

Squires et al. describe seasonal differences in flux measurements of CO, NO<sub>x</sub>, and select VOCs in Beijing during winter and summer, 2016-2017. The authors show that seasonal differences in combustion markers can be largely attributed to changing sources between the two seasons (residential vs. transportation), while changes in the fluxes of aromatic VOCs are strongly influenced by evaporative emissions during summer. The authors compare the flux measurements to inventory estimates from the MEIC, and discuss the significant discrepancies between the measurements and the inventory. The authors also use the inventory to infer how seasonal differences in emission sectors may have contributed to the observed seasonality in CO and NO<sub>x</sub> fluxes.

Overall, the manuscript is very well written and organized, and the authors tell a compelling story as to what contributes to the CO, NO<sub>x</sub>, and VOC sources observed in Beijing. It is also an important contribution to constraining emission inventories in China, which evolve quickly as sources continue to face regulations. The manuscript should be published in ACP, provided that the authors address a few comments pertaining to the comparison of the measurements and inventory.

### **Main Comments**

1. Section 3.4. I appreciate the authors' careful discussion and source attribution of the VOC fluxes, since this can be more complicated than for CO and NO<sub>x</sub> due to the variety of processes that could contribute to enhanced VOC fluxes (emissions, temperature, etc.). I am impressed by the general higher flux of aromatics in the summer and good correlation with heat flux. This seems to suggest that evaporative emissions are a driving factor in the behavior of the aromatics (as concluded by the authors), and could be the primary explanation in the seasonal differences in B/T ratios. The authors note that a detailed comparison of the VOC emissions with the inventory is beyond the scope of the work (which is reasonable), but is it possible to do a comparison between the aromatic / benzene ratios measured in this work with VOC profiles represented in the MEIC or other inventories? Li et al. (2017) tends to show that residential uses of aromatics result in higher emissions than the transportation sector. Perhaps a comparison of the B/T ratio between sectors would provide additional evidence as to what is contributing to these seasonal differences.
2. Section 3.5 and Figure 9. In this section, the authors discuss the measurements in context with the MEIC. The discussion of this section largely hinges on Figure 9, which suggests that the inventory largely overestimates NO<sub>x</sub> and CO emissions. In my opinion, this figure is misleading, since this is comparing an inventory from 2013 to measurements conducted in 2017. The authors note later in the section that the NO<sub>x</sub> and CO emissions in China change drastically on a multi-year basis. The authors discuss how these changes would have likely affected the MEIC estimates from 2013, but still conclude that the inventory is overestimated.

I think that it would be most fair to structure this section to first show how the

inventory would have changed from 2013 until 2017, then adjust the inventory estimates appropriately to account for these CO and NO<sub>x</sub> reductions, and finally compare these profiles to the measurements conducted in 2017 and discuss the discrepancies. Otherwise, a reader who skims the figures might conclude that the inventory is entirely out to lunch, which isn't truly the case.

## Other Comments

3. Page 2, Lines 25 - 30. The authors note that total emissions from inventories can differ from measured fluxes but I think it is also important to note that the distribution of VOCs can be different due to unattributed sources. Karl et al (2018) shows that there is a large, unidentified source of oxygenated VOCs (OVOCs) in European cities that isn't properly represented in emission inventories. The same conclusions were drawn by McDonald et al. (2018), who showed that these sources likely result from solvent emissions due to the use of consumer and industrial products. As China places stricter restrictions on emissions, it's likely that the relative importance of different emissions sectors will also change. Could the authors provide some context here (as they do in other places in the manuscript), and perhaps provide a brief overview of the current VOC, NO<sub>x</sub>, and CO distribution of China's emission inventory?
4. Section 2.3. How (and how frequently) were background PTR-MS measurements performed? The authors might consider referring to the instrument as a PTR-ToF-MS, since this is not a quadrupole instrument (the original PTR-MS). This distinction is important because the high time resolution of a PTR-ToF-MS is needed for flux calculations.
5. Page 5, Line 30. "Despiked" is an odd term. Would "smoothed" be a better option?
6. Page 6, Line1: "allowing the software to calculate" is very vague. Was there an algorithm applied to the data to determine instrument lag in real time?
7. Page 6, Line 23. What do the authors mean by "non-stationary periods"? Does this mean that the flux changing too rapidly over the course of the averaging period? This statement seems repetitive with the previous sentence.
8. Figure 3. It is difficult to see the grey colored traces highlighting non-stationary periods, and it's also confusing to have the contrasting periods also highlighted with grey. I would recommend adding markers for the points that were non-stationary, changing the color to clearly indicate these points, or simply remove these from the figure.
9. Page 13, Line 5 and Line 23. The authors mention that the major NO<sub>x</sub> sources are likely the same between the two seasons, which is likely true for vehicle traffic. However, wintertime fluxes appear to be higher on average (~20%), and the wintertime diurnal

patterns shows little resemblance to the summertime pattern (in line with the differences seen for CO). Furthermore, the nighttime flux of NO<sub>x</sub> seems to be substantially higher in the winter. To my eye, this suggests that there are additional important NO<sub>x</sub> sources, which are probably the same as those implicated for CO (i.e., residential heating). This seems to be supported by Fig 10, which shows that the Residential/Transportation ratio for NO<sub>x</sub> is substantially larger in the winter than compared to summer.

10. Page 13, Line 33. What is meant by “distant heating”? Is this heating using electricity?
11. Page 13, Lines 34 - Page 14, Line 6. The purpose of this paragraph isn't entirely clear. Are the authors invoking HDV emissions to explain the higher CO and NO<sub>x</sub> emissions during winter, or is this to explain the nighttime emissions during both seasons? Would one expect HDV's to be more active during winter than during summer? It does appear that the summertime morning peak in NO<sub>x</sub>, which is earlier than the CO peak, could be associated with the higher truck traffic before 06:00.
12. Figure 7: This figure is very nice. It is a little difficult to read the labels and to see the time axis on top of the darker colors of the fluxes. Could these be presented in larger fonts, and in bold?
13. Page 20, Line 30. When the authors mean that the change in B/T cannot be explained by temperature differences, I assume they mean atmospheric oxidation and not evaporation processes. This is clarified in the following sentence, but it would be good to be precise here since there is a lot of discussion about what could be affecting the B/T ratio (evaporation, chemistry, etc.) and mixing terms is a bit confusing.

## References

Li et al. (2017). MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP. *Atmos. Chem. Phys.*, 17, 935-963.

McDonald et al. (2018). Volatile chemical products emerging as largest petrochemical source of urban organic emissions. *Science*, (359) 6377, 760-764.