Anonymous Referee #1

The authors present a simultaneous field measurement dataset of BC at five sites in this paper with the aim to investigate the intra-regional transport between the south edge of North China Plain and Central China based on the variations of BC mass concentration, sources and optical properties. The dataset is important and would be with good scientific significance to help people to model the BC aerosol climate effect in East Asian by studying the changes of BC physio-chemical and optical properties during the transport. My major concern is, as the authors stated in their paper (in the introduction), one of the key purpose of this study were to quantify the regional transportation of BC at multiple observation sites in CC and SE-NCP. But the backward trajectory method used by the authors can just get some qualitative analysis of the air parcels transport as presented in their study. The paper will be greatly improved if the authors consider using models to simulate the emissions and then to quantify the intra-regional contributions at the sites based on the measured BC concentration data. In addition, there are also a lot of language issues and editing needs that have to be addressed. The authors thus need to make a careful revision and correction on the language, especially revisions on some seemingly illogical expression, to improve the overall quality of the paper for publication in the journal. I would recommend the editor to reconsider the papers after a major revision by the authors.

AR: Thanks for your comment. We have added the Geos-Chem simulation to quantify the transport contributions. However, the Geos-Chem results are not good enough and details are provided in the response to Comment 10. Additionally, we have also carefully checked and polished the language.

Other specific comments,

1. Section 3.1, the authors should more focus on discussing and comparing the different BC levels between the studied 5 sites and other regions in China or over the globe, not on North China and other regions.

AR: Thanks for your suggestion and we do the literatures review. Comparison of BC in this study and other regions was listed in the supplementary file Fig. R1:

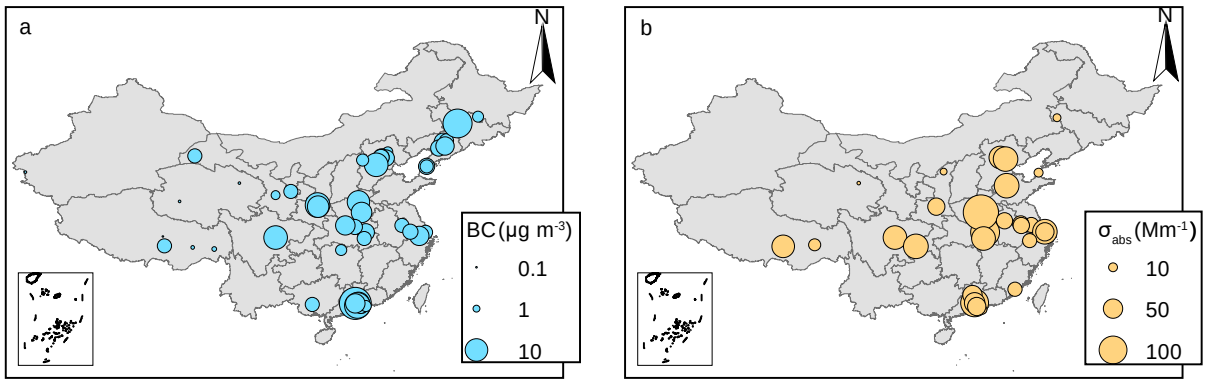

Figure. R1 Spatial distribution of BC mass concentration (a) and absorption coefficients (b) in China. More details can be found in Table S1 and S2 in the supplementary materials.

As Table S1 shown, BC was generally higher in North China and lower BC levels were found in remote areas and coastal areas as Fig. 3a shows. Wang et al, (2014b) analyzed ambient BC in an urban site in Xi'an during winter and found the average mass concentration was 8.8 ± 3.7 μg m^{-3}, which was higher than that in this study. Compared to other regions, BC levels in this study were higher than a remote area of Lulang in southeastern part of the Tibetan Plateau (0.31

± 0.55 μg m^{-3}) (Wang et al., 2018) as well as coastal areas such as Hong Kong (1.4 ± 1.1 μg m^{-3}) (Wang et al., 2017a) and a rural site in Shenzhen (2.6 ± 1.0 μg m^{-3}) (Huang et al., 2012).

References

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Wang, J., Virkkula, A., Gao, Y., Lee, S., Shen, Y., Chi, X., Nie, W., Liu, Q., Xu, Z., Huang, X., Wang, T., Cui, L. and Ding, A.: Observations of aerosol optical properties at a coastal site in Hong Kong, South China, Atmos. Chem. Phys., 17(4), 2653–2671, doi:10.5194/acp-17-2653-2017, 2017a.

Wang, Q., Cao, J., Han, Y., Tian, J., Zhu, C., Zhang, Y., Zhang, N., Shen, Z., Ni, H., Zhao, S. and Wu, J.: Sources and physicochemical characteristics of black carbon aerosol from the southeastern Tibetan Plateau: internal mixing enhances light absorption, Atmos. Chem. Phys., 18(7), 4639–4656, doi:10.5194/acp-18-4639-2018, 2018.

2. Line 214, ". . .Despite the sampling periods, site types, inlet of aerosol and instruments were different between different studies (Table S1), BC was generally higher in North China and lower BC levels were found in remote areas and coastal areas... " Here, it is not appropriate and logical expression by saying the two things using the "despite".

AR: We have corrected it as above.

3. Line 242 "..At WH, the concentration and percentage of BC_{bb} both decreased from clean to pollution, which suggested that more BCff was emitted during haze episodes...", should revise as "... both the concentration and percentage of BC_{bb} decreased from clean to pollution". . ." also, you say more fossil fuel BC was emitted. But the increased BC is probably due to the accumulation of pollutes during polluted days when the PBL is lowered.

AR: Thanks for your correction, and we have revised it.

4. Lines 248-250, ". . .the pollution episodes (Huang et al., 249 2014), and the increased secondary aerosols would be more adsorbed on the surface of BC. . .." Do you mean that more secondary aerosols will be coated on BC? "...the σabs also elevated by 11.7−254% as the air quality switched from clean to pollution (Fig. 4e). There are more secondary aerosols (i.e., sulfate, nitrate) during the pollution episodes (Huang et al., 249 2014),.." Do you have some observations of chemical composition that would support your conclusions.

AR: Yes, we have conducted the off-line low volume $PM_{1.0}$ sampling and chemical analysis at the five sites during the field sampling campaign. Here, we just show part of the chemical analysis results (which have not been published) to support that there were more sulfate and nitrate during the pollution episodes as shown in the following figure.

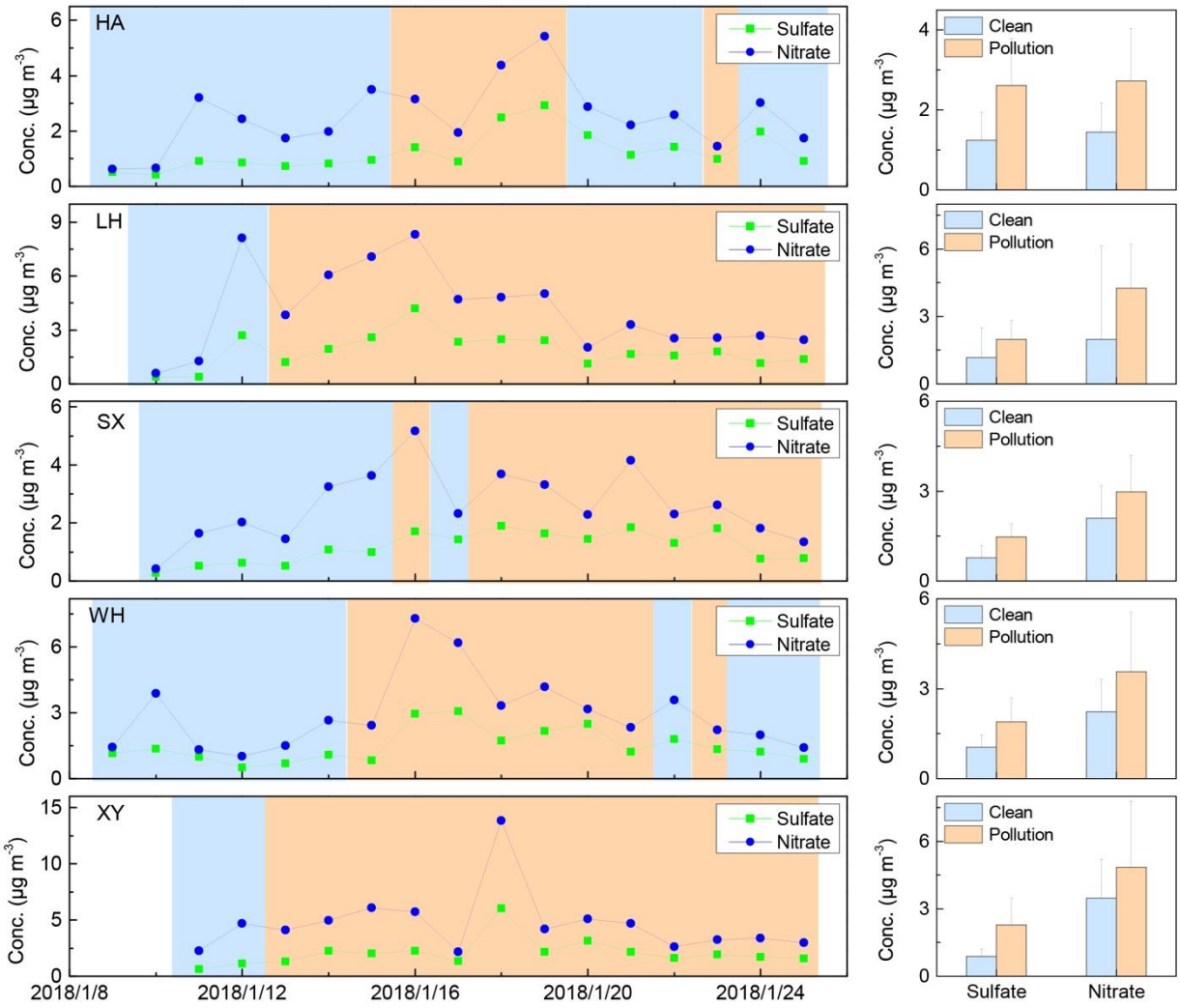

Fig. R2 Daily concentrations of sulfate (green line) and nitrate (blue line) during observation (left panel) and their average concentrations for clean days (blue) and pollution episodes (orange) (right panel).

5. Line 253, "The decreasing of AAE from clean to polluted days was also reported else- where (Zhang et al., 2015b) and it can be partly attributed to the source variation. . .", the AAE is very sensitive to particles size, so you may need to think about the particles growth due to the secondary formation processes.

AR: Thanks for your comments and we agree with you. However, we did mot measure the particle size during our observation, and we try our best to explain the decreasing of AAE during pollution episodes according to previous studies and the sentences are revised as following:

The AAE is also sensitive to other factors such as the particle size. Previous studies suggested that the particle diameter and number concentration increased from clean to pollution episodes due to several factors such as coagulation, hygroscopic growth, emissions, meteorological conditions, i.e., planetary boundary layer and wind speed (Guo et al., 2014; Zhang et al., 2017). These studies suggested that the particle diameter is generally larger during pollution days. Furthermore, the lab combustion and numeric simulation proved that BC particle with larger geometric median diameter had lower AAE value (Singh et al., 2016; Liu et al., 2018b). Therefore, lower AAE was observed during pollution episodes in this study.

References

Guo, S., Hu, M., Zamora, M. L., Peng, J., Shang, D., Zheng, J., Du, Z., Wu, Z., Shao, M., Zeng, L., Molina, M. J. and Zhang, R.: Elucidating severe urban haze formation in China, Proceedings of the National Academy of Sciences, 111(49), 17373–17378, doi:10.1073/pnas.1419604111, 2014.

Liu, C., Chung, C. E., Yin, Y. and Schnaiter, M.: The absorption Ångström exponent of black carbon: from numerical aspects, Atmospheric Chemistry and Physics, 18(9), 6259–6273, doi:10.5194/acp-18-6259-2018, 2018.

Singh, S., Fiddler, M. N. and Bililign, S.: Measurement of size-dependent single scattering albedo of fresh biomass burning aerosols using the extinction-minus-scattering technique with a combination of cavity ring-down spectroscopy and nephelometry, Atmospheric Chemistry and Physics, 16(21), 13491–13507, doi:10.5194/acp-16-13491-2016, 2016.

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6. Lines 257-267, you only show the diurnal variations of mass concentrations of BC, how about the absorption coefficient?

AR: Thanks for your suggestion, and we found that the diurnal variations of absorption coefficient and BC levels were similar as the following figure shown.

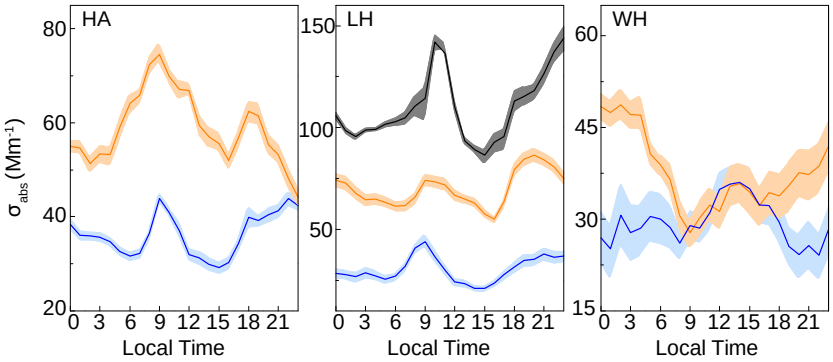

Fig. R3 Diurnal variations of BC absorption coefficients. (The figure has been added as Fig. S7 in the supplementary materials of the revised version)

In the revised manuscript, we have revised this part as following:

Figure 6 and Fig. S7 shows the diurnal variations of eBC and absorption coefficients under different air quality. The diurnal cycles of black carbon and absorption showed similar variation patterns. The BC mass concentrations were discussed here.

7. Line 271, You say ". . .combustion (traffic) and agricultural burning are higher than those from industrial emissions such as manufacturing and mineral products. But you give an example for the lower ratios from residential wood combustion, which is not an industrial source.

AR: Thanks for your comment. We have carefully checked the reference (Chow et al., 2011). The ratios of BC/$PM_{2.5}$ from mobile sources and area sources were generally higher than that from industrial sources and we have revised this part as following:

Generally, the ratios of BC/$PM_{2.5}$ from mobile sources (0.059-0.74) and area sources (0.032-0.33) were higher than that from industrial sources (0.0046-0.03). For instance, the mobile sources hold the highest ratios of BC/$PM_{2.5}$ (0.33−0.77) and the cement kiln showed lower ratio (0.03) (Chow et al., 2011).

References

Chow, J. C., Watson, J. G., Lowenthal, D. H., Antony Chen, L.-W. and Motallebi, N.: $PM_{2.5}$ source profiles for black and organic carbon emission inventories, Atmos. Environ.,, 45(31), 5407–5414, doi:10.1016/j.atmosenv.2011.07.011, 2011.

8. L291, It would be more interesting if you discuss whether the BC at downwind sites is more aged because of the transportation, because you say that ". . .the BC/CO is used to reflect the BC aging during the transport.

AR: We feel sorry that we did not discuss the aging of BC during the transport by BC/CO ratio. We have tried to discuss the BC/CO ratio using the same method in section 3.6, however, the low data resolution of CO (1-hour) would cause large uncertainty. So, we used the AAE instead of BC/CO ratio to discuss the BC aging during the transportation from upwind to downwind site. The decreasing of AAE from upwind to downwind site suggested that the BC was aged during the transportation and more details can be found in section 3.6.

9. Line 307, ". . ...The same result was also found at WH. High level of BCbb was due to more biomass burning in the southeast direction of HA and WH. . ."How do you know that more biomass burning in the southeast? Do you have some evidences to support your statement?

AR: We draw this conclusion because there were more fire spots in the southeast direction of WH during the study period as shown in Fig. R4. We have revised this part.

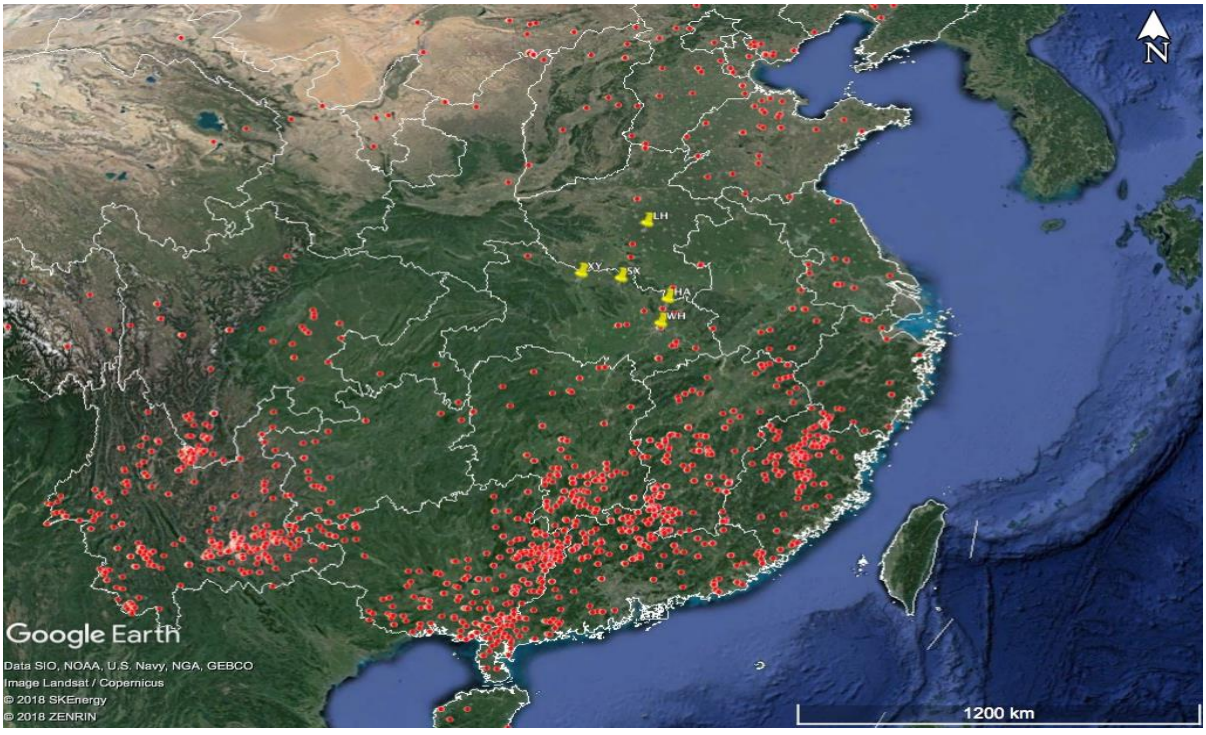

Figure R4 Locations of the fire spots downloaded from MODIS during the observation period (2018/1/08~2018/1/25).

10. Section 3.5, The paragraph may need to revise very carefully for that the current statements on the influences of the air parcels (CWT analysis) on each site are too trivial and wordy to understand. The authors are suggested to simulate the emissions and to quantify the intra-regional contributions at the sites based on the measured BC concentration data by using regional models.

AR: Thanks for your suggestion, we have tried Geos-Chem model to simulate the intra-regional transport contribution. The settings of Geos-Chem are described as the following part:

We use the nested GEOS-Chem model for China (version 11-01, http://wiki.seas.harvard.edu/geos-chem/index.php/Main_Page) to simulate the surface BC concentration. Driven by the GEOS-FP assimilation meteorology from the Goddard Earth Observing System (GEOS) of the NASA Global Modeling and Assimilation Office, the nested model has a horizontal resolution of 0.3125° longitude × 0.25° latitude with 47 vertical layers, and the lowest 10 layers are of ~130 m thickness each. The lateral boundary conditions of nested model are taken every 3 hours from a global GEOS-Chem simulation at 2.5° long × 2° lat horizontally. Spin-up time for nested model and global model are 15 days and one month, respectively. The scheme of planetary boundary layer employs a non-local scheme following Lin et al. (2010). Model convection is simulated with the relaxed Arakawa–Schubert scheme (Rienecker et al., 2008). Both the global and nested GEOS-Chem models are run with the NO_x-O*x*-hydrocarbon-aerosol-bromine tropospheric chemistry mechanism with online aerosols. Aerosols simulated by model include secondary inorganic aerosols (SIOA, including sulfate, nitrate and ammonium), secondary organic aerosols (SOA), primary organic aerosols (POA), black carbon (BC), dust and sea salts.

Monthly gridded anthropogenic emissions in China are taken from the Multi-resolution Emission Inventory for China (MEIC, www.meicmodel.org; Geng et al., 2017) of 2016 for nitrogen oxides (NO_x), carbon monoxide (CO), sulfur dioxide (SO_2), BC and POA. Following Zhang et al. (2015), emissions of anthropogenic fine dust are also included as primary $PM_{2.5}$ excluding BC and POA from MEIC in 2012. Biomass burning emissions are taken from the monthly GFED4 datasets (Giglio et al., 2013). Biogenic emissions of NMVOC follow MEGANv2.1 (Guenther et al., 2012). Soil emissions of NO_x employ the parameterization from Hudman et al. (2012).

Control and two sensitivity simulations were also done to study the regional transport contribution. The settings of control simulation were described above, and the sensitivity simulation were done with the emissions from Hubei and Henan province being closed, respectively.

The simulated and observed time series of BC at the five sites are shown in the Fig. R5. The Pearson coefficients (*r*) and NMB ranged from -0.44 to 0.07 and -39.9 to 19.8%, respectively, which suggested that the Geos-Chem is not good enough to reconstruct the BC variation in this study. There are several reasons: (1) the emission inventory uncertainty due to activity data, emission factors for energy-related combustion; burned area, fuel load and combustion completeness and emission factor for open burning emissions (Bond et al., 2013); low temporal resolution (i.e., monthly in this study); (2) the uncertainty of the input reanalysis meteorological field (i.e., in this study); (3) simple physical-chemical mechanism of BC in code, etc. More accurate and quantitative modeling for regional transportation of BC should be done after the above problems improved in the future.

Considering the poor simulation result, the Geos-Chem results were not adopted in the revised manuscript. Additionally, we carefully revised section 3.5 as the following:

Employing CWT method, the potential geographic origins of eBC for the five sites were explored (Fig. S11). Overall, CWT results of eBC at the five sites suggested that high eBC levels were found both in the north and south directions of LH and WH, while the high levels (i.e., > 4 μg m^{-3}) of eBC were only found from northeast directions of HA, SX and XY (Fig. S11). Additionally, the potential geographic source regions of BC_{bb} and BC_{ff} at HA, LH and WH were also discussed

as shown in Fig. 10. At HA, the CWT results showed that high levels of eBC (i.e., > 3 μg m^{-3}) were from north/northeast direction. However, the hot spots of BC_{bb} and BC_{ff} were different, with higher levels of BC_{bb} from both south and north directions and higher levels of BC_{ff} from the north direction. Also, higher levels of BC_{bb} and BC_{ff} were found in the south of LH. Opposite to the CWT results at HA, the hot spots of BC_{bb} was only found in the southeast direction of WH and high levels of BC_{ff} were found in the north and south directions of WH. The CWT results at WH were in line with the CBPF plots in section 3.4. The unity of CWT and CBPF results at WH suggested that there were intensive biomass burning activities in the south direction of WH during the observation period, which was verified by the MODIS fire-points distribution (Fig. S10).

We also discussed the source region differences of BC under different air quality (Fig. 11). The higher levels (>1 μg m^{-3}) of eBC, BC_{bb} and BC_{ff} were mainly from the south direction of three sites when the air was clean, while during the pollution episodes, air parcels from the north direction contributed high concentrations. For instance, at WH, high levels of eBC (> 2.5 μg m^{-3}) were found from south direction, while the source regions with high level eBC (> 3 μg m^{-3}) switched to northeast direction when the air quality was worsened. Figure 12 shows the semiquantitative results of transportation contribution results during clean and pollution episodes. At the boundary sites (HA, SX and XY), BC was mainly from south direction (accounting for 46.0−58.2%) when the air quality was clean, and it was mainly from northeast/northwest directions (51.2−76.5%) when the air quality getting worse. At SE-NCP site (LH), BC was dominantly from south direction (47.8%) during pollution episodes. At CC site (WH), BC was mainly from northeast direction (49.3−71.1%). These results suggested that northwest and northeast directions were the main transport pathways of air pollutants reaching to WH during the pollution episodes. Furthermore, to control local emissions during haze episodes, the emission sources, i.e., industry plant and open biomass burning in the upwind direction should also be controlled to prevent the further deterioration of air quality in downwind areas.

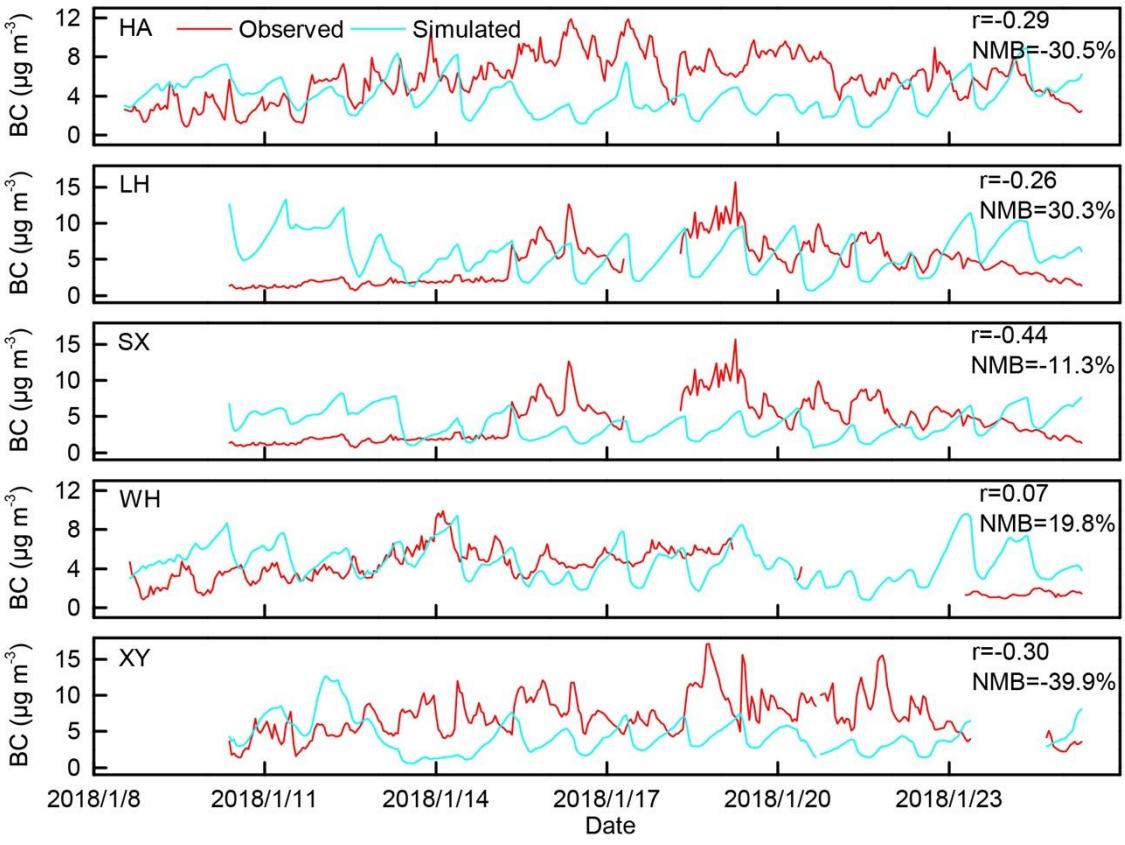

Figure. R5 Time series of observed and simulated BC concentrations from Goes-Chem model during the study period.

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11. Line 367, ". . .the travelling time (aging time) from LH to HA and from HA to LH were 28 h and 31 h, respectively, which suggested that the BC particle should be coagulated through complex atmospheric processes. Therefore, the new emission inputs along the trajectory enhanced the eBC mass concentration during the transport. . .." How do you infer that "the new emission" enhanced the eBC mass concentration from the previous sentence (longer aging time and coagulation processes) here?

AR: We have revised this part as the following:

Atmospheric removal of BC occurs in a few days to weeks via wet and dry depositions or contact with surfaces (Bond et al., 2013). In these two cases, there were no precipitation events and the transport time was short (i.e., 28 and 31h), which suggested the less removal rates. Therefore, the new emission inputs along the trajectory enhanced the eBC mass concentration during the transport

Previous study found that the BC coagulation with non-refractory materials becomes more significant when the aging timescale was greater than 10 h (Riemer et al., 2004). Chamber studies and field observations also found that the BC

absorption enhancement under polluted urban ambient air (Peng et al., 2016, Zhang et al., 2018, Wang et al., 2018c), suggesting the role of aging in modifying BC optical properties. In these two cases, the travelling time (aging time) from LH to HA and from HA to LH was 28 h and 31 h, respectively, which suggested that the BC particle should be coagulated through complex atmospheric processes. Therefore, the σ_{abs} was found increased from upwind to downwind site. On the contrary, the AAE values were found decreased during the transport. The AAE is sensitive to the particle size. A lab combustion experiment showed that the particles with smaller diameter from fresh biomass burning have lower AAE value than larger particles (Singh et al., 2016). Simulation also confirmed that the AAE of BC particle decreased with the increasing of its geometric median diameter (Liu et al., 2018b). Therefore, the diameter of BC particle increased during the transportation due to the aging processes supported by the increased absorb coefficients and decreased AAE as discussed above.

12. Line 370, "...However, slight differences found for BCbb transport: BCbb increased from LH (1.28 ± 0.06 μg m−3) to HA (2.57 ± 0.47 μg m−3), while BCbb decreased from HA (2.37 ± 0.23 μg m−3) to LH (2.14 ± 0.14 μg m−3). . ." What do you mean about this?

AR: In this sentence, we want to express the BC emission difference in these two regions. In case 1, air masses transported from LH (Henan province in north direction) to HA (Hubei province in south direction), both the eBC and BC_{bb} increased However, in Case 2, air masses transported from HA to LH, despite the eBC increased, the BC_{bb} decreased due to less BC emissions in Hubei province than those in Henan province. BC emission inventories also showed this difference (Qin and Xie, 2009, Qiu et al., 2016).

References

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