

Comments on the revised manuscript 'Surface-atmosphere fluxes of volatile organic compounds in Beijing' by Acton et al.

Reviewer: Erik Velasco

This reviewer appreciates the corrections and additions made to this second draft. The reading improved, and many of the questions pointed out in the original revision were properly answered. However, there are still four major concerns that need to be addressed before the manuscript can be considered for publication:

- A thorough analysis of the observed fluxes needs a complete description of the monitored footprint. The current description is still incomplete. An updated description of the neighborhood characteristics is needed. The information on the aerodynamic characteristics and land cover corresponds to assessments of 10-20 years ago. Are those assessments still valid considering the accelerated changes in Beijing's urban morphology in recent years?

Similarly, more information on the economic activities, vehicular traffic volume and composition, population density, number of trees and characteristics, etc. is needed to put in context the observed fluxes. This information will help to identify the error sources in the emission estimates of the gridded inventory.

- The manuscript reports fluxes collected during two field campaigns, a 28-day campaign in winter and a 40-day campaign in summer. However, after removing the averaging periods affected by lack of turbulence and instruments maintenance, as well as periods not meeting the stationarity criteria, only 25% and 70% of the data, respectively, were used for further analysis. This means that results are based on 7 and 27 net days of flux measurements. For the summer case, an almost one month of flux data might be enough for an initial assessment, but for the winter case, 7 days of data (for some VOC species even less) are not sufficient.

The authors may consider the relevance of including results of the winter campaign in this article. In an urban set up, like Beijing, seven days of flux data may represent nothing else than noise. In addition, many of the analysis performed in this study were limited to the flux data collected during the summer campaign.

- The impact of regional sources of VOCs in the observed fluxes deserves further analysis and discussion. The authors need to investigate under which synoptic conditions the loss of VOCs (i.e., negative fluxes) was observed. The use of back trajectories may help to this assessment.

Did the APHH project include measurements or modeling of the hydroxyl radical? Information on the levels of OH, NO and O₃ would make possible to estimate the residence time of each individual VOC species, and would support the proposed loss of VOCs. Also, it would help to evaluate the capacity of the eddy covariance system to measure fluxes of highly reactive VOCs. As indicated in the original revision, the time taken by an air parcel to reach the top of the tower could be higher than the time needed for the oxidation of such compounds, and thus the measured fluxes would not faithfully represent the emissions from the urban surface.

The OH concentration of 1×10^6 molecules cm⁻³ used to estimate the oxidation time of the assessed species is too low for a highly polluted atmosphere (see Shirley et al., 2006, for the case of Mexico City, www.atmos-chem-phys.net/6/2753/2006). A higher concentration would reduce

the lifetime of the VOCs, and therefore, the eddy covariance flux system mounted at 102 m of height might not be able of measuring fluxes of reactive VOCs.

- A final section providing conclusions and final remarks is missing. Scientific articles are expected to close with a short answer to the hypothesis to test. For example, is the eddy covariance a suitable method to measure urban fluxes of VOCs in Beijing? does the emission inventory is accurate for that particular sector of the city? What is needed to improve the inventory's accuracy?

Specific comments

39-40. Eddy covariance flux towers provide invaluable insight to evaluate the accuracy of gridded emission inventories, but they do not validate emission inventories.

40-49. The abstract does indicate flux measurements conducted on summer and winter, but results from the latter season are not included.

58. '... particulate matter of size smaller than 2.5 μm ...'.

62-63. This statement needs a reference.

78. I would say 'challenging' instead of 'difficult'.

96. 'Cityscape scale' is a confusing term. Eddy covariance flux towers measure fluxes at neighborhood or district scale.

97. PTR-TOF has not been defined in the text. Define acronyms the first time they are used in the manuscript.

98. Eddy covariance flux towers measures fluxes, do not estimate fluxes.

110. See previous comment about the use of flux towers to evaluate the accuracy of emission inventories

120. The description of the monitored sector of the city is still incomplete. More information on the economic activities, vehicular traffic volume and composition, population density, number of trees and characteristics, distribution and variability of building heights, etc., is needed to put in context the observed fluxes, and then be able to improve the emission estimates of the gridded emissions inventory.

121-123. This sentence is confusing. A 325-m tall tower was used as measuring platform, but the eddy covariance flux system was mounted at a height of 102 m.

124. Include the height above sea level of the location where the tower was located.

158. Explain the purpose of the system to evaluate the vertical gradient of VOC concentrations. I may be wrong, but the text does not indicate it had been used for the flux data analysis or discussion.

289. Do you mean that flux data were flagged if $u^* < 0.175 \text{ m s}^{-1}$?

340-345. Footprints at day and night are quite different. For a thorough assessment of the hourly emissions reported in the gridded emissions inventory is mandatory to evaluate the footprint across the diurnal cycle. Do not ask readers search for such information in another article. The background map provides little information. I cannot identify which sectors correspond to residential estates,

and which to commercial and institutional buildings, for instance. Also, I cannot identify which are primary and secondary roads. Can you mark the 3rd and 4th rings surrounding the location?

Table 1. Plots showing the diurnal variability of these VOC species would provide better insight in the context of the study. It does not matter, if a more detailed analysis is presented somewhere else.

422-425. This point deserves further discussion. I would say '... loss of VOCs by chemical reactions in the presence of fresh emitted NO ...', instead of '... deposition of VOCs ...'. Did the APHH project include measurements/modeling estimates of OH? Knowing the levels of OH, NO, O₃, etc. would make possible to estimate the residence time of each individual VOC species.

426-428. I do not understand this statement. Do you mean that people cook and clean less during the winter?

443. It would be useful to include in the text the variability observed in the fluxes (e.g. ± 1 standard deviation) for comparisons to fluxes reported in the literature.

Figure 5. I cannot observe any dashed line, and hardly observed the dotted ones.

637. More information about how the emission inventory was built is needed. It will help to understand the inventory's overestimations. Does the inventory estimate emissions based on activity data at city scale or use data for each individual grid? Which is the source of the emission factors used to estimate traffic emissions? Were the emission factors locally derived? How were the emission profiles determined? How were the evaporative emission estimated? Are there emission estimates for summer and winter?

338. No need to repeat the website hosting the emissions inventory. It was done in the introduction.

667-671. Check these two sentences. An overestimation of 3-4% is not equal to overestimate by a factor of 3-4.