

Response to Referee #2:

Thanks very much for your comments, suggestions and recommendation with respect to improve this paper. The response to all your comments are listed below.

General comments :

The authors presented a multi-year timeseries (2015-2018) of HCN partial columns observed at a densely populated Chinese city, Hefei, using a ground-based high spectral resolution Fourier transform infrared (FTIR) spectrometer. CO columns were also presented for comparison. The authors identified the seasonal maxima of tropospheric HCN columns at this site and compared their seasonality and interannual variations to the HCN columns obtained from other NDACC sites. Drivers of HCN seasonality and interannual variations at Hefei were determined through GEOS-Chem tagged CO simulations, as well as the global fire maps and a back-trajectory analysis technique. They also used CO measurements to estimate the HCN/CO emission enhancement ratios to quantify biomass burning emissions.

This version (manuscript-version2) has been significantly improved from the original submission in terms of analysis and presentation. The results are valuable to the community. I suggest some minor corrections, which are detailed below. The authors should perform a thorough proof-reading of the manuscript to further improve the presentation in order to meet the publishing standards of ACP.

Response: This paper has been subjected to a revision based on the comments from two referees. We also perform a thorough proof-reading of the manuscript to further improve the presentation. All your comments are appreciated and have been addressed in the revised version. Main changes/improvements are listed as follows:

Specific comments:

1) Page 1, Line 35: Are these values multi-year mean or are they the maximum values throughout the whole period? Please clarify.

Response: They are monthly means based on multi-year measurements. We have clarified this in the revised version.

2) p2, line 34-35: Could you also point out where these regions are located in China?

Response: In the revised version, we have included a map of China showing the three

most developed regions—Jing-Jin-Ji (Beijing-Tianjin-Hebei), Yangtze River Delta (including Shanghai), and Pearl River Delta (including Guangzhou and Hong Kong–Shenzhen) in Fig. S1. It looks more clear than put these regions in the world map in Figure 1.

3) p2, line 53: add “where” before “influences from biomass . . .”

4) p3, line 6: replace “can improve” with “aims to improve”

5) p3, line 17: replace “it is planned to” with “plan to”

6) p3, line 20: replace “a highest” with “the highest”

Response: The above corrections have been done in the revised version.

7) p3, line 22: “to ensure a higher signal to noise . . . and a faster. . .” compared to what?

Response: To avoid misleading, we have changed it to “ However, all mid-infrared (MIR) spectra are recorded with a spectral resolution of 0.005 cm^{-1} to follow NDACC convention”.

8) p3, line 26-27: can you spell out “&” as appropriate here.

Response: We have done this in the revised version.

9) P4, line 8: change to “Whole-Atmosphere Community Climate Model (WACCM) v6”

Response: We have done this in the revised version.

10) P4, line13-15: spell out all species names when they first appear.

Response: We have done this in the revised version.

11) P4, line 24: did you calculate the tropopause height from the NCEP reanalysis data? If so, replace “deduced” with “calculated”.

Response: Yes, and we have done this in the revised version.

12) P5, line 19: please spell out GEOS-FP.

Response: We have done this in the revised version.

13) P6, line 5: the order should be “Global Fire Assimilation System (GFAS) v1.2”

Response: We have done this in the revised version.

14) P6, line 9: change to “period of”

Response: We have done this in the revised version.

15) P6, line 11: Please be consistent in placing acronyms. Should be “the Multi-resolution Emission Inventory for China (MEIC)”.

Response: We have done this in the revised version.

16) P6, line 18-19: It is confusing here, as the GEOS-Chem OH shown in Shindell et al. (2006) is lower than the multi-model mean (Table 1). Could you clarify?

Response: We have consulted the GEOS-Chem team at Harvard university during the ACPD phase. The developer told me that what we actually used in this paper was “monthly mean OH concentrations archived from a previous full-chemistry simulation” rather than that used from Shindell et al. (2006). In the revised version, we have changed this sentence to “The OH fields were obtained from monthly mean OH concentrations archived from a previous full-chemistry simulation.”

17) P6, line 31-33: this sentence can be modified as “In this study, we only investigate the influence from the biomass burning sources. The regional definition. . .”

Response: We have done this in the revised version.

18) P6: at the beginning of section 2.3, could you briefly say the purpose of using PSCF in this study? Just add some context.

Response: We have done this in the revised version.

19) P7, line 37: delete “total” before “troposphere”

Response: We have done this in the revised version.

20) P8, line 6-10: these few lines can be re-organised to make the statements clearer. For example, “Note that the largest seasonal peak of HCN occurs in May which is 3 months later than that of CO which occurs in February, but the other two seasonal peaks for both species occur in the same months, i.e., in September and December respectively. Otherwise, their seasonal cycles show similarities.”

Response: We have done this in the revised version.

21) P10, line 11-17: could you elaborate a bit more on possible mechanisms why correlations between CO and HCN are negative or weak at these sites, especially at Bremen? Have you checked the seasonal correlation between CO and HCN at Bremen?

Response: Actually, here are the overall correlations by using all measurements from

2015 to 2018. We have double checked the programming script for this figure, and found that all fittings are correct. We have included more explanations in the revised paper, i.e., “Biomass burning is the dominant source of HCN and industrial emissions only contribute additional minor sources (Bange and Williams, 2000; Holzinger et al., 1999; Lobert et al., 1990). In contrast, anthropogenic, biomass burning, CH₄ and NMVOC oxidations are major sources of CO, and their contributions are season and location dependent. Therefore, the correlation between HCN and CO tropospheric column is also season and site dependent. High correlation of these two species is supposed to be observed if biomass burning dominates the CO variability, and vice versa.”

22) P10, line 24: how about “. . . is an effective quantity to identify biomass burning emissions. . .”?

23) P10, line 31: should be “indicating that these. . .”

24) P10, line 32: change to “throughout the year”

25) P11, line 1: change to “is located . . . , therefore emissions. . .”

26) P14, line 22: change to “both the first and the second half of the year”

27) P14, line 24: change to “in the second half of the year”

28) P14, line 25: same as above “in the first half of the year”

29) P14, line 27: change to “As shown in Figs. . .”

30) P14, line 34: replace “Drives” to “Driven”

31) P15, line 3: “meteorological conditions”

32) P15, line 7-8: change to “through large-scale atmospheric circulation”

33) P17, line 2: replace “driven” with “drove”

Response: We have done above corrections in the revised version.

34) P17, first paragraph: Are these elevated fire counts related to ENSO? Can you discuss the possible ENSO link here, rather than at the end of the section?

Response: We have moved the last paragraph here and analyzed the possible ENSO link. “Many studies have revealed that the El Niño Southern Oscillation (ENSO) can cause large scale variations in the convection, circulation, and air temperature of the global atmosphere-ocean system (Liu et al., 2017; Zhao et al., 2002), which could affect the distribution, frequency, and intensity of biomass burning emissions

(Schaefer et al., 2018). Furthermore, ENSO could also alter the destruction processes of tropospheric species through their photochemical reactions with tropospheric OH (Zhao et al., 2002). Zhao et al. (2002) found that the abnormally enhancement of tropospheric CO and HCN observed in northern Japan in 1998 were associated with the 1997–1998 ENSO events (Zhao et al., 2002). There is a close correlation between ENSO and HCN columns at Lauder (45°S) (Zeng et al., 2012; Schaefer et al., 2018), and Schaefer et al. (2018) quantified a detectable ENSO influence on biomass burning of up to 51-55% (Schaefer et al., 2018; Zeng et al., 2012). Very likely, the elevated fire counts which caused significant enhancements between September 2015 and July 2016 for tropospheric CO and HCN columns at Hefei and most selected NDACC stations were related to the 2015 – 2016 ENSO events.”

35) P17, lines 28: should it be “Zhao et al. (2000) or (2002)” here?

Response: We have changed it to Zhao et al (2002).

36) P17, line 30-31: please re-phrase this sentence as “There is a close correlation between ENSO and HCN columns at Lauder (45°S) (Zeng et al., 2012; Schaefer et al., 2018), and Schaefer et al. (2018) quantified a detectable ENSO influence on biomass burning of up to 51-55%”.

37) P17, line 32: replace “presumably” with “very likely”.

38) P17, line 37: delete “amounts” and “s” to “HCN column”

Response: We have done above corrections in the revised version.

39) In Conclusion, could you provide some further remarks on the long-term outlook of FTIR observations at this site.

Response: In the revised version, we have included the sentence “Most high resolution FTIR instruments are located in Europe and Northern America, whereas the number of sites in Asia, Africa, and South America is very sparse. As one of few FTIR stations on Asian continent, the long-term observations of trace gases at Hefei are crucial to understand global warming, regional pollution, long term transport, and contribute to the evaluation of satellite data and model simulations.”