Comments on ACP 2024-2064

Downward and upward revisions of Chinese emissions of black carbon and CO in bottom-up inventories are still required: an integrated analysis of WRF/CMAQ model and EMeRGe observations in East Asia in spring 2018

General Comments

The manuscript "Downward and upward revisions of Chinese emissions of black carbon and CO in bottom-up inventories are still required: an integrated analysis of WRF/CMAQ model and EMeRGe observations in East Asia in spring 2018" focuses on constraining an inventory based on aircraft observations and modeling. The authors used aircraft observations of black carbon and carbon monoxide from a 2018 campaign, WRF/CMAQ forward modeling, and back-trajectory tools to estimate the mismatch between predicted and observed aerosol concentrations over East and Southeast Asia. The findings suggest a potential bias in current inventories for black carbon, carbon monoxide, and carbon dioxide over the study regions and also show that the modeled aerosol concentrations produced with this model and a wildfire emissions inventory are within reasonable bounds of observed values. As such, the findings of this work are timely and can have an impact on informing emission inventory development studies for this region. The manuscript contains detailed analyses and is well-written. However, the flow can be improved with better organization. Moreover, the extrapolation of aircraft observations of a few days to an annual inventory over a large region should be justified. Specific comments are below.

Overall, I recommend the manuscript for publication after minor analyses-based revisions but more writing- and presentation-based revisions.

High-level comments

1. Consider reorganizing the manuscript for clearer flow. Current flow does not read well and may be improved by aggregating analyses based on a region, adding an Overview in the Methods, and then a bulleted list of analyses performed. Similar changes in the Results section could be useful. Also consider renaming section titles to be clearer and consistent.

2. The manuscript needs more focus on the robustness of the results when compared to aircraft observations. Do a few days of aircraft observations represent seasonal or annual magnitudes? Or could the uncertainty in the modeled concentrations be more due to the simulated meteorology and aerosol representation alone and not due to the underlying inventory?
3. While a model-observation study shows potential bias in the inventory, it does not separate those biases by source. Which sources would inventory developers need to tune up or down to match these observations? Can aircraft observations aid in identifying hotspots from a region?
4. The model-observation mismatch is attributed to uncertain emissions in the inventory. However, as the HYSPLIT back-trajectories show, the source regions could be a narrow band or can cover multiple regions. How robust are this study's findings in generalizing over the entire CHN or other regions?
5. Consider consolidating Figures 2-5 into one figure and focus on showing how the model performs in each of the cases. For example, a figure could focus on just the identification of case names based on observed back-trajectories, and another figure could focus on the flight observations and modeled concentrations. Show the relevant flight paths in the figures with back trajectories.
6. Aircraft observations have their own merits and demerits. For example, while they aid in isolating non-local sources, comparing them with modeled concentrations is difficult due to higher model uncertainties at those distances from the source region. At aircraft distances, model uncertainties may be higher than emission inventory uncertainties, for example, due to modeled rapid deposition offsetting higher emissions. How robust are aircraft observations at isolating uncertainties in emission inventories?

Specific Comments

Title: It can be made tighter; something along the lines of "Assessing uncertainty in emission estimates from China using EMeRGe aircraft observations and models".

L22: See point 6 in high-level comments. Justify this in the Introduction.

L30: "The results suggested that downward and upward revisions of Chinese emissions of BC (−50%) and CO (+20%), respectively, are required in HTAPv2.2z emission inventory." Also mention the range in other inventories such as CEDS (Hoesly et al., 2018) or the IIASA GAINS.

L34: Mention the lifetimes of SLCFs.

L50: "Our understanding of the responses of SLCF emissions to the establishment of techniques that decrease emissions in the last two decades in fast-growing Asian economies is insufficient (Chen and Chen, 2019; Kanaya et al., 2020; Ikeda et al., 2023; Zhang et al., 2022)." This point was not clear. Emissions decrease as we use more abatement (assuming activity remains the same), and the same is true everywhere.

L52: "Biomass-burning habits in Southeast Asia has become the main contributor of carbon emissions from forest fires in spring (Reid et al., 2013; Heald et al.,2003; Palmer et al., 2006; Johnston et al., 2012)." Consider rewording this to not write it as a 'habit'. People use biomass as a source of residential or heating energy in those regions and while it is a habit, it is more of a need.

L80: Good point. Agree!

L83: "Emission inventories therefore need to be tested using independent observational data." I understand this focuses on the EMeRGe dataset, but any mention of ground-based long-term monitoring and its usefulness would also be good to include in the Introduction, and then mentioning how aircraft observations can help provide another perspective, will be great to add.

L132-137: I appreciate the inclusion of underlying inventories in HTAP.

L164: Why is 1200hrs (or 5 days) a suitable time range for back-trajectory analyses in this work?

L189: Describing the common BC/CO, CO/CO2, and BC/CO2 ratios in different combustion sources would aid the reader in understanding that whenever there is a higher BC/CO ratio, it signals a source X, and a lower CO/CO2 ratio signals source Y.

L215: Shouldn't this correction factor be applied to only those grids in CHN where the emissions are coming from and were measured in the campaign? Or to the whole of CHN?

Equations 4, 5, and 6: Should this correction factor based on BC/CO and other ratios be applied to just the emission source with a known BC/CO (and other ratios)? For example, if the aircraft observed a high BC/CO ratio, it signals those emissions from source X dominated the concentrations on that day. Now, the modeled concentrations show a lower BC/CO ratio, indicating either the contribution from source X in the inventory is low, or that the contribution from other sources (with a lower BC/CO ratio at emissions) is higher. How do you differentiate between these two offsetting effects?

L225: This paragraph should be in Methods.

L260: "On the other hand, the IFS-CAMS simulation predicted the maximum CO concentration well in Deroubaix et al. (2024a), possibly because anthropogenic emissions in the IFS-CAMS simulation were taken from the CAMS-GLOB-ANTv4.2 emission inventory (Granier et al., 2019)." This brings up a good point and something that should be discussed in the end and generally --- how does the choice of inventory affect your findings?

L264: A general comment --- what does the observed/modeled ratio refer to? Is it the average observations and average modeled concentrations for the whole flight path? If so, the values closer to the source region better represent emissions uncertainty due to lesser influence by transport processes such as coagulation and deposition. Consider adding any discussion on that as well.

L282: This raises a good point about the temporal resolution in the inventory. Inventories probably do not capture diurnal or hourly emission patterns and thus the bias in aircraft observations at two time-stamps against modeled concentrations may be due to the lack of such temporal resolution in inventories. Consider mentioning that in perspective of the model-observation differences in this and other sections or in general.

Table 2: How is the R calculated here? Why is it so low in some cases? Consider adding Spearman's coefficient since the model and observed trends were similar.

L339: Even if there is no rainfall, higher moisture can lead to rapid aerosol growth from condensation and thus a faster deposition, especially farther from source regions when BC gets activated due to sulfate reactions.

L377: Similar to the comment above on Equations 4, 5, and 6 --- should not this correction be based on source-specific BC/CO ratios? This can also explain some of the differences observed in Figures 7a and 7b.

L460-465: While this is a great finding, consider including some process-specific discussion --- which specific source needs to be tuned up or down in inventories? Do the observed BC/CO/CO2 ratios help in differentiating source signals?

Figure 6: Any explanation on why the model does not produce as much variability (concentration range) as observations will be good to add.

Figure 8: How did you calculate the uncertainty in red and green boxes?

Sec. 3.2: This section touches upon some of the points I raise above but a richer discussion in identifying the sources will be good to add. Consider using information from this section to inform inventory updates in the previous section. This does not need to be big changes but a short circling back would be useful.

Sec. 4: Consider adding recommendations on how inventory-developers can directly utilize such campaigns' information to tune up or down in this section.

L653: Add discussion on whether there is any chance or reason why the BC/CO ratios in the model and observations are similar due to completely different reasons, such as rapid BC deposition in the model offset by higher BC emissions?