

## ***Interactive comment on “Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer” by P. S. Monks et al.***

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

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

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

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

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

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

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

as well as text:

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P32765, L26 – 32766, L2: “The emissions from megacities also have an effect on the surrounding region. In many cases the urban areas have been identified to be VOC-limited, where high NO<sub>x</sub> concentrations in the cities suppress ozone concentrations (Im and Kanakidou, 2012; Tie et al., 2013). For example, a modeling study conducted in the context of the MILAGRO measurement campaign in Mexico City identified that reductions in VOC emissions led to decreases in maximum ozone concentrations while NO<sub>x</sub> emission reductions led to increased maximum ozone concentrations, demonstrating that the urban core of the Mexico City metropolitan area was VOC-limited – a conclusion supported by the in-situ measurements. The surrounding mountain/rural areas were identified as mostly NO<sub>x</sub>-limited, although the range of these areas was meteorologically dependent (Song et al., 2010). This suppression of ozone by high NO<sub>x</sub> has been identified as generally more characteristic of extratropical megacities in the Northern Hemisphere (despite the Mexico City example), relative to tropical cities, because of differences in transport patterns.”

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

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

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

#### Minor Comments

I include some additional minor comments below.

1. Overall: inconsistency of units; suggest using ppb throughout (and converting all

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references using ug/m<sup>3</sup>)

Done

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

Removed from introduction and merged with section 2.1

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

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

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

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

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

Section has been rewritten.

6. Section 2.1.2d: Obvious missing reference/discussion: Kurpius and Goldstein, GRL, 2003

Reference added

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7. Page 32729 lines 26-30 & 32730 lines 1-3: Specify whether these are surface, profile or column concentrations

Clarified - surface concentrations.

8. Page 32731, lines 1-3: This list of factors should also include meteorology beyond transport (rain, radiation, temperature, etc), as well as changing surface cover (e.g. vegetation).

Text amended to reflect this.

9. Page 32733, lines 19-22: This sentence requires a reference – unclear if the citation from the previous sentence applies here.

Wilson et al, (2012) and Colette et al (2011) references added.

10. Page 32745, lines 15-17: Sindelarova et al. is not an appropriate reference here (an application, not the model description). MEGAN v2.1 also includes an algorithm for CO2 inhibition of isoprene emissions.

Reference amended and text added

11. Page 32758, lines 16, 21: Remove the repetition of line 16 of line 21. The specific reference to rice is also odd, there are many more crops affected by ozone...a list of vegetation types affected and some references would be useful.

Removed odd sentence on rice and overhauled section to make it cover crops and vegetation affected by ozone.

“Typical ozone effects on plants include reduced growth, less seed production, lower functional leaf area and earlier leaf senescence. Data compilation studies have shown that many species of plants are sensitive to ozone, including: agricultural crops such as wheat, tomato, soybean and rice and salad crops such as lettuce, spinach and onion (Mills et al., 2007a); grassland species such as clover species, buttercup and harebell (Hayes et al., 2007, Mills et al., 2007b); and tree species such as beech,

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birch and Holm oak (Karlsson et al., 2007). These effects impact on the important ecosystem services provided by plants, including food security, carbon sequestration, timber production, and protection against soil erosion, avalanches and flooding.”

12. Page 32760, line 2: Tai et al. show that climate reduced global yields by 11% (not > 20%)

Modified (10%)

13. Page 32762, line 8-9: What does “the fundamental processes involved” mean? Please elaborate.

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

14. Page 32763, line 1-2: Needs reference.

Reference added (Zhu et al, 2012)

15. Page 32763, line 28: also domestic biofuel use?

Added

16. Page 32764: lines 25-29: needs a reference (particularly for the role that evaporative emissions played in this event)

Elansky et al. reference details this. New Zvyagintsev et al. reference added

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

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

18. Section 4.3: This might also be a good place to note the potential impacts of

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biofuels on ozone concentrations, via changes to vegetation and BVOC emissions (e.g. Ashworth et al., 2012; Porter et al., 2012).

Sentence added on biofuels and ozone/AQ.

19. Section 4.5: This section does not seem to be particularly relevant to a review of tropospheric ozone. The key elements of the role of NO<sub>x</sub> have been discussed elsewhere.

The aim of this section was to demonstrate the wider linkages of ozone chemistry through Nr to the nitrogen cycle. The premise being that these are often treated in isolation.

20. Section 4.7: The basic chemistry described in this section is redundant with earlier descriptions of ozone formation. Please harmonize.

Chemistry in this section has been cut-down

21. Section 4.7: the discussion of how lightning relates to aerosols is not relevant to this review.

Removed and section restructured.

22. Page 32794, lines 1-11: In addition to several studies which show high methane leakage from fracking operations, the authors should note the Allen et al., PNAS, 2013 study which provides the counter-example of low measured leakage rates.

Added Allen reference and also recent PNAS Kang (2014) reference

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

There has been much debate as to the effect on oxidation chemistry of the radical pathways detailed on section 4.10. The authors feel there is significant value in the section in full.

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24. Page 32803, lines 20-25: A number of studies identified the role of intercontinental transport of ozone in ozone exceedances prior to the publication of the HTAP report in 2010. As written, this sentence is a bit misleading.

Changes made to remove incorrect implication.

25. Page 32805, lines 14-18: unclear what scenario this mortality reduction corresponds to – is this the result of reductions of European emissions only?

Yes, this only applies to European emission's. Text clarified.

26. Section 5.3, 5.2, and to some degree 5.1 need to be merged and redundancies eliminated.

These sections have been reviewed and there is some blurriness around the edges, they have been edited to drive greater self-consistency.

27. Section 5.4: Tai et al., GRL, 2013 recently showed the "climate penalty" has been substantially overestimated. The discussion should be modified in light of this.

Discussion modified to include "There is some debate as to the magnitude of any climate penalty e.g. Tai et al. (2013) indicates that in the presence of CO<sub>2</sub>-isoprene interaction, the projected change in isoprene emission by 2050 will be largely offset or even reverse in sign, leading to much reduced sensitivity of ozone and SOA (by >50%) to climate and natural vegetation."

28. Page 32808, lines 14-16: It is not clear why emissions that were constructed for radiative forcing purposes would be inappropriate for air quality purposes. Please justify any concerns about quality of these emission inventories, and why they might therefore still be appropriate for climate projections.

We have aimed to clarify this point in the text, added two sentences to the referenced lines. The revised text now reads: "Pioneering studies relied on projections originally designed for climate projections (SRES, Nakicenovic et al., 2000 or RCPs, van Vuuren

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et al., 2011), but the ozone precursor information in such scenarios was solely provided to assess radiative forcing and their use for surface air quality projections constitutes a deviation from their original purpose. The various scenarios make differing assumptions for future air pollution emissions and therefore, describe a wide range of future emissions over large world regions. Any downscaling in regions that exhibit large spatial heterogeneities could be problematic leading to inaccurate results (Amann et al., 2013). The use of emission projections relying on policy relevant emissions factors such as the Global Energy Assessment (Riahi et al., 2012), the ECLIPSE (Klimont et al., 2013a, b) or PEGASOS datasets are more reliable.”

29. Page 32810, lines 10-22: duplication with Section 5.4. While this is true, we have tried to make all sections self-contained and this is an important part of the AQ & CC discussion, as embedded in this forward look. My preference is to keep it.

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Interactive comment on Atmos. Chem. Phys. Discuss., 14, 32709, 2014.

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