

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

Anonymous Referee #3

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

General comments

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

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

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

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

Sections 3 and 5 might be best focused solely on Europe, but if the U.S.A. is discussed, it should be noted that the criteria for the ozone standard are reviewed periodically, and the most recent assessment was just completed, e.g. <http://www.epa.gov/ncea/isa/>

Section 4 should open with a rationale for why the selected topics are the most pressing

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ones to discuss here. A revised title, rather than "Topics", should also strive to convey their importance. Section 4.2 overlaps earlier discussion.

Specific comments:

p. 32712 The Intro here fails to communicate what the current paradigm is for what controls tropospheric ozone.

P32715. The phrasing of something well understood but remains a challenge seems a bit contradictory.

P32718 L24-25. Is this specific to deposition? Might be worth illustrating how the diurnal variability varies with geographic location, at the surface vs. at altitude to emphasize the importance of multiple spatial scales mentioned in the abstract.

P32721 L3. What does "positive and negative effects" mean here? Is deposition ever reversing and becoming a source?

P32724. Can any conclusions be drawn here about the sign of the change of climate change on long-range transport?

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

P32732. What other factors could be at play besides those put forth in the Parrish et al. "untested hypothesis"? Might the question be better posed as quantifying the relative importance of these factors at the individual locations of the measurements? A related point is on P32735, another possibility is that the measurements may reflect natural climate variability internal to the climate system that a climate model would not represent.

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

Simon, H., A. Reff, B. Wells, J. Xing and N. Frank (2014). "Ozone Trends Across the United States over a Period of Decreasing NO_x and VOC Emissions." *Environmental*

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Science & Technology 49(1): 186-195.

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

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

P32471 L20. How does Figure 14 show growth of emissions? The caption suggests that we are only shown 2005 here?

P32749. Some explanation for why the global satellite data do not provide a complete picture would be useful. It might be worth noting that there are direct tropospheric ozone retrievals, e.g.: Liu, X., et al. (2006), First directly retrieved global distribution of tropospheric column ozone from GOME: Comparison with the GEOS-CHEM model, *J. Geophys. Res.*, 111, D02308, doi:10.1029/2005JD006564.

P32762. The scientifically dubious statement seems overly strong, since it is only fairly recently that computer models have included stratospheric and tropospheric ozone chemistry to enable a combined estimate of RF from ozone.

P32767 "surprisingly small". If one considers the lower ozone production efficiency in urban plumes, is this so surprising?

P32773 "factor of five difference". Are these models all using state-of-the-art isoprene oxidation schemes? This reference may also be relevant: Ito, A., S. Sillman, and J. E. Penner (2009), Global chemical transport model study of ozone response to changes in chemical kinetics and biogenic volatile organic compounds emissions due to increasing temperatures: Sensitivities to isoprene nitrate chemistry and grid resolution, *J. Geophys. Res.*, 114, D09301, doi:10.1029/2008JD011254. How certain is the temperature-driven increase in biogenic emissions in light of CO₂-driven suppression?

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P32803. How important are plumes from one continent to the next versus diffuse background such as resulting from global methane and NO_x?

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

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

P32809. Why is IPCC 2007 cited rather than 2013?

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

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

Figure 2. Why not use the more recent ACCMIP models?

Fig 3. What is assumed here for BVOC? How sensitive is this picture to assumptions in the UKCA mechanism? Why is this referred to as a 'schematic' in the text (p 32717)? Please explain the significance of A/B/C labels in the caption.

Fig 7. Can the same colors or symbols be used for the same inventories across the
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different panels?

Fig 11. What are LDGVs? What are "real driving conditions"? How important are these differences to the ozone distributions?

Fig 13. Is this the best estimate, or the only available one? How does this differ in other world regions?

Fig 18. Is there a diurnal cycle in the fire emissions, and if so, how does that combine with the injection height variation in terms of the impact on tropospheric ozone?

Fig 41. Is this deemed by the authors to be the best approach to attributing ozone? There are numerous studies attempting to do so over the USA and elsewhere.

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

Does Table 3 repeat information in Figure 40?

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