

**Aerosol pollution
from MPCs**

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This discussion paper is/has been under review for the journal Atmospheric Chemistry and Physics (ACP). Please refer to the corresponding final paper in ACP if available.

Aerosol pollution potential from major population centers

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Received: 6 August 2012 – Accepted: 9 September 2012 – Published: 25 September 2012

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

Major population centers (MPCs) or mega-cities represent the largest of growing urban agglomerations with major societal and environmental implications. In terms of air quality they are seen as localized but strong emission sources of aerosols and trace gases which in turn affect air pollution levels in the city or in downwind regions. In the state-of-the-art atmospheric chemistry general circulation model EMAC, generic aerosol and gas phase tracers with equal emission source strengths at 46 MPC locations are used to study the balance between local pollution build up and pollution export, either vertically into the upper troposphere or horizontally, but remaining in the lower atmosphere. The insoluble gas phase tracers with fixed lifetimes are transported with the atmospheric circulation, while the aerosol tracers also undergo gravitational sedimentation as well as dry and wet deposition processes. The strength of low-level tracer export depends on the location of the emission source and prevailing meteorology, in particular on atmospheric stability and the height of the boundary layer and the mixing out of this layer. In contrast, vertical transport of tracer mass depends on the tracer's solubility: the more soluble a tracer is the less mass reaches altitudes above five kilometers. Hence, the mass of insoluble gas phase tracer above five kilometers can be up to ten times higher than the hydrophilic aerosol mass from the same source. In the case of aerosol tracers, pollution build up around the source is determined by meteorological factors which have only indirect effects on tracer lifetime, like surface wind, boundary layer height, and turbulent mixing as well as those which affect the lifetime of the tracers such as precipitation. The longer a tracer stays in the atmosphere, the lower is the relative importance of the location of the source to the atmospheric mass and thus the lower is the relative local pollution build up. We further use aerosol deposition fields to estimate regions with high deposition, that is more than 1% or more than 5% of the corresponding tracer emission deposited in this region. In doing so, we find that the high deposition areas are larger for larger aerosols, and these differ less between the MPCs than for smaller aerosols due to faster deposition. Furthermore, cities in regions with

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high precipitation rates or unfavorable geographic location, e.g. in a basin, suffer most of this high deposition. Most of the high deposition occurs over land, although about 50% of the MPCs are located along coastlines. By folding the aerosol deposition fields with geographical distributions of cropland, pasture, and forest, the impact on different land ecosystems is assessed. In general, forest are exhibited most to deposition from MPCs while pasture land is least affected. Moreover, the impact on humans, measured with a threshold exceedance of pollutant surface mixing ratios, is more dependent on population densities than on the size of the area holding a certain mixing ratio.

1 Introduction

One of the big challenges of the future is the sustainable growth of urban agglomerations. Since 2008 the majority of human population lives in urban regions with the prospect that the trend of human inflow into urban areas will continue throughout this century. The largest of these agglomerations, so called mega-cities or major population centers (MPCs), have two faces. They develop often much faster than the country in which they are located in and promote human development as well as generation of knowledge, technology, and wealth (Kraas, 2007). Furthermore, they can offer a significant chance for climate mitigation strategies as the per capita emission is smaller when many people live close together (Parrish et al., 2009). However, these human agglomerations are very heterogeneous with respect to possible lifestyle of different groups, and consequently manifest different awareness in the handling of energy, water, and waste. Moreover, both increased residential housing as well as the clustering of industry in these metropolitan areas lead to the strong localized release of air pollutants, which generally decreases the air quality for local dwellers as well as in regions downwind (Molina and Molina, 2004). The urban growth occurs on less than 3% of the land surface but with global consequences (Grimm et al., 2008). The enduring urbanization leads to major changes in land-cover and land-use with subsequent changes in surface mean temperatures (Kalnay and Cai, 2003).

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There has been considerable research on these impacts on local to regional (e.g. Par-
rish et al., 2009, 2011) and global (e.g. Lawrence et al., 2007; Butler and Lawrence,
2009; Butler et al., 2012) scales as well as as well as several large mega-city projects
(e.g. Baklanov et al., 2010; Molina et al., 2010). Lawrence et al. (2007, later referred
to as L07) studied the outflow characteristics of generic gas phase tracers with three
different lifetimes (1, 10, 100 days) from 36 MPCs to obtain insight in the balance be-
tween local pollution build-up and long-range transport. They found that low-level ex-
port is strongest in middle and high latitudes while the export into the free troposphere
is dominant in the tropics. Moreover, the vertical mixing is most responsible for reduc-
ing the surface pollution. They provide detailed insight in the outflow characteristics of
the gas phase tracers, but with a limited range of tracer properties. This study picks
where L07 left off and answers the following questions:

1. How robust are the results of L07, or do they depend strongly on the selected model?
2. To what degree do outflow characteristics depend on the lifetime of the tracer, especially if it varies in space and time, and how does it differ for gaseous and aerosol substances?
3. What more can be learned from tracer distributions and from tracer deposition fields when they are convolved with geographical population or agricultural distributions?

To investigate these aspects we use the global 3-D state-of-the-art atmospheric chemistry general circulation model (AC-GCM) EMAC (ECHAM5-MESSy Atmospheric Chemistry, Jöckel et al., 2006) in high spatial resolution with generic aerosol tracers of various sizes and different solubility as well as generic gas phase tracers equal to those used by L07. 46 cities around the world (see Table 1) have been chosen as emission hot spots to determine the balance between pollutant build up around the source and pollutant export.

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In Kunkel et al. (2012) the same set of MPCs was used to determine the remote aerosol deposition potential, i.e. the deposition beyond various distances. In particular, it was found that on average about half of the emitted mass of small ($d \leq 2.5 \mu\text{m}$) aerosol tracers is deposited beyond 1000 km. Several other studies investigated trace gas or aerosol transport from various, mostly larger sources such as countries or even continents on continental scales (e.g. Duncan and Bey, 2004; Park et al., 2005; Wagstrom and Pandis, 2010) or trans-continental scales (e.g. Betzer et al., 1988; Jaffe et al., 1999; Wild and Akimoto, 2001; Stohl et al., 2002, 2003; Liu and Mauzerall, 2007; Liu et al., 2009). More studies concerning pollutant dispersion from smaller emission points have been published for the regional scale, commonly focusing on one source point either for different meteorological conditions or with changed emission scenarios (e.g. de Foy et al., 2006; Streets et al., 2007; Wang et al., 2010; Song et al., 2010). In this study, as well as for L07, Butler et al. (2008), and Kunkel et al. (2012), the focus is directly on the comparison between MPCs all over the world.

We structured the paper to first provide a description of the EMAC model system, the metrics to characterize the outflow, and further data on population and agricultural distributions. In Sect. 3 we briefly compare EMAC results with results from L07 to assess the robustness of their analysis before the aerosol dispersion is discussed in Sect. 4. The aerosol tracers are then compared to generic gas phase tracers in Sect. 5 and finally in Sect. 6 the aerosol deposition and surface densities are used to assess the impact of MPCs on agriculture and population. The key conclusions and further open questions are summarized in the last section.

2 Model and analysis data

2.1 Model and major population centers

This study uses the same setup as Kunkel et al. (2012) with 46 major population centers distributed around the world, as listed in Table 1. Not all of them strictly fulfill the

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criteria to be called a mega-city for which, depending on definition, either more than five or ten million inhabitants are needed (UNFPA, 2007). However, we use several additional cities, e.g. Johannesburg, Sydney, Melbourne, Atlanta, Kinshasa, Khartoum, and Nairobi, due to their outstanding importance in their region or because they are expected to become mega-cities in the near future. Furthermore, we include three areas, i.e. the Rhine-Ruhr Area, the Po Valley, and the Szechuan Basin, which are agglomerations of many cities and important industrial sites. Moreover, Hong Kong includes also the Pearl River Delta (PRD) and Kinshasa in the Democratic Republic of the Congo as a representative for for agglomeration together with Brazzaville in the Republic of the Congo. All MPCs are taken as large emission point sources for generic tracers to study the dispersion in the atmosphere, and in the case of aerosol tracers subsequent deposition onto the surface. For this we apply the AC-GCM EMAC (Jöckel et al., 2006) with ECHAM5 (version 5.3.01, Roeckner et al., 2006) as a base model to calculate the meteorology. The base model is coupled to the Modular Earth Submodel System (MESSy, version 1.9, Jöckel et al., 2005, and www.messy-interface.org) which allows the inclusion of different sub-models for physical and chemical processes in the atmosphere and interactions between the atmosphere and biosphere. The model provides a good representation of gas phase tracers (e.g. Jöckel et al., 2006; Pozzer et al., 2010; Tost et al., 2007) as well as aerosols (e.g. Pringle et al., 2010; Pozzer et al., 2012). Generic aerosol tracers have been used by Burrows et al. (2009) to study the transport of biogenic aerosol particles between different ecosystems and by Lelieveld et al. (2012) to determine the global exposure risk after nuclear accidents.

2.1.1 Meteorological setup

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.

The model simulations were initialized in July 2004 and ended in January 2006, with

the first half a year used as model spin up time. Meteorological variables, namely temperature, vorticity, divergence, and the logarithm of surface pressure are weakly nudged towards analysis data from the ECMWF ERA-Interim reanalysis project (Dee et al., 2011). The year 2005 was chosen as analysis time period as it shows no strong annual modes in ENSO or NAO and coincides with the main analysis year of the MEGAPOLI project (Baklanov et al., 2010).

2.1.2 MESSy setup

The present model study comprises the following physical processes: emission, atmospheric transport, and wet and dry deposition of generic aerosol tracers and emission, atmospheric transport, or a radioactive like decay of generic gas phase tracers equal to those used in L07. For each of the 46 emission points an independent set of tracers was released at a constant, prescribed emission rate of 1 kg s^{-1} . The emission was placed in the middle of the lowest model layer in all grid boxes hosting an MPC. These boxes have a mean depth of about 60 m in the vertical and approximately 100 km in the horizontal. As long as the emissions are within the planetary boundary layer, the height of the emissions affects tropospheric chemistry close to the source but 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 (Pozzer et al., 2009). Thus, by using surface emissions, we obtain a conservative estimate for results regarding long-range transport.

Tracer transport is split into transport in the large-scale advection (Lin and Rood, 1996), sub-grid scale convection (Tiedtke, 1989; Nordeng, 1994; Lawrence and Rasch, 2005; Tost et al., 2010), vertical diffusion by turbulent mixing (Roeckner et al., 2006), and in the case of aerosols gravitational settling (Kerkweg et al., 2006a), neglecting chemical reactions and micro-physical changes. The atmospheric residence time of the aerosol tracers is determined only by four independent removal processes: (1) aerosol sedimentation onto the surface, (2) dry deposition by contact with the surface based on its

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roughness and the boundary layer mixing (Kerkweg et al., 2006a), and scavenging by (3) impaction and (4) nucleation in large-scale and convective clouds (Tost et al., 2006, 2010). We refer to (1) and (2) as dry deposition processes and to (3) and (4) as wet deposition processes. MESSy sub-models in use for this study are CLOUD (Jöckel et al., 2006), CONVECT and CVTRANS (Tost et al., 2010), RAD4ALL (Jöckel et al., 2006), ONLEM (Kerkweg et al., 2006b), DRYDEP (Kerkweg et al., 2006a), SEDI (Kerkweg et al., 2006a), SCAV (Tost et al., 2006, 2007), and PTRAC (Jöckel et al., 2008).

In this study mono-modal aerosol tracers represent aerosols with ambient diameters of 0.1, 0.5, 1.0, 2.5, and 10.0 μm with a fixed standard deviation of 1.0. Instead of using a complex, computationally expensive aerosol module to explicitly treat aerosol microphysics, we use two different aerosol solubility states which differ in the treatment of scavenging by nucleation: in one case all aerosols can undergo this process (further denoted as NS_{act}) which is tracer size dependent and in the other case this process is completely inhibited and the aerosol tracers are considered to be insoluble (further denoted as NS_{inact}) which is summarized in Table 2. These two aerosol states are considered as extrema with respect to aerosol solubility, representing an upper and lower estimate of results with the range in between being representative for most of the real atmospheric aerosol particles. Thus, ten aerosol tracers are released at each emission source point, one for each size and solubility state, all with the same source strength.

This allows us to use the various aerosol tracer properties to obtain information on various parts of the aerosol size distribution. In total, we use a set of 460 aerosol tracers (46 source points \times 5 aerosol sizes \times 2 aerosol solubility states) which do not interact with each other and have no feedbacks on clouds or the radiative balance. Interpretation of the sizes is the same as that described in Kunkel et al. (2012).

Figure 1 shows the different annual atmospheric residence times of the tracers, ordered by the longest lifetime of the NS_{inact} 1 μm aerosol tracers. The aerosol residence time is calculated with a steady state approach by dividing the annual mean aerosol load by the total annual emission (Seinfeld and Pandis, 2006). We obtain residence times for each MPC with an upper limit provided by NS_{inact} and a lower limit by NS_{act}

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aerosol tracers. As the latter are more susceptible to wet deposition, they have shorter atmospheric residence times. 80% to 95% of the mass of fine aerosols is wet deposited which is the main sink of fine aerosols in nature and in the model. All aerosol tracers with diameter $d \leq 2.5 \mu\text{m}$ are defined as fine aerosols, similar to the definition of $\text{PM}_{2.5}$, which is the mass of aerosols with an ambient diameter less than $2.5 \mu\text{m}$. For larger aerosols the difference in residence time with respect to aerosol solubility becomes smaller as dry and wet deposition contribute almost equally to the sink processes. Furthermore, the effect of nucleation scavenging is smaller for larger particles as they are removed very efficiently by impaction scavenging (Tost et al., 2006). In the following chapters we will refer to aerosol tracers with 0.1 , 0.5 , and $1.0 \mu\text{m}$ as small aerosol tracers, to those with $10.0 \mu\text{m}$ as coarse aerosol tracers.

2.2 Metrics

We use similar metrics to L07 to distinguish between pollution at low levels and in the upper troposphere, and also between local and remote pollution from MPCs (summarized in Table 2). The equal emission rate for all tracers and MPCs provides the potential to compare the pollution from urban centers only by their location and prevailing meteorology, independent of real emissions. The upper tropospheric pollution potential is characterized by the mass fraction above 5 km altitude (E_{UT}). The low level pollution potential is split into a long-range ($\text{ELR}_{1 \text{ km}}$) and a city scale ($\text{RCL}_{0.5 \text{ km}}$) fraction with $\text{ELR}_{1 \text{ km}}$ being the mass fraction below 1 km altitude and at least 1000 km away from the source, while $\text{RCL}_{0.5 \text{ km}}$ is the mass fraction retained in the first 500 km from the emission source points and below 0.5 km altitude. The first two metrics describe the outflow of an MPC, either vertically (E_{UT}) or quasi-horizontally ($\text{ELR}_{1 \text{ km}}$), the third one ($\text{RCL}_{0.5 \text{ km}}$) the pollution build up around the source. To calculate $\text{RCL}_{0.5 \text{ km}}$ and $\text{ELR}_{1 \text{ km}}$, a set of equidistant points around each emission source have to be determined. On a plane they are located on a circle with the emission source as center point, on a graticule (latitude-longitude grid) this becomes an ellipsoid. The corresponding distances from the center points are

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based on the radii of the WGS84 ellipsoid (National Imagery and Mapping Agency, <http://earth-info.nga.mil/GandG/publications/tr8350.2/wgs84fin.pdf>, 2000 or see L07). In addition, the local pollution build up can be assessed by adding up the area, or other properties in this area, in which the tracer density exceeds a given threshold A_Y , with Y a threshold value in ng m^{-3} (tracer mixing ratios are provided in mol mol^{-1} and can be converted to ng g^{-1} with molar masses for the tracers, here chosen to be 1 g mol^{-1} , and the molar mass of air 28.97 g mol^{-1} ; the surface air density, approximately 1 kg m^{-3} , is then used to convert the mass mixing ratio in the lowest layer to a concentration in ng m^{-3}).

We further focus on the concentrated deposition patterns, i.e. high relative deposition. We calculate the emission normalized deposition fraction D_X (in %), i.e. the annual mean deposition flux DEP_{accu} (in $\text{kg m}^{-2} \text{ s}^{-1}$) in each model grid box divided by the annual global mean emission flux EM_{total} (in $\text{kg m}^{-2} \text{ s}^{-1}$) from the corresponding source point:

$$D_X = \frac{\text{DEP}_{\text{accu}}}{\text{EM}_{\text{total}}} \times 100\%. \quad (1)$$

This fraction together with a threshold value X (in %) can be used to characterize the impact of an MPC on different ecosystems. We fold D_X with surface properties, namely total surface area, water or land area, crop land, pasture, or forest. Similarly, we use A_Y to assess the population number being exposed to a certain surface mixing ratio. Depending on the property, we speak of a cropland, pasture, or forest deposition or human exposure in the following sections.

Finally, it is noted that for seasonal analyses, metric results for urban centers from the Southern Hemisphere have been shifted by six months to match the seasons on the Northern Hemisphere.

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2.3 Agricultural and population data

Gridded population number for the year 2005 is used to determine the impact of aerosols on humans (see also Fig. S1 in the Supplement). Data is based on gridded population data of more than 300 000 national and sub-national administrative units to assign population values to grid cells (SEDAC, 2005).

Moreover, the fraction of three different land ecosystems, crop land, pasture, and forest data are used to determine the impact on land ecosystems (see Figure S1 in the Supplement). Crop land and pasture data are compiled using satellite-derived land cover data and agricultural inventory data (Ramankutty et al., 2008) for the year 2000. Data is originally available at about 10 km horizontal resolution <http://www.geog.mcgill.ca/~ramankutty/Datasets/Datasets.html>, as of 8 June 2012). The forest data is based on US Geological Survey Global Forest Resource Assessment (FRA 2000) and is based on images from the Advanced Very High Resolution Radiometer (AVHRR) satellite at a resolution of about one kilometer <http://edc2.usgs.gov/glcc/fao/index.php>, as of 8 June 2012). The data set originally comprises five classes: closed forest (40–100 % canopy cover), open or fragmented forest (10–40 % canopy cover), other wooded land, and water (lakes, rivers, etc.) which have been merged to one forest class for this study. All these data sets have been regridded to the model resolution and together with the metrics D_X and A_Y are used to assess the human exposure, cropland deposition exposure, pasture deposition exposure, and forest deposition exposure.

3 Model comparison of outflow characteristics from major population centers

We first summarize the results of the model comparison between the EMAC model and the MATCH-MPIC model, a semi-offline, three dimensional, chemical transport model (e.g. Rasch et al., 1997; Lawrence et al., 1999; Kuhlmann et al., 2003) which was used by L07. A more detailed analysis is given in the electronic supplement. We use the same 36 major population centers as L07 to test the reproducibility of their

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5 results. Total and upper tropospheric column and surface densities agree well between the two simulations and also the resulting metrics give comparable numbers with high coefficients of determination (R^2) of about 0.75. In general, the low level export is stronger when simulated with MATCH-MPIC, as well as the area with a mass threshold exceedance of 10 ng m^{-3} , while the upper tropospheric pollution is stronger in EMAC (see Table 3).

10 The main results from L07 are well reproduced when using EMAC which underlines the robustness of the results. Discrepancies in the metrics mainly result from the different underlying meteorological fields (from NCEP for MATCH-MPIC and from ECMWF for EMAC) and different advective and convective transport schemes which introduce the largest uncertainties in such calculations.

4 Atmospheric dispersion of aerosols from urban emission hot spots

15 In the following section we focus on aerosol transport with subsequent deposition which leads to different atmospheric residence times of single tracers. Aerosols are removed by dry and wet deposition processes, thus, aerosol weight, surface properties as well as meteorological conditions determine the distance an aerosol particle can travel in the atmosphere before being deposited to the ground. We utilize $\text{RCL}_{0.5\text{km}}$ to characterize the local low-level pollution build-up, $\text{ELR}_{1\text{km}}$ for the remote low-level pollution, and E_{UT} for upper tropospheric pollution.

4.1 Local pollution build up

20 In Fig. 2, the top panel shows annual averages of $\text{RCL}_{0.5\text{km}}$. The gray shaded area which is bounded by the solid and dashed black lines represents the MPC averages for NS_{inact} and NS_{act} aerosol tracers. The light gray shaded areas contiguous to this area denote the standard deviation, each plotted one-sided for the upper and lower bound. The overall annual mean values range between 10% and 14% for small tracers and

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15% to 19% for large tracers. Six climate classes are used equal to those in Kunkel et al. (2012), to provide more information about regional differences. As our results are mainly dependent on the prevailing meteorology, the MPCs are grouped by their Köppen-Geiger climate classification (Peel et al., 2007) which depends on temperature and precipitation (see also Table 1): tropical A (red), arid B (brown), temperate no dry season Cf (light green), temperate dry winter Cw (light blue), temperate dry summer Cs (dark green) and cold D (blue) (see Table 2).

For small NS_{inact} aerosol tracers, minimum retention of tracer mass occurs in urban centers in arid (B) and temperate dry summer (Cs) regions with $RCL_{0.5km}$ about 7%, and the maximum for MPCs in tropical (A) and cold (D) regions with values of about 11% to 12%. The same regions show the minima for NS_{act} aerosol tracers of about 9%, while tropical and temperate no dry season (Cf) means are the maxima, with values of about 17%. Large aerosol tracers from urban centers in arid regions reveal minimum values of 9% (NS_{inact}) and 10% (NS_{act}), respectively, while the maximum values are found for both solubility states in cold regions and also in the soluble case for temperate no dry season regions, with climate class means of 20% (NS_{inact}) and 22% (NS_{act}). Further, almost all mean values for arid and temperate dry summer regions show, independent of solubility state, smaller mean values than all other climate class means, which can also be seen in almost every season of the year (see Figure S4 in the Supplement). Only in temperate dry winter regions low $RCL_{0.5km}$ values appear during winter months (DJF). Thus, the relative pollution build up in these two regions is smaller than in all other climate classes. Furthermore, the pollution build up in two contrary climate regions, tropical and cold, is highest, although meteorological conditions lead to different atmospheric dispersion (see Sect. 4.2).

The retention of tracer mass is controlled by two mechanisms. On the one hand, $RCL_{0.5km}$ anti-correlates with the atmospheric residence time of the tracers. As mentioned before, the residence time is mainly governed by wet removal, thus aerosol tracers with short residence times often originate in regions with high precipitation rates, i.e. tropical regions. In contrast, for tracers from arid regions the residence time is relatively

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long due to missing precipitation. Thus, the relative contribution of the emission to the global burden of a tracer is larger for those with short lifetimes, since a large fraction of the burden is generally closer to the source with higher $RCL_{0.5\text{km}}$ values. This accounts for about 50 % of the variance in the overall $RCL_{0.5\text{km}}$ results. On the other hand, meteorological conditions that change the distribution of the tracer mass in the atmosphere, though not directly their residence time, are responsible for the missing variance. For example surface winds, turbulent mixing, or the depth of the boundary layer height can alter $RCL_{0.5\text{km}}$ values without affecting the aerosol lifetime.

The influence of precipitation also plays a major role in the seasonal variations in $RCL_{0.5\text{km}}$. The minimum climate means stay almost constant throughout the year as the minima are given by mean values from regions which are weakly affected by precipitation. In contrast, the maxima vary with the season, with the difference between $RCL_{0.5\text{km}}$ minima and maxima being largest in summer (JJA) when precipitation rates are generally larger and consequently the aerosol residence times shorter.

4.2 Outflow of pollution from urban centers

4.2.1 Remote low-level outflow

In contrast to local pollution build up, the emissions from urban centers also undergo transport which can either be horizontal or vertical. We first assess the horizontal transport focusing on the mass which is transported near the surface or which was first lifted and subsequently descended again to lower altitudes, e.g. in a dry intrusion or simply by aerosol sedimentation. Overall annual means of $ELR_{1\text{km}}$ vary between 16% and 20% for small and 12% and 15% for large tracers with larger values now given by NS_{inact} aerosol tracers (see middle panel in Fig. 2). Including climate class averages, the diversity increases with values ranging from 11% to 27% for small and from 8% to 19% for large aerosol diameters. Aerosol tracers from cold regions possess the largest remote low level mass fraction with more than 25% of their total mass (averaged over NS_{inact} and NS_{act} tracers) located in the near-surface layer downwind of the source.

In contrast to the local build-up, aerosols from temperate dry summer regions show a large downwind pollution potential. The least export occurs in temperate dry winter and tropical regions as upper level export is favored in these regions (see following Sect. 4.2.2).

5 Similarly, the smallest seasonal means appear for aerosol tracers from tropical regions, except during winter when those from temperate dry winter regions show least downwind pollution potential due to relatively enhanced transport into the upper troposphere. $ELR_{1\text{ km}}$ means peak either for aerosol tracers from cold or temperate dry summer regions. In general, the highest downwind pollution potential is found during
10 winter months (DJF) with overall means between 21 % and 25 % and the least during summer (JJA) with overall means of about 10 % to 12 %.

In winter, atmospheric low level inversions occur more frequently and aerosol tracer mass is kept more efficiently within the boundary layer which is commonly shallower during this time of the year. Together with less convective activity, the $ELR_{1\text{ km}}$ seems
15 to be dominated by the atmospheric stability. Higher stability leads to greater remote low level transport.

Similar to L07 we analyzed the response of a change in the depth of the near-surface layer and consider a metric height of 500 m. L07 found that the remote low level export decreased almost linearly with the volume, i.e. only a tenth of the $ELR_{1\text{ km}}$ numbers
20 are found in a layer which is 100 m deep. For aerosols, on annual average we find that about 50% of the $ERL_{1\text{ km}}$ mass is present in a volume half as deep as our original volume, thus the same relationship is found for aerosols as for gases in L07. We see a small seasonal variation in this number with a maximum in winter (on average 53%) and a minimum in summer (on average 47%) due to the same reason as for the seasonal variation in $ELR_{1\text{ km}}$. Additionally, the ratio is slightly dependent on aerosol solubility with higher ratios for soluble aerosol tracers but with almost no dependence
25 on aerosol size.

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4.2.2 Outflow into the upper troposphere

Aerosol mass can also be lofted out of the source region into the upper troposphere (bottom panel of Fig. 2). Overall annual means of E_{UT} are between 3% and 4% for small and 2.5% and 3.5% for large aerosol tracers and thus much smaller than for the other two metrics and especially much smaller than E_{UT} values for gas tracers reported by L07. The order of the climate means is reversed compared to the order found for $ELR_{1\text{km}}$, now with maximum pollution for aerosol tracers from temperate dry winter and tropical regions and minimum pollution for those from cold regions.

These findings rely on several facts which need to be discussed. First, the difference between NS_{inact} and NS_{act} means strongly varies between climate classes. For tropical regions which are heavily and regularly affected by convection, the different treatment of nucleation scavenging leads to strong differences in the UT pollution with NS_{act} means accounting for only about 60% of the NS_{inact} means. In contrast, for aerosols from cold regions this ratio is about 90%, thus the difference is almost negligible caused by an inefficient nucleation scavenging of snow and ice crystals according to the model formulation. Second, in L07 the strongest vertical tracer displacement occurred in tropical regions, contrary to our results with maximum vertical aerosol transport in temperate dry winter regions, especially in the case of NS_{act} means. The input of mass into the upper troposphere occurs in most cases by convective or conveyor belt lifting. Both processes are commonly associated with cloud formation and subsequent precipitation which both remove aerosol particles efficiently. Nucleation scavenging during cloud formation is the dominant scavenging process for aerosol particles with diameters larger than $0.2\ \mu\text{m}$, while impaction scavenging becomes important for large aerosol particles. With sufficient lifting activity the E_{UT} mass fraction strongly depends on the liquid and/or ice water content of the clouds and their extension, which in turn determine the scavenged mass. From the pure gas phase tracer simulations we know that the upward transport is strongest in the tropics though the moisture present there limits the upward transport of aerosols so strongly that less mass than in the

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temperate dry winter regions reaches the upper troposphere. In arid regions precipitation hardly affects lifting processes; however, the tracer distribution is affected by the development of a very deep, well mixed air layer which can reach altitudes up to 5 km such as the Saharan Air Layer (Hall and Peyrillé, 2007). The aerosols emitted at the surface are already mixed to altitudes of several kilometers in this layer and consequent lifting above 5 km can occur faster and is often associated with little or no precipitation. Third, the seasonal cycle is also reversed compared to what was reported for $ELR_{1\text{ km}}$, with minimum values during winter, overall mean of about 2%, and maximum values during summer, overall mean between 4.5 and 6.5% (see Figure S4 in the Supplement). Furthermore, the variability, indicated by the standard deviation and also by the differences between various climate classes, is largest in summer and smallest in winter. This is again attributed to the convective activity, which is stronger in summer, but also shows strong differences between different regions of the world and thus also between different climate classes.

Thus, the interplay of atmospheric moisture and lifting activity and their effect on aerosol tracers is the determining factor for transport to the upper troposphere, with relatively small aerosol mass fractions in this domain.

4.3 Relation between pollution build up and outflow

Finally, we want to see whether there are significant correlations between pollution build up and outflow for aerosols from MPCs. L07 reported that for gas phase tracers a significant relation was found, and that the upward transport, rather than the low level transport, is mainly responsible for ventilating surface pollution. Moreover, they found that the trade-off between long-range low-level export and dilution of pollutants in the region surrounding the MPCs becomes more evident for regions with similar vertical transport.

In general, we find an anti-correlation between the horizontal and vertical export (see Table 4). This relationship is expected as aerosol mass is either lifted to higher altitudes or kept at lower levels, being advected rather horizontally. However, the overall

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anti-correlation is weakened by the weak positive correlation in the tropics. In the tropics, an ambiguous situation is found for different MPCs. For instance in Jakarta and Manila, the opposing relationship between the two export pathways is still assessed, while it breaks down in other MPCs, for instance in Rio de Janeiro, Kolkata, or Lagos. A possible interpretation might be that in the first case, convective scavenging dominates the wet deposition and in the second case more aerosols are scavenged in large-scale clouds. In regions with dominant convective scavenging more aerosol mass is effectively lifted to higher altitudes but also efficiently removed by scavenging. Less mass is then left over to participate in any long-range transport, especially at low levels. This could result in an opposing relationship between vertical and near-surface long-range transport. Oppositely, more large-scale scavenging indicates that a larger proportion of aerosol mass has possibly undergone farther transport before removal. A correlation between vertical and near-surface long-range transport would then suggest that the outflow occurs at high and low levels.

Between low level export and low level retention, the correlation coefficients show a very ambivalent behavior with most values in the range of $[-0.5, 0.5]$. The sign varies between various climate classes and also between aerosol size and solubility. Only in two cases are higher correlations assessed: a positive one in temperate dry summer regions for small and large aerosol tracers and a negative one for small aerosol tracers from cold regions. In the first region, the mass fraction is rather small for $RCL_{0.5\text{ km}}$ but high for $ELR_{1\text{ km}}$. The positive correlation indicates that the low level export forms an important path to accumulate mass in the near-surface region downwind. A positive correlation might be expected for the cold regions with high $ELR_{1\text{ km}}$ and $RCL_{0.5\text{ km}}$ mass fraction. However, as the vertical export is low in this climate region, the strength of surface wind and turbulent mixing within the boundary layer determines whether more of the mass is retained close to the source or advected downwind.

As it is expected, the correlation coefficients between low level retention and vertical export reveal in most cases a negative sign. For small aerosol tracers the correlations are rather weak, only in temperate dry summer regions do we see a strong anti-correlation

and in cold regions a strong correlation. The positive correlation coefficients for cold regions reveals that if lifting takes place it occurs close to the emission source at low levels. On the other hand, an anti-correlation points more to lifting that either takes place farther away from the source or has its origin above 500 m altitude. For most climate regions this is true for the large aerosol tracers as they show stronger anti-correlations, even in cold regions. However, the sampling statistics are small and not equally distributed over the climate classes, thus the results should only be interpreted as a first indication.

5 Aerosol – gas phase comparison

Finally to conclude the analysis of atmospheric dispersion from urban emission sources, we want to show similarities and differences between the gas phase tracers with fixed atmospheric lifetime and aerosol tracers with various atmospheric residence times. Here, we mainly focus on aerosol tracers with a diameter of 1 μm which have a lifetime comparable to, though generally shorter than the lifetime of, the gas phase tracers, but also include aerosol tracers with a diameter of 10 μm in parts of the discussion. In Sect. 3 we briefly discussed similarities and differences in the dispersion from MPCs with two different model systems. Now, we compare the aerosol results discussed in Sect. 4 with gas phase tracer results from an EMAC simulation using the same configuration as used for the aerosol simulation.

Figure 3 gives a qualitative overview of how the atmospheric dispersion patterns change with respect to the removal of an atmospheric tracer. The amount of various removal processes acting on the individual tracers decreases from left to right in Fig. 3. In general, the gas phase tracers in the right column are wider spread from the source due to their longer lifetime. Close to the source points, similar values are found for gas phase and aerosol tracers but farther away the discrepancies between the various types of tracers become more apparent. The total column densities reveal higher values in the direct outflow regions of the MPCs in the case of the gas phase tracers and

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also in remote areas like the Southern Pacific with no significant tracer burden present for aerosol tracers. Tracer distributions in the surface layer vary strongly and the transport to higher latitudes is not strongest for the gas phase tracers, but rather for NS_{inact} tracers, especially in Northern Europe. In contrast, the gas phase tracers predominate in the northern latitude storm tracks where almost no aerosol mass is simulated anymore. The largest differences between aerosol and gas phase tracers are found for the column densities in the upper troposphere above 5 km. The absolute aerosol tracer peak values are about the magnitude of the background levels of the gas phase tracers in remote areas. Moreover, location and strength of peak values differ, for instance for Khartoum and Teheran, which both have a much weaker signal in the gas phase compared to the background than for the aerosol tracers. Furthermore, Santiago and Lima are clearly discernible in the aerosol phase but not in the gas phase, and also the outflow from East Asia extends farther northward for the gas phase tracers.

The question arises then how the pollution potentials differ between gas phase and aerosol tracers. Figure 4 shows the three pollution potentials used to investigate the similarities and differences in the local pollution build up and pollution export: $ELR_{1\text{ km}}$ (top panel), E_{UT} (middle panel), and A_{10} (bottom panel). Generally, the accordance between aerosol and gas phase tracer is higher when using the NS_{inact} aerosols. The greatest similarity in pollution potential is found for the low level pollution potentials.

Aerosols show slightly higher $ELR_{1\text{ km}}$ values than gas tracers, with coefficients of determination (R^2) higher than 0.9 (NS_{inact}) and 0.8 (NS_{act}), respectively. As the R^2 are a measure of how much of the variance in the correlation can be explained by the source location, the tracer properties seem to be less important to determine the mass fraction found in the lowest layers downwind of the source. Instead, the location of the source along with the prevailing meteorology account for the mass transported downwind. In contrast, the vertical transport shows gas phase mass fractions up to ten times higher than aerosol mass fractions. The correlation between gas and aerosols is still positive but weak, with an R^2 of about 0.4 (NS_{inact}) and about 0.15 (NS_{act}), respectively. The source location is in this case of minor importance compared to the tracer

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properties, especially the solubility of a tracer which determines to a great extent the mass fractions above 5 km. In the case of the area mass threshold exceedance, both the source location and tracer properties need to be considered more equally to explain the relation between gases and aerosols as positive correlations are found with $R^2 = 0.78$ (NS_{inact}) and $R^2 = 0.67$ (NS_{act}) for small aerosols and an averaged R^2 of about 0.53 for large aerosols. Hence, the source location determines the A_{10} more in case of small aerosols while the size of the aerosol seems to be more important for the large aerosols.

In summary, the findings of Sect. 4 are further supported by the comparison of gas phase and aerosol tracers, especially that the source location is most important in determining the low level pollution, while tracer properties have a substantial impact on the vertical transport.

6 MPC impact on ecosystems and humans

Aerosol particles have the ability to potentially influence their environment when they are airborne in several ways. For instance, humans can breathe in small aerosol particles deeply into the lungs and it is certain that this affects the life expectancy of humans (e.g. Dockery et al., 1993; Pope et al., 2009). Large particles are mainly responsible for decreasing visibility (e.g. Watson, 2002) and generally particles which contain toxic constituents from anthropogenic combustion can harm crops (e.g. Wiman et al., 1990; Burkhardt, 2010) and humans (e.g. Pöschl, 2005; Stanek et al., 2011). In this chapter we assess the potential impact of the urban emitted aerosol particles on the environment by using the metric D_X , which is the ratio of the total (= dry and wet) deposition flux to the global mean emission flux (defined in Sect. 2.2) and threshold values X of 1% and 5%, and we refer to this as high relative deposition. Although we are aware of the different impacts of dry and wet deposited, we discuss only their sum here and leave a further investigation open for following studies. Furthermore, we use the metric

A_Y to quantify the number of people that are exposed to a certain atmospheric mixing ratio.

6.1 Impact on ecosystems

6.1.1 Impact on land and sea surfaces

5 First, we assess the total surface area which fulfills the criterion of D_X . In Fig. 5 the total surface area is illustrated for each MPC for two aerosol diameters, $1.0 \mu\text{m}$ and $10.0 \mu\text{m}$, on the left for D_1 and on the right for D_5 . The solubility states are displayed by different gray shadings with the light gray bar representing the additional area between the two solubility states. In contrast to the atmospheric metrics, the smaller area, indicated by the dark gray bar, is not always determined by the same solubility state for each MPC but most commonly by the NS_{inact} tracers. If only a dark gray bar is drawn, then there is no difference between the two states, while a single light gray bar indicates that no area is covered at all for one state. By definition areas which are covered with at least 5% of the emissions are also covered with at least 1%.

15 In the case of D_1 , each MPC covers at least a small area, independent of aerosol size, which is not the case for D_5 . Aerosol tracers from MPCs which are known to undergo substantial long-range transport, e.g. Baghdad, Teheran, Cairo, or Khartoum, are more widely spread and thus aerosol mass is deposited in lower concentrations in each grid cell. However, other MPCs, e.g. London or Tianjin, show also zero area covered, thus long-range transport is not the only limiting factor for high relative deposition. In contrast, the maximum is found for MPCs which are either influenced by heavy precipitation rates, e.g. Lagos or MPCs in South-East Asia, or located in a basin with low ventilation, e.g. Bogotá or Szechuan. On average large aerosol tracers cover larger areas due to their shorter travel distances and the subsequent smaller total overall area which is affected by deposition. This in turn leads to higher deposition rates on the area where deposition occurs. Furthermore, the faster deposition rates for large aerosols lead to smaller differences between the areas covered by the two solubility states.

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Further, we divide the total surface area into land and sea fractions to obtain a more accurate knowledge over the location of high relative deposition. Generally, the low level outflow of aerosols is similar to the gas phase outflow reported by L07 with more low-level long-range transport over oceans. Nevertheless, the total deposition still occurs more over continents with about 55 % of the small tracer emissions and 68 % of the large tracer emissions being deposited on land surfaces. For the high relative deposition, we focus only on D_1 as not all of the MPCs cover a surface area in case of D_5 . As the land-sea fractionation is very similar for all aerosol tracers, independent of size or solubility, we choose the NS_{act} 1.0 μm aerosol tracers to show D_1 in Fig. 6. The dark gray bars represent the land fraction which outweighs the sea fraction in the light gray color bars, although almost 50 % of the MPCs are located at coastlines and several more close to the coast. In several cases the land fraction is close to 100 %, mainly for MPCs with a dominant outflow over continental regions, e.g. Moscow, Rhine-Ruhr Area, Teheran, Delhi, Szechuan. However, there are also some MPCs close to coastlines which do not have high relative deposition over seas, e.g. Santiago or Beijing. The highest marine D_1 values appear for Athens and Rome, with about 80 % sea fraction, but not surprisingly Jakarta and Manila show the highest sea fractions in terms of absolute area as each of these cities is located on a small island in a maritime environment.

6.1.2 Impact on cropland, pasture, and forest

To answer the question which of the three ecosystem – cropland, pasture, or forest – is most exposed to deposition from MPCs, we initially fold the sum of all annual accumulated MPC deposition fields with the relative fractions for cropland, pasture, and forest (see Figure S1 in the Supplement). The differences between small aerosol tracers ($d \leq 1.0 \mu\text{m}$) are minor as well as between the two solubility states. Thus, we focus the discussion on the NS_{act} 1.0 μm and 10.0 μm aerosol tracers which are shown in Fig. 7. The pure deposition patterns have their maximum around to the source location (Kunkel et al., 2012). In Fig. 7 MPCs are in most cases well detectable by the red

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5 areas in the six panels. For several MPCs these red areas exceed the MPC size as these are surrounded by large areas of an ecosystem, e.g. for cropland the corn-belt around Chicago, the region between Paris and east of the Rhine-Ruhr Area, the region north of Lagos or large areas on the Indian subcontinent and in Eastern China. In the case of pasture land only a few hot spots are detected, for instance the region around Bogotá, the Sahel region in Africa, and again large areas in Eastern China. Forests seem to be widely exposed to deposition from MPCs, e.g. in the USA in a belt from New England down to Atlanta, high forest deposition exposure is simulated as well as in Bogotá and in the region between Rio de Janeiro and São Paulo. In Central Africa the Congo basin with the African rainforest and in Asia regions in Eastern China and also in Japan reveal high exposure. In Europe, the high northern latitudes are affected due to the vast appearance of boreal forest. For the same reason the deposition of Moscow can still be detected north of Mongolia far away from its source. In contrast, one of the most forested areas, the Amazon basin, seems not to be affected by the deposition of the MPC tracers used in this study because it is not located downwind of the MPCs.

10 Using D_1 and D_5 , we can calculate the area of each ecosystem which is exposed to more than 1% or 5% of the emission. Table 5 shows the averages of small and large aerosol tracers for each ecosystem. Generally, larger areas are found for the soluble tracers and, as discussed before for the total surface area, for larger aerosol tracers with one exception in the case of forests for D_1 ($NS_{act} 1.0 \mu m$). The results for the two aerosol solubility states differ by less than 25%, in most cases even less than 20%. Maximum averages are found for forests, followed by croplands with up to 40% less and pasture with about 50% less area exposed to high relative deposition compared to forests. More detailed information for each MPC and ecosystem is found in Figure S5 in the Supplement.

25 Taken together, forests which cover large areas on each continent are most affected by the MPC deposition. In contrast, pasture areas are also widespread though they are less affected by the MPC deposition than croplands, which are mainly present in a few

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concentrated regions. However, these regions are nearly all close to or in the outflow regions of MPCs.

6.2 Population based impact

We now assess the impact of MPC aerosols on humans. For this surface densities are used instead of aerosol deposition fields, since airborne particles are potentially inhaled by humans and consequently cause health issues. Small aerosols are again represented by the $1.0\ \mu\text{m}$ aerosol tracers and for comparison and as an upper limit for efficiently inhalable aerosols we show also results for $10.0\ \mu\text{m}$ aerosols.

Figure 8 displays surface densities in the upper panels for the NS_{act} tracers, on the left for the small (also shown in the middle panel in Fig. 3 with another color-scale), on the right for the large aerosols. In the surface layer the differences between NS_{act} and NS_{inact} are almost negligible, especially in regions close to the MPCs, thus we focus our discussion on the NS_{act} aerosols tracers. The small aerosol tracers are commonly more widely dispersed due to the faster gravitational settling of large aerosols and subsequent removal out of the surface layer. MPCs are easily detectable by the red dots in the figure, even when the surface densities are convolved with the geographical population distribution (lower panels in Fig. 8). However, in addition two large domains are more accentuated: India and (East) China. Together these two countries host about one third of the global human population and thus the high population densities ensure that the highest numbers in the convolved variable of surface density and population distribution are found there. Other densely populated regions like Europe, Japan, or Eastern US show similar surface densities but lower convolved numbers.

To further investigate and quantify how many people are affected by each MPC, we add up the number of people in the area with a mass threshold exceedance (A_Y). In Fig. 9 this is shown for the $1.0\ \mu\text{m}$ and $10.0\ \mu\text{m}$ aerosol tracers and a mass threshold Y of $10\ \text{ngm}^{-3}$. Highest values are found independently of aerosol size or solubility in Delhi, Beijing, Tianjin, Karachi, and Shanghai with more than 1.3 billion people being exposed to more than $10\ \text{ngm}^{-3}$ in and around Delhi. For other MPCs like Sydney,

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Melbourne, Santiago, or Kinshasa the lowest population numbers are found, simply due to the relatively sparse population density around these cities.

In conclusion, the area A_γ is a measure of the distribution of aerosol mass in the surface layer, especially of the dense part of the outflow plume. However, A_γ does not seem to determine how many people are exposed to a certain aerosol mass. Instead, this is more dependent on the population in and around an MPC. This is also underlined when computing the coefficients of determination to see how much of the variance of the exposed population number is explicable by the area itself. For A_{10} we find only weak correlations for small ($R^2 < 0.2$) and large ($R^2 < 0.06$) aerosol tracers, and also by changing the threshold value to 1 ngm^{-3} and 100 ngm^{-3} we ascertain no substantially higher correlations.

7 Summary and conclusions

This study is an extension of Lawrence et al. (2007) to understand and compare the dispersion and deposition of air pollutants from urban centers. The primary focus is on generic aerosol tracers of different size and solubility with various atmospheric residence times. We show that high relative contributions of pollutant levels around the source are found in regions where pollutants have short atmospheric residence times which are constrained by meteorological conditions, especially by scavenging in and below clouds. Export from the cities, either by vertical mixing or low level long-range transport is determined by the stability of the atmosphere. High stability favors low level export and is most dominant in cold regions, whereas the vertical export is strongest in regions with high lofting potential, i.e. high instability. Furthermore, the export to higher altitudes is dependent on the moisture in the atmosphere and therefore cloud formation and subsequent wet removal processes, which leads to the strongest export in temperate dry winter regions and not in tropical regions, in contrast to insoluble gas phase tracers. The export at low levels is similar for aerosol and gas phase tracers but substantially different for transport above 5 km, with gas phase tracer concentrations

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up to ten times higher than the aerosol tracer concentrations due to the effective removal of aerosols in clouds. Moreover, the dispersion of generic gas phase tracers from MPCs has been compared for two models, one with online calculated meteorology and one with semi-offline calculated meteorology as well as different physical parametrizations. These differences account for only about 25 % of the variance in the results, thus a high accordance is shown in vertical and long-range transport of the tracers between the two models.

Aerosol deposition fields convolved with geographical distributions of agricultural data and a metric to account for regions with high relative deposition are used to determine the impact of MPCs on ecosystems. We assess the area in which more than 1 % and 5 % of the emissions are deposited. On average larger aerosols exceed these deposition fractions over larger areas with less variation between single MPCs. The areas vary more for smaller aerosols due to their longer lifetime and thus wider dispersion. This effect is strongest for MPCs in arid regions where aerosols have the longest atmospheric residence time while regions characterized by high precipitation or unfavorable geographic conditions reveal larger areas with high relative deposition. Most of this deposition occurs over land regions except for some MPCs like Rome, Athens, Jakarta, and Manila, with more deposition onto water surfaces. We further find that the impact on forests is generally largest while it is smallest on pasture areas. Similarly, we use an area of mass threshold exceedance folded with population to assess the impact on humans and find that high population densities favor high human exposure, more so than the size of the area containing high surface mixing ratios.

All our results are solely based on model results and thus dependent on model uncertainties. The EMAC model was extensively evaluated for trace gas and aerosol distributions in many studies. A further improvement would be to represent the aerosol tracers better with properties, size and solubility, which can evolve in time and space and interact with other atmospheric constituents including clouds and atmospheric trace gases. Moreover, the coarse model resolution is a key issue with respect to the sub-grid scale processes like the introduction of the emissions, vertical mixing, e.g. out of

the boundary layer, convection, or scavenging as these physical processes are of major importance for the distribution of tracers in the atmosphere. Higher vertical and horizontal resolution can be provided by regional scale models with the limitation of many simulations being necessary to cover all the major population centers used in this study.

5 Nevertheless, as urban centers will become even more important in the future by serving as habitats for more people than at present, the research activities which have been accomplished so far, e.g. projects like MEGAPOLI (<http://www.megapoli.info/>, Baklanov et al., 2010), CityZEN (<http://www.cityzen-project.eu/>) and MILAGRO (<http://www.eol.ucar.edu/projects/milagro/>, Molina et al., 2010), should rather be seen as first
10 initiatives which scratched the surface and provided initial results, but also left several questions open or created new question.

Appendix A

Regression lines

In Fig. 4 regression lines are drawn. These regression lines are based on the following equations. x-values are representatives of gas phase results, y-values of aerosol
15 results.

For E_{UT} :

$$- NS_{inact} 1.0 \mu m: y = 0.1392x - 0.4996$$

$$- NS_{act} 1.0 \mu m: y = 0.06542x + 0.79841$$

20 $- NS_{inact} 10.0 \mu m: y = 0.1211x - 0.8125$

$$- NS_{act} 10.0 \mu m: y = 0.06005x + 0.31090$$

For $ELR_{1 km}$:

$$- NS_{inact} 1.0 \mu m: y = 1.443x + 3.592$$

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- $NS_{act} 1.0 \mu m: y = 1.245x + 1.1950$
- $NS_{inact} 10.0 \mu m: y = 1.023x + 2.885$
- $NS_{act} 10.0 \mu m: y = 0.8722x + 1.8004$

For A_{10} :

- $NS_{inact} 1.0 \mu m: y = 1.352x - 0.6669$
- $NS_{act} 1.0 \mu m: y = 0.9642x - 0.4251$
- $NS_{inact} 10.0 \mu m: y = 0.3325x + 0.1759$
- $NS_{act} 10.0 \mu m: y = 0.2830x + 0.1589$

Supplementary material related to this article is available online at:

<http://www.atmos-chem-phys-discuss.net/12/25389/2012/acpd-12-25389-2012-supplement.pdf>.

Acknowledgements. The authors would like to thank all EMAC developers and modelers for providing code and helpful discussions. We further thank S. Borrmann for valuable advice and our MEGAPOLI partners for fruitful discussions. D. Kunkel acknowledges funding from the International Max Planck Research School. The open-source software packages Yorick (<http://yorick.sourceforge.net/>) and R (R Development Core Team, 2010) have been used for the analysis and graphics.

The service charges for this open access publication have been covered by the Max Planck Society.

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Table 1. Name, location, and climate class of all MPCs used as urban point sources. Longitude and latitude correspond to the model grid box with tracer emissions. DRC = Democratic Rep. of Congo, RC = Rep. of Congo, PRC = People's Rep. of China, ROK = Rep. of Korea. Climate classes from Peel et al. (2007): Am = tropical, monsoon, Aw = tropical, savannah, BWh = arid, desert, hot, BSk = arid, steppe, cold, Csa = temperate, dry, hot summer, Csb = temperate, dry, warm summer, Cwa = temperate, dry winter, hot summer, Cwb = temperate, dry winter, warm summer, Cfa = temperate, no dry season, hot summer, Cfb = temperate, no dry season, warm summer, Dfa = cold, no dry season, hot summer, Dfb = cold, no dry season, warm summer, Dwa = cold, dry winter, hot summer.

MPC	Lon	Lat	Climate class
Europe			
London, UK	−0.08	51.50	Cfb
Paris, France	2.33	48.83	Cfb
Rhine-Ruhr, Germany	7.00	51.50	Cfb
Moscow, Russia	37.62	55.75	Dfb
Po Valley, Italy	11.00	45.00	Cfa
Rome, Italy	12.30	41.50	Csa
Athens, Greece	23.40	37.60	Csa
Istanbul, Turkey	29.00	41.00	Csa
Africa			
Cairo, Egypt	31.25	30.03	BWh
Khartoum, Sudan	32.50	15.50	BWh
Nairobi, Kenya	36.80	−1.27	Cwb
Kinshasa, DRC, Brazzaville, RC	15.30	−4.33	Aw
Lagos, Nigeria	3.45	6.42	Aw
Johannesburg, South Africa	28.93	−26.52	Cwb
West Asia			
Baghdad, Iraq	44.70	33.33	BWh
Teheran, Iran	51.42	35.68	BWh

Table 1. Continued.

MPC	Lon	Lat	Climate class
South Asia			
Karachi, Pakistan	67.07	24.85	BWh
Mumbai, India	72.90	18.92	Aw
Dehli, India	77.20	28.63	Cwa
Kolkata, India	88.40	22.57	Aw
Dhaka, Bangladesh	90.43	23.72	Aw
East Asia			
Szechuan Basin, PRC	105.00	30.00	Cwa
Hong Kong, PRC	114.20	22.20	Cwa
Beijing, PRC	116.40	39.92	Dwa
Tianjin, PRC	117.15	39.15	Dwa
Shanghai, PRC	121.48	34.25	Cfa
Seoul, ROK	127.10	37.52	Cwa
Osaka, Japan	135.45	34.65	Cfa
Tokyo, Japan	138.75	35.67	Cfa
South-East Asia			
Bangkok, Thailand	100.50	13.70	Aw
Manila, Philippines	121.50	14.50	Aw
Jakarta, Indonesia	106.83	-6.20	Am
Oceania			
Melbourne, Australia	145.00	-38.00	Cfb
Sydney, Australia	151.20	-33.87	Cfa
North America			
Los Angeles, USA	-118.28	34.05	BSk
Chicago, USA	-87.67	42.00	Dfa
Houston, USA	-95.30	29.75	Cfa
Atlanta, USA	-84.50	33.00	Cfa
New York, USA	-74.02	40.72	Cfa

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Table 1. Continued.

MPC	Lon	Lat	Climate class
Mexico City, Mexico	−99.02	19.43	Cwb
South America			
Lima, Peru	−77.03	−12.03	BWk
Bogotá, Columbia	−74.20	4.40	Cfb
Santiago, Chile	−70.67	−33.50	Csb
Buenos Aires, Argentina	−58.33	−34.58	Cfa
São Paulo, Brazil	−46.62	−23.63	Cfa
Rio de Janeiro, Brazil	−43.20	−22.92	Aw

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Table 2. Brief description of symbols used in this study.

Symbol	Description
NS_{act}	Aerosol tracers which can be scavenged by nucleation
NS_{inact}	Aerosol tracers which cannot be scavenged by nucleation
$RCL_{0.5km}$	Fraction of total tracer mass retained in a circular volume of height 0.5 km and a radius of 500 km
$ELR_{1.0km}$	Fraction of total tracer mass exported beyond 1000 km and below an altitude of 1.0 km
E_{UT}	Fraction of total tracer mass above 5 km altitude
A_Y	Total surface area (in $10^6 km^2$) of model grid cells with a tracer mass larger than $Y = 10 ng m^{-3}$
DEP_{accu}	Annual mean deposition mass flux in $kg m^{-2} s^{-1}$
EM_{total}	Annual global mean emission flux in $kg m^{-2} s^{-1}$
D_X	Emission normalized deposition fraction, i.e. the fraction of tracer DEP_{accu} in each grid cell which is larger than X per cent of the corresponding EM_{total} , where $X = 1$ or 5 %
A	Tropical climate class
B	Arid climate class
Cs	Temperate dry summer climate class
Cw	Temperate dry winter climate class
Cf	Temperate no dry season climate class
D	Cold climate class

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Table 3. Differences between EMAC and MATCH-MPIC annual values and corresponding ranks for $ELR_{1\text{km}}$, E_{UT} , and A_{10} for the 36 MPCs: \bar{x} , mean differences; x_d , corresponding mean absolute deviations, with $\bar{x} = \sum_{i=1}^N x_i / N$, $x_d = \sum_{i=1}^N |x'_i| / N$, $x'_i = x_i - \bar{x}$, x_i metric value of an MPC, and N total number of MPCs.

	$ELR_{1\text{km}}$		E_{UT}		A_{10}	
	%	Rank	%	Rank	$\times 10^6 \text{km}^2$	Rank
\bar{x}	-2.32	0	1.05	0	-0.91	0
x_d	2.88	4.89	6.41	3.94	0.99	4.06

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Table 4. Pearson correlation coefficients for correlations between E_{UT} , $ELR_{1\text{ km}}$, and $RCL_{0.5\text{ km}}$, climate class resolved and for all MPCs (row All) for small ($1.0\ \mu\text{m}$) and large ($10.0\ \mu\text{m}$) aerosol tracers.

	E_{UT} : $ELR_{1\text{ km}}$	$ELR_{1\text{ km}}$: $RCL_{0.5\text{ km}}$	E_{UT} : $RCL_{0.5\text{ km}}$	E_{UT} : $ELR_{1\text{ km}}$	$ELR_{1\text{ km}}$: $RCL_{0.5\text{ km}}$	E_{UT} : $RCL_{0.5\text{ km}}$
$1.0\ \mu\text{m}$	NS_{inact}			NS_{act}		
A	0.00	-0.26	-0.04	0.43	-0.31	-0.30
B	-0.97	0.43	-0.43	-0.76	-0.12	-0.36
Cf	-0.75	-0.38	-0.18	-0.61	-0.43	-0.31
Cs	-0.90	0.72	-0.88	-0.96	0.73	-0.84
Cw	-0.84	0.13	-0.60	-0.58	0.02	-0.70
D	-0.98	-0.92	0.84	-0.98	-0.95	0.86
All	-0.77	-0.19	-0.27	-0.51	-0.28	-0.49
$10.0\ \mu\text{m}$	NS_{inact}			NS_{act}		
A	0.05	-0.31	-0.05	0.15	-0.38	-0.21
B	-0.92	0.39	-0.64	-0.59	-0.17	-0.63
Cf	-0.75	0.08	-0.53	-0.66	-0.09	-0.52
Cs	-0.96	0.89	-0.95	-0.97	0.90	-0.95
Cw	-0.70	0.22	-0.75	-0.49	0.09	-0.78
D	-0.97	0.45	-0.66	-0.92	0.01	-0.38
All	-0.74	0.13	-0.58	-0.50	-0.07	-0.66

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Table 5. Average area (in 10^3 km^2) covered with high relative deposition, D_1 and D_5 .

		Cropland	Pasture	Forest	Population
D_1					
1.0 μm	NS _{act}	20.26	12.61	33.21	25.85
	NS _{inact}	16.37	10.74	31.23	20.83
10.0 μm	NS _{act}	25.29	15.87	32.26	29.74
	NS _{inact}	24.48	15.12	24.56	28.66
D_5					
1.0 μm	NS _{act}	2.28	1.51	4.25	5.23
	NS _{inact}	1.88	1.25	3.28	4.03
10.0 μm	NS _{act}	5.29	3.14	6.54	9.10
	NS _{inact}	4.69	2.60	5.85	8.54

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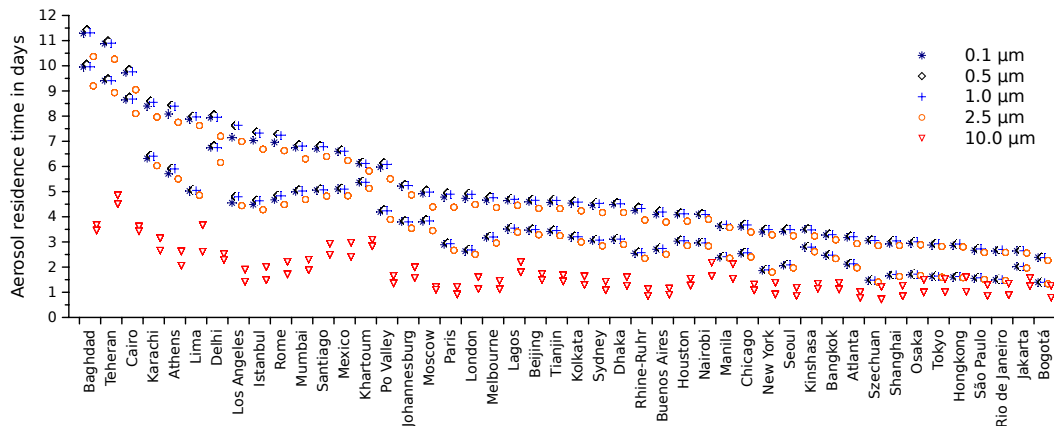


Fig. 1. Aerosol residence times for all MPCs and all aerosol tracers. The upper value is determined by the NS_{inact} tracers, the lower value by the NS_{act} tracers.

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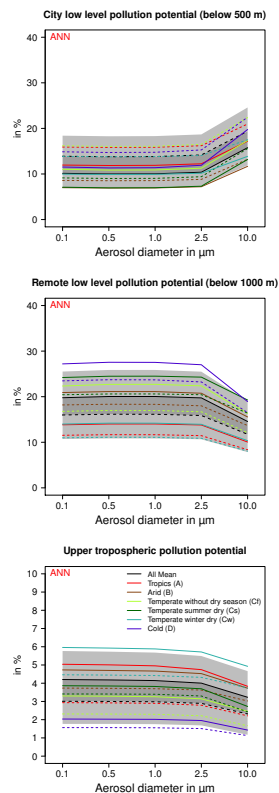


Fig. 2. Annual averages of $RCL_{0.5\text{km}}$ (upper panel), $ELR_{1\text{km}}$ (middle panel), and E_{UT} (bottom panel). Dark gray shaded areas represent the area between the total means (black lines) of NS_{act} (dashed) and NS_{inact} (solid) MPC tracers. Light gray shaded areas show one standard deviation, plotted single sided for NS_{act} and NS_{inact} mean values. Colored lines represent the mean values for different climate classes: tropical (red, A), arid (brown, B), temperate no dry season (light green, Cf), temperate dry summer (dark green, Cs), temperate dry winter (light blue, Cw) and cold (blue, D).

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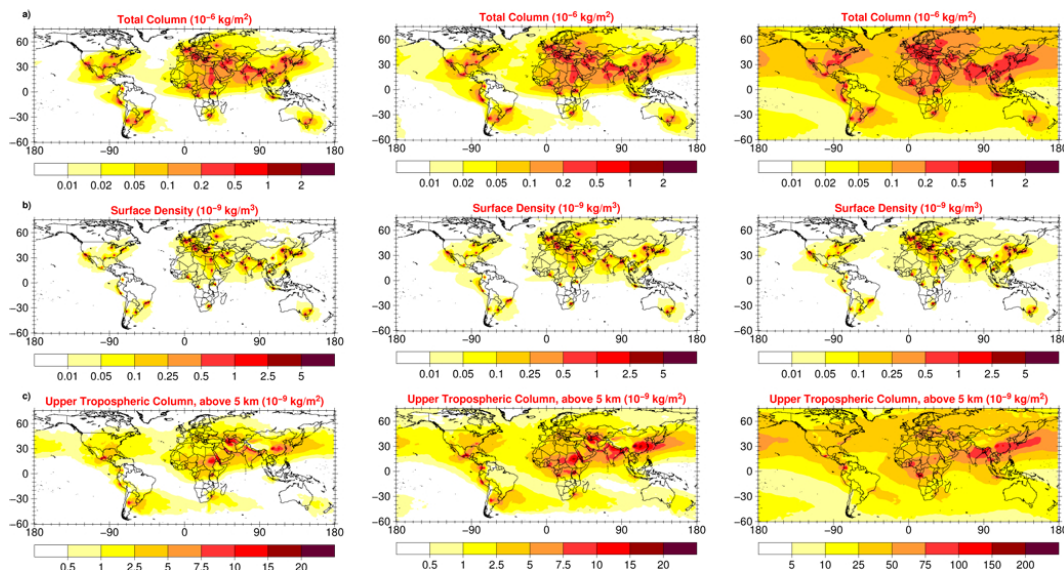


Fig. 3. Total column densities (top panel), surface densities (middle panel), and upper tropospheric column densities (bottom panel) for the sum of all 46 MPC tracer sources from the EMAC simulations. Left and central columns show the NS_{inact} and NS_{act} $1.0 \mu\text{m}$ aerosol tracers and the right column the gas phase tracers with a 10d decay lifetime. Units are $10^{-6} \text{ kg m}^{-2}$ for total column densities, $10^{-9} \text{ kg m}^{-3}$ for the surface densities, and $10^{-9} \text{ kg m}^{-2}$ for the upper tropospheric column densities. Please note the different color-scaling, a factor of ten, for the upper tropospheric column densities between the aerosol and gas phase tracers.

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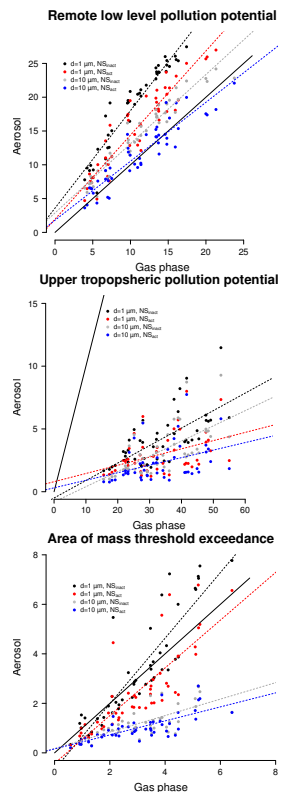


Fig. 4. Comparison between gas phase and aerosol pollution potentials: $ELR_{1\text{ km}}$ (top panel, in %), E_{UT} (middle panel, in %), and A_{10} (bottom panel, in 10^6 km^2). Colored dots represent different aerosol properties: black = $NS_{\text{inact}} 1.0\ \mu\text{m}$, red = $NS_{\text{act}} 1.0\ \mu\text{m}$, gray = $NS_{\text{inact}} 10.0\ \mu\text{m}$, and blue = $NS_{\text{act}} 10.0\ \mu\text{m}$. These aerosol tracers are considered to cover the aerosol spectrum of this study and other tracers are neglected to keep lucidity. The solid black lines represent the bisecting line, the colored dashed lines the regression lines (see equations in the Appendix).

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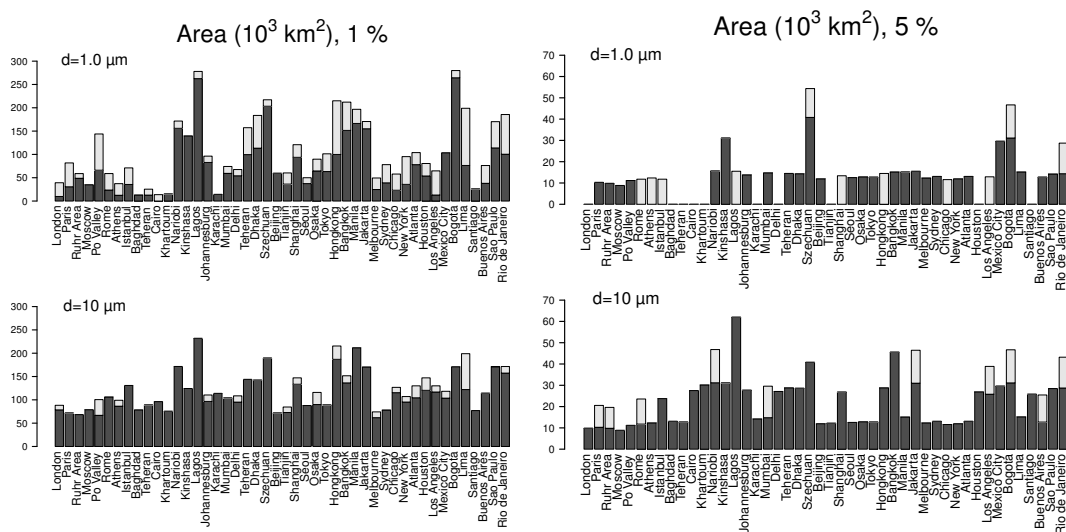


Fig. 5. Total area covered with more than 1 % (left side) and 5 % (right side) of the global mean annual emission. Top panels display the 1.0 μm, bottom panels the 10.0 μm aerosol tracers. The dark gray shaded bars show the minimum area covered by either the NS_{act} or NS_{inact} tracers, the light gray shaded bars on top the additional area covered by the tracers with the other nucleation scavenging state. Display order of the MPCs is by longitude, the area shown is in units of 10³ km².

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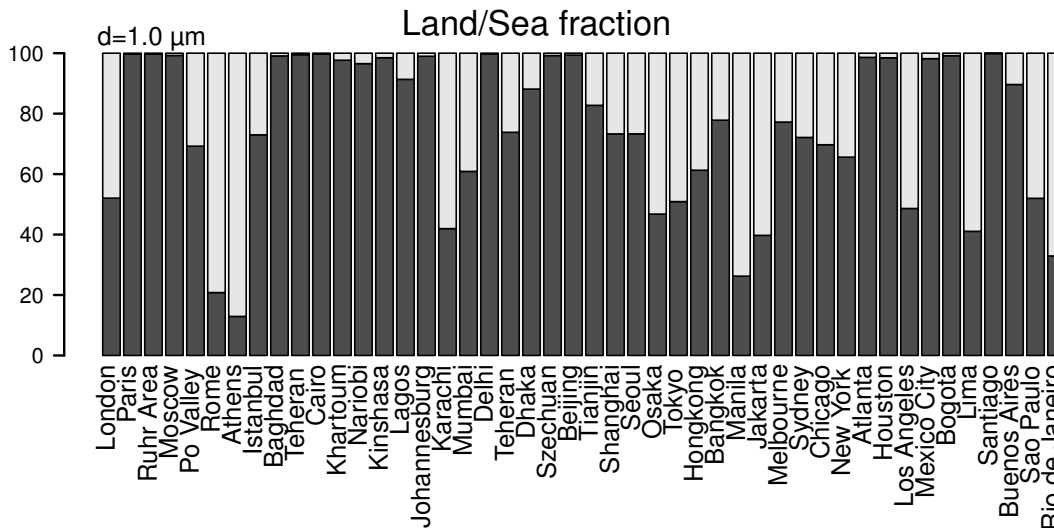


Fig. 6. Example for land-sea fractionation in % of D_1 for the NS_{act} $1.0 \mu\text{m}$ aerosol tracers. Dark gray bars represent the land fractions, light gray bars the sea fractions.

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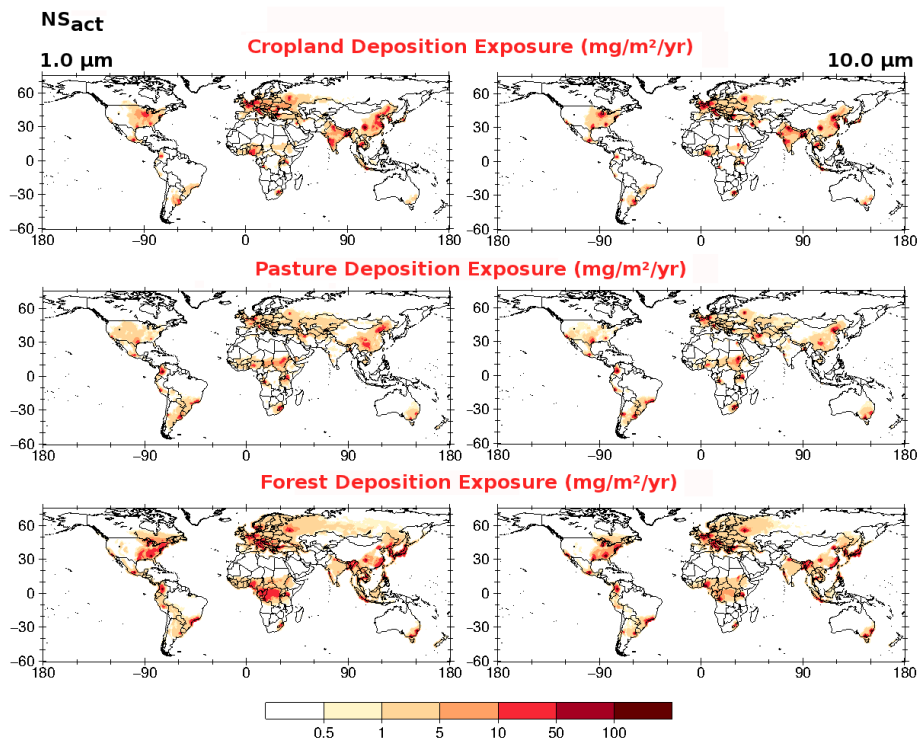


Fig. 7. Folded maps of the sum of all MPCs' annual accumulated deposition fields (in $\text{mg}/\text{m}^2/\text{yr}$) and the relative fractions of the three ecosystems (see also Fig. S1 in the Supplement). On the left side for the $\text{NS}_{\text{act}} 1.0 \mu\text{m}$, on the right side for the $\text{NS}_{\text{act}} 10.0 \mu\text{m}$ aerosol tracers.

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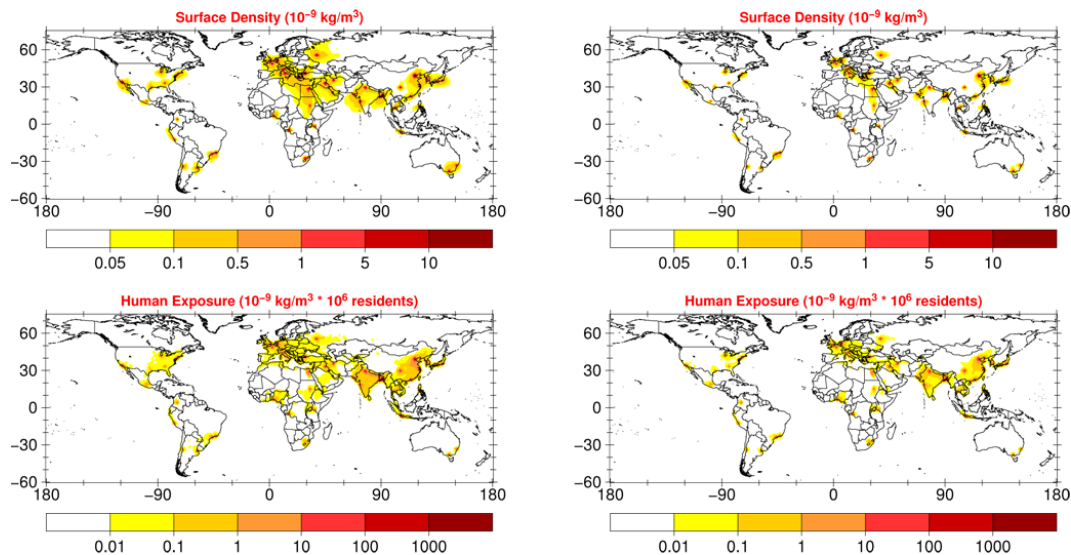


Fig. 8. Sum of all MPC surface densities in $10^{-9} \text{ kg m}^{-3}$ alone (upper panels) and convolved with the geographical population distribution (lower panels). On the left side for the $\text{NS}_{\text{act}} 1.0 \mu\text{m}$, on the right side for the $\text{NS}_{\text{act}} 10.0 \mu\text{m}$ aerosol tracers.

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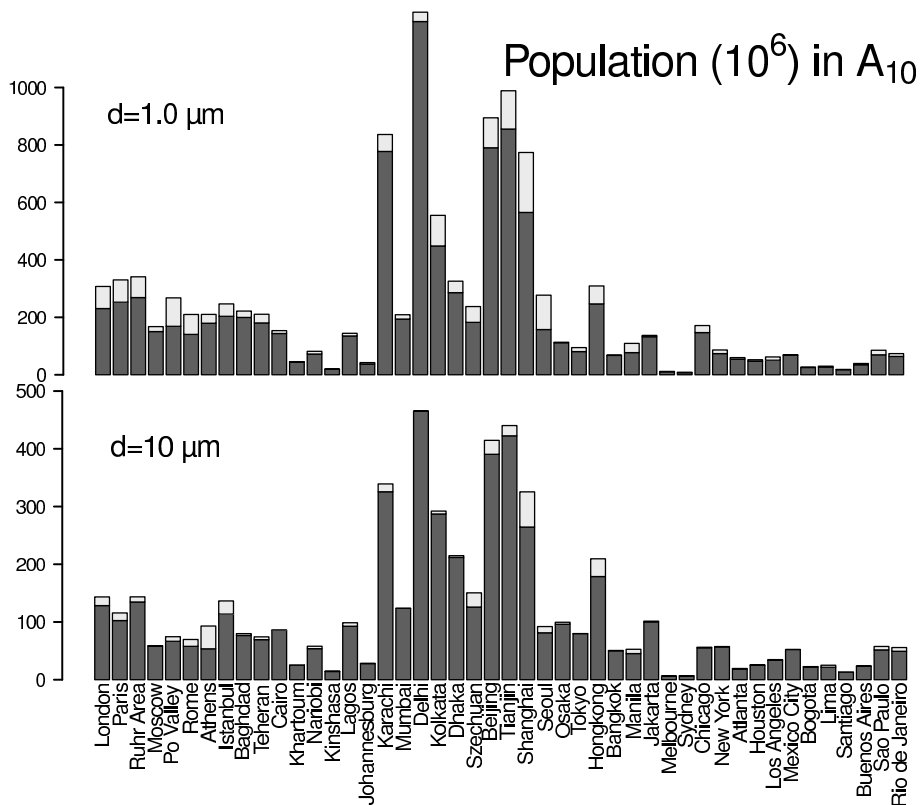


Fig. 9. Population in millions which reside in A_{10} for $1.0\mu\text{m}$ (top panels) and $10.0\mu\text{m}$ (bottom panels) aerosol tracers. Dark and light gray bars are defined as before.

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