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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Modelling the impact of megacities on local, regional and global tropospheric ozone and the deposition of nitrogen species" *by* Z. S. Stock et al.

Anonymous Referee #1

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This study quantifies the impact of megacities on surface ozone and nitrogen deposition with a global chemistry climate model through use of both emission perturbation and chemical tagging approaches. It concludes that emissions from megacities have a relatively small impact on a global scale, as previous studies have found, but that the effects can be larger on a local scale.

The paper clearly describes useful results, but it provides relatively little new insight beyond that of previous papers from the same project, e.g., Butler et al., 2012. The additional new angle that this paper promises in addressing nitrogen deposition is not sufficiently well exploited here, as the focus is principally on the magnitude of global

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deposition. Given that the lifetime of oxidized nitrogen is relatively short, and that whatever is emitted will be deposited over an annual global domain, it is unsurprising that changes in deposition closely mirror changes in emission. What would be more interesting is to explore how the changes in chemistry associated with megacity emission changes alter the speciation of deposited nitrogen, the mechanism (dry vs. wet removal), or the location of deposition (through the effects on chemical timescales; this is partly addressed in section 4.2). The authors should be in a good position to answer these questions based on their present studies, and by doing so would have stronger and more scientifically valuable results to present. I'd like to see the authors tackle these issues (to the extent that they are able to with their current results) and include these when they prepare their revised manuscript.

It is important to emphasize in the discussion of tagging vs. perturbation (particularly in section 3.5) that the techniques provide differing but complementary information. Tagging provides a quantification of the contribution of megacity emissions at the current time, while perturbation approaches provide an indication of how oxidants change when emissions are changed (and so are more useful for policy advice). The presence of megacity emissions suppresses ozone from other sources, so it is natural for tagging to attribute a larger contribution from megacities than perturbation approaches. The applicability of the different techniques depends on how the information will be used, so it is important to make this clear in the discussion.

Specific Comments

p.17677, l.27: please note that global models in their current form do not capture a wide range of scales, they represent only the large scales. Small scales are not captured, and thus a global model is only one of a set of tools needed to explore the full range.

p.17678, l.18-20: some clarification is required here. Emission perturbation approaches are appropriate for quantifying the sensitivity of atmospheric responses to emission changes, but not for source attribution. Grewe et al. (2010) quantified the

ACPD
13, C6275–C6279, 2013

errors in applying a perturbation approach to source attribution, but they remain very useful and accurate for many other purposes. Please add "for source attribution" after "perturbation method" here.

p.17682, l.24-27: the caveat about model biases is merited here, but assumes that the systematic biases are independent of the surface emission perturbations applied here. Given that these occur in the northern hemisphere, it is quite likely that they are partly related to implementation of surface emissions, and may thus be dependent on them. The perturbation studies will therefore not all contain "the same underlying biases". The sentence should be rephrased to acknowledge this.

p.17683, section 2.2: please include the total magnitude of megacity emissions here (or in Table 1).

p.17684, l.1-2: "All emissions scenarios are constructed at the 0.5x0.5 resolution" - it would be clearer to be more specific here, and state that the emissions perturbations were applied at 0.5x0.5 resolution, before aggregating to the model resolution. I assume from this that emissions data sets are prepared off-line for each scenario separately, although this is not explicitly stated.

p.17684, section 2.4: it is important to be clear about the assumptions made in this kind of simple tagging approach, and in particular that it is dependent on NOx control of O3 chemistry. This is only approximate, and while accounting for 98.4% of production is comforting, megacity chemical environments are typically VOC-sensitive, and it is therefore possible that they are less well represented by this method than the global troposphere. The final sentence of the section needs to be altered to acknowledge this.

p.17684, l.28: this is presumably 98.4% of total ozone production, not of the total ozone burden?

p.17685, l.13: is this ratio the concentration ratio or the emission ratio?

p.17686, l.7: is the NOx emission change identical to Butler et al. 2012?

ACPD
13, C6275–C6279, 2013

p.17689, section 3.3: Fig 5 is very interesting, but is difficult to interpret. It is notable that four tropical cities buck the prevailing trends, showing small moves away from NOx limitation in the redistribution scenarios and larger responses in the same direction for the 25% increase scenario. Why is this, and what do these cities have in common?

p.17691-2, section 3.5: See general point about tagging vs. perturbation made above, and the different information provided. In Fig 8, it is clear that the presence of megacities leads to a reduction in European ozone in winter of 0.5 ppb; the tagged approach indicates that megacity emissions contribute 1.2 ppb of ozone, which suggests that they also remove 1.7 ppb of ozone from other sources that would otherwise have been there if the megacities weren't. It would be useful to note this in the discussion here. The change in non-megacity ozone due to megacity emissions has largely been neglected in this paper

p.17693, section 4.1: I am surprised not to see any figure or table of results on ozone exceedances here, given that this is a central focus of the paper.

p.17694, l.4-5: the weakness here is not in the uncertainty in the emissions but in the ability to resolve the spatial scales associated with relatively fast photochemistry. The main problem is therefore systematic biases caused by numerical mixing that lead to incorrect timescales for production.

p.17696, l.27-28: note again here that perturbation approaches are often more useful for quantifying the impact of emission control policies; nevertheless, a combined tagged/perturbation approach would indeed be beneficial.

p.17711: the labels in Fig 5 are small; please consider increasing the font size (or the overall size of the figure).

Typos and minor corrections

p.17676, l.21: should "ozone burden at the surface" be "surface ozone", (i.e., concentration, not burden)?

ACPD
13, C6275–C6279, 2013

p.17676, l.22: "to increase.... by 3%" would be clearer as "to contribute 3% of...."

p.17677, l.18-21: sentence unfinished here, or perhaps misplaced ")".

p.17689, l.23: "to the formation ratio as" -> "in the formation ratio to"

p.17696, l.21-22: formatting issue.

Interactive comment on Atmos. Chem. Phys. Discuss., 13, 17675, 2013.