Summary:

This paper studied the correlation relationship between black carbon (BC) and organic aerosol (OA), and the correlations of two types of carbonaceous particles with other gaseous pollutants in both airborne observations and associated four different modeling frameworks. The airborne platforms that flew over Europe and East Asia were equipped with identical instrument payloads, which made the concentrations of different types of pollutants comparable. The flights encompass several major urban plumes, making it possible to further study the in-plume relationships of the related pollutants.

This paper presented comprehensive analyses of all detected pollutants and quantitatively analyzed the correlations and relative abundance of each atmospheric species. Observations in Europe and Asia showed variated pollutants abundances. Different levels of correlation between gaseous pollutants and particulate matters reflect dynamic natures of co-emission intensities. By comparing the observations with modeling outputs, the study found that stronger correlations between primary gaseous pollutants and particulate matters were widely presented in modeling than observations. In contrast, the spatial variance of the observed relative abundance of certain gaseous species and BC or OA in two city plumes were not reproduced by modeling. This discrepancy is partially attributed to the inaccurate ratios in anthropogenic emission inventories. The paper suggests more observational constraints including satellite-, airborne-, and ground-based measurements were incorporated into the compilation of the inventories.

General suggestions:

1. Figure 1: This figure mainly illustrates the flight trajectories of two campaigns and the portions of urban plume transects. Were those urban plume transects discretely distributed along the trajectories like it shows in the figure, or should be temporally and spatially continuous during the campaign. Given the vertical profiles of the sampled pollutants are not the main topic to be discussed in this study, a 2-D figure with trajectory lines overlaid on it should be sufficient to illustrate the idea. The urban plume portion can be highlighted in a different color. 2-D map would also help to visualize the boarders and populous regions.
2. Figure 2: This figure mainly shows the strong correlation between BC and OA among almost all frameworks. The visualization of this figure can be further improved. Now there is a high degree of data overlap in both panels. The color of data points and relative size of markers make the data difficult to distinguish. May consider present the correlation in each framework (observations, each modeling settings) in a separate subplot, and use the scatter density plot to illustrate the correlations.
3. Line 181: The higher BC/OA ratio in Asia is attributed to wildfires in Indochina. Given wildfires contribute a large amount of primary OA and secondary OA precursors as well, is there any chemical analysis of OA during the campaign (or source apportionment) to indicate the contributions of possible OA sources? HCHO was regarded as a better proxy for OA according to the correlation analysis. Any observations from the mass spectrometers can be added to infer the possible mechanisms behind this correlation?

4. Figure 3: In panel b, at high BC concentrations, concentrations of SO2, HCHO, and NO2 tend to be constant as BC increases. Any interpretation of this relationship here? Similar patterns were found in Figure 4b.
5. Table 3. Any explanation on zero R-value of CAMS-forecast O3 to OA in Europe?
6. Secondary OA takes up a large portion of mass in total OA. I suppose the ratios of emission fluxes in Figure 5 mainly quantifies the relative abundance of primary sources (BC, POA, etc.), but not considering the SOA precursors that may further form particle-phase OA. The SOA formation mechanisms depend on model selections and need to be considered in inventory evaluation. In addition, a positive matrix factorization (PMF) analysis on observed chemical composition of aerosol can also help to provide source apportionment information of OA.

Line-by-line suggestions:

1. Line-230: “R-value”
2. Line-264 to 267: These two sentences are a bit redundant in expression.