

Interactive comment on “Aerosol pollution potential from major population centers” by D. Kunkel et al.

Anonymous Referee #2

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Overview

The paper documents the efficiency of export of particulate matter from large cities around the world. The issue relates to the environmental footprint of human activities in densely populated areas onto the larger scale. It expands upon a previous study by some of the co-authors of the present paper on the efficiency of export of trace gaseous species from large cities. By comparing the finding using slightly different model setup the author support the robustness of the approach and therefore address some of the limitations presumably pointed out in the 2007 paper. Outstanding features related to the different behaviour of aerosols and gaseous trace species are also introduced in the paper. The paper is well structured, the methodology is clearly explicated, the analysis of results is thorough and the paper reveals some very relevant implications

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for environment exposure assessment. As a consequence I would strongly support the publication of this study in ACP once the following minor points have been addressed.

Major comments

In the introduction, the authors refer to some studies proposing a quantification of trace species outflow from very distinct perspective. Most relevant with the present paper would be a comparison with a Lagrangian approach. The main limitation of the paper from my perspective is that it relies on a global model (even if the comparison with L07 addresses somewhat this limitation). In the perspectives the author mention the possible comparison with regional-scale tools but I wonder if there is not already published work on environmental footprints using Lagrangian approaches that would be relevant to compare with.

The choice of the authors to ignore some important additional sources of variability and favour a synthetic setup of the experiment is legitimate as it allows highlighting some key findings. However a qualitative discussion is missing on how some of the processes ignored would bear upon the results. Of course conducting such an analysis with reactive aerosols might have proven non-practical and/or difficult to interpret. But I am sure the authors are aware that the chemical reactivity of the aerosols would have a strong impact on their results. The paper would benefit from a short discussion on the expected impact of the different chemical composition in each MPC, as well as regarding the role of secondary formation of aerosol from exported gaseous species.

The uncertainty associated to the model used in the study is slightly overlooked. To my perspective the material provided in the supplement would deserve to fit in the paper in Section 3, especially if addressing model uncertainty is presented as one of the first objective of the study in the introduction. In the same section (3) the “main results” mentioned P25400 L7 should be explicated. In addition I find that MATCH-MPIC and EMAC are presented as totally different model whereas it is not exactly the case. In the supplement it is mentioned that both rely on the same convective tracer

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transport scheme which is crucial for the present study. Therefore it cannot be argued that model uncertainty has been explored in details. On the same note convective transport should be removed from the sentence in Supp P4 L26 if it turns out to be identical in both models.

I'm surprised that low-level transport from "cold" regions (Section 4.2.1) is not discussed further as it has important consequences for instance on deposition of soot on snow and subsequent modification of the albedo of polar regions. Such low level transport attributed here to the 'stability' of the lower atmosphere has also been documented before under the framework of isentropic transport (for instance by means of Lagrangian modelling). The authors might want to add a specific subsection addressing this topic (or add another pseudo-category – snow – in the landuses explored in section 6) given the potential implication of their work for this area of research.

The conclusion is too synthetic from my perspective. In particular it lacks a reference to the key finding regarding the respective role of location and tracer properties (P25409 L 9), and discussion of the assumptions made with regards to ignoring aerosol composition (would the conclusions hold if a given location would be dominated by particles that would be particularly hydrophilic/volatile/etc.. ?) as well as formation of secondary particulate matter.

A discussion on the overall role of megacities in the global burden of pollution would also be interesting. In particular the present study could somehow contribute to the intense debate on the most appropriate scale for air quality mitigation measures (local, country, continent, ...). Whereas with regards to exposure there is always a benefit to reduce air pollution at the local scale, from a quick browsing of this paper and Lawrence et al. 2007, one shall conclude particulate matter has little impact at larger scales. Therefore the authors might want to comment on the global radiative impact of PM originating from densely populated areas.

Minor comments:

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P25391 L21: In densely populated areas, there is potentially as much scope for air pollution mitigation efficiency than for greenhouse gases. Therefore one can question whether the density of emission will really matter in the future as suggested here. However the exposure will remain high for sure. This feature could be explained in more details here as it relates also to one of the key finding of the paper mentioned in the conclusion (P25415 L19).

P25392 L23: What is referred to as high-resolution shall be considered very coarse from a non-global modelling perspective. The exact spatial resolution should given more clearly. P25395L12: Would it be possible to state whether ignoring interactions between trace species yields an upper or lower bound for the quantification of the outflows. P25395 L16-18: this sentence is confusing, please reword.

P25395L22-P25396L7: There are a few features of the model setup that are overlooked. Very little details are given on the turbulent mixing parameterization that is expected to have a very significant impact. Scavenging is presented as impacting only particulate matter whereas some gases are indeed scavenged quite efficiently. More importantly, secondary aerosols are virtually ignored from the discussion as well as re-volatilisation of aerosol that can be transported as gaseous (or semi-volatile) species and condense again in remote areas. Maybe the authors considered that such processes would be bounded by the "active" and "inactive" type of tracers, but it is worth a short discussion in the model setup.

P25396 L25: I find the discussion of Figure 1 much too short as it raises many questions. This paragraph actually addresses aggregated findings, and one shall wonder if the split by city should not be discussed at a later stage, e.g. within a synthesis once the climate types are introduced.

P25397 L3: A reference is needed to support the statement that wet deposition constitutes the main sink for PM in nature.