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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Urban source term estimation for mercury using a boundary-layer budget method" *by* Basil Denzler et al.

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Referee comment in bold, reply in plain text, *modified text for manuscript in italics*.

A revised and highlighted version of the manuscript is available in the supplementary material.

Denzler and coworkers present a top-down approach to estimate urban mercury emissions from ground-based measurements. Their approach is a nice and simple boundary-layer mass balance method applied during periods of temperature inversion and low wind speeds when the measured urban concentrations are

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Discussion paper

most sensitive to local emissions. Using two years of measurements, they calculate gaseous elemental mercury emissions in Zurich and compare it to those reported in the Swiss national emissions inventory. The authors also provide a convenient tool based on their approach to calculate gaseous elemental mercury emissions in other cities. It is important to quantify the anthropogenic emissions of mercury to understand its effects on the biogeochemical cycling of mercury and to build the knowledge needed for the success of regional and global efforts to lower the human health burden of mercury. Bottom-up emissions inventories are uncertain and need to be checked against top-down estimates, as has been done in this study. The top-down method described here will certainly be of much interest to readers of ACP who study mercury and also to those who are working to quantify emissions of similar pollutants. The study is scientifically sound, well written, and presents the relevant data supporting their conclusions.

We would like to thank the referee for the positive review and for recognizing the importance of the field of atmospheric mercury research and the need to further constrain bottom-up mercury inventories. Furthermore, we appreciate the constructive comments and are convinced that the quality of the manuscript has improved with the desired adjustments.

(i) The authors seem to have completely overlooked gas and particle-bound oxidized mercury. There is ample evidence that a significant fraction of mercury emissions are in these forms (e.g. Zhang et al., 2016). It is important that the authors discuss the effect of neglecting oxidized mercury on their emissions estimate and its comparison with estimates of (total) mercury emissions.

Referee 1 raises an important aspect of atmospheric mercury, which we so far have not discussed in the manuscript mainly due to the lack of data on swiss mercury emissions

in this regard. However, we see the need to inform the reader about our thoughts and assumption on oxidized mercury. We have thus introduced several parts addressing the topic of mercury speciation.

First of all the composition of atmospheric mercury has been addressed in the introduction (page 1, line 22).

Furthermore, a comment on the major point source in the model area has been made in the section of model parametrization. (page 5, line 2)

(ii) In section 4, the authors provide a tool in the form of a nomogram that can supposedly be adapted by nonspecialists to calculate emissions elsewhere. This is indeed useful, but I am concerned that the conditions under which this tool is broadly applicable (or not applicable) are not clearly laid out. I recommend that a more objective description of this be presented in this section. It could include for example a threshold for the meteorological parameters for which their method works, the general characteristics of the site that measures the urban background, what if the stacks of point sources are taller than 150 m, etc.

A section, discussing the limitation of the boundary-layer budget approach has been added, as has been recommended by the Referee 1. (page 11, line 17)

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Minor comments: Figure 2 shows the weekly and monthly variation of mercury concentrations and that of other species. The authors use this figure to demonstrate that mercury emissions are constant in time. However, the relatively high background concentration of mercury makes the variations in local concentrations seem small. It would be more insightful to subtract the background and then show how local mercury concentrations vary in time.

Momentarily, in Figure 2 we show a relative concentration for all the trace gases CO, CH4 and GEM. For a comparison of the three gases relative concentrations are necessary. A background subtraction does not change the variation. It only changes the scale of the y-axis. However, the proportions between CO, CH4 and GEM stay the same. We therefore argue to maintain the current representation, which has the advantage of clarity and best interoperability. Furthermore, as such we do not introduce any assumptions made regarding the GEM background concentrations into the graph.

It is not clear why the deposition, emissions from land and water, and oxidation of mercury can be neglected in the model. This needs to be better discussed with relevant citations.

Deposition, emissions from land and water, and oxidation of GEM are without a doubt important processes for the description of the atmospheric fate of mercury. The only reason we can neglect these processes is that they are relatively slow. Considering our small model area, the residence time within this box is short (1h for windspeed of 3 m/s) When comparing the fluxes produces by these processes within the small model area they are negligible in comparison to the strong advective flux. The description on this has been specified. (page 5, line 12)

In Section 2.1, a second measurement site on the outskirts of Zurich is mentioned. But those measurements are not discussed in the paper. I think they could provide valuable constraints on the spatial contrasts in mercury and help support their assumption that deposition and chemistry can be neglected.

A paragraph in the measurement results section has been added to the manuscript discussing the results from the second measurement location as suggested by the referee. (page 7, line 4)

Page 4, line 14: ". . .were identified by visual inspection of the data." Which data?

The sentence has been changed to: *Over the course of the measurement period nine episodes of day-night inversion were identified by visual inspection for the criteria of strong day/night inversion.* (page 4, line 17)

Page 3, line 30: "boundary-layer *top* is reached"

The line has been changed according to the suggestion.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2018-402/acp-2018-402-AC1-supplement.pdf

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