Opinion: Establishing a Science-into-Policy Process for Tropospheric Ozone Assessment

Richard G. Derwent[1,*], David D. Parrish[2] and Ian C. Faloona[3]

[1] rdscientific, Newbury, Berkshire, UK

[2] David.D.Parrish, LLC, 4630 MacArthur Ln, Boulder, Colorado, USA

[3] Department of Land, Air, & Water Resources, University of California, Davis, California, USA

Correspondence to: Richard G. Derwent (r.derwent@btopenworld.com); David D. Parrish (david.d.parrish.llc@gmail.com)

Abstract. Elevated tropospheric ozone concentrations driven by anthropogenic precursor emissions is an environmental issue scientifically similar to the depletion of the stratospheric ozone layer and global climate change; however, the tropospheric ozone issue lacks the generally accepted, international assessment efforts that have greatly informed our understanding of the other two issues. Here we briefly review those successful science-into-policy approaches, and outline the elements required to conduct a similar process for tropospheric ozone, especially for establishing a simplified model of the underpinning science, useful policy metrics and motivating international policy forums for regulating ozone production over the hemispheric and global scales.

1 Introduction

The global environmental policy problems involving the depletion of the stratospheric ozone layer and global climate change have been identified, fully researched and moved into their respective policy arenas over the last fifty years or so. A number of features of the ozone depletion and climate change problems that have brought them to the forefront of environmental policy can be detailed and then compared with the corresponding features of the tropospheric ozone problem. The process of science-into-policy is much

less developed for tropospheric ozone, as it seems to have fallen between the two stools of ozone depletion
25   and climate change.

While not presenting an existential crisis of the same magnitude as depletion of the stratospheric ozone layer or global climate change, tropospheric ozone is widely recognized as an important air pollutant (Monks et al., 2015). Beyond fundamentally controlling the oxidation potential of Earth's inhabited atmosphere, tropospheric ozone damages human health (Fleming et al., 2018; Cohen et al., 2017), impacts
30   crops and vegetation (Mills et al., 2018; Feng, et al., 2022) and is a man-made greenhouse gas, third in importance behind carbon dioxide and methane (IPCC, 2014; Skeie, et al., 2020).

Urban and regional ozone has been subject to policy actions for several decades to reduce air pollution and ozone episodes and these measures have been largely successful in North America (Parrish et al., 2022) and Europe (Derwent et al., 2022) and are making progress in other continents. An important
35   counter example to note here is the growth of tropospheric ozone over East Asia, where in spite of large recent reductions in air pollutant emissions, regional ozone has generally risen (Wang et al., 2020). Regardless of the policy actions, exceedances of air quality standards and guidelines set to protect human health still occur and will do so for the foreseeable future. This is because ozone episodes sit on top of a baseline that is hemispheric and even global in scale. Furthermore, as the manifold, deleterious effects of
40   ozone continue to be revealed by ongoing research, the policy targets of exposure are very likely to be reduced even further or different exposure metrics developed. In either case the relative importance of the background is going to become the dominant effect on future compliance or non-compliance.

Here, we outline a process by which understanding of the science underpinning tropospheric ozone could lead to robust international policy action with a view to simultaneously reducing the global scale
45   climate impacts of tropospheric ozone, reaching healthy air quality, and ameliorating damage to crops and vegetation.

2 The science-into-policy processes of stratospheric ozone layer depletion

Concerns were first raised in the 1970s about the possible impacts of man-made chlorofluorocarbons (CFCs) on stratospheric ozone by Molina and Rowland (1974). These concerns were put into sharp focus
50   by the discovery of the stratospheric ozone hole in the 1980s in Antarctica by Farman et al. (1985). Policy

action followed swiftly, not simply because of the importance of these discoveries, but because a number of potential policy hurdles or stumbling blocks had already been surmounted.

A 'model' had been developed describing the mechanism by which the stratospheric ozone hole formed that was widely accepted by the atmospheric science community (Solomon et al., 1986; Crutzen and

55 Arnold, 1986; Cox and Hayman, 1988). It was proposed that man-made CFCs are photolyzed in the stratosphere to form active chlorine atoms and radicals which catalyze the ozone destruction. Armed with this 'model', the process of review and assessment began in earnest under the aegis of the World Meteorological Organization (WMO). A policy metric was developed, the ozone depletion potential (ODP), for the CFCs and ultimately for all ozone-depleting substances. It should be noted that ODPs

60 cannot be 'observed'. These policy metrics required the 'model' of stratospheric ozone to be well understood so that they could be faithfully derived.

Policy actions were formulated within the Vienna Convention for the Protection of the Ozone Layer under the auspices of the United Nations Environment Program (UNEP). The Vienna Convention used the WMO reviews and assessments to build a set of Protocols identifying each ozone depleting substance

65 in turn and moved towards their phase-out, in order of importance, as determined by the products of the ODPs and the abundances.

3 Science-into-policy processes of global climate change

It has been postulated over the last two centuries by Eunice Foote, Joseph Fourier and John Tyndall that carbon dioxide would act as a greenhouse gas (Royal Institution, 2019) and Svante Arrhenius (1896)

70 quantified the global temperature increase that would result from increased CO_2 levels. Charles Keeling identified the global scale rise in atmospheric carbon dioxide levels from his observations on Mauna Loa, Hawaii and at the South Pole in late 1950s (Keeling et al., 1989). Policy-makers were first made aware of the emerging issue of global climate change in the 1980s. Scientific review and assessment began around this time under the International Panel on Climate Change (IPCC), spear-headed by Bert Bolin

75 who placed the greatest emphasis on assessment with a clear focus on science-into-policy. The IPCC was formed under the aegis of the WMO and its first scientific assessment was published in 1990 with

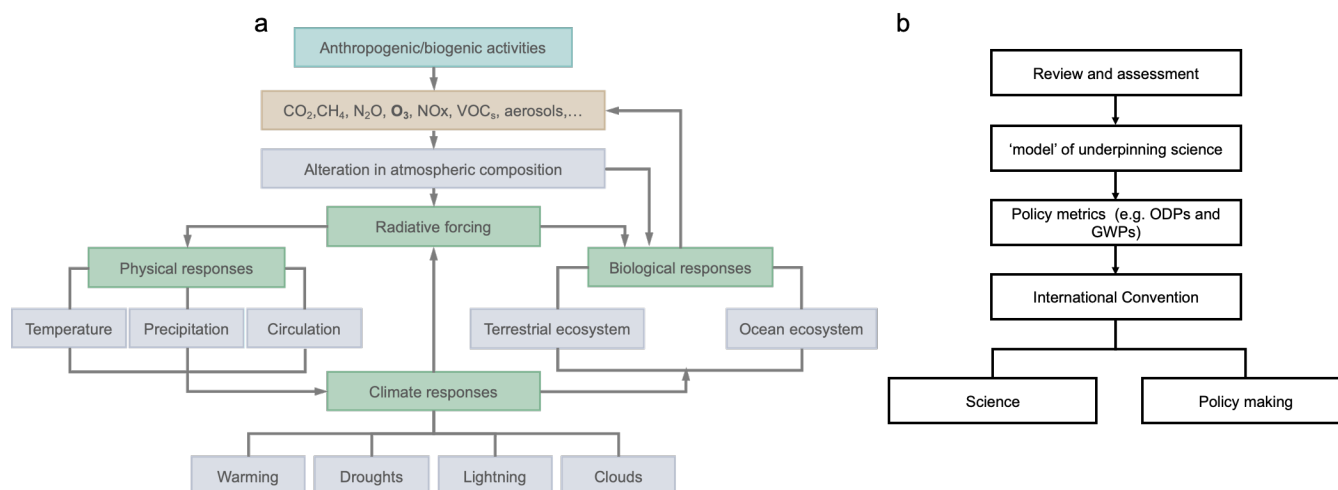
subsequent major scientific assessment reports on the physical science basis in 1995 (SAR), 2001 (TAR), 2007 (AR4), 2014 (AR5), and 2022 (AR6).

Daniel Albritton (Birks et al., 1992) conceptualized a 'model' describing the basic scientific framework for IPCC. It describes how atmospheric composition change drives radiative forcing, which in turns drives atmospheric responses in terms of changes in temperature, winds and rainfall (physical responses). These atmospheric responses drive changes in the climate (climate responses) which impact on the biosphere (biological responses) ultimately leading to ecosystem responses, as illustrated in Figure 1a. Feedbacks occur when atmospheric responses, such as melting ice, modify radiative forcing, or when ecosystem responses change atmospheric composition through, for example, changes in wetland methane emissions. In this 'model', global climate change is seen as a system of forcings and feedbacks, with man-made composition change as the ultimate driving force. This system has been represented with increasing sophistication through the development of increasingly complex earth-system models (ESMs).

The series of IPCC reports has been presented to the United Nations Framework Convention on Climate Change (UN FCCC). In response to requests from the UN FCCC, the IPCC developed a policy metric, the global warming potential (GWP), so that the different propensities of a range of different trace gases to influence climate change could be represented on a common basis in policy contexts. Using GWPs the UN FCCC put together a basket of six trace gases and began developing strategies with the aim of reducing dangerous anthropogenic climate change. Interestingly, the basket does not address tropospheric ozone, although it is the third most important man-made greenhouse gas after CO_2 and methane (Stevenson et al., 2013). Indeed, tropospheric ozone is one of the most important short-lived climate forcers (SLCF).

4 Essential elements of the science-into-policy process

From the two sections above, we can identify the essential elements of the science-into-policy process addressing ozone layer depletion and global climate change, as illustrated in Figure 1b. These are, firstly, review and assessment of the underpinning science with strict and open peer review and encouragement of research with a clear focus on the science-into-policy process. Secondly, development of an easily understood 'model' of the underpinning science, and thirdly, development of a policy metric with full

buy-in from the atmospheric science community. But fourthly, and most importantly, we can identify the
105   importance of having an international convention which brings together the policy-making and
atmospheric science communities and provides a framework for taking account of both scientific and
policy developments.

110   **Figure 1: Science-into-policy processes of Global climate change.** Panel (a) A 'model' of underpinned
science representing the key processes involved with climate change revised from Danial Albritton (Birks
et al., 1992). Forcings are represented by downwards arrows. Feedbacks are processes by which responses
lower down in the diagram drive changes further up. Panel (b) Essential elements of the science-into-
policy processes.

115   **5 Review and assessment of the tropospheric ozone**

There are open and accessible data repositories covering many country-wide or regional monitoring
networks, for example, those operated by the United States Environmental Protection Agency (US EPA),
the United Nations Economic Commission for Europe (UN ECE), the European Monitoring and
Evaluation Program (EMEP), etc. In addition, there are some data portals which provide access to network
120   data, including those operated by the Norwegian Institute for Air Research, the International Global
Atmospheric Chemistry (IGAC) Tropospheric Ozone Assessment Report (TOAR) and the WMO World

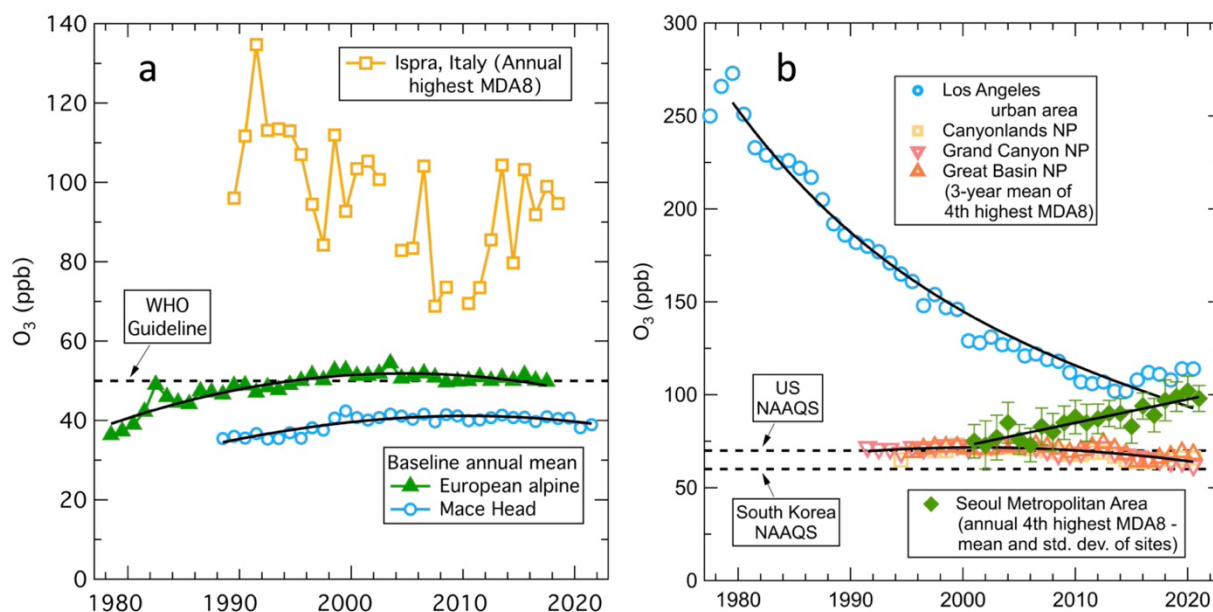
Data Centre for Greenhouse Gases. Some illustrative examples of long-term changes of both baseline and urban ozone concentrations are included in Figure 2.

125

Figure 2: Example long-term records of measured ozone. Baseline (Europe and US) and urban (Europe, US and Asia) ozone concentrations are included. Note that differing statistical metrics are used between the records.

There are reviews of urban and regional ozone compiled by the US EPA, EU and EMEP, but they are
130 generally restricted to single jurisdictions or single networks. Hemispheric and global scale reviews are compiled by the UN ECE Task Force on Hemispheric Air Pollution (HTAP) and TOAR. All these reviews, whilst providing excellent coverage, suffer distinctly from a dichotomous consideration of urban and regional scales on the one hand and the hemispheric and global scales on the other, and lack a strategic focus on science-into-policy for tropospheric ozone mitigation issues.

135 In response to the human health impacts of elevated ozone levels, policy-makers have taken extensive measures on local and regional scales to control the emissions of the main ozone precursors: oxides of nitrogen (NO_x) and reactive organic compounds (Sillman et al., 1999; Ehlers et al., 2016; Lu et al., 2010). Control of motor vehicle exhaust emissions through the mandatory fitting of exhaust gas catalysts and

140 evaporative cannisters has largely been a complete success, enabling large reductions to be achieved in ozone precursor emissions from road transport. Elevated ozone levels have declined in almost all legislative and administrative regions where ozone precursor emissions have been controlled. However, these declines have not gone far enough. Air quality targets, guidelines or standards set to protect human health have not always been achieved and, with the identification of adverse health impacts at lower ozone concentrations, there is growing pressure to further tighten standards; with some exceptions, non-

145 attainment of air quality targets remains an important policy issue. Whilst assessment of long-term changes in urban and regional ozone levels point to the huge impact of the measures taken to reduce ozone precursor emissions, they also demonstrate that reductions in exceedances have slowed during the past decade (Parrish et al., 2022; Derwent and Parrish, 2022). The situation is also reflected by the atmospheric chemistry literature which is characterized by dichotomous views separated by scales; with papers on

150 urban ozone (Ehlers et al., 2016; Nelson et al., 2021; Tan et al., 2019; Cardelino and Chameides, 1995; Brune et al., 2016; Pusede et al., 2015) often ignoring or oversimplifying the impact of the background troposphere on the urban environment and papers on the background troposphere (Parrish et al., 2009; Wang and Jacob, 1998; Gaudel et al., 2018) often ignoring or oversimplifying the impact of urban and regional pollution.

155 **6 A 'model' of the underpinning science for tropospheric ozone**

Development of a widely-accepted, simple, conceptual 'model' that intuitively explains the broad features of how ozone sources, sinks and transport processes all interact to establish the observed local, regional and large-scale spatial distributions, seasonal cycles and long-term temporal changes of ozone is urgently required. Such a 'model' would form the core of a robust assessment, would be invaluable to

160 researchers in their efforts to understand the beautifully detailed observational data and chemistry-transport model results that are presently available to the atmospheric community. Such a conceptual 'model' would provide a firm foundation upon which to conceive and organize present and future research efforts into a more comprehensive understanding of all aspects of the spatial and temporal distribution of tropospheric ozone. Such an intuitive model would be an essential component of a modeling hierarchy

165 (Held, 2005) by complementing the comprehensive numerical models that aim to simulate in full detail as much of the atmospheric chemistry and dynamics as possible.

   Figure 3 presents a schematic figure illustrating some of the basic principles of the tropospheric ozone issue updated from Derwent et al. (1998b). The diagram envisages background air containing O_3, CO and CH_4 entering an urban area or rural region on the right-hand side. Urban and regional biogenic precursor
170 emissions drive local and regional scale photochemical ozone production which elevates ozone concentrations above the global baseline levels, leading to human health effects and crop and vegetation damage. After one to several days travel downwind, the regionally-polluted air with elevated levels of O_3, CO, NO_x and unreacted organic compounds is lofted from the continental boundary layer and rejoins the global circulation. One aspect of such a 'model' requires particular attention; it should aim to bridge the
175 dichotomous views of urban pollution ignoring or oversimplifying the impact of the background troposphere on the urban environment and the background troposphere ignoring or oversimplifying the impact of urban pollution.

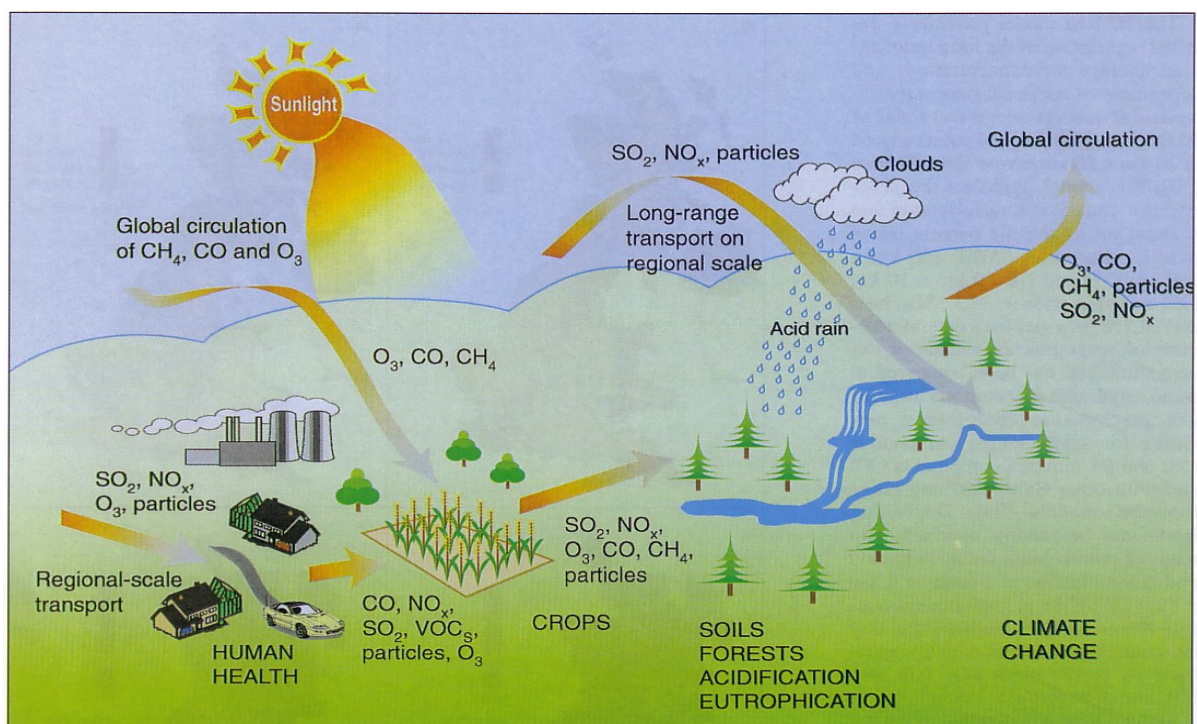



Figure 3. Schematic illustration of the basic principles underpinning the tropospheric ozone issue.
180 Note that tropospheric ozone sources include production in background, urban and rural areas as well as

injection of ozone from the stratosphere. The ozone produced in the three distinctly different source areas are mixed in the troposphere, which depicts the difficulties of tropospheric ozone control.

Since the manifold of processes determining the tropospheric ozone distribution is considerably simpler than that driving the climate system, this required 'model' will be much simpler than that developed by
185 IPCC. As examples of its utility, the 'model' should be able to provide a clear description of northern midlatitude baseline ozone, including answering 1) why ozone maximizes in late spring in the free troposphere but peaks earlier in the spring with a summer minimum in the marine boundary layer, 2) why mean concentrations approximately doubled during the first half of the 20th century but have since decreased and 3) to what extent is ozone homogeneously mixed within the prevailing zonal flow.
190 Moreover, the 'model' should explain how ozone compares between the Northern and Southern Hemispheres, both at present and in the pre-industrial atmosphere. Of great utility would be an interactive atlas, such as provided by the IPCC AR6 Synthesis Report, that quantifies background and anthropogenic contributions to ozone concentrations at any specified location on the globe, particularly if those quantities could be illustrated as a function of variable local or hemispheric precursor emissions.

195 A further important aspect of the 'model' of tropospheric ozone is a close association with the 'model' of climate change. For example, future changes in stratosphere-troposphere exchange due to an accelerated Brewer-Dobson circulation (Abalos et al., 2020) along with a changing Northern Hemisphere stratospheric ozone abundance (Woltmann et al., 2020) may raise background ozone concentrations over the next century. The observed increase in background NO_x (Qu et al., 2021) could be the result of rising
200 soil temperatures, increasing wildfire impacts and increasing lightning (Murray, 2018) production in a warming climate.

7 Policy-relevant metric

Developing a policy metric like ODPs and GWPs for tropospheric ozone is more complex but emission inventories provide a policy focus for tropospheric ozone (Figure 4). The previously developed
205 parameters such as OFPs (Ozone Formation Potentials) (Carter et al., 1995) and POCPs (Photochemical Ozone Creation Potentials) (Derwent et al., 1998a) are not considered to be quite comparable with ODPs

and GWPs as they are highly state-dependent and so their efficacy as accurate metrics depends on local conditions, which vary over time and space.

Regional air quality and global models all require emission inventories and chemical mechanisms,
210   together with observations to evaluate their performance. These models effectively convert ozone precursor emission inventory data into predictions of ozone concentrations over specific spatial and temporal scales in response to policy needs. By changing the ozone precursor emissions, whilst keeping all other input data the same, modelers can visualize the impact of particular ozone precursor emission control strategies on the tropospheric ozone distribution, for the benefit of policy-makers.

215   Almost all urban areas and regions have been inventoried in greater or lesser detail for each of the major ozone precursors. Examples include the CHIEF (Clearinghouse for Inventories and Emissions Factors by US EPA), EMEP (by UN ECE), MEIC (Multi-resolution Emission Inventory for China), SAFAR(System of Air Quality and Weather Forecasting and Research), EDGAR (Emission Database for Global Atmospheric Research, from IGAC) and CEDS (Community Emission Data System) (Hoesly et al., 2018).

220

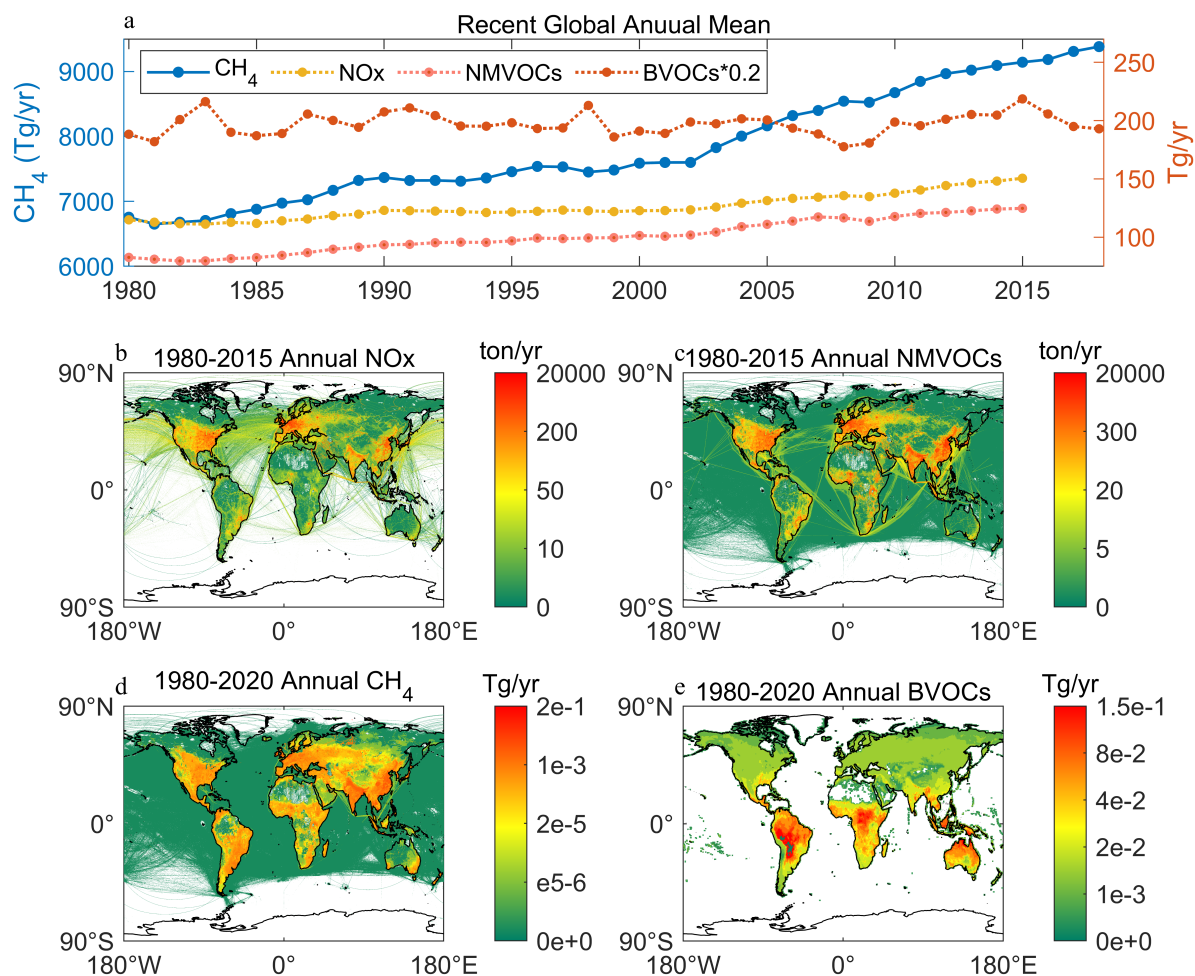



Figure 4. Emission rates of key ozone precursors. Panel a, time series of annual anthropogenic emission rate of CH$_4$ on the left axis along with the annual emission rates of anthropogenic NOx, NMVOCs (Non-methane volatile organic compounds) (EDGARv6.1) and BVOCs (Biogenic volatile organic compounds) (MEGAN-MACC) on the right axis during 1984-2020. Panels b and c, spatial distribution of annual

225   emissions of NOx and AVOCs (Anthropogenic volatile organic compounds) (tonne/yr) averaged over the period 1984–2015. Panels d and e same as b and c but for CH$_4$ and BVOCs (Tg/yr) over the period 1984–2020.

Whilst the emission factor approach is well-defined for man-made emissions, a different approach is
230   required for biogenic emissions. Accurate estimation of the biogenic emissions of isoprene, terpenes and

NO_x requires information on plant and tree species, on ecosystems and on the local meteorological conditions of temperature, radiation and soil moisture. Biomass burning is another difficult source to treat rigorously in ozone policy models. It is important to treat both agricultural biomass burning and wild-fires separately, for policy purposes.

235   In summary, we consider that ozone precursor emission inventories can provide an up-to-the-task basis for driving policy formulation for tropospheric ozone. High annual emission rates (> 100 tonne/yr/grid square) over densely populated regions (e.g. Asia, Europe and North America) predominate the total emission of global anthropogenic NO_x and NMVOCs (Figure 4b-c) which supports the conclusions that intensive anthropogenic air pollutant emissions in these regions make great contributions to the non-

240   attainment of ozone air quality standards. Moreover, CH_4 emissions increased similarly to those of NO_x and NMVOCs (Figure 4a) also indicate possible accelerated photochemical O_3 production in background air masses. Importantly, these inventories require continual updating, extension to additional species (e.g., volatile chemical products, McDonald et al., 2018), emission sectors and global regions; "top-down" evaluation is essential as emission sources evolve (McDonald et al., 2018; Smith et al., 2022).

245   **8 Implications for future policy and science**

Whilst there are many international bodies that address issues relevant to improving scientific knowledge of the sources and distribution of tropospheric ozone, there is no international convention that could readily take up this issue and make policy progress globally. HTAP considers regions covering only Europe, USA and Canada. It specifically excludes Asia, Mexico and North Africa, together with Southern

250   Hemisphere countries.

The UN FCCC has pioneered the compilation of ozone precursor emission inventories from each country globally but does not include tropospheric ozone in its basket of trace gases. The IPCC has identified tropospheric ozone as the third most important man-made greenhouse gas after carbon dioxide and methane. The latter trace gas, methane, has been established as an important tropospheric ozone

255   precursor (West et al., 2013). However, there has been little focus on tropospheric ozone within the UN FCCC.

If SLCFs could be moved up the UN FCCC agenda and policy actions focused on methane and tropospheric ozone, a rapid impact on climate forcing would result because of the relatively short global mean atmospheric lifetimes of both methane (12 years) and tropospheric ozone (~1 month). There would

260   also be a substantial improvement in urban and regional ozone air quality, bringing the possibility of achieving air quality standards and guidelines set to protect human health and crops and vegetation.

We have every confidence that if a robust and cogent, peer-reviewed policy-oriented scientific review of the tropospheric ozone issue could be assembled by the atmospheric science community, then policy progress could be made under the auspices of the UNEP. Such a review could be built on the current

265   TOAR and HTAP activities, with inclusion of participation by policy associated scientists. Coordinated global action on tropospheric ozone holds the promise of delivering acceptable air quality for ozone in all major population and industrial centers globally, a prospect that is unlikely to be achievable without such collective planning. Furthermore, future actions to reduce urban and regional ozone precursor emissions may be wasteful of resources if realistic account of the hemispheric or global baseline is not taken. In

270   such a situation the exceedance of air quality standards and guidelines will continue unchecked and the hands of local policy makers will be tied.

Acknowledgements

Discussions with Keding Lu, Alex Archibald, David Stevenson, Daniel Jacob, Tao Wang, Ken Carslaw, Paul Monks, Jim Crawford and Martin Schultz are kindly acknowledged. I.C.F.'s effort was supported by

275   the USDA National Institute of Food and Agriculture (Hatch project CA-D-LAW-2481-H).

Competing Interests

The contact authors have declared that none of the authors has any competing interests.

Author contributions

R.G.D. and D.D.P. were responsible for the overall design. R.G.D. and D.D.P. wrote the paper with

280   input from I.C.F.

Data availability

Annual highest MDA8 ozone data at Ispra, Italy and the annual mean ozone data at European alpine sites (described in detail by Parrish, et al., 2020) are available from the TOAR Surface Ozone Database via the JOIN web interface: https://join.fz-juelich.de. Baseline annual mean ozone data from Mace Head are
285   taken from Derwent et al. (2023). The 3-year mean of 4th highest MDA8 (i.e., the US ozone design value) from the four US data sets in Figure 2 are available from the US EPA AQS data archive (https://www.epa.gov/aqs). The surface monitor data for South Korea are available from https://www.airkorea.or.kr/web/; the annual 4th highest MDA8 data for the Seoul Metropolitan Area are taken from Kim et al. (2022). Globally-averaged annual mean and 0.1°×0.1° gridmaps of NOx and
290   NMVOCs data during 1984-2015 are obtained from EDGARv6.1 Global Air Pollutant Emissions database (https://edgar.jrc.ec.europa.eu/index.php/dataset_ap61). Global annual BVOC data during 1984-2020 is taken from MEGAN-MACC Biogenic emission inventory (Sindelarova et al., 2014), which available in ECCAD database (https://eccad.aeris-data.fr/).

References

295   Abalos, M. *et al.* Future trends in stratosphere-to-troposphere transport in CCMI models. Atmos Chem Phys **20**, 6883-6901, doi:10.5194/acp-20-6883-2020 (2020).
Arrhenius, S. On the influence of carbonic acid in the air upon the temperature of the ground. Philosophical Magazine **41**, 237-276 (1896).
Birks, J., Calvert, J. G. and Sievers, R. CHEMRAWN VII, The Chemistry of the Atmosphere: Its
300   Impact on Global Change, Perspectives and Recommendations. (IUPAC, 1992).
Brune, W. H. *et al.* Ozone production chemistry in the presence of urban plumes. Faraday Discuss **189**, 169-189, doi:10.1039/c5fd00204d (2016).
Cardelino, C. A. and Chameides, W. L. An Observation-Based Model for Analyzing Ozone Precursor Relationships in the Urban Atmosphere. Journal of the Air & Waste Management Association
305   **45**, 161-180 (1995).
Carter, W. P. L., Pierce, J. A., Luo, D. M. and Malkina, I. L. Environmental Chamber Study of Maximum Incremental Reactivities of Volatile Organic-Compounds. Atmospheric Environment **29**, 2499-2511 (1995).
Cohen, A. J. et al. Estimates and 25-year trends of the global burden of disease attributable to ambient
310   air pollution: an analysis of data from the Global Burden of Diseases Study 2015. Lancet **389**, 1907-1918, doi:10.1016/S0140-6736(17)30505-6 (2017).

Cox, R.A. and Hayman, G.D. The stability and photochemistry of dimers of the ClO radical and implications for Antarctic ozone depletion. Nature **332**, 796-800 (1988).

Crutzen, P. J. and Arnold, F. Nitric acid cloud formation in the cold Antarctic stratosphere: a major cause for the springtime 'ozone hole'. Nature **324**, 651-655, doi:10.1038/324651a0 (1986).

Derwent, R. G. and Parrish, D. D. Analysis and assessment of the observed long-term changes over three decades in ground-level ozone across north-west Europe from 1989 - 2018. Atmospheric Environment **286**, 119222, doi:https://doi.org/10.1016/j.atmosenv.2022.119222 (2022).

Derwent, R. G., Jenkin, M. E., Saunders, S. M. and Pilling, M. J. Photochemical ozone creation potentials for organic compounds in northwest Europe calculated with a master chemical mechanism. Atmospheric Environment 32, 2429-2441 (1998a).

Derwent, R. G., Metcalfe, S. E. and Whyatt, J. D. Environmental Benefits of NO_x Control in Northwestern Europe. Ambio **27**, 518-527 (1998b).

Derwent et al., Thirty-five years of observations of ozone at Mace Head, Ireland: Baseline, regional and COVID-19 influences, Atmospheric Environment, manuscript under review, 2023.

Ehlers, C. et al. Twenty years of ambient observations of nitrogen oxides and specified hydrocarbons in air masses dominated by traffic emissions in Germany. Faraday Discuss **189**, 407-437, doi:10.1039/c5fd00180c (2016).

Farman, J. C., Gardiner, B. G. and Shanklin, J. D. Large losses of total ozone in Antarctica reveal seasonal ClOx/NOx interaction. Nature **315**, 207-210 (1985).

Feng, Z. Z. et al. Ozone pollution threatens the production of major staple crops in East Asia. Nat Food **3**, 47-+, doi:10.1038/s43016-021-00422-6 (2022).

Fleming, Z. L. et al. Tropospheric Ozone Assessment Report: Present-day ozone distribution and trends relevant to human health. Elementa: Science of the Anthropocene **6**, doi:10.1525/elementa.273 (2018).

Gaudel, A. et al. Tropospheric Ozone Assessment Report: Present-day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation. Elementa-Sci Anthrop **6**, doi:10.1525/elementa.291 (2018).

Held, I. M. The gap between simulation and understanding in climate modeling. B Am Meteorol Soc **86**, 1609-+, doi:10.1175/Bams-86-11-1609 (2005).

Hoesly, R. M. et al. Historical (1750-2014) anthropogenic emissions of reactive gases and aerosols from the Community Emissions Data System (CEDS). Geosci Model Dev **11**, 369-408, doi:10.5194/gmd-11-369-2018 (2018).

Intergovernmental Panel on Climate, C. Climate Change 2013 – The Physical Science Basis: Working Group I Contribution to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. (Cambridge University Press, 2014).

Keeling, C.D. et al. A three-dimensional model of atmospheric CO_2 transport based on observed winds: 1. Analysis of observational data. Geophysical Monograph **55**, 305-363 (1989).

Kim, S.-W., et al., Changes in surface ozone in South Korea on diurnal to decadal time scale for the period of 2001-2021, Atmos. Chem. Phys. Disc., doi.org/10.5194/acp-2022-788, 2022.

Lu, K. et al. Oxidant (O_3 + NO_2) production processes and formation regimes in Beijing. Journal of Geophysical Research-Atmospheres **115**, doi:10.1029/2009jd012714 (2010).

McDonald, B. C. et al. Volatile chemical products emerging as largest petrochemical source of urban organic emissions. Science **359**, 760-764, doi:10.1126/science.aaq0524 (2018).

355   Mills, G. et al. Tropospheric Ozone Assessment Report: Present-day tropospheric ozone distribution and trends relevant to vegetation. Elementa: Science of the Anthropocene **6**, doi:10.1525/elementa.302 (2018).

Molina, M. J. and Rowland, F. S. Stratospheric sink for chlorofluoromethanes: chlorine atomc-atalysed destruction of ozone. Nature **249**, 810-812, doi:10.1038/249810a0 (1974).

360   Monks, P. S. et al. Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer. Atmos. Chem. Phys. **15**, 8889-8973, doi:10.5194/acp-15-8889-2015 (2015).

Murray, L. T. An uncertain future for lightning. Nat Clim Change **8**, 191-192, doi:10.1038/s41558-018-0094-0 (2018).

365   Nelson, B. S. et al. In situ ozone production is highly sensitive to volatile organic compounds in Delhi, India. Atmos Chem Phys **21**, 13609-13630 (2021).

Parrish, D. D., Millet, D. B. & Goldstein, A. H. Increasing ozone in marine boundary layer inflow at the west coasts of North America and Europe. Atmos Chem Phys **9**, 1303-1323, doi:DOI 10.5194/acp-9-1303-2009 (2009).

370   Parrish, D. D., Derwent, R. G., Steinbrecht, W., Stubi, R., Van Malderen, R., Steinbacher, M., Trickl, T., Ries, L., and Xu, X.: Zonal similarity of long-term changes and seasonal cycles of baseline ozone at northern mid-latitudes. Journal of Geophysical Research Atmospheres 125 (13). Doi:10.1029/2019JD031908, 2020.

Parrish, D. D., Faloona, I. C. and Derwent, R. G. Observational-based assessment of contributions to
375   maximum ozone concentrations in the western United States. Journal of the Air & Waste Management Association **72**, 434-454, doi:10.1080/10962247.2022.2050962 (2022).

Pusede, S. E., Steiner, A. L. and Cohen, R. C. Temperature and Recent Trends in the Chemistry of Continental Surface Ozone. Chemical Reviews **115**, 3898-3918, doi:10.1021/cr5006815 (2015).

Qu, Z. et al. US COVID-19 Shutdown Demonstrates Importance of Background NO2 in Inferring NOx
380   Emissions From Satellite NO2 Observations. Geophysical Research Letters **48**, doi:10.1029/2021GL092783 (2021).

Royal Institution. Who discovered the greenhouse effect? https://www.rigb.org/explore-science/explore/blog/who-discovered-greenhouse-effect (2019).

Sillman, S. The relation between ozone, NOx and hydrocarbons in urban and polluted rural
385   environments. Atmospheric Environment **33**, 1821-1845 (1999).

Sindelarova, K. et al. Global data set of biogenic VOC emissions calculated by the MEGAN model over the last 30 years. Atmos Chem Phys **14**, 9317-9341, doi:10.5194/acp-14-9317-2014 (2014).

Skeie, R. B. et al. Historical total ozone radiative forcing derived from CMIP6 simulations. Npj Clim Atmos Sci **3**, doi:10.1038/s41612-020-00131-0 (2020).

390   Smith, S. J., McDuffie, E. E. and Charles, M. Opinion: Coordinated development of emission inventories for climate forcers and air pollutants. Atmos Chem Phys **22**, 13201-13218, doi:10.5194/acp-22-13201-2022 (2022).

Solomon, S., Garcia, R.R., Rowland, F.S and Wuebbles, D.J. On the depletion of Antarctic ozone. Nature **321**, 755-758 (1986).

395   Stevenson, D. S. et al. Tropospheric ozone changes, radiative forcing and attribution to emissions in the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP). Atmos Chem Phys **13**, 3063-3085, doi:10.5194/acp-13-3063-2013 (2013).

        Tan, Z. F. et al. Daytime atmospheric oxidation capacity in four Chinese megacities during the photochemically polluted season: a case study based on box model simulation. Atmos Chem

400   Phys **19**, 3493-3513 (2019).

        Wang, Y. H. and Jacob, D. J. Anthropogenic forcing on tropospheric ozone and OH since preindustrial times. Journal of Geophysical Research-Atmospheres **103**, 31123-31135 (1998).

        Wang, Y. et al. Contrasting trends of PM2.5 and surface-ozone concentrations in China from 2013 to 2017. National Science Review, doi:10.1093/nsr/nwaa032 (2020).

405   West, J. J. et al. Co-benefits of mitigating global greenhouse gas emissions for future air quality and human health. Nat Clim Change **3**, 885-889, doi:10.1038/Nclimate2009 (2013).

        Wohltmann, I. et al. Near-Complete Local Reduction of Arctic Stratospheric Ozone by Severe Chemical Loss in Spring 2020. Geophysical Research Letters **47**, doi:10.1029/2020GL089547 (2020).

410