

## Response to Referee #1

We thank referee #1 for the comments on our paper. In the following, we repeat these comments in italics, and provide our responses in normal font.

- *Unfortunately, there is no measurement data used at all in the study,*

We fully agree to the importance of measurement data, and in fact, most of our studies do involve measurement data. In particular, our dispersion model FLEXPART has been validated with many different data sets, ranging from tracer experiments, to station measurements, aircraft measurements, etc. Notice that FLEXPART was quite successfully used in previous studies of megacity emissions (e.g., Stohl et al., 2003; Pisso et al., 2012). However, measurement data would not help for this study at all. First of all, we are considering all megacities in the world, and how should we collect comparable measurement data from all these cities in a systematic way? These data simply do not even exist. On the other hand, using measurement data from one or two megacities would not really make a difference, and would rather look artificial in the context of this paper. Even more, we study dispersion characteristics of emission plumes across many scales, and such plumes cannot easily be isolated in measurement data from emissions from larger regions with sufficient accuracy to validate aspects of this paper.

- *and the results are presented as if simulations were reality.*

It was not our intention to present the simulations as if they were reality, and we believe this has not been done in the paper. However, we made a further effort to better clarify that we are dealing with modeling simulations and two artificial tracers based on BC emissions, see also the response to the comment below.

- *The results for the passive tracer (BC<sub>tr</sub>) and for the tracer with deposition turned on (BC<sub>dp</sub>) are presented as if they were separate species that actually existed – for example as if we were looking at data for CO and CH<sub>4</sub> – whereas in fact they are both numerical representations of different things.*

We treat these tracers as different species, but computational species. There is an extremely large range of pollutants (both gases and aerosols) emitted by megacities and it would be impossible to compare megacity dispersion characteristics for all of them. This reviewer would obviously have liked to have several “real” chemical species to be treated in detail and with fewer metrics to be studied (see next comments). This would certainly have been a possibility, but we took exactly the opposite approach: We concentrated on two “artificial” tracers, intended to be representative of groups of species with roughly comparable characteristics with respect to removal processes and lifetimes. This allowed us to derive several different metrics addressing different aspects of dispersion of megacity air pollutants.

- *The 4 different sub-projects are then described in shallow terms with a considerable amount of vague discussion which regrettably leads me to recommend against publication.*

We disagree with the reviewer on this aspect. We consider it the major strength of our paper that it addresses several different “sub-projects” in a coherent way using a consistent modeling framework and for all the megacities. Certainly, given length limitations, compromises have to be made on how deep individual issues can be discussed, but the point here is to make megacities comparable with each other and rank them in terms of several quite different aspects. This hasn’t been done before to our knowledge.

- *Fig. 2: Wouldn’t a plot by latitudinal bands be clearer to interpret? Also, maybe some bar charts comparing megacity emissions with the other main categories would help situate the significance of the study.*

We think that the figure is quite straightforward to interpret. The significance of the overall emissions from megacities relative to the total emission is evident from figure 2, there is not a specific reason in this study to further distinguish the total emissions in main categories.

- *The transition from hydrophobic to hydrophilic is a crucial part of the deposition process for aerosols. This should be discussed in greater detail, and possibly some attempt made at accounting for it in the analysis.*

This is indeed important for BC, but it is not important for all aerosols. There are many ways how aerosols behave in the atmosphere, including formation of secondary species, and it is virtually impossible to account for all of these, even in a simplified way, without requiring many more tracers. We wanted to keep things simple and assigning constant properties to our aerosol “species” seemed the most logical way to do this.

- *The separation of lifetimes into logarithmic and linear seems suspect - are there references that serve as a precedent, or could you expand on the explanation as well as the justification for doing this?*

We do not have any specific references for this. We simply considered that knowing that an exponential decay fits reasonably well the ratio of total mass content,  $BC_{dp}/BC_{tr}$ , as a function of time, then taking an exponential fit on a linear scale ( $T_{li}$ ) should minimize the influence of the smaller values while a linear fit in a logarithmic scale ( $T_{lo}$ ) weight all data in a similar manner. Therefore  $T_{li}$  should give an idea of the

time scale needed for the ratio to be a negligible fraction of its initial value. This is clearly more influenced by what happens close to the source and it is remarkably different between sources located in arid and wet regions. The linear fit of logarithmically transformed data ( $T_{10}$ ) gives instead a view of how the scalar decays to a fraction of its current value, but since the scalar is quite quickly spread over large regions (and the age classes extend up to 48 days) it is less related to the behavior close to the source, while it is more connected to the average (modeled) decay rate of a scalar dispersed over the global atmosphere. We could have used different methods to define a local decay time scale such as imposing an arbitrary threshold to the spatial region of interest or setting an arbitrary threshold to discard lower value of the ratio of BCdp/Bctr. However, we think that the method used is less arbitrary.

- *pg64-In24: Similar behavior for the lower atmosphere + within 1000km, not beyond. Is this an example of confusing BCtr and BCdp with reality? The part of BCdp that has survived 10 days behaves like BCtr – almost by definition? Although does it continue to? It shouldn't. Along these lines, what does Fig 6 say that is noteworthy?*

It seems that the reviewer misunderstood the discussion. The decay discussed in that section of the manuscript is an exact exponential decay with a ten days time scale (as originally simulated by L07) and it is not related in any way to deposition processes. In fact the scalar subject to this particular decay is afterwards specifically renamed BC10 (page 26365 line 4). We believe that this is clearly explained in the text, and BCdp is never mentioned in that specific discussion. Since this exponential decay studied by L07 is homogeneous in space, and depends only on the age of the scalar (contrary to the deposition processes influencing BCdp), it acts in the same way below or above 1km elevation, and within or outside a 1000km radius (we remind that the discussion concerned the measure ELR1km originally proposed by L07). As a consequence we commented in the paper (page 64, line 23-27): “that all tracers with a decay time scale larger than about one week will behave similarly to a purely passive tracer without decay. This simply reflects the partitioning of tracer between the lowest 1 km and the rest of the atmosphere, as basically all of the tracer will be exported beyond 1000km after a week.”

Moreover, specifically about the questions/answer of the reviewer “*The part of BCdp that has survived 10 days behaves like BCtr – almost by definition? Although does it continue to? It shouldn't.*” We note that as shown in figure 8(a-b) the part of BCdp that has survived 10days does not behave like BCtr, we note also that BCdp does not in general behave like BC10 as shown in table 2 (page84), and we conclude remarking again that BCdp was never mentioned in the discussion on page 64-line24.

Although we believe that the discussion in the paper is understandable as it is, to avoid any misunderstanding we have further clarified that L07 report values of ELR1km for a scalar with an exact exponential decay with a ten days time scale.

- *What is the purpose of the distance calculation? There is a lot of data in the graphs, but it is not clear what the significance of it is.*

The distance (as a function of time) gives an idea of how far (and fast) the emitted mass travels away from the source. This is a very significant measure for the early age classes since the geometrical factor linked to the source latitude (see page 62) does not play any significant role in this case. We think that the transport distance and elevation together characterize well the advection of the plumes and are

useful to understand the subsequent results in term of the physical processes in the atmosphere. They are used for example in the discussions at page 66, line 15-29 and page 67, line 16-27.

- *The discussion of deposition is glossed over but is crucial to the results. This should be described in greater detail, and the explanation should include a consideration of the uncertainties. Small changes in the simulation of BC<sub>dp</sub> would presumably make large differences in the results. This could even be tested in the model, with for example simulations evaluating the role of wet and dry deposition as well as convection for different megacities. At the moment, there is just a vague description of Jakarta as being wetter and Lima being drier, for example.*

The reviewer correctly points out that changes in the deposition model would modify the results. However, we discussed the effects of deposition in detail for every megacity and we described the deposition model used. We think that a test of the effects of all the model variables influencing the deposition is outside the scope of the present work; here we compare consistently for every megacity two artificial scalars and we calculate various metrics. It would make the paper extremely long (and difficult to read) to also consider the effects of the various variables controlling the deposition in a consistent way.

- *pg62-ln5 There is a fair amount of work on dispersion that could add precision to these vague comments instead of saying that latitude circles become larger near the equator.*

It is not clear to us what the relevance of this comment is. The phrase “latitude circles become larger near the equator” on line 5, page 62 is part of an articulate sentence, reported below:

“The reason for this latitude-dependent behavior is influenced by three factors: (1) the plumes are transported preferentially zonally before spreading meridionally due to latitudinal transport barriers, and the length of a circle of latitude is largest at the equator, (2) cities at lower latitudes have a larger asymptotic distance possible for a uniform scalar distribution on the hemisphere and (3) the tropics are characterized by lower near-surface wind speeds but a high frequency of deep convection, so that the dispersion is at first slower than for higher-latitude cities but after a few days significant amounts of tracer in the tropics are transported by deep convection to the upper troposphere, where winds are faster. This is illustrated in Fig. 5a, c, which shows that tropical cities and particularly south-east Asian cities (e.g. Manila) are associated with a very fast increase of the plume average elevation.”

If the reviewer has any specific suggestion about literature that would help us to better explain the observed results, we would be happy to follow his suggestions and explicitly acknowledge his useful comment.

- *pg68-ln8: What do these other studies show? What is the relationship to the present work?*

The studies referenced at page 68, line 8 are: Stohl (2006), Hirdman et al. (2010) and Stohl and Sodemann (2010).

What these studies show in relation to the present work has been explained not far from page 68-line 8. More precisely it is explained at page 68, lines 15-19, for Stohl (2006); at page 71, lines 5-10, for Hirdman et al. (2010); at page 73, lines 14-19, for Stohl and Sodemann (2010).

- *The exposure study would not pass as a standalone paper, and seems to be squeezed in without much justification.*

We agree, the exposure study would not pass as a standalone paper, but we never intended to write such a paper. As said before, we consider the diversity of metrics (of which the exposure metric is related to human health) a strength, as this makes several aspects of megacity dispersion comparable. It is quite obvious that a given city can score quite differently compared to all other cities for the various metric considered.

#### **Technical Comments:**

- *Table 1 caption: refer lifetime calculations to Fig 3 and to text so reader knows which items in the table are input data, and which are derived parameters.*

We added the reference to figure 3.

- *64-10 define a. t. l.*

It was defined at page 56 line 10.

- *Check English usage for “noteworthy.” Further proofreading / editing is required, especially during the introduction. BC described as a “compound”*

We have re-checked the English.

#### **Further references not in the manuscript**

Pisso, I., Patra, P., Takigawa, M., Machida, T., Matsueda, H., and Sawa, Y.: Anthropogenic CO<sub>2</sub> flux constraints in the Tokyo Bay Area from Lagrangian diffusive backward trajectories and high resolution in situ measurements, Atmos. Chem. Phys. Discuss., 12, 10623-10649, doi:10.5194/acpd-12-10623-2012, 2012.