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

This manuscript presents eddy covariance flux measurements of volatile organic compounds (VOCs) over a district of Beijing, China. The eddy covariance method has been widely used to measure urban fluxes of carbon dioxide (CO_2) during the last 1-2 decades, but only a handful of studies has measured fluxes of VOCs. In this context, the material presented in this manuscript gains relevance and should contribute to improve current knowledge on emission patterns of precursor species, as well as provides unique data to evaluate the accuracy of gridded emission inventories. However, before the manuscript can be considered for further revision and potential publication, the authors need to address the following issues. Details are provided in the specific comments.

- The authors need to explain the advantages and disadvantages of the eddy covariance method with respect to other methods to evaluate urban emissions. The conditions in which eddy covariance flux towers can be installed need a close revision. A full understanding of the eddy covariance assumptions is also needed.
- The facilities used in this study to deploy the eddy covariance system to measure fluxes of VOCs have been previously used to measure fluxes of CO_2 (e.g., Song et al., 2013; Liu et al., 2012; Song and Wang, 2012). A review of such studies is recommended.
- Previous articles describing VOC flux measurements by eddy covariance in Mexico City (Velasco et al., 2005, 2009) and Innsbruck (Karl et al., 2018) could be used as references to analyze and present the results of this study. The fluxes reported for Mexico City provide valuable information to compare the fluxes observed in Beijing considering that both are large cities of developing nations. Similarly, the way Karl et al. (2018) analyzed and presented a large number of VOC species could be followed.
- The discussion is biased to VOC studies in cities from UK. The manuscript will be strengthened if the findings are compared to results of previous studies conducted in China and other large cities. For example, the relationship between mixing ratios of selected VOC species has been widely used as a mean to evaluate the characteristics of urban emissions (e.g., Parrish et al., 2009; von Schneidemesser et al., 2010; Velasco et al., 2007).
- The authors need to demonstrate that the eddy covariance set up faithfully captures the entire range of energy-carrying eddies through inspection of the (co)spectra of the measured variables, and therefore is capable of measuring meaningful and representative turbulent fluxes. Similarly, a comprehensive description of the monitored footprint is needed to explain the observed fluxes.
- The writing needs some improvement.

Specific comments (page, line)

2,35. The abstract can be shorter. The first paragraph provides generic information. For example, the severe air pollution problem of Beijing is well known, and there is no need to specify in which institute the measurements were conducted.

2,51-52. In which aspects, magnitude or/and temporal distribution?

2, 68-69. Urban emissions have been widely evaluated through a number of direct and indirect methods. A brief discussion on the emissions data provided by those methods in comparison to the data obtained from eddy covariance flux towers will put in context the information presented in this study.

3, 72-74. Be more specific, vehicular exhaust no. 1 and vehicular exhaust no. 2 do not provide major information.

3, 78-79. One of the main goals of the quality monitoring is to evaluate the effectiveness of the control measures in place. Many cities around the world have made important investments to build air quality monitoring networks to measure and report hourly concentrations of key airborne pollutants, including particulate matter and speciated VOCs.

3, 93. Activity data?

3, 95-97. The application of the eddy covariance method to evaluate the accuracy of gridded emission inventories of VOCs in urban areas was done by first time in Mexico City (see Velasco et al., 2005, 2009).

3, 99-106. A statement of the hypothesis to test including the study objectives in context of the APHH project will help to analyze the flux observations and reach strong conclusions.

4, 100-119. Why was the AIP site selected to deploy an eddy covariance system? Does it meet the eddy covariance assumptions of homogeneity in terms of land cover and emissions distribution? A much more comprehensive description of the monitored site is needed. Information of the land cover distribution, urban morphology, roughness elements, trees characteristics, vehicular traffic, population density, etc. is needed. Please, consider the Local Climate Zone classification (see Stewart and Oke, 2012).

4, 117-118. Urban background in terms of ambient concentrations or urban fluxes?

4, 126. '… at a height of 102 m above ground level' was mentioned in the previous paragraph.

5,155. The legend should guide readers to understand the sketch of the sampling system presented in this figure without need of going back to the text.

5, 162-163. You may consider moving this table to the main article since it is the only material presented as Supplementary Information.

7, 200. The pioneering studies in Mexico City mentioned in a previous comment were done using the disjunct eddy covariance method and the original PTR-MS instrument.

7, 201. A detailed description of the flux data postprocessing? The eddy covariance method does not estimate fluxes, does measure fluxes.

7, 203-204. Turbulent fluxes are computed using instantaneous deviations or fluctuations of the vertical wind velocity and VOC mass density (e.g., mg m^{-3}).

7, 211-212. What about the time taken by the instrument to analyze the sample?

7, 228-229. Why did Squires et al. (2020) choose this method?

7, 240-241. Do you mean stationarity?

7, 242. Explain briefly the stationarity test.

7, 245-247. There are methodologies available to fill gaps in time series of turbulent fluxes measured by eddy covariance. However, depending on the study purposes it may not be necessary to fill such gaps. Consider that in urban environments VOC fluxes respond mainly to anthropogenic and not natural variables like in a forest. Human actions can be random at times (e.g., traffic jams), thus very low or very high fluxes are possible.

7, 238-249. Please, indicate the number of periods excluded for further analysis due to lack of stationarity and/or enough turbulence. How many averaging periods were affected by rain/snow and instrument maintenance?

8, 254-255. This information was already provided.

9, 263-264. This figure and the description of the dominant winds during the field observations are useless without a proper analysis of the land cover and distribution of the measured fluxes by wind sectors.

9, 265-266. This information was already provided.

10, 279-281. Is this true at nighttime and daytime? Atmospheric stability evolves during the diurnal cycle, thus significant changes in the footprint can be expected.

11, 292-296. This figure is incomplete. A color gradient to indicate the flux contribution is missing. No spatial scale was included neither a north indicator. The article is in English, but the map includes text in Chinese. The legend refers to a grid of cells, but no cells are included. Do these footprints correspond to nighttime or daytime?

11, 302-303. Define 'sawtooth cycle'. Although the paper is about fluxes, it will be good to provide a short description and a figure about their diurnal cycle.

12, 322-330. This paragraph should be part of the methodology. Do not repeat information.

13, 349-351. Turbulent fluxes measured by eddy covariance represent the VOC exchange within the observed footprint. Periods affected by advection were removed through the stationarity criterion, isn't it?

13, 351-352. This is unclear. Do you mean that turbulent fluxes cannot be measured during winter in Beijing, and therefore half of the study was useless?

14, 366. Table 2. A complete list of the 50 VOC species identified and evaluated in this study should be included in the Supplementary Material. You may use as reference how Karl et al. (2018) analyzed and presented an even larger list of VOC species.

14, 378-382. This section discusses fluxes no mixing ratios. Mixing ratios were discussed in section 3.3.

14, 380-382. London and Manchester are only two cities. There are many papers reporting mixing ratios of oxygenated VOCs for cities of China and the rest of the world.

15, 387. Vehicular traffic is a major source of isoprene in urban areas (e.g., Borbon et al., 2001; Jaimes-Palomera et al., 2016).

17, 400. Do you mean less than 1%? Figure 6 has not been cited yet in the text.

17, 403. Be more specific on the type of combustion sources (e.g., vehicular traffic, industry, biomass burning).

17, 409. A vehicular emission control program?

17, 412-415. A comparison against the fluxes reported by Velasco et al. (2005, 2009) for two locations of Mexico City will help to put in context the fluxes observed in Beijing with respect to other large city of a developing nation.

17, 416-418. How do these ratios between mixing ratios and fluxes compare to those reported in the literature for other cities?

17, 420-425. Propene is dominated by emissions from vehicular traffic in locations with heavy traffic (see Velasco et al, 2005, 2009). Averaging periods affected by large plumes are expected to be removed by the stationarity criterion. If stacks of large industries were within the monitored footprint and their impact was not filtered by the stationarity criterion, the turbulent fluxes reported in this study are suspicious.

17, 430-432. See Fig. 4 in Velasco et al. (2005) and Fig. 3 in Velasco et al. (2009).

19, 445. The acronyms POCP and PPCP have not been defined in the text yet.

20, 446-449. A similar figure showing the flux correlations between species during the winter period is needed.

20, 466-467. See previous comment on the vehicular contribution to urban isoprene.

21, 481-482. How and who determined this emission factor for Beijing's urban vegetation?

22, 511-514. Based on the comprehensive set of measurements conducted during the APHH study, is it possible to adjust the POCP and PPCP factors for Beijing's conditions? If it is not possible, wouldn't be better to evaluate only the OH reactivity of each compound?

22, 514-516. Light alkanes can be important players if their emissions are high like in Mexico City (see Jaimes-Palomera et al., 2016).

24, 540. The VOC classification used here is 'strange'. Be more specific on the species included in the group 'other hydrocarbons'.

24, 543. Consider a different title for this section. Why did the evaluation of the accuracy of the gridded emissions inventory was limited to the summer period?

24, 544-546. This was already mentioned in the introduction (lines 90-94). Indicate the emissions inventory's year reference.

25, 553-554. Were any power plant or large industry within the monitored footprint? If the answer is positive, the fluxes reported here might not be valid. Review the basic assumptions for measuring turbulent fluxes by eddy covariance (Velasco and Roth, 2010).

25, 575-576. The poor agreement between measured fluxes and estimated emissions might respond to the size of the grids in the emissions inventory. Cells of 9 km^2 might be too large to capture the emissions observed by the flux tower.

27, 590. According to the industrial emissions reported in the inventory developed for air quality modelling and presented in this figure, the site selected to conduct flux measurements by eddy covariance in Beijing is not adequate. Review the conditions needed to perform representative eddy covariance flux measurements in urban areas (Velasco and Roth, 2010).

28, 597-599. If it was known in advance that the inventory did not include the biogenic component, then which was the purpose of comparing the measured fluxes of isoprene? The emissions estimated by Megan should be added to those reported by the MEIC inventory.

28, 608-610. Indeed, the presence of methanol, aldehydes and organic acids can be explained by local atmospheric chemistry, however, it was previously proved that the time taken by an air parcel to reach the top of the tower was much less than the time needed for the oxidation of such compounds. If it was not the case, fluxes of those compounds cannot be measured by eddy covariance in Beijing.

29, 617. Instead of presenting conclusions and final remarks, this section is a summary of results.

29, 618. Rewrite this sentence. For example: 'Fluxes of speciated VOCs at district scale were measured by first time in Beijing ….'.

29, 625. 'Stable atmospheric conditions' may sound better.

29, 626-627. It may be true for VOC species emitted by evaporation of fossil fuels and solvents, but not for species emitted by other emission sources. It is not possible to talk about advection of VOCs in the case of fluxes measured at district level by eddy covariance, see previous comments.

29, 630-633. See previous comments about comparing the fluxes reported in this study with those observed in Mexico City.

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