radicals drive atmospheric
3714 composition change (Monks, 2005)(Monks, 2005). The inorganic HO_x radicals (OH, HO₂) are regarded
3715 by many in the field of atmospheric chemistry as the most influential of all radicals, as such a great
3716 amount of effort has gone into understanding their impacts and fate in the atmosphere (Heard and
3717 Pilling, 2003;Monks, 2005;Stone et al., 2012)(Heard and Pilling, 2003;Monks, 2005;Stone et al.,
3718 2012).

3719 Recent laboratory and modelling studies have shifted attention to improved understanding of the
3720 fate and role of organic radicals. The organic radicals of importance to atmospheric chemistry can be
3721 classified as organic peroxy radicals (RO₂) (section 2.1.1), organic oxy radicals (RO) and the so called
3722 Criegee intermediates (CI), a class of bi-radical compounds believed to be formed mainly from the
3723 reaction of O₃ with alkenes.

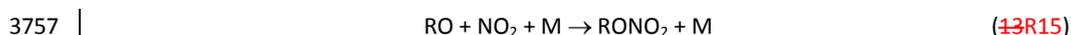
3724

3725 In the following section we briefly review the main aspects of the chemistry of organic radicals and
3726 highlight the latest discoveries in their chemistry. Significant use of the review of Orlando and Tyndall
3727 (Orlando and Tyndall, 2012)(Orlando and Tyndall, 2012) is made and for further details we refer the
3728 reader to their work.

3729 RO₂ are produced in the atmosphere by the OH (and NO₃) initiated oxidation of VOCs. Once formed
3730 RO₂ are lost *via* reactions with NO, HO₂ and other RO₂. Whilst reactions with halogens (X) and
3731 halogen oxides (XO) have been known about for several decades, their importance for inclusion in
3732 studies of tropospheric O₃ chemistry has traditionally been expected to be small (see section 4.4 for
3733 more details). The RO₂ + NO reaction is known to have two product channels. The first forming NO₂,
3734 hence propagating tropospheric O₃ formation; the second channel leads to the production of an
3735 organic nitrate (RONO₂) *via* a complex rearrangement, the exact mechanism of which is still uncertain
3736 (Dibble, 2008)(Dibble, 2008). We will not dwell on the chemistry of RONO₂ here, but suffice it to say
3737 these moieties allow O₃ precursors to be transported over great distances owing to their much
3738 longer atmospheric lifetime than NO₂. The kinetics of the RO₂ + NO reaction limit the RO₂ lifetime to
3739 only a few seconds when [NO] > 1 ppbv (see section 3.1 of Orlando and Tyndall 2012 for references).
3740 Broadly speaking the RO₂ + NO reaction can be classed as the most important of the RO₂ loss
3741 reactions to include for modelling O₃ production in the troposphere. The products of the reaction
3742 between RO₂ and HO₂ depend strongly on the structure of the RO₂ (Orlando and Tyndall,
3743 2012)(Orlando and Tyndall, 2012). Traditionally the reaction between RO₂ and HO₂ has been thought

3744 of as a radical sink, hence limiting the propagation of RO₂ + NO reactions and so reducing the
3745 potential O₃ production. The major product of this reaction for an alkyl RO₂ (R'CHOO) is an organic
3746 hydroperoxide (ROOH), a compound that is predicted to be lost from the atmosphere *via* deposition
3747 or aqueous uptake faster than its photodissociation can reform precursor radicals. The kinetics of the
3748 self (RO₂ + RO₂) and cross reactions (RO₂ + R'O₂) of RO₂ (see section 3.5 of Orlando and Tyndall 2012
3749 for references) limit the importance of these reactions to laboratory studies and parts of the
3750 atmosphere where concentrations of RO₂ are high (e.g. high BVOC emission regions).

3751 For RO₂ to have a big impact on the composition and chemistry of the atmosphere they must
3752 propagate radical production. RO are produced almost exclusively as products of the reactions of RO₂
3753 with NO. In general RO are very reactive and either undergo unimolecular decomposition (on the
3754 time scale of milliseconds) or react with O₂ producing HO₂ radicals (see section 2.1.1). Archibald et al.,
3755 ~~(Archibald et al., 2007)~~(Archibald et al., 2007) used a box model to investigate the importance of the
3756 reaction:



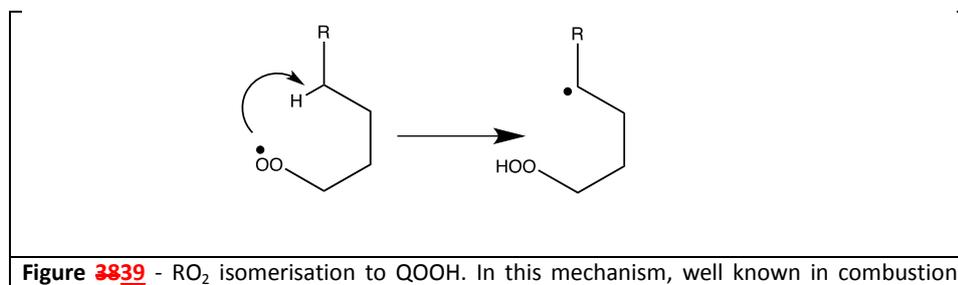
3758 and concluded that for CH₃O, reaction 13R15 could be a significant source of CH₃ONO₂ under the high
3759 NO₂ conditions found in many megacities, but that the unimolecular decomposition and reaction of
3760 O₂ with RO limits the importance of reaction 13R15 to CH₃O only.

3761 Owing to very significant disagreement between model simulations and observations of HO_x radicals
3762 in pristine environments (Lelieveld et al., 2008;Whalley et al., 2011)(Whalley et al., 2011;Lelieveld et
3763 al., 2008), a number of recent studies have focused on trying to better understand the role of NO_x
3764 free radical propagating chemistry for RO₂. In pristine environments the loss of RO₂ was traditionally
3765 thought to be dominated by the radical terminating reaction between RO₂ and HO₂. However, for
3766 acyl RO₂ (RC(O)OO) recent work has shown that the reaction of RO₂ with HO₂ has a major radical
3767 propagating product channel generating OH and RO₂ (see section 3.4 of Orlando and Tyndall 2012 for
3768 references). Based on this evidence Lelieveld et al., (2008) postulated that the reaction of RO₂ with X
3769 (where X reacted with a rate coefficient similar to that for RO₂ + NO) propagated radicals and was
3770 able to reconcile the model measurement disagreement for HO_x in the Amazon. However, Lelieveld
3771 et al. (2008) were unable to provide direct evidence for the structure of the unknown reaction
3772 partner, X. In much earlier modelling work, Frost et al., ~~(Frost et al., 1999)~~(Frost et al., 1999)
3773 speculated that near-IR absorption by RO₂ could lead to intramolecular conversion and yield HO_x
3774 radicals. Whilst a great deal of work has been performed identifying the absorbance features of RO₂
3775 in the near IR (e.g. (Kline and Miller, 2014)(Kline and Miller, 2014)), to date there has been almost no
3776 laboratory evidence of HO_x formation following absorption by RO₂ in the near-IR. However,
3777 Maccarone et al ~~(Maccarone et al., 2013)~~(Maccarone et al., 2013) have recently shown that
3778 arylperoxy radicals (RO₂ derived from aromatic hydrocarbons) are able to photo-dissociate in the
3779 visible spectrum to yield O(³P), and hence produce O₃ in the troposphere, without the need for NO_x.
3780 As highlighted by the work of Lelieveld et al. (2008) and Frost et al (1999), modelling experiments are
3781 increasingly being used to help direct laboratory studies in the search for radical propagating
3782 reactions. Archibald et al. ~~(Archibald et al., 2009)~~(Archibald et al., 2009) investigated the possible
3783 reaction between RO₂ and OH using a simple box model of the marine boundary layer (MBL). In their
3784 study Archibald et al. (2009) suggested three different mechanisms for the RO₂ + OH reaction yielding
3785 three unique product sets, two propagating radicals and the third leading to the formation of

3786 alcohols. The reaction they modelled was, at the time, very speculative and as such no kinetic studies
3787 had been performed. As such Archibald et al. (2009) used the kinetics of $RO_2 + X$ and $RO_2 + XO$
3788 reactions as analogy. Their model calculations concluded that the $RO_2 + OH$ reaction would have little
3789 impact on HO_x under conditions encountered in the MBL. Recently Bossolasco et al., (~~Bossolasco et~~
3790 ~~al., 2014~~)(Bossolasco et al., 2014) have measured the direct kinetics for the reaction between RO_2
3791 and OH and have shown that it is extremely fast ($k \approx 2.8 \pm 1.4 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$), potentially twice as fast as
3792 the upper limit used by Archibald et al. 2009. The importance of this reaction under conditions
3793 similar to those found by Lelieveld et al. (2008) is yet to be explored.

3794 Whilst unimolecular RO_2 isomerisations have been known about in combustion chemistry (where $T >$
3795 600 K) for several decades (e.g. (~~Hughes et al., 1992~~)(Hughes et al., 1992)), the importance of this
3796 process for the loss of RO_2 in the troposphere was thought insignificant - until recently. The RO_2
3797 isomerisation proceeds *via* internal H atom abstraction from the terminal oxygen of the $-OO$ group
3798 to produce what in combustion chemistry is referred to as a QOOH (see Figure 3839). Using *ab initio*
3799 calculation, Peeters et al. (~~Peeters et al., 2009~~)(Peeters et al., 2009) and Da Silva et al. (~~Silva et al.,~~
3800 ~~2009~~)(Silva et al., 2009) independently suggested a mechanism for RO_2 isomerisations in the
3801 oxidation of isoprene that they postulated could help improve the model-measurement mismatch
3802 reported by Lelieveld et al. (2008) for HO_x in pristine conditions. The focus on isoprene peroxy
3803 radicals has led to a number of new discoveries in the lab as well as from a theoretical point of view.
3804 Thanks, largely to developments in mass spectrometry and other analytical techniques, new products
3805 (isoprene epoxydiols – IEPOX; hydroperoxyaldehydes - HPAlds) have been elucidated, and their
3806 mechanisms and impacts on reconciling the “ HO_x problem” tested (~~Crouse et al., 2011; Paulot et~~
3807 ~~al., 2009~~)(Crouse et al., 2011; Paulot et al., 2009). In their work on updates to the chemical
3808 mechanism of isoprene oxidation, Archibald et al. (~~Archibald et al., 2010~~)(Archibald et al., 2010)
3809 reviewed several of the proposed mechanisms and concluded that whilst the isomerization reactions
3810 proposed by Peeters et al (2009) had the most promise in reconciling the model-measurement
3811 disagreement, the reported parameters could not be fully reconciled with atmospheric observations
3812 of other species and existing laboratory data without some degree of parameter refinement and
3813 optimization, which would probably include a reduction in the peroxy radical isomerisation rates and
3814 a consequent reduction in the OH enhancement. Indeed, Crouse et al. (~~Crouse et al.,~~
3815 ~~2011~~)(Crouse et al., 2011) provided the first laboratory evidence for the RO_2 isomerisations in the
3816 isoprene system but derived rate coefficients for the processes that were much smaller than those
3817 calculated by Peeters et al. (2009). However, modelling studies using the kinetics derived from their
3818 study suggested that 10-20% of isoprene RO_2 would undergo unimolecular isomeriations (Crouse et
3819 al., 2011) and as such this is an important process to include.

3820



chemistry, the terminal O atom of the RO₂ abstracts a labile H (through the formation of a ring structured intermediate).

3821

3822 An astonishing amount has been learnt about the atmospheric chemistry of the Criegee
3823 Intermediates (CI) over the last few years. Pioneering work carried out at the [synchrotron](#) Advanced
3824 Light Source ([Taatjes et al., 2008](#))([Taatjes et al., 2008](#)) provided the first detection and subsequent
3825 direct measurements of the kinetics of the CI ([Welz et al., 2012](#))([Welz et al., 2012](#)). Traditionally CI
3826 have been thought to have been formed exclusively from the ozonolysis reaction of alkenes (as
3827 originally proposed by ([Criegee, 1948](#))([Criegee, 1948](#))). However, the recent laboratory studies on
3828 small CI have utilized novel chemical routes, which avoid the reaction with O₃. For example, a large
3829 number of groups have used CH₂I₂ as a reagent for the formation of CH₂OO (*via* photolysis and
3830 reaction with O₂). It should be noted that CH₂I₂ one of the major iodine containing VOCs emitted in
3831 the marine boundary layer ([Saiz-Lopez et al., 2011](#))([Saiz-Lopez et al., 2011](#)) and may be a direct
3832 source of CI in this environment. McCarthy et al. ([McCarthy et al., 2013](#))([McCarthy et al., 2013](#)) have
3833 shown that CH₂OO can also be produced *via* passing a mixture of CH₄ and O₂ through an electric
3834 discharge (through what remains an as yet unknown mechanism). Based on this evidence McCarthy
3835 et al. (2013) have postulated that it is likely that CH₂OO can be formed in the upper and free
3836 troposphere, given the large mixing ratios of CH₄ and O₂ in the upper troposphere and the relatively
3837 high density of electrical discharge (in the form of lightning flashes). Given that many of the reactions
3838 between O₃ and alkenes have small rate coefficients ($\sim 10^{-17} \text{ cm}^3 \text{ s}^{-1}$) it is important that these non-
3839 ozonolysis routes to CI production be quantified using model studies in order to better understand
3840 the burden of CI in the troposphere- (see [Figure 40](#)).

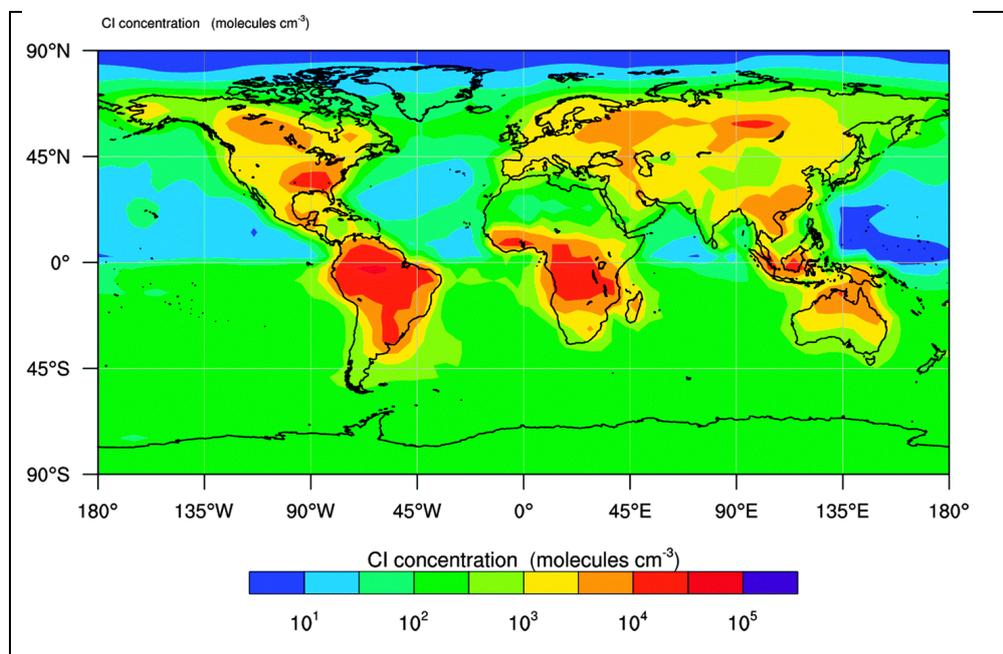


Figure 3940 – Global surface averages model concentrations of the Criegee Intermediate

concentration ~~(Taates et al., 2014)~~(Taates et al., 2014).

3841

3842 Of fundamental importance to understanding the chemistry of CIs is knowledge of their physical
3843 properties. McCarthy et al. (2013) have used a combination of methods to determine the geometry
3844 of CH₂OO, in good agreement with the work of Su et al. ~~(Su et al., 2014)~~(Su et al., 2014). As well as
3845 now having great insight into the geometry of CH₂OO, a number of studies have shed light on the
3846 absorption spectrum of CH₂OO in the UV/vis (e.g. ~~(Beames et al., 2012; Sheps, 2013)~~(Beames et al.,
3847 ~~2012; Sheps, 2013~~) Beames et al., (2012) and Sheps (2013) have shown that there is strong
3848 absorption by CH₂OO in the 320-400nm range. Whilst this may have implications for additional loss
3849 of CH₂OO by photolysis, the large cross section in this region has the benefit of making cavity
3850 ringdown spectroscopy laboratory studies of the kinetics of CH₂OO (and other CIs) possible – opening
3851 up a number of avenues for greater understanding of these ephemeral but crucial intermediates in
3852 the oxidation of VOCs in the atmosphere.

3853

3854 What seems to be emerging from the plethora of laboratory studies on the kinetics of CI reactions is
3855 that they react very fast! For example, Su et al. ~~(Su et al., 2014)~~(Su et al., 2014) have shown that the
3856 bimolecular self-reaction of CH₂OO has a rate constant near the gas kinetic limit ($k = 4 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$).
3857 Su et al. (2014) determined that the reaction proceeds *via* a CH₂OO dimer – where the zwitterionic
3858 character of the CH₂OO allows for barrierless addition of the terminal O atoms with the central C
3859 atom. This dimer is predicted to dissociate to produce two CH₂O (formaldehyde) and O₂ in its
3860 excited electronic state (O₂(¹Σ_g)). Such a fast self-reaction has implications for the analysis of previous
3861 laboratory studies in this system. Similarly fast kinetics have been observed for CIs reacting with
3862 organic acids. Using a combination of time resolved laboratory experiments, Welz et al. (2014) ~~(Welz~~
3863 ~~et al., 2014)~~(Welz et al., 2014) have very recently shown that the reactions of the C1 and C2 CIs with
3864 HCOOH and CH₃COOH are several orders of magnitude faster than previously inferred from alkene
3865 ozonolysis reactions ($k \sim 1.0 \times 10^{-10} \text{ cm}^3 \text{ s}^{-1}$). Although products of the reaction were not detected, it is
3866 likely that highly condensable products will form, potentially contributing to SOA formation and
3867 growth. Although it is interesting to note that the CI seem more reactive than RO₂ with a number of
3868 compounds, it is paramount to understand the dominant loss processes of the CI in order to
3869 understand their abundance. Before many of these direct kinetic experiments were performed it was
3870 widely assumed that in the troposphere the reaction with H₂O would dominate over other loss
3871 reactions, in spite of slow (although uncertain) kinetics. Welz et al. ~~(Welz et al., 2012)~~(Welz et al.,
3872 ~~2012~~) reported an upper limit for the CH₂OO + H₂O reaction rate coefficient ($k = 4 \times 10^{-15} \text{ cm}^3 \text{ s}^{-1}$).
3873 Relative rate experiments have yielded a number of other estimates of the rate coefficient for this.
3874 Ouyang et al. ~~(Ouyang et al., 2013)~~(Ouyang et al., 2013), who studied the reaction of NO₂ with
3875 CH₂OO, estimated $k = 2.5 \times 10^{-17} \text{ cm}^3 \text{ s}^{-1}$. In their study, Ouyang et al. were able to show that the
3876 reaction produces NO₃ – the most important oxidant at night. So far inclusion of this reaction into
3877 modelling studies has not been performed and estimates of the importance of this process for night-
3878 time chemistry are needed. Based on detection of HCHO, Stone et al. ~~(Stone et al., 2014)~~(Stone et al.,
3879 ~~2014~~) were able to put an upper limit on the reaction of the CI with H₂O of $k = 9 \times 10^{-17} \text{ cm}^3 \text{ s}^{-1}$,
3880 significantly smaller than the estimates for this reaction by Welz et al. (2012). They suggested that
3881 this much lower reactivity may mean that previous conclusions from modelling studies where H₂O

3882 was calculated as the dominant loss process may need to be revised. Combining the results of Welz
3883 et al. (2014) and Stone et al. (2014) we speculate that it is possible that in many environments where
3884 organic acids are present at the ppb level, reaction with these acids may be the dominant loss
3885 process for Cl. Interestingly, Taatjes et al. ([Taatjes et al., 2013](#))([Taatjes et al., 2013](#)) have shown that
3886 the *anti*-CH₃CHOO Cl reacts with H₂O very fast ($k = 1.0 \times 10^{-14} \text{cm}^3 \text{s}^{-1}$), suggesting that the lack of
3887 reaction between CH₂OO and H₂O may not be representative of all Cl.

3888

3889 5 Policy context

3890

3891 Within the policy context, much of the focus on ozone has been on ozone as an air pollutant (e.g.
3892 [\(OECD, 2012; EEA, 2007, 2009, 2011; Royal Society, 2008; Fowler et al., 2013a\)](#)([OECD, 2012; EEA, 2007,](#)
3893 [2009, 2011; Royal Society, 2008; Fowler et al., 2013a](#)). As recently stated the aim of much policy with
3894 respect to ozone and air quality is “to achieve levels of air quality that do not result in unacceptable
3895 impacts on, and risks to, human health and the environment” ([Fowler et al., 2013a](#))([Fowler et al.,](#)
3896 [2013a](#)) (see also section 3) within some measure of reasonable cost. This process requires an
3897 understanding of the messages from research and monitoring activities to develop new insights.
3898 Policy, to date, requires methods of assessing compliance to metrics that quantify impact. Owing to
3899 the transboundary nature of ozone, much focus has been on regional and hemispheric impact of
3900 metrics and mitigation.

3901 Throughout this section certain metrics will be detailed, the main two of which are AOT40, the
3902 seasonal accumulated exposure above 40 ppb (80 µg/m³) during daylight hours, this is normally
3903 expressed as a cumulative exposure (ppb h or ppm h) and SOMO35 the sum of the amounts by which
3904 maximum daily 8-hour concentrations of ozone (in µg/m³) exceed 70 µg/m³ (35 ppb) on each day in a
3905 calendar year.

3906 5.1 Policy Metrics for ozone

3907

3908 Ozone is a powerful oxidant which can cause adverse effects on human health, and vegetation. As a
3909 result, air quality standards for ozone have been established to mitigate these effects. Ozone can also
3910 damage some materials, particularly rubber and plastics, but no standards specifically address these
3911 effects. Ozone is a unique pollutant in that different areas of the ozone concentration frequency
3912 distribution are affected by different mechanisms and hence could require significantly different
3913 policy responses. The averaging times of different ozone metrics are also of crucial importance for
3914 policy (see e.g. [\(Pappin and Hakami, 2013; Lefohn et al., 2010\)](#)([Pappin and Hakami, 2013; Lefohn et al.,](#)
3915 [2010](#))).

3916 Health-related standards have up to now been expressed as one hour or eight-hour averages, the
3917 latter arising originally from chamber studies of human exposures where the maximum effects were
3918 observed over exposures of around eight hours. Such short term peak ozone concentrations based
3919 on averaging times of the order of hours, such as those observed in ‘smog’ episodes, are formed
3920 from the well-known VOC/NO_x chemistry (see section 2.1.1). These reactions occur typically over
3921 timescales of hours to a few days, in conditions of low wind speed and strong sunlight, and hence the

3922 spatial scale of such episodes and the policy response area, is of the order of 100s to ~1,000
 3923 kilometres. Controls on precursor emissions across Europe are therefore required to mitigate these
 3924 peaks within European nations. Figure 4041 shows a comparison of a number of national and
 3925 international ozone air quality standards.

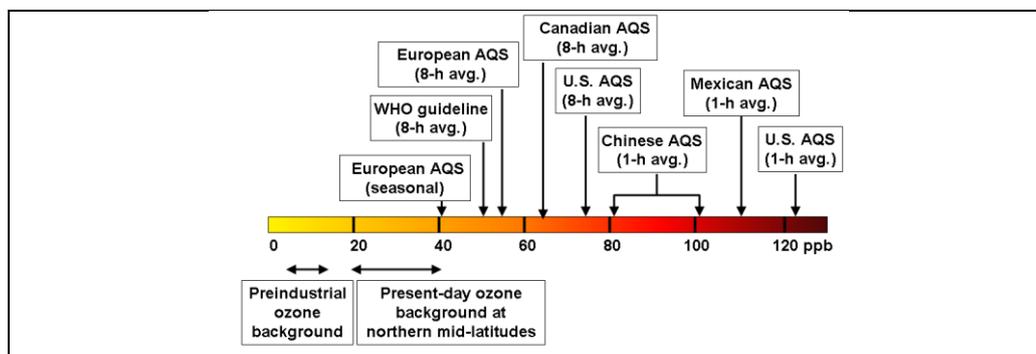


Figure 4041 – Ozone Air Quality Standards (AQS) in ppb. Different national and international standards are noted as well as estimates for northern midlatitudes of the preindustrial background (i.e., O₃ abundances with all anthropogenic emissions of NO_x, CO, VOC, and CH₄ switched off, and before current climate and stratospheric O₃ change) and the present-day baseline abundances (i.e., the statistically defined lowest abundances of O₃ in air flowing into the continents, typical of clean-air, remote marine sites) from (Council, 2009a)(Council, 2009a).

3926

3927 Effects on vegetation however are more closely related to longer term exposures and in the recent
 3928 EU Ambient Air Quality Directive (Directive 2008/50/EC) for example, the Target Value (see below)
 3929 for the protection of vegetation is expressed in terms of AOT40 as an average accumulated value
 3930 from May to July (defined in more detail later). This longer exposure period and averaging time, with
 3931 a threshold close to the tropospheric baseline ozone level means that emission controls would be
 3932 required over a much wider area than those to mitigate the health-related one/eight hour average
 3933 concentrations discussed above, and could also involve other, more long-lived precursors such as
 3934 methane.

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3935
 3936 There are several levels of ‘standard’ in use, with differing legal status. For example, the fundamental
 3937 health effect evidence is used to inform the setting of WHO Air Quality Guidelines, below which the
 3938 pollutant in question will not have adverse effects on public health. The Guidelines are set without
 3939 regard to the technological, economic or social issues which might affect their achievability. In setting
 3940 legally based standards however, some flexibility is often introduced to allow for difficulties of
 3941 achieving the levels and/or to allow for year-to-year meteorological variability. For short-term
 3942 standards this usually takes the form of allowing a number of exceedences of a given concentration
 3943 over a year, and/or averaging over several years as in the case of the USA. Some health standards in
 3944 place around the world are shown in Table 3.

3945
 3946 **Table 3** – Comparison of world health standards for ozone

Country/region	Ozone level (ppb)	Averaging time	Nature of level
----------------	-------------------	----------------	-----------------

WHO	50	Daily max. 8-hr mean	Guideline ¹
EU	60	8 hour running	Target value ²
	60	Average	Long Term Objective
USA	75	8 hour	National standard ³ primary and secondary
California	90	8 hour	State standard not to be exceeded
	70	1 hour	
UK	50	8 hour	Objective ⁴
Canada	63	8 hour	2015
	62	8 hour	2020 Both 'voluntary guidelines'
Mexico	80	8 hour	5 th maximum taken over 1 year
	110	1 hour	
China	80	1 hour	Grade I
	100	1 hour	Grades II and III (Liu et al., 2013b) (Liu et al., 2013b)
India	50	8 hour	
	90	1 hour	

3947

3948 Although in some cases the 'headline' concentration value in different countries' standards may be
3949 similar, the number of allowed exceedences of short term levels is crucially important in determining

¹ WHO also quotes 120ppb daily max. 8 hr mean as having 'significant health effects' and gives 80ppb daily max. 8 hr mean as an interim target.

² EU value not to be exceeded on more than 25 days per calendar year averaged over 3 years.

³ USA value – the 3-year average of the fourth-highest daily maximum 8-hour average measured at each monitor in an area must not exceed 160µg/m³/0.075 ppm

⁴ UK Objective maximum 8-hour running mean in a day, not to be exceeded more than 10 times per year

3950 their relative stringency. These criteria are shown on the footnote to the Table 3. Moreover, the
3951 status of the standards is also very important. In the USA, the National Ambient Air Quality Standard
3952 (NAAQS) for ozone has the same mandatory legal status as those for the other criteria pollutants. In
3953 the EU however this is not the case as it was recognised that no single Member State could control
3954 the ozone levels measured within its territory because of the transboundary nature of ozone
3955 formation during 'smog' episodes (Royal Society, 2008)(Royal Society, 2008). In the EU Directives
3956 therefore, the ozone standard is a Target Value as opposed to a mandatory Limit Value in the case of
3957 the other pollutants. A Limit Value is 'to be attained within a given period and not to be exceeded
3958 once attained'. Target Values in contrast, are 'to be attained *where possible* over a given period.' The
3959 mandatory aspects of ozone control in the EU are contained within the National Emission Ceilings
3960 Directive which sets mandatory emission ceilings for individual Member States for NO_x and VOCs,
3961 which are designed, *inter alia*, to achieve reductions in ozone levels.

3962 There are also standards in place to protect against damage to vegetation. In the EU Directive there
3963 is a Target Value of 18,000 µg/m³ · hours for AOT40, defined as the sum of the difference between
3964 observed hourly concentrations greater than 80 µg/m³ (≈ 40 parts per billion) and 80 µg/m³ over a
3965 given period using only the one-hour values measured between 8.00 and 20.00 Central European
3966 Time (CET) each day. In the US the 8- hourly NAAQS in Table 3 is also designed to protect against
3967 damage to vegetation, so the spatial scale of policy responses to protect against vegetation damage
3968 in the EU and the USA are potentially quite different- (see <http://www.epa.gov/ncea/isa/>). In the last
3969 decade, the LRTAP Convention (Convention on Long-range Transboundary Air Pollution) has
3970 introduced stomatal flux-based critical levels for vegetation that are species-specific and relevant for
3971 protecting against effects of ozone on food security, carbon sequestration and timber production,
3972 and biodiversity (see Section 3.2 and Mills et al. (Mills et al., 2011a)for further details).

3973 Two further recent developments have important implications for policy responses for mitigating
3974 ozone concentrations. First, the work of the CLRTAP (Convention on Long-range Transport of
3975 Atmospheric Pollutants) Task Force on Hemispheric Transport of Air Pollution has ~~shown~~highlighted
3976 that intercontinental transport of ozone and its precursors can make significant contributions to the
3977 exceedance of air quality standards and can even cause exceedances in their own right (HTAP,
3978 2010).(HTAP, 2010). This has already led to challenges to air quality standard enforcement in
3979 California (Hand, 2014)(Hand, 2014). In the case of Europe this suggests that emission reduction
3980 strategies should take into account measures and policies in North America. Likewise, plans to
3981 reduce ozone in North America should take into account emissions in Asia.

3982 The second development relates directly to potentially new metrics to protect human health from
3983 adverse effects of ozone and the existence or otherwise of a no-effect threshold. The recent review
3984 of the health effect literature by WHO (WHO, 2013)(WHO, 2013) has concluded that there is now
3985 evidence showing associations between long term (summer mean) ozone concentrations and
3986 respiratory mortality, and weaker associations with cardiorespiratory mortality. WHO recognised the
3987 uncertainties in this area but nonetheless suggested that long-term average WHO Guidelines and a
3988 long-term (possibly a summer mean) Target Value should be considered by the European
3989 Commission.

3991 In parallel with these conclusions, the REVIHAAP report (WHO, 2013)(WHO, 2013) also noted that
3992 evidence for a no-effect threshold for short-term impacts was inconclusive but recommended that

3993 SOMO10 as well as SOMO35 should be used in health impact assessments. These findings are
3994 significant for future policy on ozone. Long-term (summer) averages, as noted earlier, are partly
3995 determined by hemispheric or global emissions; to attain a threshold of 10 ppb hourly average in
3996 SOMO10 would also imply reductions of emissions on a global scale. If the evidence behind these
3997 emerging conclusions strengthens then the geographical scale of ozone reductions strategies will
3998 need to be extended beyond the regional level to hemispheric or global scales (~~Hsu et al., 2013~~)(Hsu
3999 et al., 2013).

4000

4001 5.2 Ozone mitigation and baseline ozone

4002

4003 Observational evidence suggests that baseline ozone concentrations as they effect Europe have been
4004 rising up until about 2000 (~~Wilson et al., 2012; Parrish et al., 2009; Simmonds et al., 2004; Jenkin,~~
4005 ~~2008; Derwent et al., 2006~~)(Wilson et al., 2012; Parrish et al., 2009; Simmonds et al., 2004; Jenkin,
4006 2008; Derwent et al., 2006), though currently they appear to be falling (Logan et al., 2012; Parrish et
4007 al., 2012). An increasing background contribution to European ozone levels could represent a
4008 substantial future challenge to the attainment of ozone limit values (~~Derwent et al., 2010~~)(Derwent
4009 et al., 2010). With an increasing background contribution there is a requirement to control more
4010 ozone of anthropogenic European origin to achieve the limit. Models have shown (~~Derwent et al.,~~
4011 ~~2010; Szopa et al., 2006~~)(Derwent et al., 2010; Szopa et al., 2006) that the benefit to European
4012 emission controls can be significantly counterbalanced by increasing background ozone. These
4013 results were confirmed ~~for in~~ a long-term perspective (2050) ~~in by~~ Colette et al. (2013) ~~that who~~
4014 emphasized that air quality legislation was indeed anticipated to bear its fruit in reducing ozone
4015 exposure in the future, even if under a business as usual scenario such as the RCP8.5, climate change
4016 and long range transport ~~constituted a heavy penalty~~.

4017 Similarly, in the US context there has been much debate as to the contribution and quantification of
4018 background ozone and its effect on the attainment of standards (~~Lefohn et al., 2014; Fiore et al.,~~
4019 ~~2014~~)(Lefohn et al., 2014; Fiore et al., 2014; Prather et al., 2003; Fiore et al., 2002). Lefohn et al (2014)
4020 have explored the concept of an emissions-influenced background in an attempt on a continental
4021 scale to partition the controllable ozone (see Figure 4142). Fiore et al (~~Fiore et al., 2014~~)(Fiore et al.,
4022 2014) noted that the largest model differences in the calculation of North American Background
4023 ozone, in this case a model construct, were linked to differences in contributions from the
4024 stratosphere, wildfires, lightning and isoprene chemistry. Prather et al. noted in a model study that
4025 the background ozone might be a northern hemispheric problem given future emission scenarios
4026 (Prather et al., 2003).

4027 The recent results of the 2013 revision (~~Amann et al., 2013~~)(Amann et al., 2013a) of the European
4028 Thematic Strategy on Air Pollution (TSAP) indicate that the reduction of 10% of the 25,000
4029 anticipated ~~death~~deaths attributed to ozone ambitious by the 2005 TSAP for European emission
4030 reductions should be safely achieved in 2025 with about 18,000 annual premature ~~death~~deaths
4031 under the scenario currently planned.

4032 Fowler et al, in their recent report on research findings in support of the EU air quality policy argued
4033 that “more attention needs to be given to treating ozone as a hemispheric transboundary issue”
4034 ([Fowler et al., 2013a](#))([Fowler et al., 2013a](#)) (see section 5.3).

4035

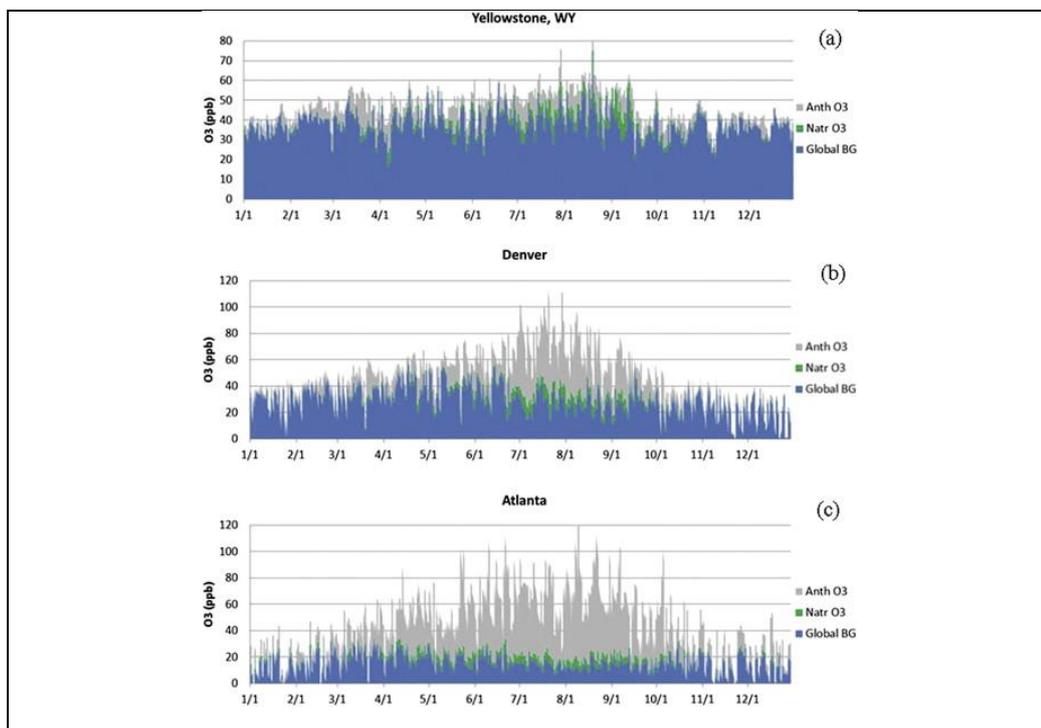


Figure 4142 - Modeled contributions of hourly GBO₃ (global tropospheric O₃ plus stratospheric O₃), natural (Natr O₃), and anthropogenic (Anth O₃) adding to total hourly O₃ for a) Yellowstone NP, b) Denver and c) Atlanta ([Lefohn et al., 2014](#))([Lefohn et al., 2014](#)).

4036

4037 5.3 Hemispheric transport of ozone and it precursors in the policy context

4038

4039 Substantial efforts have been undertaken to quantify the importance of long-range transport of
4040 ozone from distant source regions as part of UNECE HTAP ([HTAP, 2010](#))([HTAP, 2010](#)). More than 20
4041 global models conducted idealised source-receptor experiments to quantify hemispheric transport of
4042 ozone and its precursors and the key findings are reported in Fiore et al ([Fiore et al., 2009](#))([Fiore et
4043 al., 2009](#)) and HTAP ([HTAP, 2010](#))([HTAP, 2010](#)). 20% reductions in anthropogenic precursor
4044 emissions from North American, East Asian and South Asian sources reduce annual mean EU O₃ by
4045 about 0.4, 0.2 and 0.1 ppbV, respectively ([HTAP, 2010](#))([HTAP, 2010](#)). The influence of LRT has a
4046 seasonality, with the largest impact in spring (March-April: ~0.8 ppbV) and minimum impact in late
4047 summer (July-September: ~0.45 ppbV) ([HTAP, 2010](#))([HTAP, 2010](#)). This influence may appear small,
4048 but the equivalent O₃ response to a 20% emission change over Europe itself is 1.5-2.0 ppbV in
4049 summertime, and is close to zero on a regional mean basis in winter when titration is important. It is

4050 worth noting that the HTAP emission reductions (20%) have been exceeded in reality over Europe for
4051 the 1990-2009 period (EEA, 2011)(EEA, 2011). By scaling the 20% emission changes to the actual
4052 regional emission changes and accounting for the nonlinearity in ozone responses, (Wild et al.,
4053 2012)(Wild et al., 2012) derived the ozone trend over Europe from 1960-2000 along with the
4054 contributions from the EU, external sources (i.e. LRT) and changing atmospheric CH₄. An increase of
4055 about 6.5 ppbV is calculated between 1960 and 1990, and a slight decrease from 1990-2000. More
4056 than half of the 1960-1990 trend came from non-European sources (2.1 ppb) and CH₄ (1.6 ppb), with
4057 only 2.8 ppb from changes in EU emissions. Interestingly trans-Eurasian transport of air pollutants has
4058 been implicated in the enhancement of ozone in western China (Li et al., 2014b).

4059 A number of studies have focussed on the contribution of increasing precursor emissions over Asia to
4060 ozone over North America. Reidmiller et al. (Reidmiller et al., 2009)(Reidmiller et al., 2009) have
4061 used the HTAP simulations to demonstrate that precursor emissions from both East Asia and Europe
4062 influence the policy-relevant maximum 8-hour average (MDA8) ozone metric, but note that regional
4063 emission controls over North America are 2-10 times as effective at reducing this ozone metric as the
4064 equivalent controls in these foreign regions. However, Lin et al., 2012, However, Lin et al. (Lin et al.,
4065 2012a) demonstrate that Asian emissions may contribute as much as 8-15 ppb ozone to MDA8 in the
4066 south-western US in springtime on days when ozone exceeds 60 ppb, indicating the important role
4067 that long-range transport may play in ozone exceedences. More recent work by Lin et al. (Lin et al.,
4068 2014a)(Lin et al., 2014a) has shown that transport of the export-related Chinese pollution
4069 contributed 0.5-1.5% of ozone over the western United States in 2006. This Chinese pollution also
4070 resulted in one extra day or more of noncompliance with the US ozone standard in 2006 over the Los
4071 Angeles area and many regions in the eastern United States. Equivalent studies focussing on air
4072 quality metrics in Europe have yet to be performed, but are likely to show smaller impacts given the
4073 greater transport distances from regions showing substantial emission increases.

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4074 Transport within Asia has been considered in a recent HTAP study showing that O₃ from East Asian
4075 sources affects the most densely populated parts of South Asia (Chakraborty et al., 2015).

4076 5.4 Impacts of climate change

4077

4078 Jacob and Winner (Jacob and Winner, 2009)(Jacob and Winner, 2009) have undertaken a
4079 comprehensive review of the impact of climate change on air quality, including ozone. Despite
4080 regional differences, the dominating signal is one towards an increase of ozone levels induced by
4081 global warming (Stevenson et al., 2006), that led Wu et al (Wu et al., 2008) to coin the term "climate
4082 penalty". The possible pathways for such a penalty include increasing continental temperatures,
4083 changing atmospheric humidity and changes in the prevalence of stable, anticyclonic conditions
4084 trapping pollutants in the boundary layer and possibly leading to higher surface ozone even without
4085 changes in anthropogenic precursor emissions. These meteorological factors may be supplemented
4086 by climate driven changes in biogenic emissions of isoprene and in dry deposition of ozone. It is
4087 worth noting that in the lower free troposphere and in remote surface regions ozone is expected to
4088 decrease owing to increased water vapour in a warmer world (Jacob and Winner, 2009), and more
4089 recently von Schneidmesser et al (von Schneidmesser et al., 2015) have undertaken a
4090 comprehensive review of the impacts of climate change on air quality, including ozone. Despite
4091 regional differences, the dominating signal is one towards an increase of ozone levels induced by

4092 global warming (Stevenson et al., 2006), that led Wu et al (Wu et al., 2008) to coin the term “climate
4093 penalty”. The possible pathways for such a penalty include increasing continental temperatures,
4094 changing atmospheric humidity and changes in the prevalence of stable, anticyclonic conditions
4095 trapping pollutants in the boundary layer and possibly leading to higher surface ozone even without
4096 changes in anthropogenic precursor emissions. These meteorological factors may be supplemented
4097 by climate-driven changes in biogenic emissions of isoprene and in dry deposition of ozone. It is
4098 worth noting that in the lower free troposphere and in remote surface regions ozone is expected to
4099 decrease owing to increased water vapour in a warmer world (Jacob and Winner, 2009). There is
4100 some debate as to the magnitude of any climate penalty e.g. Tai et al. (Tai et al., 2013) indicates that
4101 in the presence of CO₂-isoprene interaction, the projected change in isoprene emission by 2050 will
4102 be largely offset or even reverse in sign, leading to much reduced sensitivity of ozone and SOA (by
4103 >50%) to climate and natural vegetation

4104 Bloomer et al. (~~Bloomer et al., 2009~~)(Bloomer et al., 2009) estimated the magnitude of these effects
4105 from surface ozone observations over the US at about 2.2 ppb per degree C- and Pfister et al have
4106 shown increasing summertime ozone levels (Pfister et al., 2014) under various climate scenarios.
4107 These effects have been quantified over Europe in several model studies (~~Colette et al., 2013;Meleux~~
4108 ~~et al., 2007;Langner et al., 2012a;Langner et al., 2012b;Andersson and Engardt, 2010;Manders et al.,~~
4109 ~~2012;Hedegaard et al., 2008;Hedegaard et al., 2013b;Katragkou et al., 2011~~)(Colette et al.,
4110 2013;Meleux et al., 2007;Langner et al., 2012a;Langner et al., 2012c;Andersson and Engardt,
4111 2010;Manders et al., 2012;Hedegaard et al., 2008;Hedegaard et al., 2013b;Katragkou et al., 2011).
4112 The general order of magnitude of the climate penalty over Europe is a few ppb by the middle of the
4113 century. Langer et al (~~Langner et al., 2012c~~)(Langner et al., 2012b) point out a larger increase at the
4114 95th percentile of hourly ozone, suggesting that the effects of climate change may be particularly
4115 important during high ozone events. The effects of changes in isoprene emissions and dry deposition
4116 have been quantified separately by Andersson and Engardt (~~Andersson and Engardt,~~
4117 ~~2010~~)(Andersson and Engardt, 2010) who find increases in mean ozone of up to 5 ppb by 2050 in
4118 some parts of western Europe, with as much as 60% of this change attributed to decreased dry
4119 deposition, and with increased isoprene emissions contributing up to 1 ppb. As noted above large
4120 uncertainties remain regarding the impact of climate change on biogenic emissions, with up to a
4121 factor five difference in isoprene change reported in a multi-model ensemble (~~Langner et al.,~~
4122 ~~2012b~~)-(Langner et al., 2012c).

4123 There are few studies that proposed quantitative comparisons of the relative impact of climate
4124 change and air pollution mitigation strategies on surface ozone. Existing investigations all agree on a
4125 larger impact of air quality policies(~~Tagaris et al., 2007;Hedegaard et al., 2013a;Colette et al., 2013~~)
4126 (~~Tagaris et al., 2007;Hedegaard et al., 2013a;Colette et al., 2013~~), endorsing to the efficiency of
4127 currently planned mitigation strategies, despite external penalties brought about by climate change
4128 and long range transport.

4129 It should be noted however that studies focusing on the ~~impacts~~impacts of climate change use long-
4130 term projections (typically 2050) which have substantive precursor mitigation. Given the magnitude
4131 of the response to precursors changes, the sensitivity to the choice of the scenario is worth noting
4132 (~~Fiore et al., 2012a~~)(Fiore et al., 2012a). Pioneering studies relied on projections originally designed
4133 for climate projections (SRES (~~Nakicenovic et al., 2000~~)(Nakicenovic et al., 2000) or RCPs (~~van Vuuren~~
4134 ~~et al., 2011~~)(van Vuuren et al., 2011), but the ozone precursor information in such scenarios was

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4135 ~~solely provided to assess radiative forcing and their use for surface air quality projections constitutes~~
4136 ~~a deviation from their original purpose. The use of emission projections relying on policy relevant~~
4137 ~~emissions factors such as the Global Energy Assessment (Riahi et al., 2012), the ECLIPSE (Klimont et~~
4138 ~~al., 2013a; Klimont et al., 2013b) or PEGASOS datasets are more reliable.~~

4139 , but the ozone precursor information in such scenarios was solely provided to assess radiative
4140 forcing and their use for surface air quality projections constitutes a deviation from their original
4141 purpose. The various scenarios make differing assumptions for future air pollution emissions and
4142 therefore, describe a wide range of future emissions over large world regions. Any downscaling in
4143 regions that exhibit large spatial heterogeneities could be problematic leading to inaccurate results
4144 (Amann et al., 2013b). The use of emission projections relying on policy relevant emissions factors
4145 such as the Global Energy Assessment (Riahi et al., 2012), the ECLIPSE (Klimont et al., 2013a) or
4146 PEGASOS datasets are more reliable.

4147 While the implications of these effects for surface ozone and air quality policy are substantial, there
4148 is still considerable uncertainty in the magnitude of these indirect climate effects, and improved
4149 understanding of the processes controlling these atmosphere-land surface interactions is needed.

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4152 6 The future – Air Quality and Climate

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4154 Recently, pollutants that typically fall under the ‘air quality’ categorization have been receiving
4155 increasing attention for their role in climate change and their impact on radiative forcing. More
4156 specifically, significant focus has been on ozone, methane (as a GHG and an ozone precursor), and
4157 aerosol constituents (mostly the black carbon (BC) component). Significant potential exists for co-
4158 benefits with coordinated air quality and climate policies. A number of reviews have recently
4159 addressed various aspects of the linkages between air quality and climate, from the chemistry
4160 interactions to the policy implications (~~von Schneidemesser and Monks, 2013;Isaksen et al.,~~
4161 ~~2009;Fiore et al., 2012b;Unger, 2012).~~(~~von Schneidemesser and Monks, 2013;Isaksen et al.,~~
4162 ~~2009;Fiore et al., 2012b;Unger, 2012).~~ For more details, see these reviews. Here, a brief overview of
4163 the topic will be given, with a focus on ozone and the latest developments.

4164 In addition to being an air pollutant with significant adverse health effects, ozone is also a
4165 greenhouse gas. A recent estimate of ozone’s radiative forcing effect (from pre-industrial times to
4166 the present day) is $+0.40 \text{ W m}^{-2}$ (~~Myhre et al., 2013~~)(~~Myhre et al., 2013~~), other estimates include
4167 $+0.44$ and $+0.23$ (~~Unger, 2012;Fiore et al., 2012b).~~(~~Unger, 2012;Fiore et al., 2012b~~). For comparison,
4168 the RFs attributed to methane and CO_2 are $+0.48 \text{ W m}^{-2}$ and $+1.66 \text{ W m}^{-2}$, respectively (~~IPCC,~~
4169 ~~2007~~)(~~Myhre et al., 2013~~). The ozone precursors NO_x , NMVOCs, and CO, have little to no direct
4170 effect on climate, but influence climate and radiative forcing through their effects on ozone and
4171 methane, as well as atmospheric oxidant concentrations and indirect ecosystem effects [~~von~~
4172 ~~Schneidemesser and Monks, 2013~~](~~von Schneidemesser and Monks, 2013~~) and references therein].
4173 Reductions in NMVOCs and CO tend to be synergistic and result in overall decreases in RF, although
4174 the effects can be fairly minor, especially for NMVOCs (~~Collins et al., 2013~~)(~~Collins et al., 2013~~). The
4175 effect of NO_x on climate/radiative forcing is much less straightforward. A variety of interactions,
4176 feedbacks, and ecosystem effects confound the picture for NO_x , resulting in significant uncertainty
4177 and often times competing effects on climate. A variety of modelling studies have evaluated the
4178 various direct and indirect effects (~~Collins et al., 2010;Collins et al., 2013;Shindell et al., 2009;Fry et~~
4179 ~~al., 2012;Colette et al., 2011)~~(~~Collins et al., 2010;Collins et al., 2013;Shindell et al., 2009;Fry et al.,~~
4180 ~~2012;Colette et al., 2011~~) and a summary can be found in von Schneidemesser and Monks (~~von~~
4181 ~~Schneidemesser and Monks, 2013~~)(~~von Schneidemesser and Monks, 2013~~). More research is needed
4182 on this topic.

4183 Methane, an important greenhouse gas in its own right, is also an ozone precursor. With a lifetime of
4184 approximately a decade in the atmosphere, methane is well-mixed globally and therefore has a
4185 significant influence on background ozone levels. Methane is a distinct win-win possibility for air
4186 quality and climate in that reductions in methane emissions would decrease ozone, including
4187 baseline ozone, and thereby lessen adverse impacts on vegetation and human health, but also
4188 benefit climate by reducing 2 GHGs simultaneously (~~Fry et al., 2012;Shindell et al., 2012;Isaksen et~~
4189 ~~al., 2014)~~(~~Fry et al., 2012;Shindell et al., 2012;Isaksen et al., 2014~~). A model study investigated the air
4190 quality (O_3) and climate benefits of methane reductions and found that O_3 reductions were relatively
4191 linear with respect to reductions in methane emissions. In addition, the CH_4 emission reductions did
4192 not depend strongly on location, as the climate and air quality benefits were realized globally, which

4193 | would allow for the most cost effective emission controls to be implemented (~~Fiore et al.,~~
4194 | ~~2008~~)([Fiore et al., 2008](#)).

4195 | However, the interactions go both ways. Not only does ozone affect the climate, but changes owing
4196 | to climate change will also influence ozone production. This effect is known as the 'climate penalty'.
4197 | A variety of effects resulting from a changing climate will potentially influence ozone concentrations,
4198 | some increasing ozone, some decreasing ozone (~~Rasmussen et al., 2013~~)([Rasmussen et al., 2013](#)).
4199 | Many of these effects are associated with a significant amount of uncertainty. Climate changed
4200 | induced increases in methane emissions from wetlands, stratosphere-troposphere exchange of
4201 | ozone, lightning NO_x, and regional stagnation all consistently lead to increased ozone when
4202 | investigated, while increases in dry deposition and humidity consistently lead to decreases in ozone
4203 | (~~Fiore et al., 2012b; Isaksen et al., 2009~~)([Fiore et al., 2012b; Isaksen et al., 2009](#)). Other climate change
4204 | induced effects such as increased wildfires have a much more uncertain effect on ozone. For
4205 | example, for a regional study in California, climate-related perturbations (temperature, biogenics and
4206 | water vapour) led to combined peak 1 h ozone increases of up to 11 ppb (~~Millstein and Harley,~~
4207 | ~~2009~~)([Millstein and Harley, 2009](#)).

4208 | In a model ensemble study by Colette et al. (~~Colette et al., 2012~~)([Colette et al., 2012](#)) future (2030)
4209 | air quality in Europe owing to just air quality policy or air quality and climate policy was compared.
4210 | The base case included all current and planned air quality measures up to 2030 and was compared to
4211 | a 'sustainable' case with the air quality as well as climate measures (2° target and energy efficiency
4212 | improvements). Depending on the scenario annual mean O₃ was found to slightly increase over NO_x
4213 | saturated areas but the overall O₃ burden would decrease substantially, including exposure to
4214 | detrimental levels of O₃ for health (~~Kinney, 2008~~)([Kinney, 2008](#)). The air pollution measures were
4215 | responsible for the main improvements in ozone, but an additional co-benefit of at least 40%
4216 | (depending on the indicator) was attributed to the climate policy (~~Colette et al., 2012~~)([Colette et al.,](#)
4217 | [2012](#)). A study by Crawford-Brown et al. (~~Crawford-Brown et al., 2012~~)([Crawford-Brown et al., 2012](#))
4218 | focused on the co-benefits to O₃- and PM-related health effects resulting from implementation of
4219 | climate policy in Mexico. A base case scenario was compared to a decarbonisation scenario where
4220 | CO₂ emissions in Mexico would be reduced by 77% by 2050. The co-benefits to air quality were
4221 | reductions in O₃ of 11-13% by 2050. The reduction in non-fatal diseases from O₃ and PM related
4222 | health co-benefits was valued at \$0.6 billion per year (~~Crawford-Brown et al., 2012~~)([Crawford-Brown](#)
4223 | [et al., 2012](#)). Both of these studies show that there are significant benefits to pursuing coordinated
4224 | policies for air quality and climate, and not only for the improvement of O₃ related air quality. Rypdal
4225 | et al have commented on the challenges for putting tropospheric ozone in climate agreements
4226 | (~~Rypdal et al., 2005~~)([Rypdal et al., 2005](#)).

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4230 7 Conclusions

4231

4232 Ozone remains central to atmospheric chemistry as the initiator, propagator and product of
4233 photochemistry. Its influence is felt on human health, ecosystems and climate. Ozone though
4234 ubiquitous remains an enigma. In many places in the Northern Hemisphere mid-latitudes ozone in
4235 some senses remains a paradox, the high summertime peak levels decreasing but the regional
4236 background levels rising ([Fishman et al., 2014](#);[Parrish et al., 2012](#))([Fishman et al., 2014](#);[Parrish et al.,](#)
4237 [2012](#)). Globally this points to the need to treat ozone across the range of scales, a transboundary
4238 issue, but with an emphasis on the hemispheric scales ([Fowler et al., 2013a](#);[Simpson et al.,](#)
4239 [2014](#))([Fowler et al., 2013a](#);[Simpson et al., 2014](#)). Recent air pollution episodes in Europe have
4240 pointed to the continuing need to think about the how climate change policies interact with air
4241 quality policy and what can be learnt from current episodes ([Monks, 2014](#))([Monks, 2014](#)). There
4242 remain a number of clear challenges for ozone such as explaining surface trends, incorporating new
4243 chemical understanding, ozone-climate coupling as well as a better assessment of impacts.

4244

4245 With respect to the future, studying the atmospheric chemistry of ozone relies on the continuing
4246 interplay of laboratory studies of fundamental parameters being integrated with our best theories
4247 using numerical models and evaluated against in situ observations. This “three-legged” stool
4248 approach is generic, but is a vital model to use to understand not only ozone but the wider chemistry
4249 in the atmosphere ([Abbatt et al., 2014](#))([Abbatt et al., 2014](#)).

4250 One area implicit in this review, which hasn't been discussed explicitly, has been the role of
4251 observations in understanding, quantifying and describing ozone across the scales. Observations of
4252 ozone and its precursors underpin and drive the development of our knowledge base ([Laj et al.,](#)
4253 [2009](#))([Laj et al., 2009](#)). Bowman has argued that the global nature of pollution and climate change
4254 requires a new observing system for ozone ([Bowman, 2013](#))([Bowman, 2013](#)). A key feature of the
4255 system should be the ability to predict and attribute ozone to emissions. A combination of satellites,
4256 ground-based remote sensing and ground-based observations are needed with geostationary
4257 observations ([Lahoz et al., 2011](#))([Lahoz et al., 2011](#)) being an observational anchor (see Figure [4243](#)).

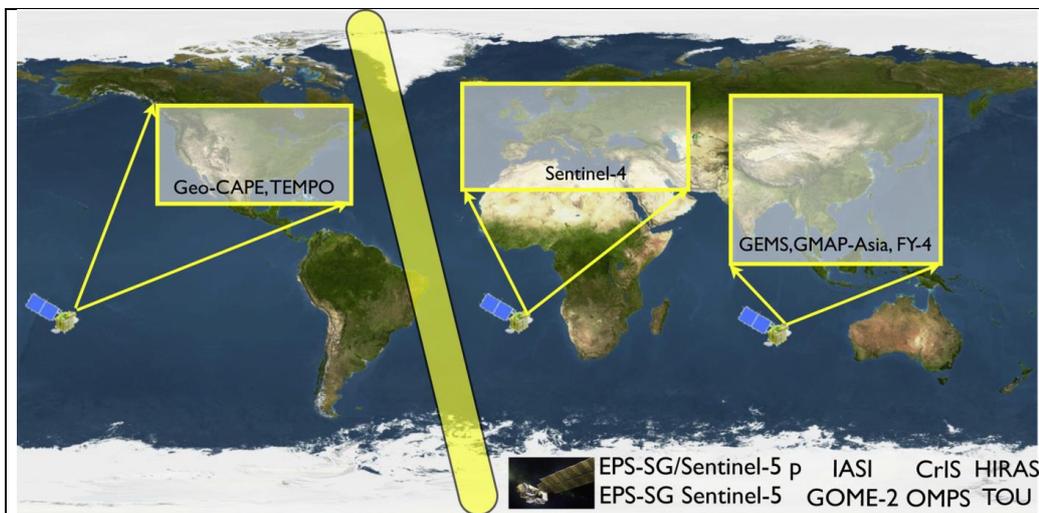


Figure 42 – Constellation of geostationary and low-earth orbiting satellites planned for the coming decade that will measure tropospheric ozone. Current instruments that measure tropospheric ozone, e.g., TES or OMI, but will not be continued in the future are not listed. A complete description of planned satellites is available at <http://www.wmo-sat.info/>. (From (Bowman, 2013))

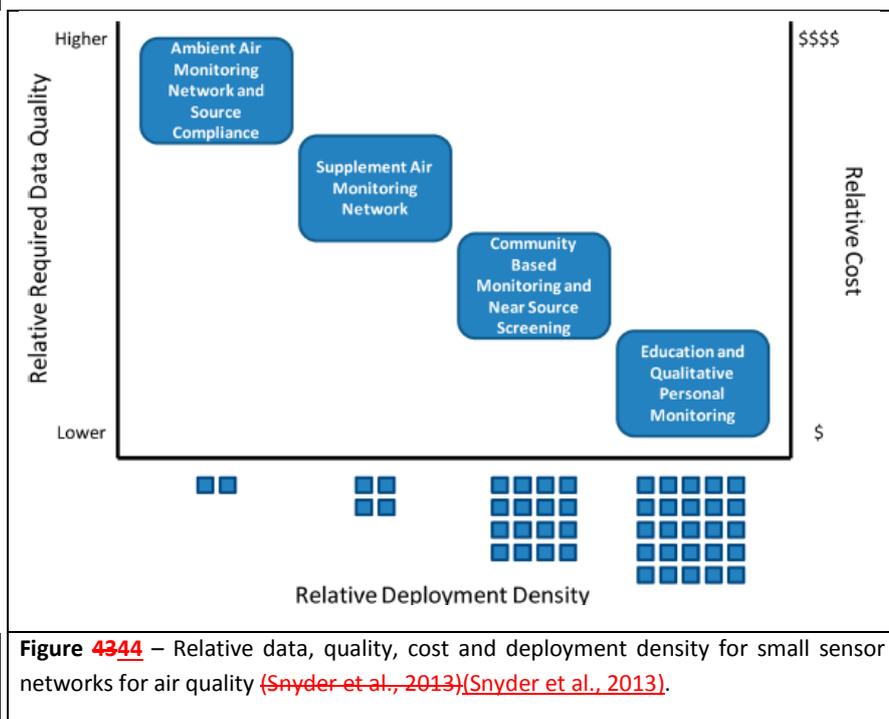
Figure 43 - Constellation of geostationary and low-earth orbiting satellites planned for the coming decade that will measure tropospheric ozone. Current instruments that measure tropospheric ozone, e.g., TES or OMI, but will not be continued in the future are not listed (From (Bowman, 2013)).

4258

4259 It is clear that space observations offer the global view and repeatability not easily available from
 4260 other observing systems. Measurement of ground-level ozone from space still remains a significant
 4261 challenge (Foret et al., 2014; Duncan et al., 2014)(Foret et al., 2014; Duncan et al., 2014). There is
 4262 future potential for global observations of tropospheric ozone from space either directly (Sellitto et
 4263 al., 2013)(Sellitto et al., 2013) or through a combination with models (Zoogman et al., 2014; Martin,
 4264 2008)(Zoogman et al., 2014; Martin, 2008). Duncan et al (Duncan et al., 2014)(Duncan et al., 2014)
 4265 have recently reviewed the application of satellite data to air quality application including common
 4266 mistakes to avoid.

4267 The growth of small sensors for a range of trace species including ozone has the potential to offer
 4268 insights on unheralded spatial and temporal timescales (Snyder et al., 2013; Mead et al.,
 4269 2013; Piedrahita et al., 2014)(Snyder et al., 2013; Mead et al., 2013; Piedrahita et al., 2014). As
 4270 illustrated in Figure 4344, there are potential new avenues opened up for air quality monitoring by
 4271 the deployment of extensive networks of these sensors: all the way down to personal sensing (e.g.
 4272 (Wheeler et al., 2011; Andersen et al., 2010)). The absolute measurement of ozone remains a
 4273 challenge with some of these sensors owing to interferences (Mead et al., 2013)(Mead et al., 2013).
 4274 New developments suggest these configurations of sensor technologies can give long-term
 4275 atmospheric performance for the measurements of ozone (Williams et al., 2013; Bart et al.,
 4276 2014)(Williams et al., 2013; Bart et al., 2014).

4277 There may yet be new links and biospheric feedbacks driven by ozone to be explored. For example,
 4278 the effect of elevated O₃ and whitefly herbivory significantly increased tomato volatiles, which
 4279 attracted *E. Formosa* wasps and reduced the whitefly feeding on tomatoes (Cui et al., 2014)(Cui et
 4280 al., 2014).



4281

4282 There are interesting attempts to make public outreach on the effects of ozone on plants and crops.
 4283 Fishman et al, have described a establishment of an “Ozone Garden” (Fishman et al., 2014)(Fishman
 4284 et al., 2014). The garden provides real-time measurements of O₃ concentrations as well as firsthand
 4285 observations of the detrimental effects of this pollutant. Meteorological data, as well as the O₃
 4286 concentrations from the monitor, are recorded and publicly disseminated in near-real time *via* the
 4287 internet.

4288 Looking forward it is clear that levels and patterns of global ozone will continue to change, impacting
 4289 global warming, air quality, global food production and ecosystem function. There remains a need for
 4290 continuing research to quantify impacts and interactions across all the scales. The issues around
 4291 tropospheric ozone are not “solved” (Guerreiro et al., 2014)(Guerreiro et al., 2014) and new data
 4292 continues to shed light on more aspects of ozone and its interactions in the global atmosphere.

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4298

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4302

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