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Atmospheric Chemistry and Physics Discussions

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

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Major comments

1) Comparison with Lagrangian approaches

To our knowledge there are no specific Lagrangian model approaches in the literature to study the outflow of particulate matter from such a number of MPCs on global scale. Stohl et al. (2002) investigated the outflow of tracers from different continents, which is referenced in the introduction (P25393, L8). However, only six emission sources were used and the tracers represented gaseous CO. Thus, this study is more comparable to Lawrence et al. (2007).

More footprint studies with Lagrangian techniques for anthropogenic emission source points have been conducted for example for the release of radioactive species after nuclear reactor accidents (e.g., see www.flexrisk.boku.ac.at, Stohl et al., 2012).

2) On the role of the aerosol reactivity and consequent variability

The passive tracers should be regarded as proxies for different types of aerosols. We further refer to an earlier publication where the passive aerosol tracers were interpreted as follows:

"The small aerosol tracers can be interpreted as primary solid aerosol particles like black carbon or soluble secondary organic or inorganic particles. The aerosol tracers with a diameter of 2.5 μm represent the largest aerosols that are still classified as fine particulate matter used for air quality regulations (compare PM2.5 = aerosol mass of particles with an aerodynamic diameter smaller 2.5 μm). These aerosols are distinguished from larger particles, here the 10.0 μm tracers, which represent fractions of dust, sea-salt, and sometimes minor amounts of organic matter present in urban centers. " (Kunkel et al. 2012).

This has also been quoted on L24P25396 L24. Since we consider the nucleation scavenging state as extrema with respect to aerosol solubility, we expect all further -more detailed- representation of the aerosol reactivity to be in-between our results. Moreover, a discussion on the expected impact of the different chemical aerosol composition in each MPC would go far beyond the scope of this paper where we explicitly wanted to answer the question of the potential impacts of particulate matter in general. The impact of different aerosol species will be the topic of a follow-up paper (see also the answer to the synthetic conclusions).

3) MATCH-MPIC and EMAC model comparison

The convective transport routine is the only similarity between MATCH-MPIC and EMAC. However, the convective transport strongly depends on the underlying convec-

tion parametrization which determines the mass fluxes and in the supplement (chapter 3) it is discussed that these are different in the two models. Consequently, the convective transport is also different in the two models, only the transport algorithm is the same. We further think that the material in the supplement about the comparison between MATCH-MPIC and EMAC should not be discussed in the paper in too much detail since the goal was not to conduct a model comparison study. For interested readers we added the material in the supplement and chapter 3 in the manuscript should only summarize the most important results.

The comparison was only intended to increase the confidence that our results mainly depend on the tracer properties and the source locations (see also reply to Anonymous Referee #1).

4) Low-level transport from "cold" regions

This is for sure an interesting point, especially in light of the deposition on snow. We also calculated the deposition on snow in the same way as we have done it for the other three ecosystems (forest, pasture, crop-land).

However, it was found that only one MPC (Moscow) has a more substantial impact on snow from which we conclude that the overall impact from MPCs on snow is generally low and is not further discussed in the paper.

5) Synthetic conclusions

A too detailed discussion for specific aerosol components from various MPCs would go beyond the scope of this paper since all MPCs have a different emissions in terms of composition, time of release, and emission strength.

The key findings regarding the respective role of location and tracer properties were drawn from Chapter 5 in the manuscript (see P25409 L9).

We added a sentence to refer to the follow-up study which focuses on different aerosol types from MPCs in the conclusions (P25415 L28): "This will be the scope of a follow-

up study which investigates the impacts from different anthropogenic emissions from MPCs on the burdens of various aerosol types." .

6) Overall role of megacities

A discussion on the overall impacts of MPCs in the global burden of pollution would go far beyond the scope of this paper which has the goal to discuss the potential impacts from MPCs.

However, this discussion will be the topic of the before mentioned follow-up study where individual aerosol types from MPCs are investigated in light of their impact on the global and regional aerosol budget.

Minor comments

1) Air pollution mitigation and greenhouse gases

We agree that populated areas have the potential of air pollution mitigation strategies (see P25391 L16). Moreover, it is well documented that the density of the emissions has an effect on the individual exposure (e.g., Dockery et al., 1993). However, our results point out that the number of people exposed to a certain threshold value (resulting from a unit mass release) is larger in regions with high population densities.

2) Spatial resolution is exactly given on P25394 L24: "The spectral base model is truncated at wave number 106 (T106), which corresponds to a horizontal resolution of the quadratic Gaussian grid of about 1.125° or circa 100 km in longitude and latitude with 31 sigma hybrid levels in the vertical, mainly distributed in the troposphere from the surface up to 10 hPa.". For -Eulerian- global atmospheric chemistry circulation models this resolution can be considered as high.

3) Interactions between trace species

We conducted simulations (not further discussed here) with a lower spatial resolution (T42, 2.8°, ∼300 km) but with a more complex representation of the aerosols, coagu-

lation and coating could change the size and solubility of the passive aerosol tracers (passive in a sense that they do not alter the background aerosol). We found that the impacts of coagulation and coating do not change the results of the pollution potentials substantially. The results are in the order of the results for the NSact or NSinact tracers in the respective size range. From this we concluded that the passive tracers can well represent the ambient aerosols.

4) P25395 L16-18 rephrased to:

"Pozzer et al. (2009) found that the emission height affects tropospheric chemistry close to the source if the emissions are within the planetary boundary layer; the overall effect on the chemical composition is rather low. However, potential transport distances might increase with injections into the free troposphere, e.g. from industrial stacks. "

5) Model setup

Vertical diffusion is one transport process (see also reply to Anonymous Referee #1) which is important for the transport at low levels. However, providing details on the vertical diffusion parametrization would go beyond the scope of this study. Since we use the standard vertical diffusion parametrization of ECHAM5, we refer to Roeckner et al. 2006 and references therein, where a more detailed description is provided. Moreover, the focus of this study was on particulate matter and not on gas-phase species. Trace gases are scavenged depending on their Henry's Law coefficient, which depends on several factors such as concentration in the gaseous and aqueous phase or temperature. We would expect that results for reactive gases would most probably be similar to the results for the "active" tracers which are most efficiently removed from the atmosphere.

6) Aerosol lifetimes

Figure 1 shows already first qualitative indications of the results that are found later together with the differences in the single climate classes. The main point that should

be addressed here is that the aerosol lifetime is within observed values for all tracer from all MPCs and that the differences represent the differences due to the size and the activation state of the aerosols and also to the different locations. The aerosol lifetimes in our study are also of comparable order to those recently reported by Kristiansen et al. (2012).

7) P25397 L3: We added reference for the main sink of fine PM: Kunkel et al., (2012) and Monks et al. (2009).

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