

This paper reports the BC and CO concentrations and their correlation based on continuous measurements made at the summit of Mt. Huang in eastern China between 2007 and 2008. The authors investigated the seasonal variation of the $\Delta BC/\Delta CO$ ratio and the linkage of such variation to fuel type uses in eastern China. They also performed detailed back trajectory cluster analysis to look at the effects of pollution plumes from different regions of eastern China on the $\Delta BC/\Delta CO$ ratios observed at the site. The study should provide important datasets for future studies on the EC and OC emissions, chemical aging and transformation, and climate impacts over the region. While the results appear interesting in general, some explanations of measurement data are too arbitrary without addressing fundamental physical and chemical processes that control the lifetimes of EC and OC and thus the EC/OC ratio. Existing emission estimates for the regions, especially the varying source types that may have different EC/OC emissions ratios, are not well taken into account in their discussions. Therefore, the manuscript needs major revisions before being accepted for publication in Atmospheric Chemistry and Physics. My specific comments and suggestions are below.

Specific comments

- 1) In Abstract, Page 4448, Line 6: regarding to '*between 2006-2009*', is the CO data available for the years 2006 and 2009?
- 2) In Introduction, Page 4451, Line 13, the author states '*There have been few investigations on the $\Delta BC/\Delta CO$ ratio in China*'. There have been many papers on BC and CO in China, including measurements at the rural sites.
- 3) In Introduction, Page 4451, Line 17-19: how could the API in the cities be used to determine the regional pollution events?
- 4) In Sect. 2.1, Page 4452, Line 7: I did not find 'Fig. 1a' in the text before. It is difficult to see the distribution of BC in Fig. 1b. It would be nice to add a plot for BC/CO emission ratio with a unit the same as used for measurement data.
- 5) In Sect. 3, Page 4454-4455: does the $\Delta BC/\Delta CO$ ratios estimated from emission inventory have a same unit as those calculated from measurement data? Or both have been changed to the STP values? Here the authors spend one section describing the uncertainties in the emission inventory. However, they just use the emission distributions of this inventory to explain their observational results in a qualitative way. Except for stating that the $\Delta BC/\Delta CO$ ratio for urban plumes is similar to those

obtained from an emission inventory in the conclusion, no evaluation on the emission inventory based on their measurements is given in the manuscript.

- 6) In Sect. 4.1, Page 4455, Line 22-24, the authors states '*Urban pollution resulting from the industrial and residential burning of biofuel for cooking and heating are the most probable explanations for such phenomena*'. Do you have sufficient data to support your explanation? Why did you exclude the emissions from the traffic? How about the influence of seasonal variation in PBL? Similar 'explanations' appear in several places of the manuscript. There have been a few papers on CO measurements at the rural sites of North China published in international journals.
- 7) In Sect. 4.2, Page 4457, Line 20-26, the authors states '*BC₀ and CO₀ (baseline concentrations of BC and CO) were determined as mean values of the 1.25 percentile of data.....*'. How was BC₀ and CO₀ determined in other studies? Does the definition of BC₀ and CO₀ (especially CO₀) influence the calculated $\Delta BC/\Delta CO$ ratio greatly? How can the measured $\Delta BC/\Delta CO$ ratio be used to compare with the $\Delta BC/\Delta CO$ ratio estimated from the emission inventory, the latter defining BC₀ and CO₀ as zero? Could you use the different BC₀ and CO₀ for different clusters?
- 8) In Sect. 4.2, Page 4457, Line 26-28, the authors states '*This declination mainly reflected the baseline of clean air masses from the continent interior because more than 90% of air mass came directly from the Mongolian Plateau*'. I don't understand this. Mt. Huang should be further away from Mongolian Plateau than Beijing? Can air mass from the Mongolian Plateau be transported to Mt. Huang without passing the polluted areas in North China?
- 9) In Sect. 4.2, Page 4458, Line 7-14, the authors states '*An airborne single-particle soot photometer..... at high altitude (2–5 km) over Mexico, similar to our results. The high $\Delta BC/\Delta CO$ ratio at the urban site from heavy vehicles Nevertheless, these explanations are difficult to apply in our case*'. These discussions are confusing.
- 10) In Sect. 5, Page 4458, Line 16-18 and Page 4459, Line 3-5: what is the grid resolution of NCEP meteorological data you used? Is it fine enough to distinguish an urban plume?
- 11) In Sect. 5, Page 4458, Line 18-20: how about the data used in Sect. 4, with all RH

conditions included?

- 12) In Sect. 5, Page 4460, Line 3-8, the authors states '*For the whole period, about 46% air masses back trajectories were classified into Cluster #1,, the values are similar to measurements in urban areas (Kondo et al., 2006; Spackman et al., 2008; Han et al., 2009). Simple footprint analysis (supplementary Fig. 2) indicated that urban plumes in Southern Anhui province mostly resulted in relatively high BC and CO concentrations, and domestic and industrial emissions were the major contributions*'. I doubt if simple footprint analysis can separate the urban plumes from biomass burning sources and further industrial emissions from traffic emissions.
- 13) In Sect. 5, Page 4460, Line 18-21, the authors states '*However, our results were higher than airborne measurements by Single Particle Soot Photometer in the altitude range of 2.7–4.1 km around Mexico City with a mean $\Delta BC/\Delta CO$ ratio of $2.80 \text{ ngm}^{-3} \text{ ppbv}^{-1}$ for fresh emissions and $3.3 \text{ ngm}^{-3} \text{ ppbv}^{-1}$ for 1-day-old emissions (Subramanian et al., 2010)*'. The results of this study have been compared with the work of Subramanian et al. (2010) several times (see Comment 9 above) with different conclusions, sometimes 'similar' and sometimes 'higher'. The authors should make it clear when the results are comparable and what the comparison results imply.
- 14) In Sect. 5, Page 4461, Line 1-4: I doubt if the emission source attribution in the Yangtze Delta is the same as it is in Beijing. There have been many studies on air pollution in the Yangtze Delta, including the development of a regional emission inventory (e.g., Zheng et al.: *A highly resolved temporal and spatial air pollutant emission inventory for the Pearl River Delta region, China and its uncertainty assessment, Atmos. Environ.*, 43, 5112-5122, 2009). The authors may also have a look at of the INTEX-B inventory.
- 15) In Sect. 6, Page 4461, Line 24-25, the author states '*Here daily averaged BC concentrations over $25 \text{ } 1000 \text{ ngm}^{-3}$ and RH less than 50% were taken as the criterion of high BC pollution*'. Did you consider RH of air masses on the day before?
- 16) In Sect. 6.1, Page 4463, Line 12-14, the authors states '*We believe BC and CO were well mixed and had constant $\Delta BC/\Delta CO$ ratios soon after emission in certain areas, notwithstanding variations in emission strengths for different industry types*'. Can the

production of CO via oxidation of VOCs and its loss affect the $\Delta BC/\Delta CO$ ratios in the plume? What is the BC loss rate in unit of ngm^{-3} per day estimated by its dry deposition and collision sinks?

- 17) In Sect. 6.2, Page 4464, Line 14-16, the authors states '*According to the emission inventory, BC emission factors for biomass burning were about 0.47–0.98 g kg⁻¹ owing to incomplete combustion and much higher than those for urban gasoline vehicle emissions*'. It may be meaningless to just compare only the emission factors without considering the amounts of gasoline and biomass burned.
- 18) In Sect. 7, Page 4464, Line 24-Page 4465, Line 1: why did you doubly count the data (the RH ranges are overlapped) when dividing the data into different groups?
- 19) In Sect. 7, Page 4464-4465: I would suggest that the BC and CO concentrations also be presented in Table 3. Could all the decrease in the $\Delta BC/\Delta CO$ ratio be attributed to BC loss by dry deposition? Would the CO concentration be kept constant? How about the changes in meteorological conditions associated with RH?
- 20) In Sect. 7, Page 4465, Line 17-21, the author states '*According to the INTEX-B anthropogenic emission inventory,(Supplementary Fig. 8),'*'. There have been many measurements of aerosol chemistry in the regions.
- 21) In Sect. 8, Page 4465-4466: the authors merely give a summary of the $\Delta BC/\Delta CO$ ratios measured in different regions by different studies. What new (or different) concepts, ideas, and methods does this study present compared to these previous studies? Is the photochemical processing important for the variations of the $\Delta BC/\Delta CO$ ratio in eastern China? Can the quality of an emission inventory, e.g., INTEX-B inventory, be evaluated based on the analyses of this study? Maybe Sect. 3 should be moved here for discussion.
- 22) The authors may make necessary revisions in Abstract and Conclusions when having addressed the comments and suggestions above.