4 **Opinion: Papers that shaped Tropospheric Chemistry**

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14 **Abstract**

15 Which published papers have transformed our understanding of the chemical processes in the
16 troposphere, and shaped the field of atmospheric chemistry? By way of expert solicitation and
17 interactive peer-review, this paper explores the influence of the ideas in peer-reviewed articles based
18 on the input from our community of atmospheric scientists. We explore how these papers have
19 shaped the development of the field of atmospheric chemistry, and identify the major landmarks in
20 the field of atmospheric chemistry through the lens of those papers' impact on science, legislation and
21 environmental events. We also explore the ways in which one can identify the papers that have most
22 impacted the field and discuss the advantages and disadvantages of the various approaches.

1. Introduction

Air quality and anthropogenic climate change are two environmental issues of current importance to society. Atmospheric composition is central to both these issues. The atmosphere, and its components, supports life on Earth. In turn, the atmosphere is affected by human population growth and industrialization, as well as all the consequences of those changes. The changes in atmospheric composition also influence the ecosystem on which humans rely.

Air pollution (née composition) and its impacts have a history stretching back to antiquity – see for example the expositions in (Brimblecombe, 1987;Fuller, 2018;Jacobson, 2002;Stern, 1968;Sportisse, 2010;Preining and Davis, 1999;Fowler et al., 2020) and others. Changes in atmospheric composition, with their negative impact particularly on human health (Lelieveld et al., 2015), ecosystems (Fowler et al., 2009) and latterly links to their climate (see for example, (Fiore et al., 2012;von Schneidemesser et al., 2015)), have become primary global concerns during the 20th century. As an academic subject, air pollution has mostly been systematically studied only since the mid-late 20th Century. There have been several recent reviews, (e.g., (Brasseur et al., 2003;Monks et al., 2009;Ravishankara et al., 2015;Ravishankara, 2003)), which have mapped the growth of atmospheric chemistry, but it is not only peer-reviewed papers that provide relevant overviews. It is important to note that when dealing with the development of this subject (or any scientific subject for that matter), much of the baseline knowledge is embodied in textbooks, which for many are the entry point to and the primary reference for the topic (e.g. (Jacob, 1999;Wayne, 2000;Finlayson-Pitts and Pitts, 2000;Seinfeld and Pandis, 2006;Brasseur et al., 1999).

Figure 1 shows the number of peer-reviewed by year that mentioned the phrase "atmospheric chemistry" in the text, as catalogued by the Scopus bibliographic database (https://www.scopus.com/). It shows a growth in the later 70s from around a hundred papers a year to approximately 4,000 a year currently, with a large increase especially over the past two decades. Of course, many more papers discuss atmospheric chemistry, or are relevant to it, without explicitly mentioning these words!
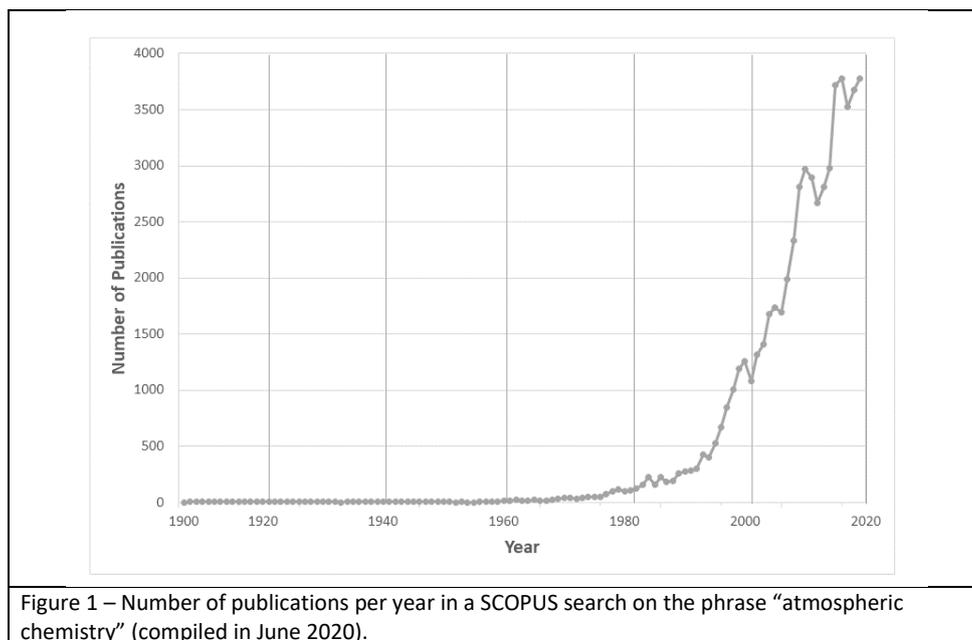
Figure 1 – Number of publications per year in a SCOPUS search on the phrase "atmospheric chemistry" (compiled in June 2020).

52 This paper aims to recognize and highlight some of the most influential peer-reviewed articles that
53 have shaped this field. There were many pivotal scientific discoveries and there were many papers
54 that spurred action and further research. What were the significant discoveries that shaped the
55 atmospheric chemistry of today? And how do we narrow down the list contributions to highlight the
56 most impactful ones?

57 There are many ways to choose the papers that described discoveries and influenced atmospheric
58 composition and chemistry. Here we have assembled an initial[1] compilation of influential articles. Our
59 goal is not to show what makes a 'great' paper, which depends not only on the science, but also the
60 quality of the writing, readability, and structure of the written work – all criteria that are highly
61 subjective. Nor are we aiming only for those papers that led to policy and management actions.
62 Instead, we try to reflect on *the papers' science and content* and the influence of the ideas in those
63 papers on our community of scientists and on the field's development. Our approach is to present our
64 thoughts – informed by the solicitation for input from colleagues in the field – and share what we
65 think are the fundamental discoveries and developments, start a discussion, and allow others to build
66 on, reinforce or critique our work.

67 In addition to peer-reviewed papers, and the above mentioned textbooks, we have of course other
68 mediums through which we communicate (have communicated) with our peers. These include
69 scientific reports, conferences, and meetings. In addition, we have scientific assessments and
70 evaluations, which often get more scrutiny and reviews than the peer-reviewed papers they include.
71 These days, other communication media, such as social media, have also become prevalent as formats
72 for exchange both within the scientific community and with a broader audience. However, the entire
73 community cannot attend all conferences and meetings, the scientific reports are not always
74 accessible and often not peer-reviewed, and the assessments are often driven more by policy needs

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[1] Initial as it is intended to be informed by the community through an open review process as outlined later.

75 rather than by scientific discoveries. Publishing peer-reviewed papers is the closest we come to
76 reaching the whole community. We do note that, despite its known issues, peer-reviewed literature
77 is still considered the gold standard for quality and reliability. For these reasons, we discuss only peer-
78 reviewed papers here, although we aim to communicate the overarching scientific advances that
79 shaped the field.

80

81 *1.1. How were the papers selected?*

82 Easily measurable criteria, like the number of citations, are one metric. However, this approach
83 favours papers of a particular vintage and not necessarily the earlier or later papers. Besides, there
84 are several drawbacks to these simple and objective criteria. Citations tend to go down when
85 something is assumed to be common knowledge and makes it into textbooks or compilations. For
86 example, nobody cites Priestley for discovering oxygen or Schönbein for discovering ozone, whenever
87 atmospheric composition is mentioned. Indeed, some of the central concepts of atmospheric
88 chemistry and physics are considered to be common knowledge, and their origins are taken for
89 granted. The number of citations will also be influenced by the journal in which a paper is published,
90 and quite often (we hate to say this) also depends on who else cited them and in which journal they
91 were cited. Citations also depend on how many people are otherwise researching a particular subject.
92 Furthermore, critical assessments and expert data evaluations suppress the citation of the original
93 papers. This is particularly the case, for example, for papers on chemical kinetics and photochemistry,
94 whereby people tend to simply cite the data evaluations such as NASA/JPL
95 (https://jpldataeval.jpl.nasa.gov/) or IUPAC (http://iupac.pole-ether.fr/) panel reports. Similarly,
96 people often cite the quadrennial ozone layer depletion and IPCC assessments, thereby obfuscating
97 the underlying original papers. Other types of papers, such as reviews, tend to get an excessive
98 number of citations (for understandable reasons). Lastly, we cannot overlook the influence of journal
99 availability in different parts of the world. This availability is exacerbated when journal costs go up,
100 and not everybody can access new papers. Nevertheless, there is still a relevance to the number of
101 citations of a paper. We show, for example, the 10 most-cited papers if we were to search on the
102 combination of words "atmospheric and chemistry" in Table 2.

103 For all the above reasons, we decided to use a different approach here. We solicited the scientific
104 community to obtain input from the experts in the field. To accomplish this, we put out a call through
105 the International Global Atmospheric Chemistry (IGAC) (Melamed et al., 2015) project to its contacts
106 and thereby engaged a broad audience. Despite the broad audience of IGAC, the vast majority of
107 responses came from scientists in North America and Europe. An initial list of influential papers was
108 established by combining the replies received from the expert solicitation to evaluate the most
109 nominated papers. In addition, a variety of perspectives were assembled for the writing team,
110 including different career stages, nationalities, and genders. Despite all these efforts, the selection
111 methods will still inevitably create bias that cannot be escaped. Therefore, in many respects, the
112 chosen papers are not supposed to be a definitive list, but rather a compilation that allows researchers
113 to discuss and reflect on what makes impactful science, and maybe ponder on the landmarks in our
114 subject. Furthermore, we hope that the end product can provide an interesting history and context to
115 those who are joining the community and document the current "perception" of what are the most
116 important papers.

117 We have noted the drawbacks in our methodology simply to present upfront some of the limitations
118 of what we did in this paper. However, we hope that others will find this work relevant and engaging,
119 and actively participate during the peer-review process, so that we can obtain the perspective of a

broader community more reflective of the global composition of the field. To facilitate this, we submitted the paper in an open-access and interactive review that includes a public comment period. We hope that this approach will overcome some of the limitations and reservations we expressed earlier. We **invite and encourage you, the reader,** to weigh in on the choices and help determine the final shape of this overview. We shall seek to reflect this discussion in the final version.

1.2. Scope of work

As with the selection method, one can debate the scope and the methodology for a work such as this. Still, the boundaries we have drawn encompass studies that have shaped our understanding of the atmosphere and the underlying chemical and physical processes, focusing mostly on the troposphere. This includes modelling, field measurements, remote sensing, and laboratory studies (Abbatt et al., 2014). We have also included atmospheric interactions with the biosphere, cryosphere, and hydrosphere.

We selected 2010 as the cut-off year. Our rationale is that for a paper to have been influential in the whole field it must be at least ten years old and thus had time to accrue recognition. We recognize that important papers in newer areas of endeavour are disadvantaged by this criterion. Examples include the subjects of SOA formation (Ehn et al., 2014;Crounse et al., 2013), the chemistry of Criegee intermediates (Welz et al., 2012), galactic rays induced aerosol nucleation (Kirkby et al., 2011), and air pollution-climate connections (Shindell et al., 2012). Influential assessments such as bounding black-carbon (Bond et al., 2013) are also missed. However, these areas will undoubtedly be recognized in the coming decades. The ten-year window also allows the scientific community to have had extensive input on a paper's validity, i.e., meeting the criterion of "standing the test of time."

The papers have been grouped into the following general categories and are presented as such in Section 2.

1. *Foundations*
2. *Aerosols and Clouds*
3. *Secondary Organic Aerosols*
4. *Chemical Kinetics, Laboratory Data and Chemical Mechanisms*
5. *Heterogeneous and Multiphase Chemistry*
6. *Chemical Models*
7. *Tropospheric Ozone*
8. *Nitrogen Chemistry*
9. *HOx Chemistry*
10. *Nightime Chemistry*
11. *Halogen Chemistry*
12. *Volatile Organic Compounds*
13. *Biogenic Emissions & Chemistry*
14. *Biomass Burning*
15. *Emissions and Deposition*
16. *Chemical Transport*
17. *Satellites and the Troposphere*
18. *Stratospheric Chemistry*
19. *Other issues that influenced tropospheric chemistry*

164

165   The groups were chosen to reflect the main areas of research or endeavour, recognizing that this
166   division could be done in several different ways. There is no assumed equivalence in these groups
167   regarding their perceived or real importance or impacts. In the following, we discuss the papers in
168   each group to show why they have been nominated and to put them in the historical context of the
169   development of atmospheric chemistry as a discipline.
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172

173 **2. Survey of Areas**

174

175 *2.1. Foundations*

176 Atmospheric chemistry has some long-standing and deep roots. However, it blossomed in the second
177 half of the 20th century following concerns about the ozone layer depletion and various forms of
178 tropospheric pollutions, including such as Los Angeles smog, London smog and acid precipitation
179 (Table 1). Many note John Dalton's early contributions on the proportion of gases in the atmosphere
180 (Dalton, 1805) and John Tyndall's Bakerian lecture on radiation and gases (Tyndall, 1861) as among
181 the first studies in this field. The work of Arrhenius "On the Influence of Carbonic Acid in the Air upon
182 the Temperature of the Ground" (Arrhenius, 1896) and the subsequent paper of Callendar, "The
183 artificial production of carbon dioxide and its influence on temperature" (Callendar, 1938), laid the
184 groundwork for the linkage between atmospheric chemistry and climate. Concerning aerosols, the
185 seminal work of John Aitken (Aitken, 1888) "On the number of dust particles in the atmosphere"
186 details early work to count the number per cubic centimeter in various indoor and outdoor
187 environments. It is interesting to note that physiologists looking at the number of live germs in the air
188 stimulated Aitken's work. The later work of Köhler (Köhler, 1936) which explored cloud droplet
189 nucleation remains the basis for later work (see the *Aerosols and Clouds* section). The start of
190 atmospheric chemistry as a distinct discipline probably arrived with Chapman's chemical theory of the
191 stratospheric ozone layer in 1930 (Chapman, 1930), which will be further discussed in the
192 *Stratospheric Chemistry* section. This study heralded the importance of atmospheric chemistry on a
193 global scale.

194 In analyzing the influential papers on atmospheric composition, one cannot help but note the
195 relationship between these papers and the most significant contemporary environmental issues
196 (Table 1). The first of these was the Los Angeles smog, which had its European counterpart, the London
197 "Pea-Soup" (Brimblecombe, 1987). The two events, which in chemical terms have no equivalence, had
198 comparable impacts on public health and opinion. The oft recognized work of Haagen-Smit (Haagen-
199 Smit and Fox, 1954;Haagen-Smit, 1952;Haagen-Smit et al., 1953) in the early 1950s on the Los Angeles
200 smog was the first to coin the term "air pollution" in the modern era. Haagen-Smit showed that
201 automobile exhaust gases can form ozone in the air and should, therefore, be considered a definite
202 source of smog. Figure 2, redrawn from Haagen-Smit (1952), shows a schematic presentation of the
203 reactions in polluted air leading to smog. Notably, the basic features of tropospheric chemical
204 processes, as we understand them today, were already recognized in these early papers, and they
205 already showed how ozone could be chemically produced in the troposphere.

206 It is widely recognized that both Crutzen (Crutzen, 1973a;Crutzen, 1973b) and Chameides and Walker
207 (1973) found that similar "smog reactions" oxidize methane and carbon monoxide to produce
208 substantial amounts of ozone in remote regions of the atmosphere. They estimated chemically
209 produced ozone to be much greater than that transported from the stratosphere, which was believed
210 to be the primary source of this chemical in the troposphere at that point. A few years earlier, in 1970,
211 Hiram Levy II had suggested that the hydroxyl radical, which provides the dominant oxidation
212 mechanism in the troposphere, was formed in unpolluted air by the same mechanism that had been
213 described as occurring in polluted air (Levy, 1971). This paper by Levy (1971) is recognized by many as
214 the first description of the chemistry of the lower atmosphere involving hydroxyl radical reactions of

215 methane and carbon monoxide, hydroperoxyl radicals, and the photolysis of ozone and formaldehyde
216 as radical sources. In particular, he recognized that the very short-lived electronically excited oxygen
217 atom (O^1D) is a possible source of the OH radical, an idea now well established.

218 Around the same time, Weinstock (1969) explained how cosmic rays lead to the production of
219 radiocarbon dioxide ($^{14}CO_2$), which is incorporated into living plants. This process requires a rapid
220 turnover of radiocarbon monoxide (^{14}CO), which was unexpected because the lower atmosphere was
221 thought to be a "chemical desert". Instead, carbon monoxide appeared to have a turnover time of
222 about one-tenth of a year, primarily driven by hydroxyl radical oxidation. To some, this paper kicked
223 off the research which led to our present understanding of the atmospheric chemistry of the lower
224 atmosphere.

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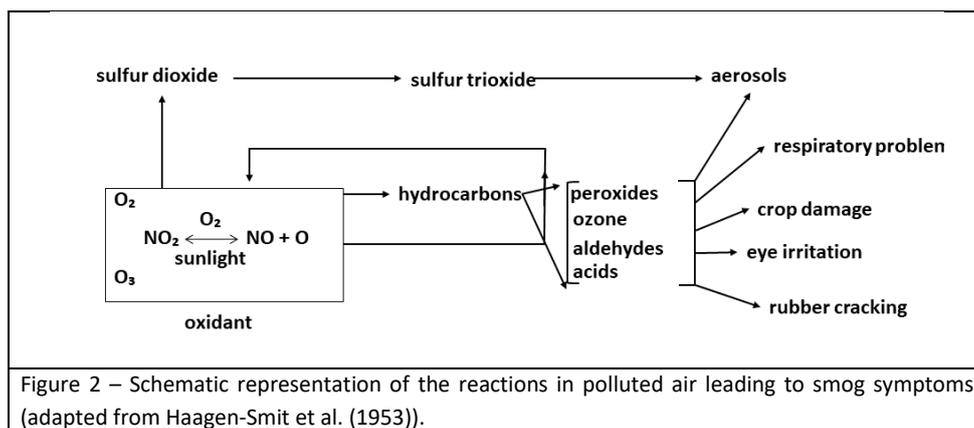
Figure 2 – Schematic representation of the reactions in polluted air leading to smog symptoms (adapted from Haagen-Smit et al. (1953)).

226

227 It has been claimed that "acid rain was one of the most important environmental issues during the
228 last decades of the twentieth century" (Grennfelt et al., 2019) (see Table 1). One of the reasons is that
229 acid rain first demonstrated that air quality was not merely a local issue but a regional issue and
230 showed that the atmosphere has no definite boundaries (Fowler et al., 2020). Although the case of
231 acid rain and its effects had been noted and reported by some earlier papers e.g., Odèn (1968), for
232 some, it is the paper by Likens and Bormann (1974) that made this issue known to the science
233 community at large. Other early papers (for example, from Urone and Schroeder (1969) and Penkett
234 et al. (1979)) also recognized the vital role of liquid-phase oxidation of SO_2 by oxidants such as H_2O_2
235 and O_3. Current estimates suggest that roughly 50% of the SO_2 oxidation in the lower troposphere
236 occurs through liquid-phase reactions.

237 There is little doubt that one of the most impactful series of papers is that of the eponymous curve by
238 Keeling (Keeling, 1960;Keeling et al., 1979;Pales and Keeling, 1965), showing the steady rise in CO_2
239 measured at Mauna Loa observatory (this work has continued uninterrupted by NOAA/ESRL/CML over
240 the past few decades). Keeling's work was built on the previously mentioned work of Callendar (1938)
241 who compared measurements of CO_2 at Kew, UK (1891-1901) with those in the Eastern USA (1936-
242 1938) and noted an increase in concentration. Although the gas in question is CO_2, which is often seen
243 only as climate gas, changes in its levels reflect the changing composition of the atmosphere and the

244   effects that it can have, and shows that the two subjects cannot be easily separated. Furthermore, the
245   increase in CO_2 is of immense importance to ocean acidification, a topic not touched upon here but
246   nevertheless very important. The seminal paper by Ramanathan et al. (1985) that highlighted the role
247   of methane, chlorofluorocarbons, and N_2O for climate strengthened the case for the inclusion of
248   chemistry in the climate issue. In many respects, this close-coupling between atmospheric chemistry
249   and climate change was brought to the forefront with the 1995 Nobel prize being awarded to Paul
250   Crutzen, Mario Molina and F. Sherwood Rowland "for their work in atmospheric chemistry,
251   particularly concerning the formation and decomposition on ozone"[2] (see *Stratospheric Chemistry*
252   section) and, later, with the Nobel prize to the Intergovernmental Panel on Climate Change, IPCC.

253

254   *2.2. Aerosols and Clouds*

255   Aerosols in the atmosphere greatly influence both air quality and climate change; they are also a
256   significant media for composition change in the atmosphere. In this section we discuss three main
257   areas of aerosol related research: (1) Understanding the mechanisms and atmospheric chemistry
258   processes that influence aerosol formation, nucleation, and growth, and how aerosols affect
259   composition; (2) The role of aerosols as cloud condensation nuclei and the influence that this process
260   has on climate; and (3) The impact of particulate matter on human health. These areas are however,
261   related and there is not always a clear division. *Secondary Organic Aerosols* (SOA) and *Heterogeneous*
262   *and Multiphase Chemistry* are discussed in the corresponding sections (2.3 and 2.5).

263   The roots of modern aerosol science lie, as previously discussed (see the 2.1. *Foundations* section), in
264   the works of Aitken (1888) and Köhler in 1936 (Köhler, 1936) on the cloud droplet. Twenty years after
265   Köhler's research, Junge (1955) provided the power-law describing aerosol number and identified the
266   stratospheric aerosol layer, now dubbed the 'Junge layer'. Junge concluded: "A real step forward in
267   the understanding of the basic processes in air chemistry can be gained only if aerosol and gases are
268   measured simultaneously but separately, and if the aerosols, in turn, are separated according to size."
269   This suggestion has been a clarion call for atmospheric scientists ever since.

270   In 1958, Junge and Ryan (1958) attempted to elucidate the formation of particles from gas-phase
271   reactants, particularly SO_2 and NH_3, while Fitzgerald (1974) investigated the variation in aerosol
272   composition with particle size. They showed that cloud droplet size distribution was insensitive to the
273   specific soluble constituents. Twomey (Twomey, 1977, 1974) suggested that air pollution gives rise to
274   the whitening of clouds and influences the planet's radiative balance. He also indicated that there is a
275   connection between pollution aerosols and cloud reflectance (albedo). This concept is now often
276   referred to as the "Twomey effect." Twomey (1977) expanded on the 1974 work, exploring the
277   balance between the scattering vs. absorption effect on the incoming solar radiation. It is on this basis
278   that much of the current research on the role of aerosols *via* their direct and indirect effect on climate
279   has been built. Bolin and Charlson (1976) estimated that anthropogenic sulfate aerosol from the US
280   and Europe would lead to a global temperature decrease of 0.03-0.06 °C. They recognized early on
281   that "we are already approaching the time when the magnitude of the indirect effects of increasing
282   use of fossil fuel may be comparable to the natural changes of the climate over decades and
283   centuries."

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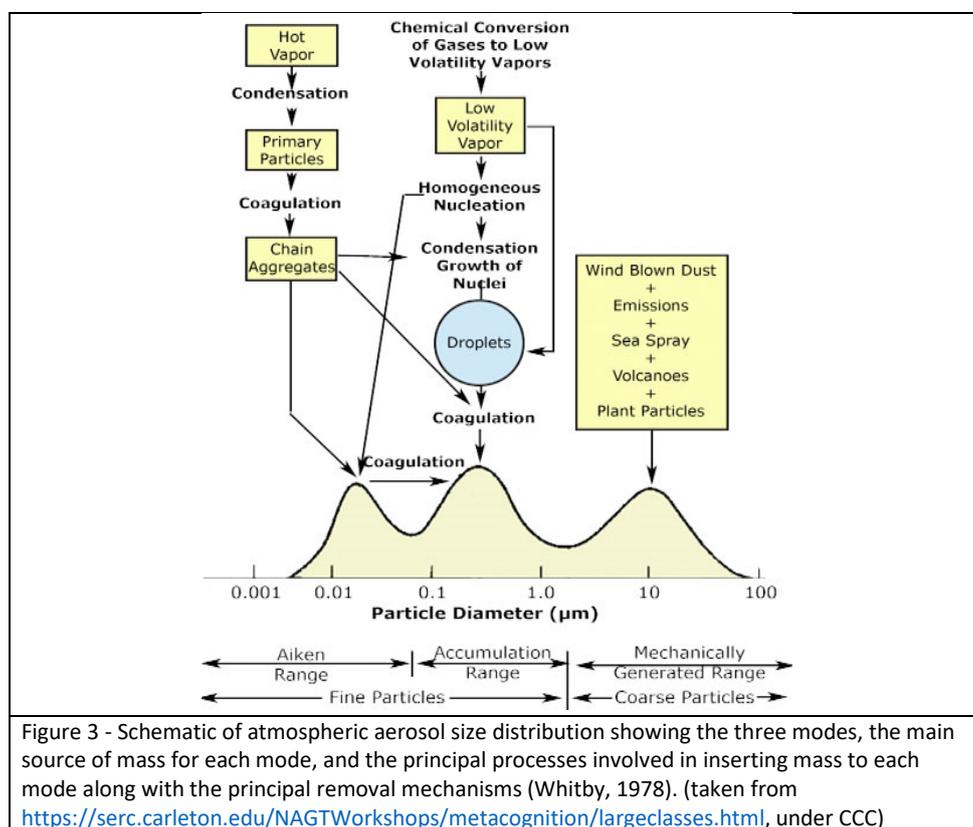
[2] https://www.nobelprize.org/prizes/chemistry/1995/press-release/

284   In the early 1970s, Whitby and Knutson developed an instrument to measure particle size distribution
285   in the nm to μm range (Knutson and Whitby, 1975) – the well-known aerosol mobility analyser. They
286   used the measurements from this instrument to introduce a new formulation of the formation and
287   growth of atmospheric particles and aerosol size modes – the Whitby diagram which is now a common
288   text book figure and is shown in Figure 3. The outcomes of this work show the importance and
289   influence of the development of new instruments that probe the atmosphere.

290

Figure 3 - Schematic of atmospheric aerosol size distribution showing the three modes, the main source of mass for each mode, and the principal processes involved in inserting mass to each mode along with the principal removal mechanisms (Whitby, 1978). (taken from https://serc.carleton.edu/NAGTWorkshops/metacognition/largeclasses.html, under CCC)

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292   The CLAW hypothesis (the acronym taken from the surnames of the proposers Charlson, Lovelock,
293   Andreae, and Warren) (Charlson et al., 1987) further connected aerosol science to gas-phase
294   chemistry, specifically focused on the feedback loop between ocean ecosystems and Earth's climate.
295   Although the conclusions of the paper have been questioned (Quinn and Bates, 2011), this paper
296   highlighted the interconnections within atmospheric sciences, and environmental sciences in general.

297   The work of Friedlander and co-workers (Stelson et al., 1979) further highlighted the role of liquid-
298   phase chemistry leading to aerosol particles. A key milestone in our understanding of sulfate
299   formation was the recognition that the reaction of $HOSO_2$ with O_2 is exothermic (Calvert et al., 1978)
300   and leads to gas-phase SO_3, contrary what was accepted at the time. Prior to this finding, there were
301   major difficulties in understanding the formation of gas-phase H_2SO_4 formation (Davis et al., 1979)

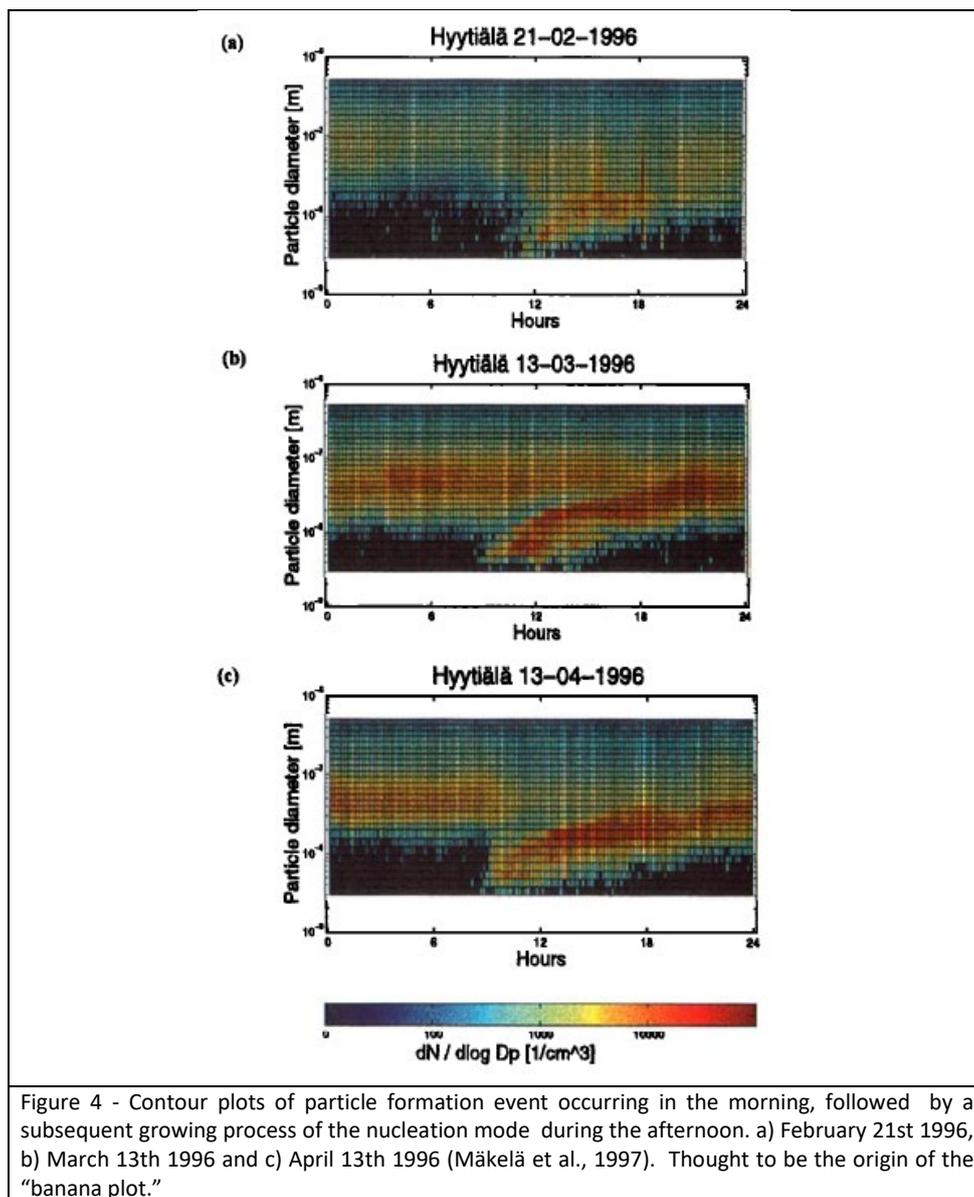
302   from SO₂ oxidation, an essential step for the nucleation of new particles from the gas-phase in the
303   atmosphere.

304   This area of research was further developed by Robbin and Damschen (1981) who investigated the
305   role of peroxide in the liquid-phase in oxidising SO₂ which was key to understanding the phenomenon
306   of acid rain. Graedel and Weschler (1981) reviewed the chemical transformations in atmospheric
307   aerosols and raindrops and extended the idea of Martin and Damschen. Stelson and Seinfeld (1982)
308   evaluated the thermodynamics of ammonium, nitrate, and sulfate aerosols, which was a significant
309   step in understanding particle formation and growth. Pankow's 1994 work (Pankow, 1994b, a) on the
310   absorption model of gas/particle partitioning of organic compounds in the atmosphere is of
311   fundamental importance for models to calculate the amounts of PM formed and their growth in the
312   urban and regional air, and in the global atmosphere.

313   Charlson et al. (Charlson et al., 1990;Charlson et al., 1991) produced the first global estimate of the
314   direct aerosol effect that subsequently had a large impact on climate modelling. The role that aerosols
315   would have on CCN and cloud albedo was also acknowledged, concluding that it may be substantial.
316   How substantial, however, was not quantified at that point because of a lack of knowledge on the
317   relationships involved. A few years later, Boucher and Lohmann (1995) provided an estimate of the
318   indirect effect of anthropogenic aerosols on climate. After many additional years of study based on
319   these foundations and analyses of radiative balance, the total radiative forcing by anthropogenic
320   aerosol is now estimated to be roughly -1.1 W/m², thereby solidifying the importance of aerosols in
321   climate change.

322   Building on the work of Whitby, Mäkelä et al. (1997) conducted continuous monitoring of particles at
323   a forest site in Finland. Beyond confirming the existence of 3 submicron particle size modes (the
324   nucleation, Aitken, and accumulation modes, see also Covert et al. (1996)), they also observed new
325   particle formation events. These events have been subsequently observed by others and are often
326   depicted in the literature using the famous "banana plots" (Figure 4).

327

Figure 4 - Contour plots of particle formation event occurring in the morning, followed by a subsequent growing process of the nucleation mode during the afternoon. a) February 21st 1996, b) March 13th 1996 and c) April 13th 1996 (Mäkelä et al., 1997). Thought to be the origin of the "banana plot."

328

329   There has been ample recognition for the research on process representations, such as the mole-
330   fraction-based thermodynamic models (Clegg et al., 1998b, a) and one-parameter model for
331   hygroscopic growth and CCN activity (Petters and Kreidenweis (2007)). Facchini et al. (1999) presented
332   experimental work aimed at understanding the role of surface tension in droplet growth and the
333   subsequent effect on cloud albedo and radiative forcing (RF), while Knipping et al. (2000) used a
334   simplified experiment to investigate the role of reactions of gases with ions at the air-water interface.
335

336 More specifically, the role of organics in the formation and growth of aerosols particles has been a
337 significant area of research (Kulmala et al., 2000). In addition to the natural hydrocarbons noted
338 above, it has become clear that anthropogenic hydrocarbons such as aromatic compounds are also
339 involved in new particle formation and their growth (Odum et al. (1997).
340
341 *2.3. Secondary Organic Aerosols*

342 Since the mid-2000s, secondary organic aerosols (SOA) have been the focus of much research,
343 addressing their abundance, sources, and production pathways. One of the foundational works in this
344 area is the recognition of the role of natural and anthropogenic hydrocarbons, and in particular
345 isoprene chemistry in the formation of SOA (Claeys et al., 2004).
346
347 The chemical composition of SOA across the globe is still poorly understood (Zhang et al., 2007),
348 although ways to describe the growth of SOA have advanced significantly (Kalberer et al., 2004).
349 Donahue et al. (2006) developed an approach based on the volatility of organics, a concept termed
350 "volatility basis set." This concept has been extended to a host of volatilities and their classifications.
351 For example, as shown in Figure 5, Robinson et al. (2007) postulated that a large amount of SOA mass
352 is unexplained by current models, and methods used to estimate SOA production do not capture what
353 is measured/observed in the field.

354 Jaenicke (2005) was the first to suggest that biological particles are an important fraction of
355 atmospheric aerosol. This paper prompted the development of a new and exciting field within
356 atmospheric sciences.

357

Figure 5 - Results from the photochemical oxidation of diesel exhaust in an environmental chamber. The grey area indicates the primary aerosol (POA + other species). The red area shows the upper-bound estimate of the contribution of known SOA precursors to the suspended aerosol mass leaving behind a large fraction that is not accounted for (blue area) (Robinson et al., 2007).

358

359 The introduction of the aerosol mass spectrometer (AMS) by Worsnop and colleagues (Canagaratna
360 et al., 2007) along with the pioneering instruments of Prather (Gard et al., 1997) and Murphy et al.
361 (2006) have established that organic compounds are ubiquitous in aerosols. Zhang et al. (2007) and
362 later Jimenez et al. (2009) explored the chemical composition of PM at different sites across a part of
363 the globe, and their work has been extended by a large number of groups.

364 Aimed at addressing some of the 'missing urban SOA' in models, Surratt et al. (2010) investigated SOA
365 production from isoprene and Virtanen et al. (2010) showed the amorphous solid state of biogenic
366 secondary organic aerosol particles,challenging the traditional views of the kinetics and
367 thermodynamics of SOA formation and transformation.

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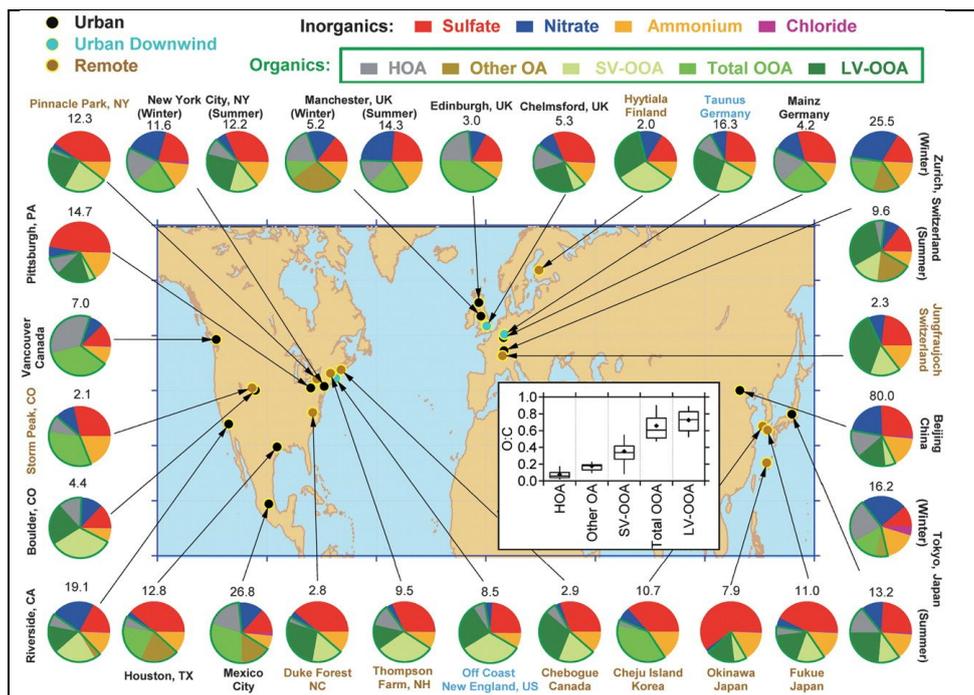


Figure 6 - Total mass concentration (in micrograms per cubic meter) and mass fractions of
nonrefractory inorganic species and organic components in submicrometer aerosols measured with
the AMS at multiple surface locations in the Northern Hemisphere at mid-latitudes. The organic
components were obtained with FA-AMS methods (Zhang et al., 2007). In some studies, the FA-
AMS methods identified one OOA factor, whereas in other locations, two types, SV-OOA and LV-
OOA, were identified. HOA is a surrogate for urban primary OA, and Other OA includes primary OAs
other than HOA that have been identified in several studies, including BBOA. Inset: distributions of
O:C for the OA components identified at the different sites (Jimenez et al., 2009).

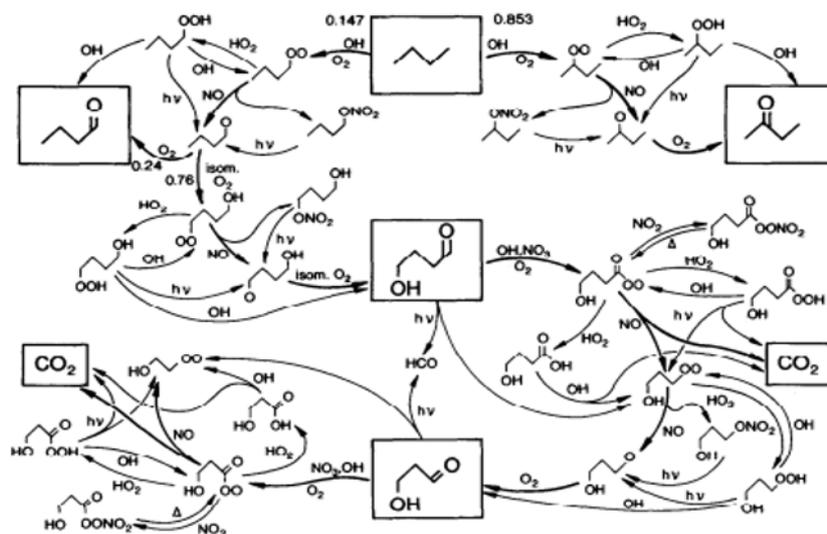
369

370

371

2.4. Chemical Kinetics, Laboratory Data and Chemical Mechanisms

Chemical kinetics is one of the foundations of atmospheric chemistry (Abbatt et al. (2014). This includes a number of different areas: investigation of individual chemical reactions; theoretical understanding of elementary reactions; evaluations and compilation of kinetics data; development and compilation of chemical mechanisms for use in models (see the 2.6. *Chemical Models* section); testing and simplification of the models for use in numerical models. Demerjian et al. (1974) is considered by many in the community to be one of the cornerstones of chemical mechanism development and it has been influential in a number of other research areas as well. This paper provided an explicit chemical mechanism for the troposphere in which all the chemical reactions were written as numerically integrated stoichiometric equations to predict photochemical ozone production rates. Previously, all chemical mechanisms had been highly "reduced" (into simple mechanisms) and/or parameterised, with non-stoichiometric equations. Using Demerjian's approach, many explicit atmospheric chemical mechanisms have been derived, including one of the most widely used, the Master Chemical Mechanism (MCM, Figure 7) (Jenkin et al., 1997;Jenkin et al., 2003). Currently, there are a variety of tropospheric chemistry mechanisms that capture the scope of chemical reactions that are used in a number of models including the 1990 Carter mechanism (Carter, 1990), RADM/RACM (Stockwell et al., 1997;Stockwell et al., 1990), SAPRC-07 (Carter, 2010) and the aqueous-phase mechanism CAPRAM (Ervens et al., 2003).

There is no doubt that the chemical kinetic data compilations have been the backbone of providing much needed experimental data to all chemical mechanisms and models (see the 2.6. *Chemical Models* section). The comprehensive reviews of Atkinson starting around 1986 (Atkinson (1986)) and followed by many others, provided a consistent description of the reaction pathways of the alkyl, peroxy and alkoxy radicals produced by the reactions of hydroxyl radicals with a wide range of organic compounds. These papers led the way for the compilation of the IUPAC and JPL chemical kinetic data evaluation of tropospheric reactions (Atkinson et al., 1989;Crowley et al., 2010;Atkinson et al., 1992;Atkinson et al., 2004;Atkinson et al., 2006;DeMore et al., 1997;Burkholder et al., 2020). (Note that compilation of kinetics data for stratospheric reactions dates back to mid-1970s (Hudson and Reed, 1979)) These works have been the foundation for the development of all chemical mechanisms and have led to the standardisation and improvement of condensed chemical mechanisms used in all chemical models.

403

404   Figure 7. Oxidation mechanism of butane in the Master Chemical Mechanism (Jenkin et al., 1997).

405

406   The recognition of reactions with negative activation energies and the role of weakly bound adducts
407   were other steps in improving our understanding of chemical kinetics. When the adduct is strong, we
408   term it an association reaction, which exhibits negative activation energies and pressure dependence.
409   Understanding and representing these type of reactions in atmospheric chemistry has been a major
410   step. In particular, the pioneering work of Troe and coworkers have enabled realistic and simpler
411   representation of these reactions based on the Rice-Ramsperger-Kassel-Marcus (RRKM) theory (Troe,
412   1979, 1994).

413   Atmospheric chemistry is often termed atmospheric photochemistry since the initiator for many of
414   the reactions is the production of free radicals, which are directly or indirectly the result of solar
415   radiation. Over the decades, the representation of photochemical processes has been well
416   established. A key element is the calculation of the "j-value" (i.e. the photolysis rate) for a
417   photochemical process, which depends on radiative transfer to obtain the solar flux and laboratory
418   measurements of absorption cross sections and quantum yields. The pioneering works on methods
419   for quickly and accurately calculating j-values are those of Madronich and Flocke (1999) and of Prather
420   and colleagues (Wild et al., 2000).

421   Moving towards individual reactions, the work of Howard and Evenson (Howard and Evenson, 1977)
422   on the reaction between HO_2 + NO has been recognised as a scrupulously careful study that
423   overturned conventional wisdom on this key reaction in photochemical smog/ozone formation (and
424   in stratospheric chemistry).

425   Three papers nearly a decade apart address the fundamental importance of robust laboratory
426   measurements to underpin model-led interpretation of experimental data. The seminal work
427   demonstrating the long-wavelength tail on the ozone photodissociation quantum yield (Ball et al.,
428   1993;Ravishankara et al., 1998) and the related work on the O^1D + N_2/O_2 reactions (Ravishankara et

429 al., 2002), key processes in the formation of OH radicals in the troposphere. Prompted by the findings
430 of Lelieveld et al. (2008) (see the 2.13. *Biogenic Emissions & Chemistry* section), a pair of laboratory
431 papers published in 2009, about HO_x radical regeneration in the oxidation of isoprene (Peeters et al.,
432 2009) and unexpected epoxide formation in the gas-phase photooxidation of isoprene (Paulot et al.,
433 2009) have changed the way we understand the gas and aerosol products and impacts of isoprene
434 chemistry (Kleindienst, 2009).

435

436   *2.5. Heterogeneous and Multiphase Chemistry*

437 Earth's atmosphere contains various amounts of condensed matter suspended in air. The most visible
438 condensed matter is, of course, clouds. One can also see aerosols when their concentrations are large;
439 examples include smog, wildfires, and volcanic eruptions. In addition to clouds, snow and ice provide
440 different mediums that can alter chemistry.

441 Many chemical reactions occur on the surfaces of particles suspended in air, ice/snow on the ground,
442 and within liquid drops. In general, these processes catalyse reactions that would be very slow in the
443 gas-phase, such as those between closed-shell molecules, and/or can produce products that do not
444 form in the gas-phase. For these reasons, heterogeneous and multiphase reactions are of immense
445 interest, although distinction between heterogeneous and multiphase chemistry is not always clear
446 cut (Ravishankara, 1997).

447 The unique contribution of reactions in/on condensed matter burst into the limelight due to their role
448 in stratospheric ozone depletion (Solomon et al., 1986). However, such reactions had been recognized
449 to be important before the ozone hole research, in the oxidation of SO_2 (Urone and Schroeder (1969)
450 and Penkett et al. (1979)). Since the 1990s, the roles of heterogeneous and multiphase reactions have
451 been highlighted in many tropospheric processes, as noted here in various sections (see for instance
452 sections 2.1. *Foundations* and 2.4. *Chemical Kinetics, Laboratory Data and Chemical Mechanisms*).

453 Chameides and Davis (1982) studied the free radical chemistry of cloud droplets and its impact upon
454 the composition of rain showing that the radical chemistry in water droplets could drive peroxides
455 production, which has the ability to rapidly oxidise sulphur species – a strong link to acid rain. Akimoto
456 et al. (1987) work on the photoenhancement of nitrous acid formation in the surface reaction of
457 nitrogen dioxide and water vapour demonstrated the existence of an additional radical source in smog
458 chamber experiments and challenged our understanding of the role of such heterogeneous reactions
459 in the atmosphere. The work of Mozurkewich et al. (1987), Hanson et al. (1992) and subsequently
460 Thornton and Abbatt (2005), on the measurements of HO_2 uptake to aqueous aerosol was highly
461 influential in the debate on the aerosol loss of HO_x, a question that has vexed many modelling studies.

462 A pioneering paper in tropospheric cloud chemistry is the study by Jacob et al. (1986) in the San
463 Joaquin valley that used a multiphase measurements and modelling approach to study the formation
464 of acid fog. Two further papers have brought heterogeneous chemistry to the fore: Dentener et al.
465 (1996) in their original paper on the role of mineral aerosol as a reactive surface in the global
466 troposphere showed the potential role of mineral dust on the SO_x, NO_y and O_3 chemistry, and Jacob
467 (2000) who reviewed in more detail at heterogeneous chemistry and ozone *via* HO_x and NO_y chemistry
468 in relation to aerosols and cloud droplets producing a highly cited series of recommendations for
469 future studies.

470 Another similar area is the role of chemical reactions on/in snow and ice. Such reactions were
471 highlighted by Barrie et al. (1988), specifically with regard to the role of halogens at the surface layer
472 in the troposphere, and hinting at the role of the cryosphere as a source of chemical species to the
473 atmosphere see the 2.11 *Halogen Chemistry* section). Given the extent of the cryosphere and in
474 particular of snow (Grannas et al., 2007), results in the late 1990s demonstrated its role in promoting
475 heterogeneous reactions as a significant source of unusual and unexpected chemical species to the
476 atmosphere. One of the most nominated works in this area was that by Honrath et al. (1999)
477 investigating NO_x production from the illuminated snowpack. Davis et al. (2001) pioneering work on
478 the unexpected production of NO_x in the pristine Antarctica is also worthy of note.

479

480   *2.6. Chemical Models*

481 Chemical models are the conduit to represent our knowledge of the chemical and physical processes
482 in the atmosphere within a mathematical framework that allows prediction and testing against
483 laboratory and field studies. They also help explain atmospheric observations and predict what could
484 happen under different conditions. Early, simple chemical models (with no chemical transport) were
485 useful tools to elucidate and test the basic theory of photochemical ozone formation (Levy, 1971).

486 The recognition that one cannot treat the chemical transformation without considering atmospheric
487 transport came early. The original simple 1-D models, often designed with a parameterized vertical
488 transport in terms of an "Eddy Diffusion" concept, were superseded by 2-dimensional models and
489 have now been largely replaced by complex 3-D models. Also, chemical transport models (CTMs),
490 which use analysed winds, are often used to separate transport from chemistry. Now, free-running
491 on-line 3-D models, which include chemistry, have been implemented, and the continued
492 enhancements in computing capabilities have greatly improved our modelling capabilities. Logan and
493 co-authors are recognised by many contributors as providing the basic model description of global
494 tropospheric chemistry (Logan et al., 1981).

495 A series of three papers from the Seinfeld group (Reynolds et al., 1973;Reynolds et al., 1974;Roth et
496 al., 1974) provided the earliest complete descriptions of an air quality policy model. They linked
497 together emission inventories, meteorological data, chemical mechanisms and air quality network
498 data to evaluate model performance. All subsequent air quality policy models have followed the same
499 general approach and basic formulation.

500 One of the earliest detailed chemical modelling studies that integrated highly instrumented intensive
501 field campaigns data was that of Harriss (1988) for the ABLE 1A campaign in the Amazon Boundary
502 Layer. Bey et al. (2001) first detailed GEOS-Chem, a global, three-dimensional, tropospheric, chemical
503 transport model. Though not the only global tropospheric model, as an open-source model with a
504 large user community and flexibility, it has become a very influential global model. In recent years
505 WRF-Chem has also been used extensively (Grell et al., 2005). Earlier stratospheric chemical transport
506 models were advanced by for example, Chipperfield and Pyle (1988).

507 Multi-model ensembles of the troposphere as epitomised by Stevenson et al. (2006) and Fiore et al.
508 (2009) (see the 2.16. *Chemical Transport* section) are a powerful tool for generalising the model
509 "understanding" of the atmosphere. This modelling approach makes use of many different models to
510 achieve a more accurate representation of the observations than it would be possible by using only

511   one model, thus producing more reliable outcomes for assessments and policies on a global scale. In
512   addition, multiple runs of the same models with slightly different initial conditions are used to examine
513   the range of outcomes. This approach is akin to the use of multiple models and model runs in weather
514   predictions.

515

516   *2.7. Tropospheric Ozone*

517   Ozone is one of the central molecules of atmospheric chemistry and runs through much of the
518   foundations of the discipline, from its role in the stratosphere as UV shield, to its role as a major
519   greenhouse gas, to its pivotal part in the troposphere as the start and end-product of oxidation
520   chemistry, and its detrimental influence as an air pollutant harmful to human health and ecosystems.
521   Much of the early thinking on ozone was focused on the question of whether tropospheric ozone was
522   a small subset of stratospheric ozone, see for example Galbally (1968) and Fabian and Pruchniewicz
523   (1977). The latter paper showed the value of observational networks based on standardised
524   instrumentation and calibration techniques, together with consistent siting criteria, and raised the
525   issue of seasonal variations in tropospheric ozone and the nature of the processes that drive them.
526   The vertical structure of a layer of high O_3 concentrations in the stratosphere, where O_2 could be
527   directly photolyzed to make oxygen atoms and hence ozone, and declining concentration in the
528   troposphere was indicative of a stratospheric source and a tropospheric sink and was the prevalent
529   theory prior to late 1970s (see also the 2.18. *Stratospheric Chemistry* section). A major breakthrough
530   were the two papers by Chameides and Walker, and Crutzen (Chameides and Walker, 1973;Crutzen,
531   1973b) that showed that ozone can be photochemically made across the troposphere, just like it is
532   made in smog *via* the reactions involving hydrocarbons and nitrogen oxides.

533

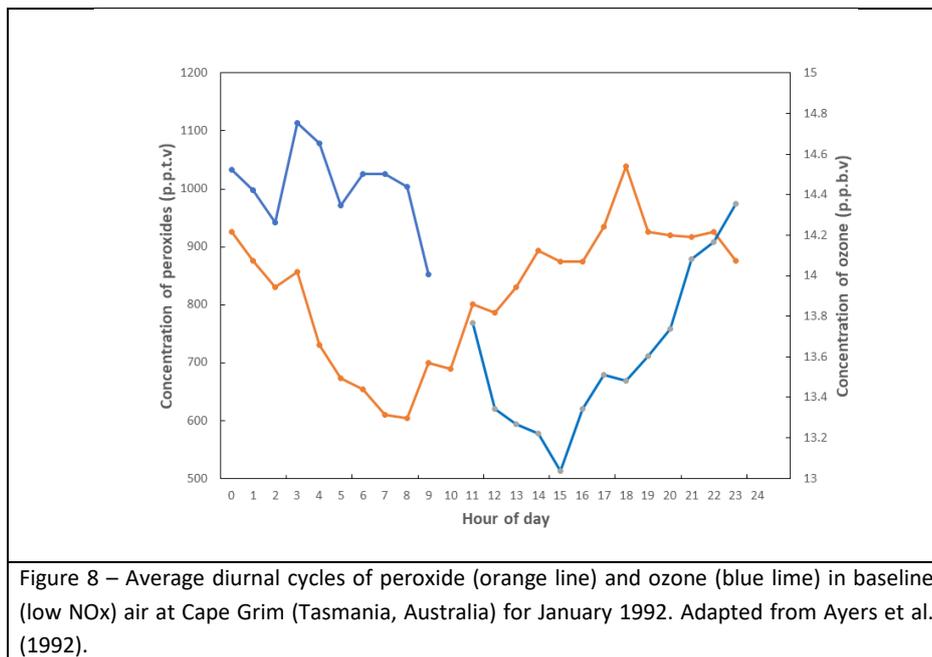
534   The importance of ozone as a radiative gas has been known for a long time, with a significant fraction
535   of heating in the stratosphere coming from ozone photolysis followed by its reformation and thus
536   converting sunlight to heat. Fishman in 1979, (Fishman et al., 1979) identified that tropospheric ozone
537   is also a greenhouse gas. Hence, a change in tropospheric ozone will perturb the radiative energy
538   budget of the Earth-atmosphere system which will in turn perturb the climate system. Ozone thus
539   became the second trace gas after carbon dioxide to be implicated in global warming and climate
540   change.

541

542   Large scale mapping of global tropospheric ozone was first undertaken by Logan (Logan, 1985) who
543   looked at seasonal behaviour and trends with a view to understanding anthropogenic influence. This
544   was later complemented by a paper exploring the photochemical origins of tropospheric, rather than
545   stratospheric, ozone in the rural United States (Logan, 1989). As the understanding of the
546   photochemistry of ozone developed, measurements at Niwot Ridge, Colorado (Liu, 1987) aimed to
547   quantify the elements of the ozone budget by season, bringing forward the concept of ozone
548   production efficiency. Lin et al. (1988) explored the non-linearity of tropospheric ozone production
549   with respect to NMHCs and NOx. Though this chemistry had been outlined much earlier – e.g.
550   Demerjian et al. (1974) – this work explored it in the background atmosphere with models and
551   measurements. A powerful demonstration of the low-NOx ozone destruction chemistry came from
552   measurements made at Cape Grim, Australia, a background station, where Ayers and Penkett (Ayers

553   et al., 1992) and their team(s) used measurements of ozone and peroxides (Figure 8) to show further
554   experimental proof for the photochemical control of ozone in remote locations.

555

Figure 8 – Average diurnal cycles of peroxide (orange line) and ozone (blue lime) in baseline (low NOx) air at Cape Grim (Tasmania, Australia) for January 1992. Adapted from Ayers et al. (1992).

556

557   Measurements have always been a critical driver in tropospheric chemistry, and the idea to use in-
558   service commercial aircraft as a platform for programs such as MOZAIC/IAGOS (Thouret et al., 1998)
559   has been recognised for the enormous amount of high quality data, which would otherwise be difficult
560   to regularly obtain from the upper troposphere and lower stratosphere. Using such measurements
561   Newell et al. (1999) combined dynamical and chemical tracers to further delineate ozone origin and
562   budgets. In the same year, Logan published a synthesis of ozone sonde data (Logan, 1999) which gave
563   an unprecedented look at the seasonal and vertical distribution of ozone and became a reference
564   point for the subject. A year later, Thompson et al. (2000) used a combination of shipboard and
565   satellite views of a tropospheric ozone maximum to suggest the occurrence of a tropical Atlantic ozone
566   "paradox". The "Atlantic paradox" refers to a greater tropospheric ozone column amount over the
567   South Atlantic than the North Atlantic during the West African biomass burning season. This
568   phenomena was further explored using an expanded network of ozonesondes in the southern
569   hemisphere (SHADOZ) (Thompson et al., 2003). In combination with the earlier work of Logan these
570   became the basis for the measurement description of ozone in the troposphere.

571   A decades worth of knowledge on the relationship between ozone and its precursors was pulled
572   together by Sillman (Sillman, 1999), cementing the concepts of NOx- and VOC-sensitive (or NOx
573   saturated) chemical regimes. The paper introduced a generation of researchers to isopleth diagrams
574   (the famous Sillman plot Figure 9) and ozone production efficiencies (OPEs).

Figure 9 - . Ozone isopleths (ppb) as a function of the average emission rate for NO$_x$ and VOC (10^{12} molec. cm^{-2}s^{-1}) in 0-D calculations. The isopleths (solid green lines) represent conditions during the afternoon following 3-day calculations with a constant emission rate, at the hour corresponding to maximum O$_3$. The short blue dashed line represents the transition from VOC-sensitive to NO$_x$-sensitive conditions. Adapted from Sillman and He (2002).

575

576   The power of models to explore global tropospheric ozone distributions, budgets and radiative forcing
577   was fully demonstrated in the study by Stevenson et al. (2006), that brought together 26 atmospheric
578   chemistry models to explore both the air quality and climate roles of ozone (see also the 2.6. *Chemical*
579   *Models* section). As discussed in the 2.16. *Chemical Transport* section, a similar approach was used by
580   Fiore et al. (2009) to explore the relationship between inter-continental transport and ozone.

581

582   *2.8. Nitrogen Chemistry*

583   Nitrogen oxides are an integral part of tropospheric processes. Nitrogen oxides are released into the
584   troposphere from a variety of biogenic and anthropogenic sources including fossil-fuel combustion,
585   biomass burning, microbial activity in soils and lightning. The concept of the Leighton photostationary
586   state (Leighton, 1961) between NO, NO$_2$, and O$_3$ was well established by mid-1990s, and early work
587   from Singh and Hanst (1981) highlighted the potential role of peroxyacetyl nitrate (PAN) to be a
588   reservoir for NO$_x$ in the unpolluted atmosphere. A landmark paper in the area of nitrogen chemistry
589   was that of Logan (1983) that brought together global and regional budgets for the nitrogen oxides
590   (Table 3). Later, a paper that focused more narrowly on a specific source of NO$_x$ was that of Yienger
591   and Levy II (1995) who produced an empirical model of global soil-biogenic NO$_x$ emissions. Higher up
592   in the atmosphere, the work on sources and chemistry of NO$_x$ by Jaeglé et al. (1998) is recognised for
593   its contribution to the understanding of the NO$_x$ cycle in the upper troposphere.

594   These works were complemented by a more holistic view of the nitrogen cycle and in particular the
595   concept of reactive nitrogen (Nr) from Galloway et al. (2004) that clearly showed the linkages between
596   the terrestrial ecosystem and the atmosphere and how the nitrogen budget had and would change.

597 In more recent times, extensive work on vehicle NOx sources from exhaust remote sensing data, as
598 epitomised in Carslaw (2005) should be highlighted. This paper pointed out the trends that can be said
599 to have led to the denouement of the Volkswagen emissions scandal.

600

601   *2.9. HOx Chemistry*

602 There is no doubt that the chemistry of OH and HO_2 (known together as HO_x) has a central role in the
603 atmosphere as well as holding a certain fascination to atmospheric scientists owing to the significant
604 challenges involved in measurement and understanding its impact locally to globally. Much of the
605 history of the measurements of OH and HO_2 is covered in the review of Heard and Pilling (2003). As
606 they wrote "clearly, OH plays a central role in tropospheric chemistry. The in situ measurement of its
607 concentration has long been a goal, but its short lifetime and consequently low concentration provide
608 a serious challenge."

609 In order to assess the global impact of OH chemistry in the absence of direct measurements, reactive
610 proxies have been used. Singh (Singh, 1977) used methyl chloroform to estimate OH abundance since
611 methyl chloroform is exclusively anthropogenic and its emissions are known. This type of work
612 provided a comprehensive picture of the global distribution of OH and, hence, a first overall look at
613 the oxidative capacity of the atmosphere. It was followed, using halocarbon measurements by the
614 AGAGE network, by a global OH determination while also introducing the atmospheric chemistry
615 community to formal inverse modelling (Prinn et al., 1995). Spivakovsky and co-workers expanded on
616 this work to derive 3-D distributions of OH and used this information to assess the wider impact on
617 the lifetimes of halocarbons, which have implications for stratospheric ozone (Spivakovsky et al.,
618 2000). Thanks to the availability of long term observations of halocarbons from the AGAGE and NOAA
619 Networks, later work using a similar approach found evidence for substantial variations of
620 atmospheric hydroxyl radicals in the previous two decades (Prinn et al., 2001), thus providing a broad
621 overview not only of the global distribution but also of the temporal variability of this crucial species.
622 Such estimates allowed for the quantification of the lifetime of important chemicals such as methane
623 and HCFCs.

624 The in-situ OH detection in the troposphere has proven elusive for a long time. The use of laser-
625 induced fluorescence provided some of the first clues to its atmosphere concentrations in the 1970s
626 and early 1980s (Davis et al., 1976;Wang et al., 1975), but any of these early measurements were
627 found to have significant artefacts. The study by Eisele (1994) at the Fritz Peak Observatory in
628 Colorado, was the first intercomparison experiment of different measurement techniques and
629 provided much needed confidence in the observations of this key molecule. Stevens et al. (1994)
630 developed the low-pressure laser-induced-fluorescence (LIF) instrument, which quikly became one of
631 the most successful and widely used techniques for ambient measurements of OH and HO_2. As
632 ambient observations of HOx became available, they were found useful to test our understanding of
633 the tropospheric chemical processes, by comparing them with the results of chemical models (see the
634 2.6. *Chemical Models* section). Recognised as a foundational paper in this area, Ehhalt (1999)
635 explained with clarity the role of radicals in tropospheric oxidation and what controls their
636 concentrations, using both ambient measurements and calculated concentrations of OH. The OH
637 radical is particularly suited to test our understanding of chemical processes and this was clearly
638 demonstrated in 2009, when the discrepancies between observed and calculated OH and HO_2 in the

639 polluted region of Southern China led Hofzumahaus and co-workers to propose a regeneration
640 pathway for OH, which does not involve NOx and thus does not produce O_3 (Hofzumahaus et al., 2009).
641 This, in turn, prompted a major reassessment of the isoprene oxidation mechanism by Peeters et al.
642 (2009) who suggested that isomerisation of hydroxyperoxy radicals from isoprene oxidation could be
643 fast enough to regenerate HOx in highly forested, low NOx environments (see the 2.4. *Chemical*
644 *Kinetics, Laboratory Data and Chemical Mechanisms* section) and led to a major revision of isoprene
645 chemistry and its role in the troposphere (see the 2.13. *Biogenic Emissions & Chemistry* section).

646

647 The sources and sinks of HOx radicals have always been a major research focus (Finlayson and Pitts,
648 1976) and the work of Paulson and Orlando (1996) on the reactions of ozone with alkenes as a source
649 of HOx in the boundary layer is widely recognised. Radical chemistry is highly sensitive to the levels of
650 NO and NO_2 and Kleinman's modelling work on hydrogen peroxide (H_2O_2) concentrations in the
651 boundary layer is recognised for its simple elegance in describing how the HO_x cycle chemistry is
652 influenced by NOx and in giving insight into the differing fates of OH/HO_2 radicals under different NOx
653 regimes (Kleinman, 1991) (Figure 10).

654

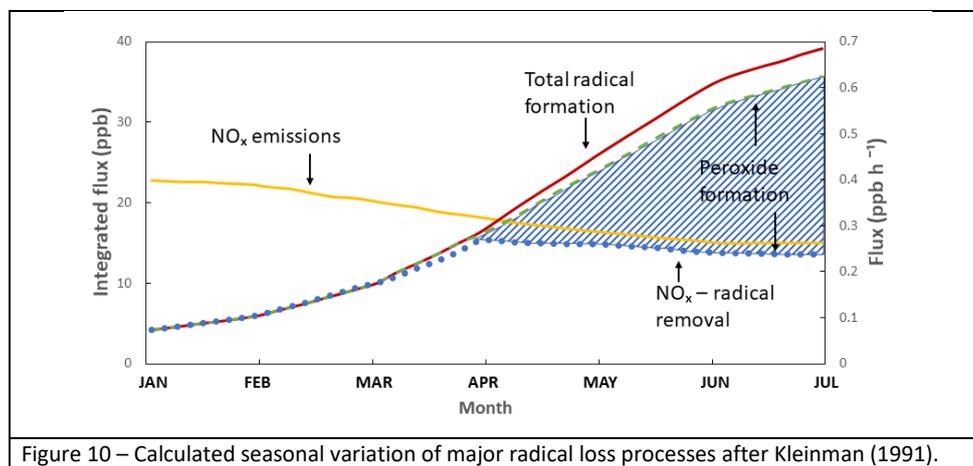



Figure 10 – Calculated seasonal variation of major radical loss processes after Kleinman (1991).

655

656 The first direct measurements of OH lifetime (Di Carlo et al., 2004) provided evidence of missing
657 reactivity, i.e. that not all sinks of the OH radical are known, which relates to earlier work by Lewis et
658 al. (2000) on unmeasured volatile organic compounds (see the 2.12. *Volatile Organic Compounds*
659 section).

660

661   2.10.   *Nighttime Chemistry*

662 There is widespread recognition that the atmosphere's oxidative chemistry is active during the night
663 as well during the day. Evidence of nighttime chemistry driven by the nitrate radical (NO_3) as well as
664 by ozone was first observed in the (polluted) troposphere in 1980 by Platt and co-workers (Platt et al.,
665 1980). Much of the early NO_3 work, including laboratory and field studies, is summarised in Wayne's

666 seminal review (Wayne et al., 1991). Platt and colleagues and Plane and colleagues' groundbreaking
667 work based on long-path absorption had indicated the importance of NO_3 in the troposphere (Allan et
668 al., 1999;Platt et al., 1979).

669 Two papers that have been highly influential in shaping our view of nocturnal chemistry are "Nitrogen
670 oxides in the nocturnal boundary layer: Simultaneous in situ measurements of NO_3" (Brown et al.,
671 2003) and "Variability in Nocturnal Nitrogen Oxide Processing and Its Role in Regional Air Quality"
672 (Brown et al., 2006). Both these papers showed the power of state of the art measurements coupled
673 with models to assess the impact of nocturnal and heterogeneous chemistry on regional air quality.
674 In particular, the paper by Brown et al. (2006) was a powerful demonstration of the role of
675 heterogeneous chemistry and aerosol composition in controlling N_2O_5 and, therefore NO_3,
676 concentrations (Figure 11).

677

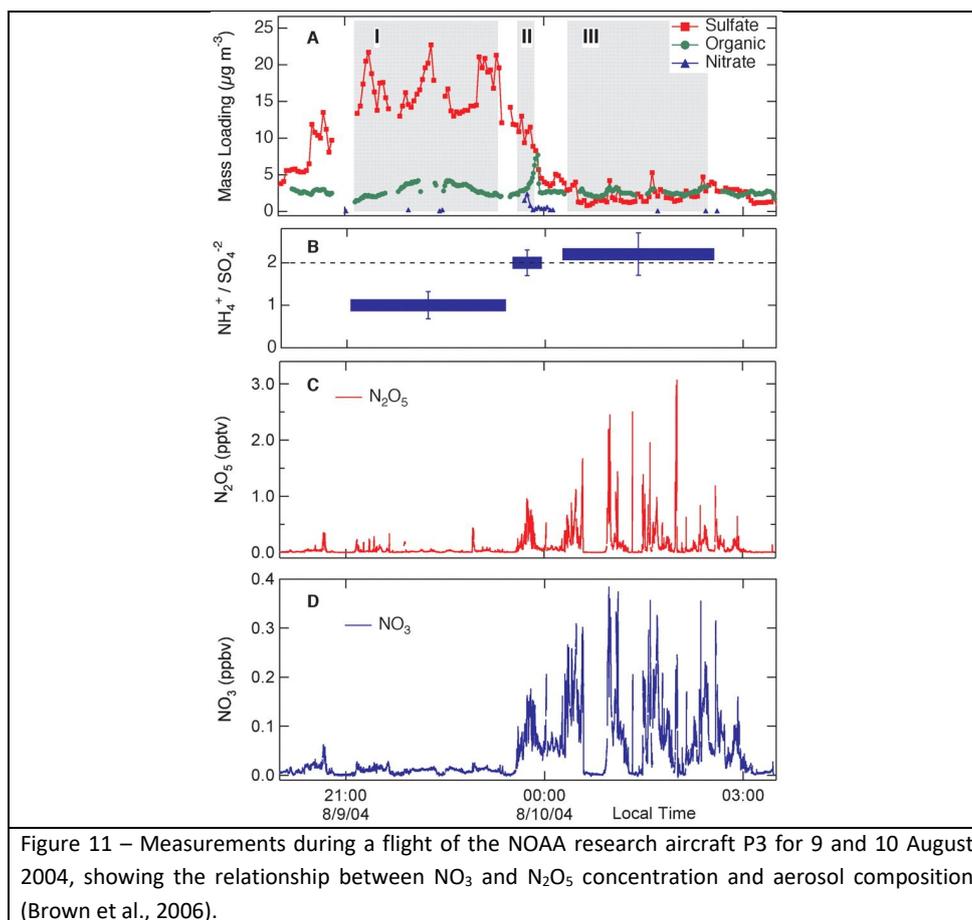



Figure 11 – Measurements during a flight of the NOAA research aircraft P3 for 9 and 10 August
2004, showing the relationship between NO_3 and N_2O_5 concentration and aerosol composition
(Brown et al., 2006).

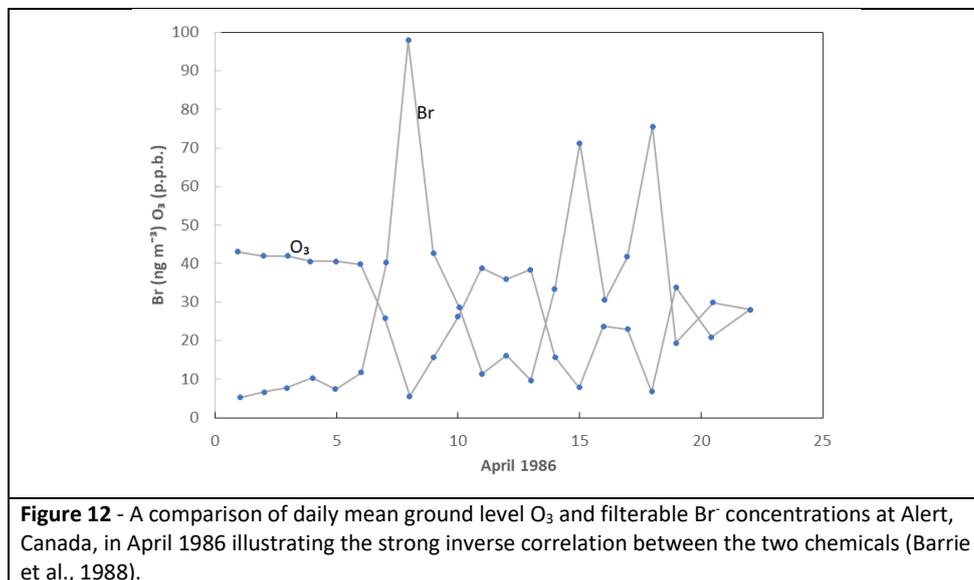
678

679   2.11.   *Halogen Chemistry*

680 In comparison to the atmospheric chemistry in the stratosphere, where halogen chemistry has been
681 well known and characterized for a long time (see the 2.18. *Stratospheric Chemistry* section), the
682 recognition of the role of halogen species in the oxidative chemistry of the troposphere occurred much
683 later. Reviews of the earlier work can be found in Platt and Hönninger (2003),Monks (2005) and the
684 extensive review by von Glasow and Crutzen (von Glasow and Crutzen, 2007).

685 The role of halogens in the troposphere has been discussed going back to the 1970s (e.g. (Graedel,
686 1979)). The potential importance of iodine in the troposphere was highlighted by a seminal paper by
687 Chameides and Davis in 1980 (Chameides and Davis, 1980). An important early paper is that from
688 Barrie in 1988 (Barrie et al., 1988) that demonstrated the dramatic impact of bromine chemistry on
689 Arctic boundary layer ozone (Figure 12). The occurrence of ozone depletion events in the polar
690 boundary layer suggested that halogens could have a significant impact on atmospheric chemistry at
691 low altitudes and not just in the stratosphere. This work brought together halogen and heterogeneous
692 chemistry and led to the discovery of bromine catalyzed ozone depletion on ice-covered surfaces. (see
693 2.5. *Heterogeneous and Multiphase Chemistry*).

694

Figure 12 - A comparison of daily mean ground level O₃ and filterable Br⁻ concentrations at Alert, Canada, in April 1986 illustrating the strong inverse correlation between the two chemicals (Barrie et al., 1988).

695

696 One of the foundational papers in the area of halogen chemistry is the modelling study by Vogt et al.
697 (1996) which set the theoretical framework for the sea-salt activation mechanism for halogen release
698 and linked halogen chemistry with the sulphur cycle. While the initial research focus was on chlorine
699 and bromine, Alicke et al. (1999) reported the first iodine oxide observations in the marine boundary
700 layer at Mace Head, Ireland, and proved that iodine can also be an important player in the chemistry
701 of the troposphere. Further investigation found evidence that biogenic iodine species can be
702 responsible for the formation of marine aerosol and cloud condensation nuclei (O'Dowd et al., 2002)
703 recognizing the potential for wide scale impact of iodine chemistry in particle formation. Finlayson-
704 Pitts and her colleagues had suggested the importance of chlorine in tropospheric chemistry based

705 on laboratory data (Finlayson Pitts, Nature 1989), but it wasn't until 2008 that Ostoff and co-workers
706 (Osthoff et al., 2008) – and the related comment "When air pollution meets sea salt" by von Glasow
707 (2008) – brought attention to the potential for nitryl chloride (ClNO$_2$) chemistry to impact ozone
708 formation, nitrogen recycling and VOC oxidation, with the first ambient observations of this molecule.
709 Also in 2008, the work from Read et al. (2008) clearly showed the global importance of halogens on
710 tropospheric ozone using long-term observations of iodine and bromine oxides (IO, BrO) made at
711 the Cape Verde Observatory.

712

713 *2.12.   Volatile Organic Compounds*

714 Volatile organic compounds (VOC) embraces a wide variety of species emitted from man-made and
715 natural sources. In many respects VOC are the fuel of the oxidative chemistry in the atmosphere,
716 involved in many gas- and particle-phase processes.

717 Ehhalt (1974) brought together the details of the methane sources and sinks and put them into a
718 consistent framework that described the life cycle of methane. This conceptual framework has
719 subsequently been developed for a wide range of trace organic gases. The original understanding of
720 the life cycle of methane has remained largely unchanged over the subsequent 40 years and has
721 formed the basis of the IPCC science assessments on the role of methane in global warming and
722 climate change. Methane itself has been long recognised as important for tropospheric chemistry, but
723 also for climate change as a greenhouse gas. Specifically, the work of Blake and Rowland (1986)
724 documented the global increase in methane and its implications for climate change.

725 The large differences in reactivity among the individual VOC have always been a feature of their
726 chemistry. Darnall et al. (1976) produced a reactivity scale for atmospheric hydrocarbons based on
727 their reaction with the hydroxyl radical, an idea that is still influential to the present day. The concept
728 was further advanced by Carter and Atkinson (1989) who looked at incremental hydrocarbon
729 reactivity, where knowledge of the reactivities of organics with respect to ozone formation in the
730 atmosphere can provide a useful basis for developing appropriate control strategies to reduce
731 ambient ozone levels. It was the beginning of an approach that is often now used in regulation to
732 determine which organic compounds would have the greatest effect in reducing ozone.

733 VOC transformation can be important in a number of different atmospheric processes. One highly
734 cited early example is the work of Pitts et al. (1978) on the atmospheric reactions of polycyclic
735 aromatic hydrocarbons and their ability to form mutagenic nitro derivatives under typical atmospheric
736 conditions.

737 While measurement techniques rarely seem to get a mention as being influential, the discipline relies
738 on observations as a critical part of the oeuvre. Already mentioned was the huge impact that accurate
739 techniques to measure the OH radicals had on the development of the field (see the sections 2.9. *HOx*
740 *chemistry)*. Another example is the development of Proton-Transfer Reaction Mass-Spectrometry,
741 which has revolutionised the measurement, in particular, of VOCs (Lindinger et al., 1998).

742 Research is ongoing as to how many VOCs there are in the atmosphere and what the consequences
743 are of not being able to measure them all. The work of Lewis et al. (2000) used novel VOC
744 measurements (GC x GC) to find that there was a larger pool of ozone-forming carbon compounds in
745 urban atmospheres than previously posited (Figure 13). The later paper by Goldstein and Galbally

746   (2007) expanded on this work hypothesizing that thousands of VOCs are still unmeasured and
747   unknown, with potentially huge consequences for the carbon budget of the atmosphere.

748   Continuing work in this area, de Gouw et al. (2005) produced a landmark study that combined analysis
749   of organic carbon in the both the gas- and particle-phase in the polluted atmosphere as part of the
750   New England Air Quality study by looking at the evolution of VOCs from their emission sources The
751   study showed that most of the organic carbon in the particle-phase was formed by secondary
752   anthropogenic processes and that an increasing fraction of the total organic mass was constituted of
753   oxygenated VOCs as a result of the air masses being processed/aged.

754

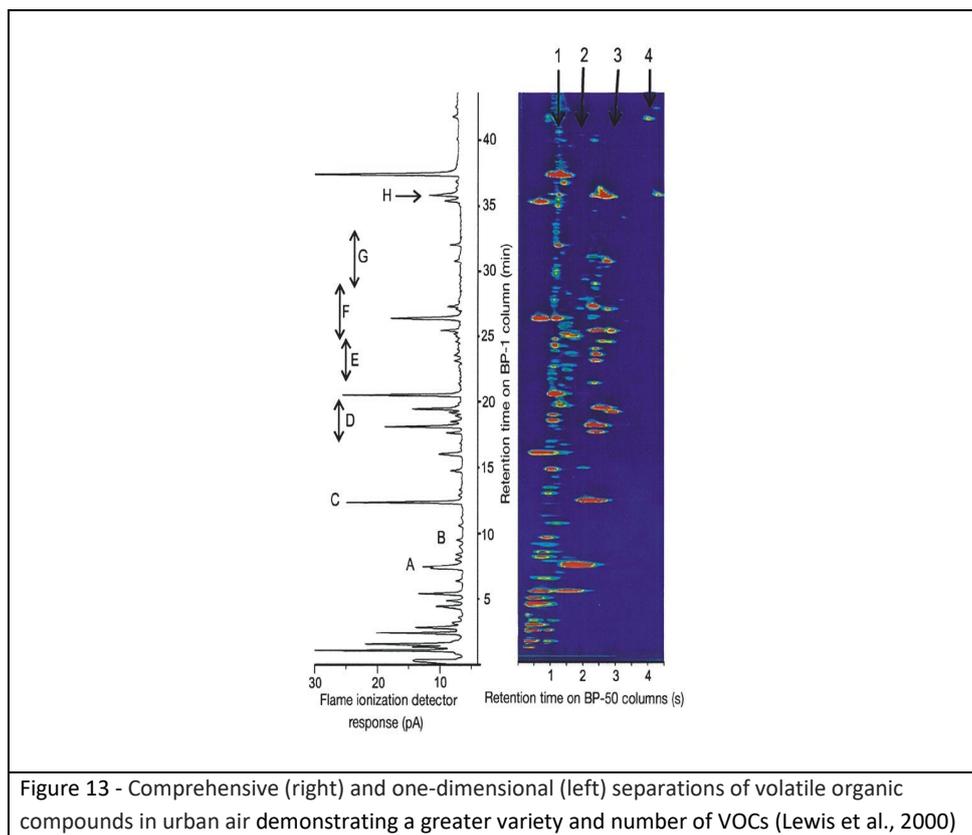



Figure 13 - Comprehensive (right) and one-dimensional (left) separations of volatile organic
compounds in urban air demonstrating a greater variety and number of VOCs (Lewis et al., 2000)

755

2.13.   Biogenic Emissions & Chemistry

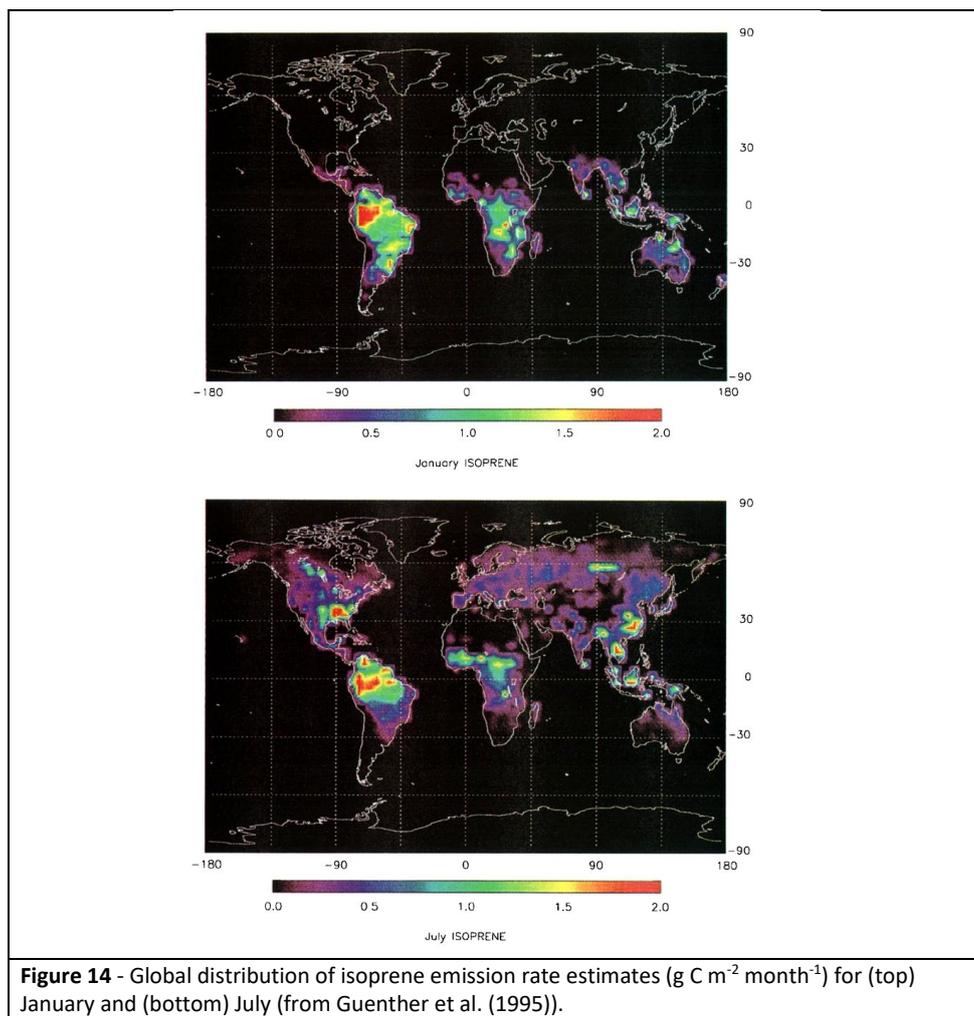
757   Although it has been known for a long time that plants emit organic compounds, the relevance of
758   biogenic VOC for atmospheric chemical processes was not immediately recognized. Detection of
759   isoprene and α-pinene in forested environments was first reported in the 1960s-1970s (Rasmussen
760   and Went, 1964;Rasmussen, 1970;Went, 1960). One of the first studies, a paper from Went (1960)
761   hypothesised that "volatilisation of terpenes and other plant products results in the production of,
762   first, blue haze, then veil clouds … ". The biological mechanisms that control the release of organic

763 compounds from vegetation were investigated by Baldwin and Schultz (1983) and many others that
764 followed.

765 Before the late 1980s, it was largely assumed that all reactive volatile organic compounds in the
766 atmosphere came from pollution sources and that biogenic sources were trivial and inconsequential.
767 The fundamental work by Chameides and co-workers (Chameides et al., 1988) was one of the first to
768 show the ubiquity of biogenic VOCs and their role in ozone formation even in urban areas. Later,
769 Fehsenfeld et al. (1992) extensively reviewed the state of the knowledge of sources and biochemical
770 processes of VOCs, bringing the subject to the attention of the wider atmospheric chemistry
771 community and opening a new branch of atmospheric chemistry. Another influential review of
772 biogenic VOC emissions, including the physiology of plants, was that by Kesselmeier and Staudt (1999).
773 These studies were pivotal for major policy formulations to abate ozone pollution.

774 Based on this work, relatively simple functions to predict the emissions of biogenic VOC were
775 developed early on by Guenther et al. (1993) and soon evolved into more sophisticated high resolution
776 global models (Guenther et al., 2000;Guenther et al., 1995), allowing for the emissions of biogenic
777 compounds to be included in atmospheric chemistry models across all scales, and is still used in
778 modern Earth system models today. Eventually, this work took the form of the widely used MEGAN
779 model (Model of Emissions of Gases and Aerosols from Nature) (Guenther et al., 2006) (Table 2 and
780 Figure 14).

781

Figure 14 - Global distribution of isoprene emission rate estimates (g C m^{-2} month^{-1}) for (top) January and (bottom) July (from Guenther et al. (1995)).

782

783 Terrestrial vegetation is not the only source of biogenic emissions. Aneja et al. (1979) discussed the
784 importance of biogenic sulphur compounds and their role in stratospheric chemistry, while Charlson
785 et al. (1987) connected marine biology, atmospheric chemistry and climate into the already
786 mentioned CLAW hypothesis (see the 2.2. *Aerosols and Clouds* section).

787 Marine aerosol formation was thought for a long time to be dominated by inorganic components,
788 mainly sea-salt and non-sea salt sulphate, but O'Dowd and co-workers (O'Dowd et al., 2004) showed
789 that biological activity of plankton blooms can enhance the concentration of cloud condensation
790 nuclei (CCN), a key aspect of the chemistry-climate feedback mechanism. A similar mechanism is also
791 active in relation to biogenic halogen compounds (O'Dowd et al., 2002) which also affect aerosol
792 formation as well as the ozone, nitrogen and sulphur cycles, as discussed in the *Aerosols and Clouds*
793 section.

794 A paper that has been defined as "controversial but set off a huge amount of activity" is "Atmospheric
795 oxidation capacity sustained by a tropical forest" by Lelieveld et al. (2008). In their work the authors
796 proposed a new chemical mechanism for low NOx, high VOC regions (such as tropical forests), based
797 on modelling studies of a field dataset. Although further studies contradicted this hypothesis, Lelieveld
798 et al. (2008) was instrumental in prompting a large amount of laboratory and theoretical studies in
799 the past 10 years; these studies resulted in a major revision of our understanding of biogenic VOC
800 chemistry, as discussed in the *HOx chemistry* section.

801

802 ### 2.14. *Biomass Burning*

803 Biomass burning, particularly in the tropics, affects terrestrial vegetation dynamics, soil erosion,
804 movement of organic carbon, hemispheric atmospheric composition, air quality and more broadly the
805 radiative forcing *via* emissions of trace gases and aerosols (Monks et al., 2009). Crutzen (Crutzen et
806 al., 1979) was the first to highlight biomass burning in the tropics as an important source of
807 atmospheric gases, such as CO, H_2, N_2O, NO, CH_3Cl and COS. The importance of biomass burning, based
808 on the observations of a small set of fires, and the appreciation of its potential role was a major step
809 in our understanding of the role of biomass burning in air quality, climate change, and the composition
810 of the troposphere. It is, however, the later paper "Biomass Burning in the Tropics: Impact on
811 Atmospheric Chemistry and Biogeochemical Cycles" by Crutzen and Andrea (Crutzen and Andreae,
812 1990) – one of the top 10 most cited Atmospheric Chemistry paper (Table 2) – that has had the
813 greatest impact on this research area . Hao and Liu (1994) made a further step forward, looking at
814 where and when biomass burning and thereby the related emissions occur. They developed an
815 improved database of the amount of biomass burned owing to deforestation, shifting cultivation,
816 savannah fires, fuel wood use, and clearing of agricultural residues, focused on tropical America, Africa
817 and Asia during the late 1970s.

818 Simoneit et al. (1999) introduced the important concept that "the monosaccharide derivatives (e.g.
819 levoglucosan) are proposed as specific indicators for cellulose in biomass burning emissions." They
820 showed that levoglucosan is emitted at such high concentrations that it can be detected in air pollution
821 filter samples at considerable distances from the original combustion source, allowing for source
822 apportionment.

823 The 2001 paper "Emission of trace gases and aerosols from biomass burning" by Andreae and Merlet
824 (2001), which is also one of the top 10 most cited paper (Table 2 and Figure 15), pulled together
825 emission factors for a large variety of species emitted from biomass fires and is considered a key
826 reference for biomass burning emission factors. Further work in the biomass burning area was later
827 presented by Reid et al. (2005) in a review paper where they looked at measurements of smoke
828 particle size, chemistry, thermodynamic properties, and emission factors from a variety of sources,
829 including laboratory burns, in-situ experiments, remote sensing and modelling. They brought together
830 information from the 'milieu of small pieces of the biomass-burning puzzle' and showed that there are
831 large differences in measured particle properties and particle carbon budgets across the literature.
832 van der Werf et al. (2006) investigated interannual variability and the underlying mechanisms
833 regulating variability at continental to global scales using a time series of 8 years of satellite and model
834 data. Total carbon emissions was driven by burning in forested areas, while the amount of burned

835   area was driven by savannah fires, which are influenced by different environmental and human factors
836   than forest fires.

837

Figure 15 - Vertical profiles of O_3 in the tropical troposphere. The profile over the equatorial Pacific shows no influence from biomass burning, whereas the profile over the Pacific off South America suggests O_3 enhancement due to long-range transport from the tropical continents. The O_3 profiles over Brazil and the Congo show high O_3 concentrations at altitudes between 1 and 4 km due to photochemical production in biomass burning plumes. At higher altitudes, O_3 concentrations are also substantially enhanced, possibly also because of O_3 production by reactions in the emissions of biomass burning (Crutzen and Andreae, 1990).

838

839   *2.15.   Emissions and Deposition*

840   Non-chemical sources and sinks of various species are critical components of atmospheric processes
841   and therefore are particularly essential for global and regional models. An early advocate for such was
842   the work of Olivier and the team that created the EDGAR (Emission Database for Global Atmospheric
843   Research) database (Olivier et al., 1994).

844   Emissions from vehicles and power plants have always been an essential aspect of air quality related
845   policies, and therefore an area where more focused inventory work was needed and done. This
846   approach was pioneered in California in the early 1990s, and the studies by Calvert et al. (1993),
847   Lawson (1993) and Singer and Harley (1996) helped define and verify the California Smog Control
848   Program, providing a solid scientific basis with reliable emissions data. Techniques such as the remote
849   monitoring of traffic generated carbon monoxide (Chaney, 1983) are also essential to recognise the
850   role they played in understanding vehicle emissions.

851   The deposition of gases and aerosol to the surface is another critical process in the atmosphere.
852   Chamberlain (1966) is credited with the first exposition of the resistance network approach to describe
853   the uptake of gases on surfaces and the identification of transport through the atmospheric boundary

854   layer, through the surface layer, and through the stomata on plants, as important elements of surface
855   uptake. Building upon this work, a comprehensive and widely adopted, parametrization of the dry
856   deposition process for regional and global models was presented in the late 1980s by Wesely (1989).
857   Currently, there has been renewed interest in quantifying and understanding deposition process. Yet,
858   a systematic description based on fundamental independently measureable physico-chemical
859   parameters are lacking.

860

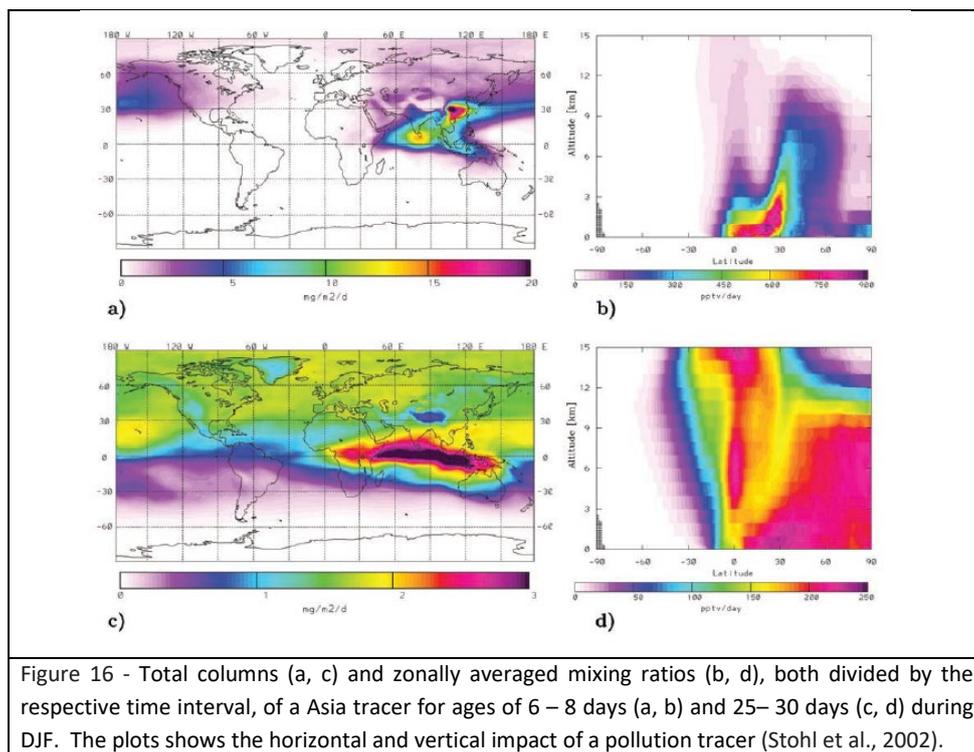
861   2.16.   *Chemical Transport*

862   Transport is an integral part of atmospheric processes and influence atmospheric composition across
863   a range of spatial scales. As early as 1975, Junge (1975) pointed out the importance of atmospheric
864   residence time of a constituent with respect to global transport and dispersion. Prather's work
865   (Prather, 1994, 1996) provided new insights onto timescales for atmospheric oxidation chemistry.

866   Stratospheric-tropospheric exchange (STE) has always been recognised as a key mechanism in
867   determining tropospheric composition. Early chemical dynamics were demonstrated by Danielsen
868   (1968) that laid the foundations for 3-dimensional modelling of chemical transport looking at
869   stratosphere-troposphere exchange based on radioactivity, ozone, and potential vorticity. Later,
870   Holton and co-authors (Holton et al., 1995) proposed an approach that placed stratosphere-
871   troposphere exchange in the framework of the general circulation and helped clarify the roles of the
872   different mechanisms involved and the interplay between large and small scales, by the use of
873   dynamical tracers and potential vorticity. This work is recognised as a big step forward for the
874   understanding of the tropospheric ozone budget.While the regional nature of air pollution has always
875   been recognised, that is less the case for the impact of trans-continental emissions on air quality. Jacob
876   et al. (1999) showed that there was a need for a global outlook for understanding regional air quality
877   and meeting pollution reduction objectives. This perspective spawned a decade of intense work on
878   intercontinental air pollution and transport. Recognised in this area is the work by Stohl and colleagues
879   (Stohl et al., 2002) who mapped out the pathways and timescale of intercontinental air pollution
880   transport and brought life to the subjects of atmospheric dynamics and transport of air pollution
881   (Figure 16).

882

883

Figure 16 - Total columns (a, c) and zonally averaged mixing ratios (b, d), both divided by the respective time interval, of a Asia tracer for ages of 6 – 8 days (a, b) and 25– 30 days (c, d) during DJF. The plots shows the horizontal and vertical impact of a pollution tracer (Stohl et al., 2002).

884

885 The study Moody et al. (1998) explored atmospheric transport history using back trajectories for the
886 Harvard Forest experiment demonstrating the power of trajectory methods at the regional scale. Stohl
887 and colleagues (Stohl et al., 2003) brought together what has been viewed by many as the
888 authoritative work on stratosphere-troposphere exchange. Key tools in the development of this area
889 were the HYSPLIT (Draxler and Hess, 1998) a forerunner of various particle trajectory and dispersion
890 models, which developed into a widely used particle dispersion model , and the FLEXPART model, a
891 Lagrangian particle dispersion model designed for calculating the long-range and mesoscale dispersion
892 of air pollutants (Stohl et al., 2005).

893 The ideas of intercontinental source-receptor relationships as embodied in the aforementioned earlier
894 works by Jacob et al. (1999) and Stohl et al. (2002). The concept was developed in an effort to explore
895 the source-transport relationships that drive ozone in regions farther away from the emission regions
896 (Fiore et al., 2009) (see the 2.6. *Chemical Models* section). This large community collaboration
897 provided valuable insights into the sensitivities of the hemispheric regional background of ozone and
898 how this is controlled by emissions from continental source regions.

899 Another critical area for atmospheric chemistry is boundary layer dynamics and meteorology. This is
900 particularly important since most emissions are emmited in the boundary layer. Atmospheric
901 dynamics in this important region has been mostly expressed as parameterizations in numerical
902 models (Stull, 1988). The spatial and temporal scales involved in the processes in this region range
903 over a very wide range. The understanding of this region has been mostly based on meteorological
904 and energy/water vapour balance points of view. However, the chemical transformation and

905   dispersion in this region are crucial for how much chemicals actually get out of this region to influence
906   the regional and global atmosphere. Furthermore, the process of dry deposition, a critical loss
907   processes for chemicals, is mostly limited to the boundary layer.

908

909   *2.17.*   *Satellites and the Troposphere*

910   The importance of satellites on the discipline of atmospheric chemistry centres on the ability to give
911   a self-consistent global view of a selected set of tropospheric trace species. The beginning and first
912   demonstration of the effective application of these attributes for the troposphere were the data and
913   the retrievals from the GOME instrument (Burrows et al., 1999) on ERS-2 and SCIAMACHY on Envisat
914   (Bovensmann et al., 1999). Historically, the roots of these early instruments are in stratospheric
915   chemistry, with GOME being deployed to be able to track stratospheric ozone and its key controlling
916   chemical species.

917   The most nominated paper in this area, and one that demonstrated the power of such observations
918   for tropospheric composition research, was "Increase in tropospheric nitrogen dioxide over China
919   observed from space" by Richter et al. (2005), which showed the capability of the satellites to track
920   the build-up of air pollution over vast regions from space (Figure 17). The importance of the work lead
921   by Palmer and co-workers in establishing a method to convert satellite observations to vertical
922   columns for comparison with e.g. models was also widely recognised (Palmer et al., 2001).

923

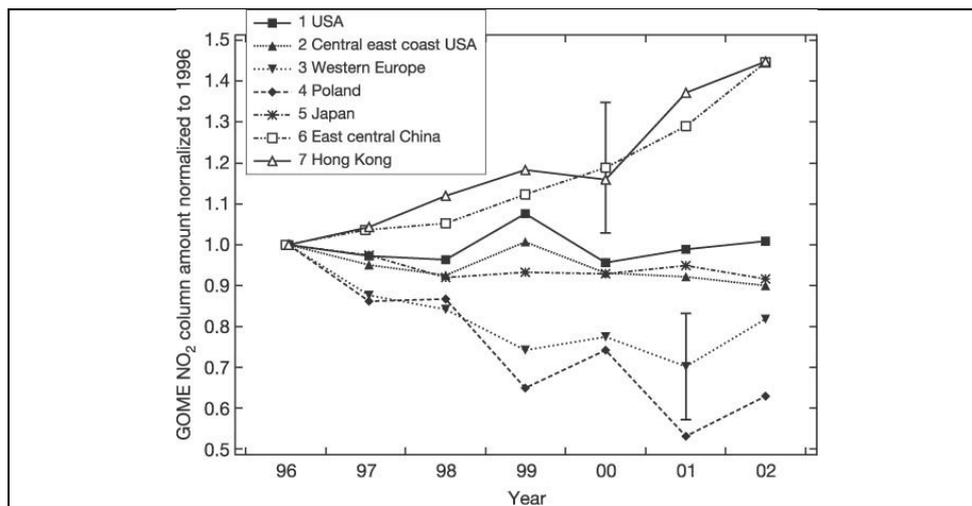



Figure 17 - The mean annual NO_2 column amount normalized to that in 1996 for the geographical regions USA, Central East Coast USA, Western Europe, Poland, Japan, East Central China, and Hong Kong showing a marked increase in NO_2 over China and decrease over Europe (Richter et al., 2005).

924

925   Satellite-based instrumentation can measure not only gas-phase trace species but also dust and
926   aerosols; the combination of SEAWIFs and TOMS to track Asian dust events (Husar et al., 2001) and
927   the development of the MODIS aerosol algorithm (Remer et al., 2005) provided convincing

928   demonstrations of this capability. A step-change in this area was made with the paper "Estimating
929   ground-level PM2.5 using aerosol optical depth determined from satellite remote sensing" by van
930   Donkelaar et al. (2006) which was the first description of the derivation of surface PM2.5 from satellite
931   AOD, and which has been extensively used to estimate the global impact of particulate matter (both
932   PM2.5 and PM10) on health.

933

934   *2.18.   Stratospheric Chemistry*

935   Tropospheric chemistry has always been influenced by the study of stratospheric chemistry. As
936   mentioned earlier (see the 2.1. *Foundations* section), the basis of stratospheric ozone chemistry were
937   laid in the 1930s (Chapman, 1930), whereas tropospheric chemistry followed a couple of decades
938   later. Interest in stratospheric ozone chemistry increased substantially following the works of
939   Johnston and Crutzen on the role of nitrogen oxides in the stratosphere (Crutzen, 1970;Johnston,
940   1971). The impact of supersonic transport (Johnston, 1971) and of CFCs (Molina and Rowland, 1974)
941   were important events for stratospheric ozone chemistry. While Crutzen (1970) showed that the
942   nitrogen oxides in the stratosphere come from the nitrous oxides from the ground, Johnson suggested
943   that a fleet of supersonic aircraft could release large amounts of nitrogen oxides into the lower
944   stratosphere causing substantial ozone loss (Johnston, 1971). The potential threat of supersonic
945   transport highlighted the importance of gas-phase catalysis in the atmosphere and, in particular, of
946   the catalytic ozone destruction by HO_x and NO_x. These works opened the world's eyes to the potential
947   for global environmental change from human activities. Soon after, Lovelock (1974) identified CFCs in
948   the troposphere and showed that practically all the CFCs emitted to date were still in the atmosphere.
949   The significant contributions of Hampson, Crutzen, and Johnston, and the recognition of chlorine-
950   catalysed ozone destruction by Stolarski and Cicerone (1974), paved the way for the seminal work of
951   Molina and Rowland (Molina and Rowland, 1974) linking chlorofluorocarbons to ozone layer
952   depletion. The recognition that bromine compounds can also destroy stratospheric ozone (McElroy et
953   al., 1986) further refining the story. The potential role of iodine in the stratospheric ozone depletion
954   has been raised (Solomon et al.1994), but it is still somewhat unsettled.

955   The ozone hole (Farman et al., 1985) was an unanticipated shock that awoke the world to the global
956   nature of ozone layer depletion. In an historic set of studies over a relatively short five-year period,
957   the cause of the ozone hole was understood. First was the insightful and seminal work of Solomon et
958   al. (1986) that showed that chlorofluorocarbons and other ozone-depleting gases were the key
959   anthropogenic ingredient for the ozone hole. The confluence of cold temperatures that lead to the
960   formation of polar stratospheric clouds (PSCs) and the winter vortex formation over Antarctica
961   provided the opportunity for the massive ozone depletion that resulted in the ozone hole. This work
962   confirmed the suggestion of Farman that the ozone hole was due to the increasing abundances of
963   CFCs. In particular, Solomon and co-workers (Solomon et al., 1986) recognized that stable molecules
964   such as $ClONO_2$ and HCl could react on solid (and indeed liquid). Along the way, during this intense
965   investigative period, the detection and quantification of the role of ClO as a catalyst by Anderson et
966   al. (1991), as well as De Zafra et al. (1988), was the "smoking gun" that linked the CFCs with the ozone
967   hole. The entire set of field measurements, from the ground, aircraft, and balloons, solidified this
968   linkage.

969 Less heralded, but equally important, were the laboratory studies that showed that chlorine nitrate
970 (ClONO$_2$) and HCl did indeed react on PSCs (Hanson and Ravishankara, 1994;Tolbert et al., 1987;Leu,
971 1988;Molina, 1991;Molina et al., 1987) and determined the critical rate coefficients for the self-
972 reaction of ClO, the rate-limiting step in the unique catalytic cycles in Antarctica (Cox and Hayman,
973 1988;Sander et al., 1989;Trolier et al., 1990). Much was learned in later years by studying the Arctic
974 and from the continued observations over the Antarctic. It should be noted that the termolecular
975 reaction of ClO was suggested to be important for high chlorine chemistry by Molina and Molina, and
976 the history of this reaction goes back to Norish's work at Cambridge (Norrish and Neville, 1934). One
977 of the lessons from this episode is that natural factors, in this case the formation of a vortex and the
978 occurrence of polar stratospheric clouds, can lead to unexpected consequences when an
979 anthropogenic ingredient (ozone-depleting chemicals) is added to the mix.

980 The numerical modelling of the stratosphere was an important ingredient for the success of mitigating
981 polar ozone loss, along with the theories of the ozone layer depletion and the ozone hole, the
982 laboratory studies of key processes, and the measurements in the atmosphere. Over the years, these
983 models have enabled a great deal of understanding of the coupling between chemistry and climate.
984 The development of stratospheric chemical transport models (Chipperfield and Pyle, 1988) was a
985 pivotal advancement that enabled quantitative understanding of the ozone layer depletion, including
986 the ozone hole (see the 2.6. *Chemical Models* section).

987 The weight of science led to the Vienna convention, the Montreal Protocol, and the Protocol's many
988 amendments and adjustments that have, by now, phased out the ozone-depleting gases. The
989 Montreal Protocol is the first international treaty on an environmental issue to be universally ratified
990 and is regarded as one of the most successful. That said, the ozone layer depletion story is not
991 complete. For example, the recognition that nitrous oxide is the remaining major ozone-depleting gas
992 emission (Ravishankara et al. 2009) has connected food production (tropospheric nitrogen cycles) to
993 ozone layer depletion and highlighted the importance of a holistic approach to environmental issues.

994 *2.19 Other issues that influence tropospheric chemistry*

995 Atmospheric chemistry advances have been advanced by growth in other areas. The importance of
996 anthropogenic climate change has been instrumental in invigorating atmospheric chemistry studies
997 because many of the major climate forcing agents are chemically active and climate change, in turn,
998 influence tropospheric chemistry (von Schneidemesser et al., 2015). This issue was noted earlier. In
999 addition to climate change, other adjacent discoveries and findings have influenced tropospheric
1000 chemistry studies.

1001 The global atmospheric and climatic consequences of nuclear war were investigated by both Crutzen
1002 and Birks (1982) and Turco et al. (1983). Using models developed for looking at the impact of volcanic
1003 eruptions, Turco et al. (1983) concluded that "enhancement of solar ultraviolet radiation due to ozone
1004 depletion, long-term exposure to cold, dark, and radioactivity could pose a serious threat to human
1005 survivors and to other species." Similarly (Crutzen and Birks, 1982) concluded that "the screening of
1006 sunlight by the fire-produced aerosol over extended periods during the growing season would
1007 eliminate much of the food production in the Northern Hemisphere".

1008 Air quality has an obvious direct impact on people, and this connection was recognized very early (it
1009 was in fact the primary motivation behind the fundamental work of Haagen-Smit). In 1993, Dockery
1010 et al. (1993) presented a study of six US cities showing a direct association between air pollution and

1011 mortality rates. This paper a great example of an adjacent field influences another, in this case
1012 atmospheric chemistry and public health.   Though association between air pollution and health
1013 stretch back to the Los Angeles and London smog, the 'Six Cities Study' was a landmark as it
1014 demonstrated the association between air pollution and mortality extended to much lower
1015 concentrations than those observed in the smog days.

1016

1017 **3. Discussion and Summary**

1018 A mixture of the history of the discipline and its the landmark ideas emerges in this exercise of asking
1019 the community to point to what they consider has shaped their research field. Table 1 seeks to bring
1020 these elements together to look at the evolution of the leading scientific concepts, their relevance to
1021 the environmental legislation (in this sense, we acknowledge an Euro-/US-centric bias, which the
1022 community can perhaps correct during the discussion of this paper), and the most notable
1023 environmental events that have shaped the discipline. Atmospheric science often sits at an interesting
1024 intersection between the societal outcomes (e.g., acid precipitation, air quality, ozone layer depletion,
1025 and climate) and its scientific venture. Monks and Williams (2020) have recently explored how
1026 environmental events in air quality drive policy and how a scientific and societal paradigm shift occurs
1027 once the emergency phase has passed.

1028 From an overview of all the nominated papers, several general features are apparent. Ambient
1029 measurements are one of the cornerstones of atmospheric science (Abbatt et al., 2014). It is clear
1030 that the atmosphere is under-sampled but over time we have found many ingenious ways to build
1031 different measurement strategies from the ground, airborne, balloon, sonde and space. With a focus
1032 on chemistry, it is clear that one needs to be able to measure with surety, specificity and speed in the
1033 troposphere.   Many of the nominated papers reflect the importance of instrument developments.
1034 Examples include the electrostatic sizers in aerosol science (Knutson and Whitby, 1975), various
1035 techniques to measure the hydroxyl radical (Eisele, 1994;Stevens et al., 1994;Perner et al., 1976), the
1036 development of chemical ionization mass spectrometry (Lindinger et al., 1998), and the application of
1037 the GCxGC-MS technique (Lewis et al., 2000), and aerosol mass-spectrometry measurements, e.g.,
1038 Zhang et al. (2007). Often, the science underlying the development of these instruments, such as ion-
1039 molecule chemistry, is not necessarily acknowledged.

1040 Another common theme is the critical importance and impact of long-term observations, often termed
1041 monitoring, of key atmospheric components, from CO_2 (the "Keeling" curve) to chemically active
1042 molecules such halocarbons (the Antarctic ozone hole, ozone layer depletion and climate change),
1043 methane (the changes in the global OH field, background ozone production), and NO_x (catalyst for
1044 tropospheric ozone production, vehicle emissions and acid precipitation). On the other hand, many
1045 breakthroughs in understanding the observations emerged because of basic laboratory information
1046 on kinetics and photochemistry (e.g., the reaction of HO_2 + NO, the determination of $O(^1D)$ quantum
1047 yields, and the reactions of $ClONO_2$ and HCl on PSCs.) It is noteworthy that both laboratory studies and
1048 long-term observations are currently under funding stress, a situation that is already worrying the
1049 community (see, for example, the discussion in Burkholder et al. (2017)).

1050 There is no doubt that atmospheric chemistry is an integrative science: one of the recurring themes
1051 in the papers discussed here is the tight relationship between ambient observations, laboratory
1052 experiments, and modelling. The integrative power of models has been recognised from the early

studies by Levy (1971) to the development of highly sophisticated global transport models by
Chipperfield and Pyle (1988) and Bey et al. (2001) up to the more recent demonstrations of the power
of model ensembles (Stevenson et al., 2006;Fiore et al., 2009). Much of this progress has parallels in
stratospheric chemistry. It is evident in the community that models are a powerful tool to map, test,
and predict the atmosphere's past, present, and future. The predictions and projections from these
models play essential roles in policy, planning, and management of environmental issues.

Another form of integrative or meta-analysis brings together a range of individual studies to produce
a more significant outcome, such as new insights or models. There are some notable examples of this
approach in the works on biomass burning by Crutzen and Andreae (1990) and more recently, by van
der Werf et al. (2010). Similarly, the work to produce isoprene emissions models brought together
several global isoprene flux measurements (Guenther et al., 1995;Guenther et al., 2006). Other
examples include the work of Zhang et al. (2007) and Jimenez et al. (2009) who integrated various sets
of AMS observations to give insight into land distributions of SOA.

There have been developments in fields adjacent to atmospheric chemistry that have shaped
atmospheric chemistry progress. Examples include developments in epidemiology (e.g., the Six Cities
Study by (Dockery et al., 1993)), atmospheric dynamics (the role of transport in determining chemical
composition, the role of the Antarctic vortex), in ocean science (pertaining to deposition to the ocean
surface and emissions from the oceans) and in biospheric/plant science (e.g. (Kesselmeier and Staudt,
1999)). Integrating atmospheric chemistry with these adjacent fields is not only essential but also
fruitful and was for many years embodied in the IGBP (Seitzinger et al., 2015), World Climate Research
Program (WCRP), and Earth-system science programs (now Future Earth). Wider contexts such as
paleoclimate has allowed an understanding of climate and atmospheric history over the 100,000 year
timescales (e.g. (Petit et al., 1999)). That work set the understanding that the present-day atmospheric
burdens of carbon dioxide and methane as important greenhouse gases are unprecedented during
the past 420,000 years. They also allow us to estimate the composition of the troposphere in those
ancient times.

Similarly, there are concepts that have their roots in tropospheric chemistry that have gone onto have
wider impact. The concept of the Anthropocene, first proposed by Crutzen and Stroemer in 2000,
indicates that we are in a new geological epoch driven by human activities. The idea was more fully
expounded in Crutzen (2002) the geology of mankind. There is little doubt that this has been a key
idea that has influenced much thinking as well as wider work beyond atmospheric science (Table 1).
As discussed in detail in the introductory text, we opted here to assemble this compilation of papers
by reaching out to the community for nominations, rather than using the number of citations as a
primary measure. We will not go into the details again here, suffice it to say that there are inherent
advantages and drawbacks to any method one might consider for such a work. In that sense, we would
re-iterate our call for input during the open peer-review phase for this manuscript.

The papers highlighted capture a substantial scope of the atmospheric research endeavour over the
last 60 years. The challenge now for you, the reader, is to reflect on this preliminary version of included
papers, critique the choice and the reasoning, and propose other suggestions. Again we request you
to engage in this discussion and help shape the final compilation of papers to make it a truly
community effort.

4. Author Contributions

1095

1096 PSM developed the concept and led the writing, PSM, ARR, RS, and EvS solicited input, ARR, RS, EvS
1097 contributed to writing and editing.

1098

1099 **Table 1 – Science, Regulatory and Environmental Landmarks of the 20th and early 21st Centuries**

Decade	Science Landmark	Regulatory Landmarks[1]	Environmental Events
1930	Chapman Cycles and Stratospheric Chemistry		
1940			1943 – LA Smog
1950	Air Pollution	1956 - UK Clean Air Act	1952 – Great Smog of London
1960		1963 - US Clean Air Act	
1970	SST Stratospheric Chemistry Tropospheric Chemistry Air Pollution and Clouds	1978 – Ban of Lead in Petrol (USA) 1979 - CLRTAP (UNECE)	1974 – Observations of acid rain
1980	Ozone Hole Chemistry Halogen Chemistry Biogenic Chemistry	1987 - Montreal Protocol 1980 - SO_2 directive (EU)	1985 – Observations of the ozone hole
1990	Air Pollution and Health Satellite Observations of the Troposphere Long-Range Transport of Air Pollutants	1992 – Euro 1 Emission standard 1992 - Ozone Directive (EU) 1997 - Kyoto Protocol 1999 - Goteborg Protocol 1999 – Ban of lead in petrol (EU)	1991 - Mt Pinatubo eruption
2000	SOA Concept of Anthropocene	2001 - NEC Directive (EU)	
2010	Air Pollution and Climate		2015 - Dieselgate
2020			2020 - COVID 19

1100

1101   [1] For more details on the UK/EU perspective see (Williams, 2004;Maynard and Williams, 2018) and
1102   for the USA perspective see (Jacobson, 2002); see also (Monks and Williams, 2020).

Table 2 – Top 10 Cited Atmospheric Chemistry Papers (Atmospheric+Chemistry)

(Scopus, 27/3/20)

#	Paper	Title	Citations
1	Guenther et al. (1995)	A global model of natural volatile organic compound emissions	2760
2	Andreae and Merlet (2001)	Emission of trace gases and aerosols from biomass burning	2350
3	Guenther et al. (2006)	Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature)	2175
4	Jimenez et al. (2009)	Evolution of organic aerosols in the atmosphere	1909
5	Atkinson (2000)	Atmospheric chemistry of VOCs and NOx	1773
6	Crutzen and Andreae (1990)	Biomass burning in the tropics: Impact on atmospheric chemistry and biogeochemical cycles	1686
7	van der Werf et al. (2010)	Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997-2009)	1578
8	Atkinson and Arey (2003)	Atmospheric Degradation of Volatile Organic Compounds	1502
9	Grell et al. (2005)	Fully coupled "online" chemistry within the WRF model	1436
10	Lelieveld et al. (2015)	The contribution of outdoor air pollution sources to premature mortality on a global scale	1425

(Web of Science, 27/3/20)

#	Paper	Title	Citations
1	Ramanathan et al. (2001)	Atmosphere - Aerosols, climate, and the hydrological cycle	2278
2	Andreae and Merlet (2001)	Emission of trace gases and aerosols from biomass burning	2168
3	Hallquist et al. (2009)	The formation, properties and impact of secondary organic aerosol: current and emerging issues	1988
4	Jimenez et al. (2009)	Evolution of organic aerosols in the atmosphere	1844
5	Crutzen and Andreae (1990)	Biomass burning in the tropics: Impact on atmospheric chemistry and biogeochemical cycles	1603
6	Atkinson (2000)	Atmospheric chemistry of VOCs and NOx	1596
7	Atkinson et al. (1992)	Evaluated Kinetic and Photochemical Data for Atmospheric Chemistry: Supplement IV. IUPAC Subcommittee on Gas Kinetic Data Evaluation for Atmospheric Chemistry	1488
8	Grell et al. (2005)	Fully coupled "online" chemistry within the WRF model	1332
9	Lelieveld et al. (2015)	The contribution of outdoor air pollution sources to premature mortality on a global scale	1292

10	Bey et al. (2001)	Global modeling of tropospheric chemistry with assimilated meteorology: Model description and evaluation	1212

1107

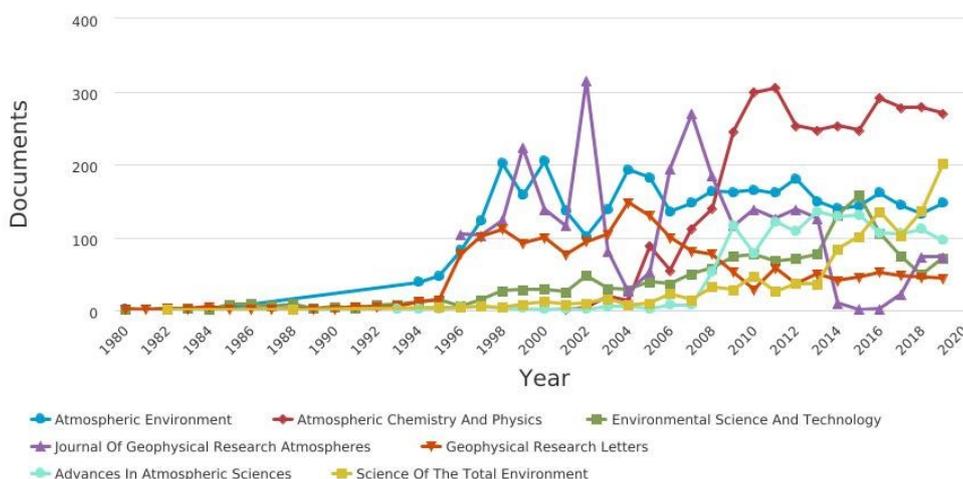
1108

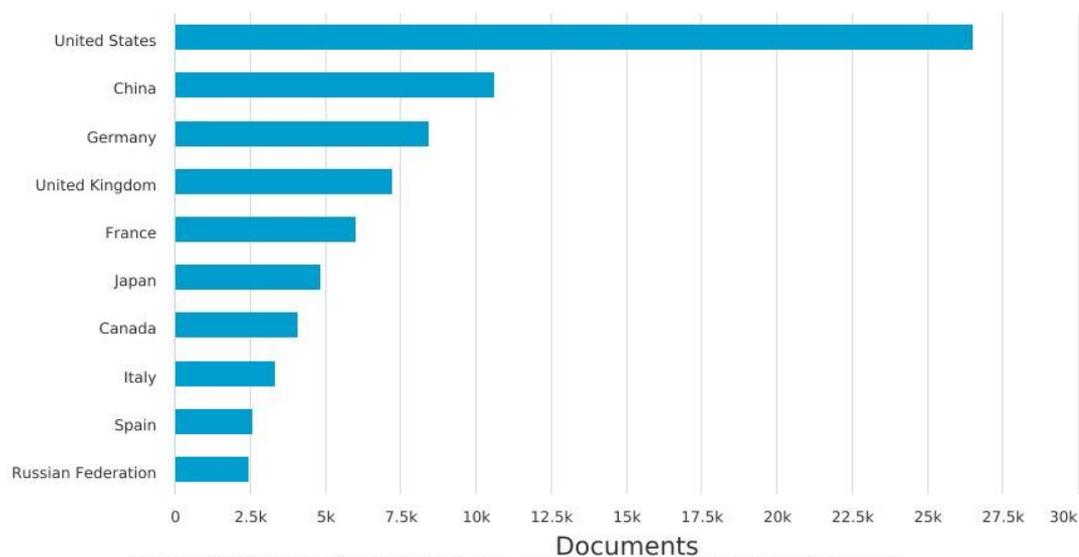
1109 **Table 3** - A global budget for NOx (Logan, 1983)

		10^{12} gm N yr^{-1}
Sources		
	Fossil fuel combustion	21 (14-28)
	Biomass Burning	12 (4-24)
	Lightning	8 (8-20)
	Microbial activity in soils	8 (4-16)
	Oxidation of ammonia	1-10
	Photolytic or biological processes in the ocean	<1
	Input from the Stratosphere	≈ 0.5
Total		25-99
Sinks		
	Precipitation	12-42
	Dry Deposition	12-22
Total		24-64

1110

1111   **Appendix 1**

1112

1113

1114

1115

1116   **Appendix 2 - Acronyms**

ABLE	Amazon Boundary-Layer Experiment
AGAGE	Advanced Global Atmospheric Gases Experiment
AMS	Aerosol Mass Spectrometer
AOD	Aerosol Optical Depth
BBOA	Biomass-Burning Organic Aerosol
CAPRAM	Chemical Aqueous Phase Radical Mechanism
CCN	Cloud Condensation Nuclei
CFC	Chlorofluorocarbon
CLAW	Charlson-Lovelock-Andreae-Warren
CTM	Chemical Transport Model
EDGAR	Emission Database for Global Atmospheric Research
FA-AMS	Factor Analysis of Aerosol Mass Spectrometry
FLEXPART	FLEXible PARTicle dispersion model
GC	Gas Chromatography
GC-MS	Gas Chromatography-Mass Spectrometry
GEOS-Chem	Goddard Earth Observing System - Chemical Transport Model
GOME	Global Ozone Monitoring Experiment
HCFC	Hydrochlorofluorocarbon
HOA	Hydrocarbon-like Organic Aerosol
HYSPLIT	Hybrid Single-Particle Lagrangian Integrated Trajectory model
IAGOS	In-service Aircraft for a Global Observing System
IGAC	International Global Atmospheric Chemistry
IGBP	International Geosphere-Biosphere Programme
IPCC	Intergovernmental Panel on Climate Change
IUPAC	International Union of Pure and Applied Chemistry
JPL	NASA Jet Propulsion Laboratory
LIF	Laser Induced Fluorescence
LV-OOA	Low-volatility Oxygenated Organic Aerosol
MCM	Master Chemical Mechanism
MEGAN	Model of Emissions of Gases and Aerosols from Nature
MODIS	Moderate Resolution Imaging Spectroradiometer
MOZAIC	Measurements of OZone and water vapour by in-service Airbus airCraft
NASA	National Aeronautics and Space Administration
NMHC	Non-Methane Hydrocarbon
NOAA	National Oceanic and Atmospheric Administration
OA	Organic Aerosol
OOA	Oxygenated Organic Aerosol
OPE	Ozone Production Efficiency

PAN	Peroxyacetyl Nitrate
PM	Particulate Matter
POA	Primary Organic Aerosol
PSC	Polar Stratospheric Clouds
RACM	Regional Atmospheric Chemistry Mechanism
RADM	Regional Acid Deposition Model
RF	Radiative Forcing
RRKM	Rice-Ramsperger-Kassel-Marcus
SAPRC	Statewide Air Pollution Research Center
SCIAMACHY	SCanning Imaging Absorption spectroMeter for Atmospheric CartograpHY
SeaWIFS	Sea-Viewing Wide Field-of-View Sensor
SHADOZ	Southern Hemisphere ADditional OZonesondes
SOA	Secondary Organic Aerosol
STE	Stratospheric-Tropospheric Exchange
SV-OOA	Semi-volatile Oxygenated Organic Aerosol
TOMS	Total Ozone Mapping Spectrometer
UV	Ultraviolet
VOC	Volatile Organic Compound
WCRP	World Climate Research Program
WRF-Chem	Weather Research and Forecasting model coupled to Chemistry

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