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Atmospheric Chemistry and Physics Discussions

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

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

General comments 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 pollu-

tion 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 NOx 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?

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

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)?

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

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.

New figures have been added to balance the chemistry section.

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

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.

ACPD
14, C13209–C13218, 2015

This would be a review in itself. There is an undoubted need for what you are asking. Some of this is covered in the framework on GEIA (http://www.geiacenter.org/), but there is a need for more critical evaluation.

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/

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

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

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

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

Specific comments:

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

At the suggestion of referee#1 the introduction was restructured and this material moved to section 2.1. This has now been integrated to give a better overview of the required paradigm.

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

Agreed, added in practice to give the correct emphasis to the challenge.

ACPD

14, C13209–C13218, 2015

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.

The climatological view of ozone is dealt with in section 2.4. There is clear merit in this idea, but it probably is beyond the scope of what can be achieved in this review.

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

Sentence modified to “For example, surface water has been found to both enhance and reduce deposition rates.”

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

HTAP (2010) says “Changes in climate will affect meteorological transport processes as well as the chemical environment and lifetime of the transported pollutants and hence the concentrations of pollutants arriving at downwind continents.” Of the papers mentioned Glotfelty et al (2014) suggest a larger impact on the US from East Asian Emissions and Doherty et al (2013) showed stronger chemistry than transport positive climate feedbacks on ozone. This has been added to text.

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

The following sentence can be added to the end of line 13 on page 32727 "The influence on ozone of low frequency climate variability on time scales longer than 30 years has not yet been assessed due to continuous ozone data sets being limited to durations shorter than 30-40 years."

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?

ACPD
14, C13209–C13218, 2015

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

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.

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

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

....

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

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

Text has been changed (as per referee#2) to note magnitudes rather than "growth"

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.

ACPD
14, C13209–C13218, 2015

The section is dealing with ozone at a climatological scale. Details of the Liu et al reference has been added but a longer discourse on satellite ozone seems to detailed within this context. References are present to guide reader on this topic.

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.

This has been changed to “fraught”.

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

Removed surprising, it is often a truism as to what one finds “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?

This is a good question, a recent paper by Squire et al, (ACP, 2015) has highlighted this as an issue. A comment has been added to this section. The Ito work was already discussed in this section.

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

The best answer to this questions is given in HTAP (2010) - Part D ’Answers to Policy-Relevant Science Questions’ (http://www.htap.org/publications/2010_report/2010_Final_Report/HTAP%202010%20Part%20D%20110407.pdf)

ACPD
14, C13209–C13218, 2015

"To quantify the relative importance of emission changes outside each of these regions, as compared to emission changes inside each of these regions, we defined the Relative Annual Intercontinental Response (RAIR) metric. RAIR is defined as the sum of the changes in the annual, regionally-averaged concentration within a region due to a 20% decrease in emissions in the three other regions divided by the sum of the changes in concentration within a region due to a 20% decrease in emissions in all four regions. The value of the metric ranges from 0%, indicating no intercontinental influence, up to 100%, indicating that air quality in a region is completely dominated by intercontinental sources."

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

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

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.

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

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.

ACPD
14, C13209–C13218, 2015

Reference and discussion added on future directions.

P32809. Why is IPCC 2007 cited rather than 2013?

Reference updated and changed to chapter lead.

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.

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

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?

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

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

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

Figure 38 (old 37) – shown as dramatic example of methane emissions. Text details other work on NMVOC emissions from fracking.

Figure 39 (old 38) – is to help the reader with the chemistry.

ACPD

14, C13209–C13218, 2015

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

Figure 2 is a good representation to match the text. There is good referencing to the 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.

It is not schematic – text changed.

A/B/C –added caption

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

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

Figure redrawn and colours and symbols matched.

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

Caption amended to explain these are light-duty good vehicles under real world driving conditions rather than test-cycle. In relationship to the importance, in the UK it has been shown that the failure of these standards on the older vehicles has impacted measured NO2.

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

Figure 13 is an example (as it says in text). Caption has been expanded to include key point re. VOC speciation.

Fig 18. Is there a diurnal cycle in the fire emissions, and if so, how does that combine

ACPD

14, C13209–C13218, 2015

with the injection height variation in terms of the impact on tropospheric ozone?

There is indeed a study of the diurnal variation of the fires injection height in the Sofiev et al. paper. They indicate for example: "Diurnal variation of the injection height is huge: one can practically consider two independent datasets – one for daytime and one for nighttime, with transition during morning and evening." This has been added to the text.

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.

Figure 42 (old 41) is illustrative of an ozone measurement system. There is no implicit endorsement as to the approach.

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

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

Does Table 3 repeat information in Figure 40?

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

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

ACPD

14, C13209–C13218, 2015