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.

We would like to thank both reviewers for their comments. We provide a response to comments and details of any changes to the manuscript below.

Anonymous Referee # 1

General Comments

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 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.

- The work on nitrogen deposition here is only one aspect of this study, with the results presented on chemical regime changes and the comparison to tagging techniques also providing new contributions beyond the original perturbation studies of Butler et al., 2012. We appreciate the reviewer's interest in the nitrogen deposition changes and we have therefore investigated this further, based on data currently available from our model integrations. We find small changes in the speciation of deposited nitrogen. For example, megacity emissions were found to increase the annual average dry deposition of N_2O_5 by ~6% and HNO_3 by ~4% compared to the ~3% average change across all species. When comparing the change in mechanism (dry vs wet removal), we find that globally there is little change in the ratio of dry to wet deposition with the removal of megacity emissions (<0.2% increase). Even on regional scales, for example over Europe, the change in ratio of dry to wet deposition is <0.5%. We have therefore added the following sentences to section 4.2:
  - p.17694, l.23: Further analysis revealed that megacity emissions contribute a higher percentage to the dry deposition of certain species, e.g. the annual average dry deposition of N_2O_5 is increased by ~6% and the dry deposition of HNO_3 by ~4% when megacities are present.
  - P.17695, l. 4: When comparing differences in dry versus wet deposition, we found that globally there is little change in the ratio of dry to wet deposition with the removal of megacity emissions (less than 0.2% increase). Even on regional scales, for example over Europe, the change in ratio of dry to wet deposition is less than 0.5%.

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.

- The tagging and perturbation techniques are both clearly useful and we agree with the reviewer that it is important to stress their differing but complementary contributions. We therefore alter the first paragraph of section 3.5 to further emphasise the different uses of the methods and the benefits of each technique:
  - p.17691, l.20: In this section we discuss the merits of the two different approaches used to investigate the effect of megacity emissions and compare their results. Both approaches make differing but complementary contributions to understanding the effect of megacities. The emission perturbation method is a useful tool to investigate changes in megacity emissions. These types of experiments can provide guidance to policy relevant questions such as what would be the impact on oxidants and other pollutants when altering megacity emissions. In contrast, tagging techniques are best suited to separate the contribution of megacity emissions on pollutants and oxidants, relative to that of other emission sources.

    In section 3.1, the impact of megacities was quantified using an annihilation method, showing that removal of megacity emissions results in a 0.27% reduction of total tropospheric ozone.

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.

- We agree with the reviewer and global models are certainly only one of a set of tools available. Here, the intention was to simply highlight the use of a global model in this study and we rephrase our final sentence on p17677 L27 to clarify this.

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 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.

- We accept the reviewer's suggestion and alter p.17678, l.18-20 accordingly.

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.

- We rephrase this sentence to clarify that the biases are not identical, but are similar between the runs.

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

- We make the assumption here that the reviewer is referring to anthropogenic megacity emissions. The total magnitude of megacity anthropogenic NO_x emissions is ~6Tg/yr. This is ~6% of the global total anthropogenic NO_x emissions. For NMVOC, the total magnitude of megacity anthropogenic emissions is ~1.5$Tgyr^{-1}$ (~5% of the global total anthropogenic NMVOC emissions). We add this information to section 2.3.

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.

- The reviewer is correct that all emission data sets are prepared offline for each scenario separately. We reword p17684 L1-2 to make it clear that the perturbations are applied before interpolating the emissions to the model grid.

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 NO_x control of O_3 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.

- We appreciate that our original sentence could be misleading and agree there is a need to stress that the megacity environment may be represented less well due to being generally VOC-sensitive. We are careful to highlight the limitations in using a simple NO_x tagging approach in sections 2.4 and 3.5. We also remove the final sentence in section 2.4.

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

- The 98.4% is calculated from the summation of all the tagged ozone from separate NO_x sources, therefore it refers to the percentage of the total ozone burden. We alter p17684 L28 to make this clearer.

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

- We refer to the concentration ratio, although we diagnose the chemical environment based on the ratio of hydrogen peroxide to nitric acid formation as described in section 2.5, p17685 L18-22.

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

- Butler et al. 2012 note a 6% change in anthropogenic NO_x emissions and 4% change in anthropogenic plus natural emissions, which is the same as in this study. The same anthropogenic emissions dataset and megacity mask are applied in both studies, however there are small differences in soil and lightning emissions. Note that once the emission perturbations are applied in each study the emissions are aggregated to different model grid resolutions.

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 NO_x 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?

- In figure 5 we decided to plot individual grid cells to display a clearer signal in the chemical regime changes. Therefore for megacities that span more than one grid cell, each cell is represented separately. In this case Hong Kong is made up of 4 grid cells which are the unusual cases that the reviewer refers to here. In Section 3.3 we explain how Hong Kong is not included in Figure 5d analysis, as in the redistribution scenario the city and country boundaries are almost identical. To avoid detracting from figure 5 we decided not to include Hong Kong in this analysis and remove the relevant grid cells. We leave the explanation for the removal of Hong Kong in the text in section 3.3.

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.

- Although the reviewer makes an interesting observation here, in the comparison of tagging and perturbation approaches we have been deliberately cautious in the discussion of figure 8. Due to using a NO_x-tagging scheme for megacities, as noted by the reviewer earlier, there are much larger uncertainties at smaller scales, due to the typically VOC-limited environments being less well represented by the tagging method. Therefore we do not attempt to make assumptions about the removal of ozone from other sources here.

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.

- In section 4.1 we discuss ozone exceedances as an addition to the ozone changes discussed earlier in the paper. This section is intended as an extension of the results to build on the application of the findings here rather than a focus. We decided not to add a table of results or figures in this section due to the large caveats with applying global models to quantify ozone exceedances in this way. We are careful to stress the limitations in section 4.1 and believe this caveat is strong enough to warrant only taking generalisations from latitudinal megacity groups rather than displaying tables of individual megacity results.

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.

- We believe we meant the same thing here and thank the reviewer for bringing it to our attention. By talking of the 'uncertainty associated with' we were referring to the spatial averaging rather than the uncertainty in emissions themselves. Therefore we rephrase p.17694, l.4-5 to make this clear.

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.

- We alter the final paragraph on p17696 to emphasise this point.

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

- We are aware of the detail in this figure and have increased the size of the font. We will check the final figure size is large enough in the revised manuscript for readers to view clearly.

Typos and minor corrections

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

- We thank the reviewer for bringing this to our attention, this should simply be "ozone burden".

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

- We agree with the reviewer and make this change.

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

- The ")" is misplaced, we correct this.

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

- We agree with the reviewer and make this change.

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

- We will check this issue doesn't occur in the revised manuscript.