Dear editor,

Thank you for your kind considerations on our manuscript entitled "Aerosol-meteorology feedback diminishes the trans-boundary transport of black carbon into the Tibetan Plateau" (egusphere-2023-252). We appreciate that you gave us a chance to improve our manuscript to a level suitable for publication in ACP. We also want to express our deep thanks to the reviewers of the positive comments. Those comments are all valuable and very helpful for revising and improving our paper. We have studied comments carefully and have made corrections, which we hope meet with approval. The main corrections in the paper and the responds to the reviewer's comments are as following:

Answers to reviewers:

Reviewer #1:

The manuscript reported a record-breaking aerosol pollution event in the TP. Cross-boundary transport along the south slope the Himalayas from the surrounding regions was clearly the cause, implying glacier melt and ecological environment disturbance for the TP. Due to the extremely scarce availability of field observations, however, cross-boundary transport flux, meteorological pattern delivering aerosol, and the aerosol-meteorology feedback have rarely been discussed. The manuscript clearly shows strong aerosol-meteorology feedback on the transport flux of aerosols. The strong feedback on meteorology and aerosol distribution are also discussed in details. I hence recommended this manuscript for publication in ACP.

Firstly, we appreciate that you gave us a chance of revision to improve our manuscript to a level suitable for publication in Atmospheric Chemistry and physics. We also want to express our deep thanks to your positive comments. The comments are replied as follows:

1. The measurement distinguishes this manuscript and also a previous one, i.e., zhang et al., 2020, written by the key authors from a pure model simulation. However, these major results on transport flux and the aerosol-meteorology feedback are merely model simulations? Whether do the observation here or in other places reflect the pattern of transport flux or the aerosol-meteorology feedback pattern on transport flux?

Response: Thank you for your valuable suggestion. As the reviewer stated, the major results on transport flux and the aerosol-meteorology feedback are from model simulations. Because known as the 'Third Pole', the Himalayas and the Tibetan Plateau (TP) have very limited observational dataset due to harsh environment, limited access for fieldwork, and the sparsity of fixed instrumental stations.

Through the literature research, it is found that there are studies using observational dataset to reflect the transport flux of aerosols. For example, using ground-based multi-axis differential optical absorption spectroscopy (MAX-DOAS) at the Nancheng site in suburban Beijing on the southwest transport pathway of the Beijing-Tianjin-Hebei (BTH) region, Hu et at. (2022) estimated the vertical profiles of transport fluxes in the southwest-northeast direction. The results showed that the maximum net transport fluxes per unit cross-sectional area, calculated as pollutant

concentration multiply by wind speed, of aerosol extinction coefficient (AEC), NO_2, SO_2 and HCHO were 0.98 $km^{-1}m\ s^{-1}$, 24, 14 and 8.0 μg $m^{-2}\ s^{-1}$ from southwest to northeast, which occurred in the 200–300 m, 100–200 m, 500–600 m and 500–600 m layers, respectively, due to much higher pollutant concentrations during southwest transport than during northeast transport in these layers. The average net column transport fluxes were 1200 $km^{-1}\ m^2\ s^{-1}$, 38, 26 and 15 mg $m^{-1}\ s^{-1}$ from southwest to northeast for AEC, NO_2, SO_2 and HCHO, respectively, in which the fluxes in the surface layer (0–100 m) accounted for only 2.3%–4.2%.

However, in terms of the influence of aerosol-meteorology feedback on transport flux of aerosols, it is found that no matter in regions with abundant observational data or in regions with sparse observational data, the influence of aerosol-meteorology feedback on the transport flux of aerosols was evaluated by means of model simulation, because sensitivity tests are involved in such studies. For instance, Huang et al. (2020) suggested that the aerosol-meteorology interaction and feedback enhanced the trans-boundary transport of pollutants between the North China Plain and the Yangzi River Delta regions and thus exacerbated the haze levels in these two regions simultaneously, which was published in nature geoscience.

2. Although observation-model comparison on BC concentration and AOD have been conducted, these results do not fully justify the simulated transport flux pattern? More regious comparisons also transport flux among observations/reanalysis and models, or intermodel comparison on transport flux, might be helpful?

Response: Thank you for your good advice. The reviewer made a very good point here. According to the reviewer's suggestion, we not only validated the model performance on temporal variation in AOD at different stations by comparing the simulated AOD with the ground-based and satellite-based observational AOD (Figure S4), but also verified the model performance on the spatial distribution of AOD over the study area by comparing the simulated AOD with the satellite-based and reanalyzed AOD (Figure S5). However, for BC, the comparison between reanalysis and simulation was only conducted because we have very limited in-situ observed BC (the observed BC is only available at the QOMS station). According to the reviewer's suggestion, we further validated the model performance by conducting the inter-comparison among in-situ observation, simulation, and MERRA-2 reanalysis, as shown in Figure A1. The results show that the temporal variation in simulation is very close to that of simulation. Moreover, the correlation coefficient between the simulation and observation is 0.867, passing the 99% confidence level. Therefore, the model configuration used in this study presented a reasonable performance on BC. For the spatial distribution of BC over the study area, we compared the simulation with reanalysis from MERRA-2 and simulation from CAM_Chem. The results show that the spatial pattern of the WRF-Chem simulated BC is similar to that of the reanalyzed BC (Figure S6); however, the spatial pattern of BC from CAM_Chem is not reasonable (Figure A2).

Additionally, in terms of the BC transport flux, as the spatial pattern of BC from CAM_Chem is not reasonable, we can't further verify the transboundary transport flux of BC with CAM_Chem data. Also, the BC from MERRA-2 has no vertical information,

resulting in the inability to provide vertical profile of BC transport flux. Therefore, the transport flux of BC was not verified by inter-model comparison.

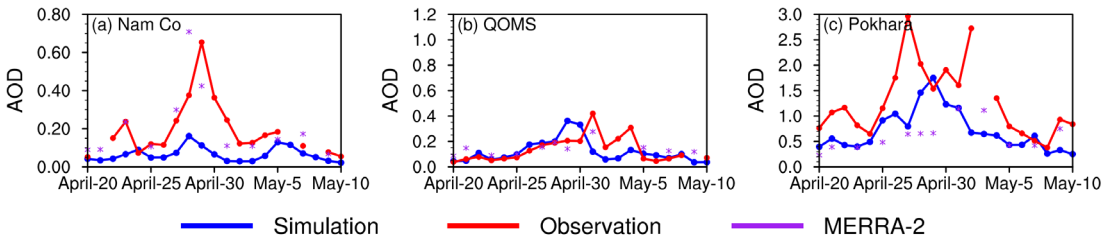

Figure S4. Inter-comparison of temporal variations in simulated AOD and ground-based as well as satellite-based AOD at (a) Nam Co, (b) QOMS, and (c) Pokhara stations for the period from April 20 to May 10, 2016.

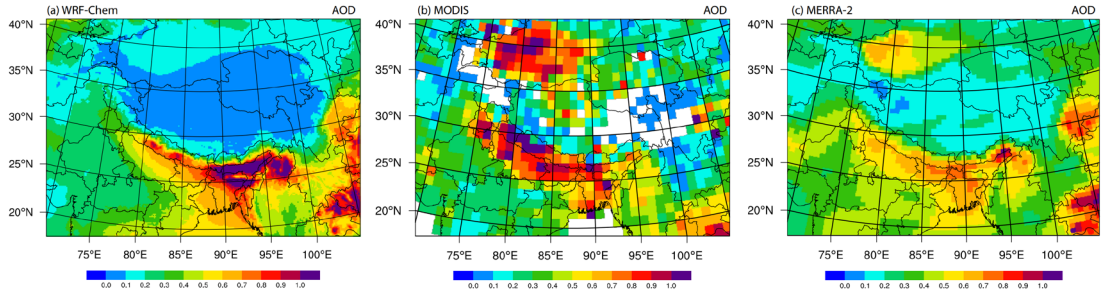

Figure S5. Inter-comparison of spatial distribution of simulated mean daily AOD and satellite-based as well as reanalyzed mean daily AOD from April 20 to May 10, 2016, over the study area.

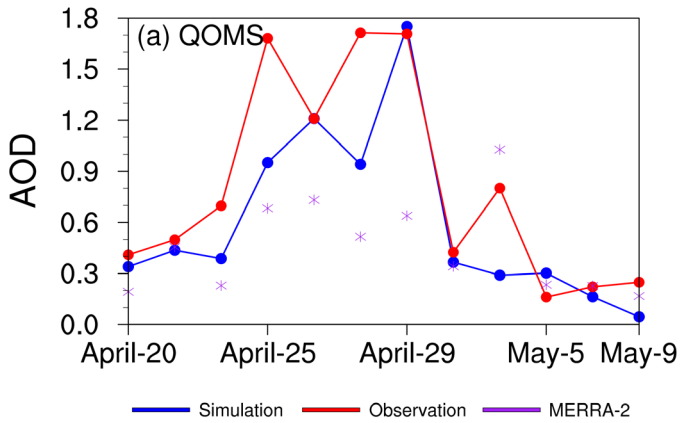

Figure A1. Inter-comparison of temporal variations in simulated BC and in-situ observed BC as well as reanalyzed BC at QOMS station for the period from April 20 to May 10, 2016.

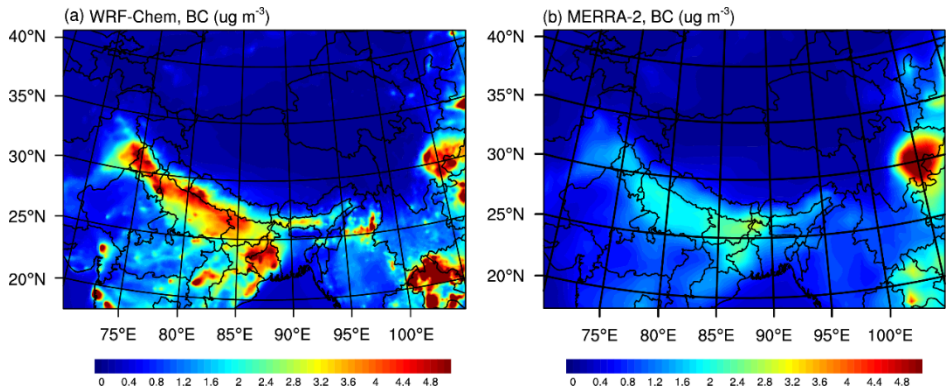

Figure S6. Spatial distributions of simulated and reanalyzed daily mean BC concentrations over the domain averaged for the period from April 20 to May 10, 2016.

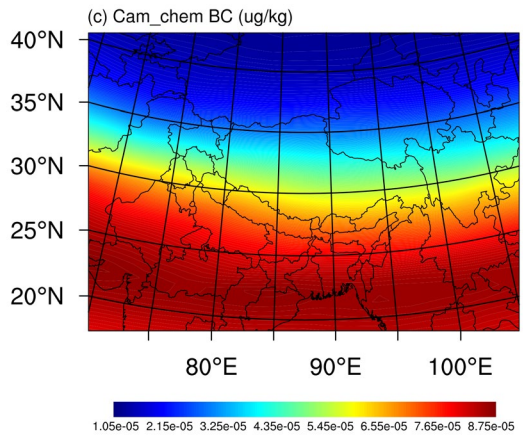

Figure A2. Spatial distribution of daily mean BC from CAM_Chem data averaged for the period from April 20 to May 10, 2016.

3. The WRF-Chem experiments take advantage of the record-breaking aerosol pollution event and run with aerosol-meteorology feedback on or off. Is there any observational evidence that aerosol-meteorology feedback changes the distribution or transport flux of BC? Are there model evidences that aerosol-meteorology feedback model better capture the observation of BC concentration or transport flux?

Response: Thank you very much for your valuable advice. Aerosol-meteorology interactions can change surface aerosol concentration via different mechanisms such as altering radiation budget or cloud microphysics. Although most previous works associated with the effect of aerosol-meteorology interaction on air pollution were mainly based on model simulation, there are observational evidence that aerosol-meteorology feedback could change the distribution or transport flux of air pollutants as well as model evidence that aerosol-meteorology feedback model better capture the observation of BC concentration or transport flux. For instance, based on multiyear measurements and reanalysis meteorological data, Huang et al. (2018) gave observational evidences on aerosol-meteorology interaction and its impact on pollution aggravation. They found a significant heating in upper planetary boundary layer with maximum temperature change about 0.7 °C on average and a substantial dimming near

surface with a mean temperature drop of 2.2 °C under polluted condition. Both observations and simulations using multiple models suggested that light-absorbing aerosols, like black carbon, exert crucial parts on such interaction. Moreover, both observations and simulations imply that increased stability caused by aerosol-meteorology interaction may continue to influence the atmospheric stratification and deteriorate the pollution on the next day. Additionally, Zhang et al. (2018) quantified the enhancement of $PM_{2.5}$ concentrations by aerosol-meteorology feedback in China in 2014 for different seasons and separate the relative impacts of aerosol radiation interactions (ARIs) and aerosol-cloud interactions (ACIs) by using the WRF-Chem model. They found that ARIs and ACIs could increase population-weighted annual mean $PM_{2.5}$ concentrations over China by 4.0 μg/m^3 and 1.6 μg/m^3, respectively. Also, Huang et al. (2020) reported that long-range transport and aerosol–meteorology feedback may interact rather than act as two isolated processes as traditionally thought by investigating typical regional haze events in northern and eastern China. This interaction can then amplify transboundary air pollution transport over a distance of 1,000 km and boost long-lasting secondary haze from the North China Plain to the Yangtze River delta. The results show an amplified transboundary transport of haze by aerosol–meteorology interaction in China and suggest the importance of coordinated cross-regional emission reduction with a focus on radiatively active species like black carbon. The study was performed by designing sensitivity experiment with WRF-Chem model. Taken together, there are observational and model evidences that aerosol-meteorology feedback could change the distribution or transport flux of BC.

4. Zhang et al., 2020 suggest that model resolving more valleys and mountains better capture valley transport and overall cross-Himalayan transport. As the authors have discussed potential weakness in current 15 km resolution model, will the current model be satisfying in simulating the aerosol-meteorology feedback?

Response: Thank you very much for your valuable advice. Although the WRF-Chem model with a horizontal resolution of 15 km×15 km used in this study is coarser than that of the study conducted by Zhang et al. (2020), the horizontal resolution of 15 km is overall satisfying in simulating aerosol-meteorology feedback. Because numerous previous modeling studies on aerosol-meteorology feedback have a horizontal resolution of 20 km or even coarser (Hu et al., 2022;Zhang et al., 2018;Li et al., 2022;Bharali et al., 2019;Gao et al., 2015;Huang et al., 2020). Considering that the topography of the TP is more complex than that of other regions, we use a relatively finer resolution of 15 km other than 20 km or even courser of other studies. Moreover, the WRF-Chem model with a horizontal resolution of 15 km had already been used to investigate the aerosol-meteorology feedback over the TP and its surrounding regions in a previous study (Yang et al., 2017). Therefore, a horizontal resolution of 15 km used in this current study is overall satisfying.

5. From Figure 7 & Figure 13, the aerosol-meteorology feedback does not simply lower the transport flux as written in the title?

Response: Thank you for your valuable suggestion. Figure 7 shows the longitudinal distribution of vertically integrated BC mass flux along the cross section in Figure 2 from simulation with aerosol-meteorology feedback, while Figure 13 depicts the difference in longitudinal distribution of vertically integrated BC transport flux along the cross section in Figure 2 from simulations with and without aerosol-meteorology feedback. Therefore, the impact of aerosol-meteorology feedback on BC transport flux is presented in Figure 13. Because northwestern South Asia contributes more BC to the TP via cross-Himalayan transport during the severe aerosol pollution event and the largest BC transport flux occurs at mountain valley in western Himalayas. In other words, the transboundary transport of BC towards the TP mainly occurred in the central and western Himalayas. Moreover, the interaction between aerosol and meteorology mainly occurred in the atmospheric planetary boundary layer. Therefore, from Figure 13, it is obvious that, from 72 °E to 92 °E in the central and eastern Himalayas, the BC transport flux induced by aerosol-meteorology feedback is almost negative, indicating that the aerosol-meteorology feedback in the central and eastern Himalayas does reduce the transport flux of BC towards the TP.

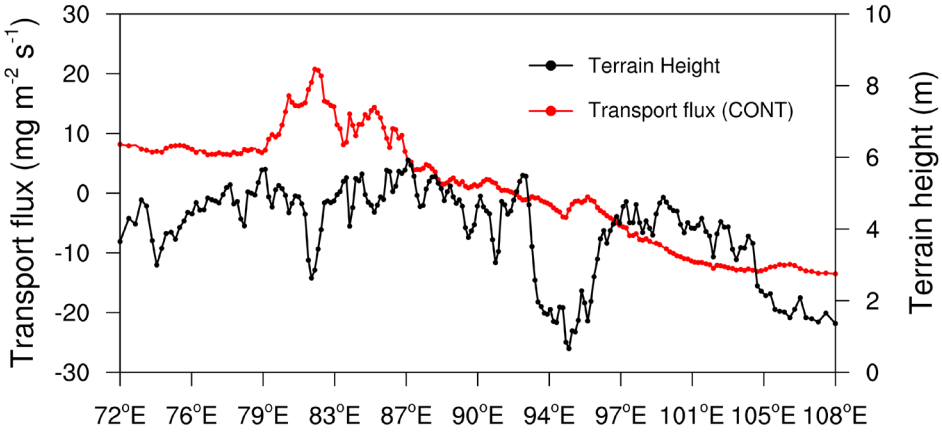

Figure 7. Longitudinal distribution of vertically integrated BC mass flux (red line) along the cross section in Figure 2 from simulation with aerosol-meteorology feedback. The black line represents the terrain height.

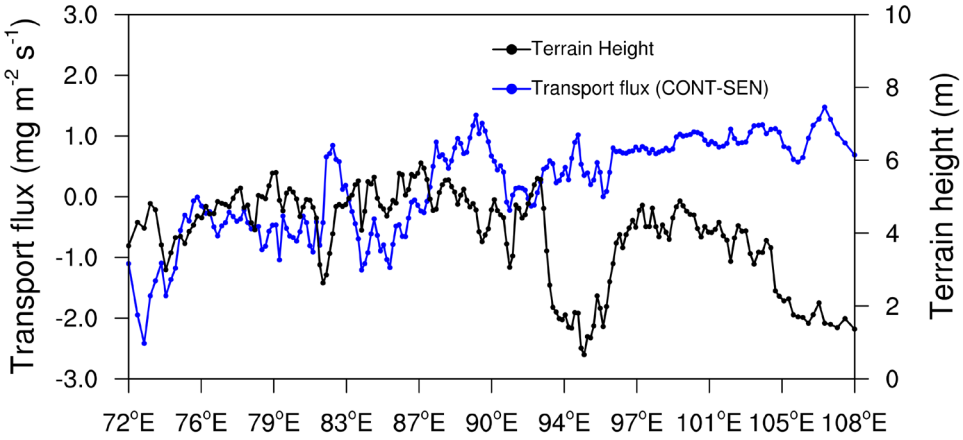

Figure 13. Difference in longitudinal distribution of vertically integrated BC transport flux along the cross section in Figure 2 from simulations with and without aerosol-meteorology feedback. The black line represents the terrain height.

6. The authors know clear the uncertainties in a pure model simulation (in lines 906-917). A discussion on wanted future experiments to constrain these uncertainties would be nice.

Response: Thank you very much for your valuable advice. Aerosol direct radiative forcing (DRF) depends critically on many assumptions about the aerosol mass concentration, size, shape, optical properties, and mixing state that affect aerosol optical depth (AOD), single scattering albedo (SSA), and asymmetry parameter. SSA variations of 11% may change the sign of DRF from negative to positive (Jethva et al., 2014). The most important factor of uncertainty in the calculation of AOD and SSA is the assumption of the aerosol mixing state (Curci et al., 2015). Curci et al. (2019) compared an ensemble of regional models over Europe and North America and found that the absolute error in simulating SSA is a few percent, but the sign of the bias has a certain dependence on the aerosol mixing state assumption. Therefore, the aerosol direct effect is very sensitive to the mixing state between scattering aerosols and absorbing aerosols. The representation of how chemical species are mixed inside the particles (the mixing state) is one of the major uncertainty factors in the assessment of these effects. It is thus recommended to focus further research on a more accurate representation of the aerosol mixing state in models, in order to have a less uncertain simulation of the related optical properties. Generally, there are three aerosol mixing assumptions, including external, internal (BC-core surrounded by well mixed scattering-shells) and partially internal mixtures (32.2% of sulfate and nitrate, 35.5% of BC and 48.5% of OC were internally mixed). Previous study indicated that core-shell internal mixing representation produces the most accurate absorption AOD and SSA at Aerosol Robotic Network (AERONET) Sun photometers site observations dominated by carbonaceous absorption (Tuccella et al., 2020). Therefore, in the future, we plan to improve the simulation accuracy by modifying the aerosol mixing state in the model. In addition, our results are based on a severe aerosol pollution event over a short period, and studies with longer duration are desirable in the future to test whether the results obtained from this severe aerosol pollution event are universal.

The last paragraph has been revised as follows:

There are still uncertainties in this study. Because the aerosol direct effect is very sensitive to the mixing state between scattering aerosols and absorbing aerosols and the aerosol feedback derived from the aerosol radiative effect has large impacts during the daytime. By analyzing the model performance on aerosols, we find that the WRF-Chem model exhibited an underestimation for AOD in this study. This underestimation may have important effect on aerosol feedback during the most polluted period. Similarly, the BC transport flux quantified by WRF-Chem model also has bias to some extent. However, with very limited observational data over the TP, numerical model is the best tool for this study. Therefore, we plan to focus further research on a more accurate representation of the aerosol mixing state in models, in order to have a less uncertain simulation of the related optical properties. Also, to improve the model performance, emissions with higher resolution and model with finer horizontal resolution will be used.

In addition, we note that our results are based on a severe aerosol pollution event over a short period, and studies with longer duration are desirable in the future to test whether the results obtained from this severe aerosol pollution event are universal.

Once again, special thanks to you for your good comments.
Best Regards.

Yuling Hu and Shichang Kang on behalf of all co-authors.

Reviewer #2:

The interaction between aerosols and meteorology, and its impact on the cross-boundary transport flux of BC (black carbon) over the Tibetan Plateau (TP), has received limited attention in previous research. This paper presents a comprehensive investigation of the aerosol-meteorology feedback and its influence on BC transport flux during a period of heavy aerosol pollution. The study utilizes WRF-Chem simulation to thoroughly analyze the phenomenon. Additionally, the paper elucidates the meteorological factors that contribute to the occurrence of severe aerosol pollution events over the TP. The concept introduced in this article is characterized by its novelty, and the study's findings hold significant implications for the preservation of the TP's ecological environment. Hence, I recommend that this manuscript be revised and considered for publication in ACP. Please find below some specific comments for further improvement:

Firstly, we appreciate that you gave us a chance of revision to improve our manuscript to a level suitable for publication in Atmospheric Chemistry and physics. The comments are replied as follows:

1. The authors validated the model performance on BC and AOD by comparing the simulation and observation. Although the comparison results are basically satisfactory, the data used to validate the model performance is still simple and I suggest inter-model comparison should be considered, which might be more convincing.

Response: Thank you very much for your valuable advice. The reviewer made a very good point here. According to the reviewer's suggestion, we not only validated the model performance on temporal variation in AOD at different stations by comparing the simulated AOD with the ground-based and satellite-based observational AOD (Figure S4), but also verified the model performance on the spatial distribution of AOD over the study area by comparing the simulated AOD with the satellite-based and reanalyzed AOD (Figure S5). However, for BC, the comparison between reanalysis and simulation was only conducted because we have very limited in-situ observed BC (the observed BC is only available at the QOMS station). According to the reviewer's suggestion, we further validated the model performance by conducting the inter-comparison among in-situ observation, simulation, and MERRA-2 reanalysis, as shown in Figure A1. The results show that the temporal variation in simulation is very close to that of simulation. Moreover, the correlation coefficient between the simulation and

observation is 0.867, passing the 99% confidence level. Therefore, the model configuration used in this study presented a reasonable performance on BC. For the spatial distribution of BC over the study area, we compared the simulation with reanalysis from MERRA-2 and simulation from CAM_Chem. The results show that the spatial pattern of the WRF-Chem simulated BC is similar to that of the reanalyzed BC (Figure S6); however, the spatial pattern of BC from CAM_Chem is not reasonable (Figure A2).

Additionally, in terms of the BC transport flux, as the spatial pattern of BC from CAM_Chem is not reasonable, we can't further verify the transboundary transport flux of BC with CAM_Chem data. Also, the BC from MERRA-2 has no vertical information, resulting in the inability to provide vertical profile of BC transport flux. Therefore, the transport flux of BC was not verified by inter-model comparison.

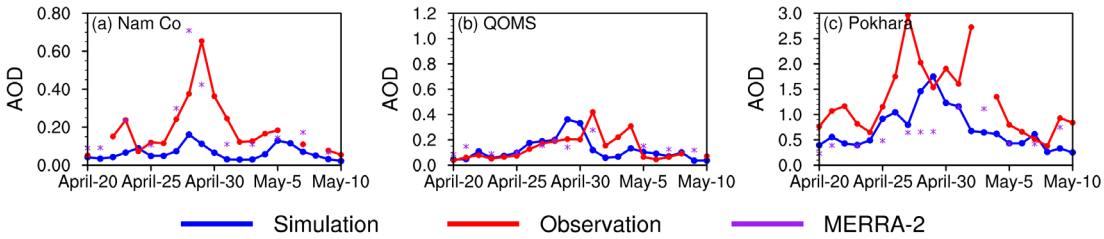

Figure S4. Inter-comparison of temporal variations in simulated AOD and ground-based as well as satellite-based AOD at (a) Nam Co, (b) QOMS, and (c) Pokhara stations for the period from April 20 to May 10, 2016.

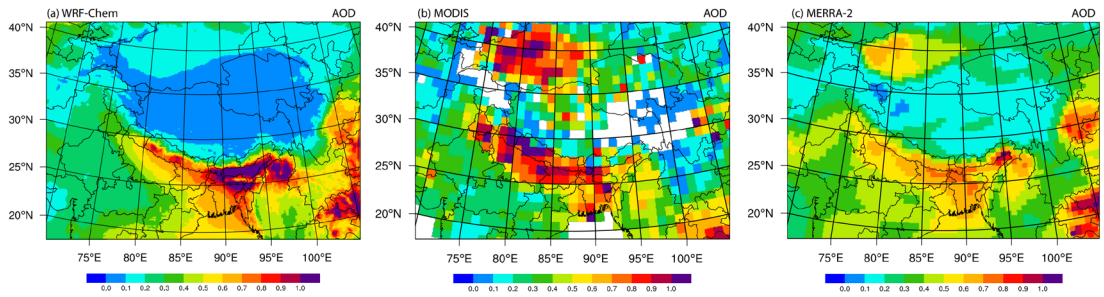

Figure S5. Inter-comparison of spatial distribution of simulated mean daily AOD and satellite-based as well as reanalyzed mean daily AOD from April 20 to May 10, 2016, over the study area.

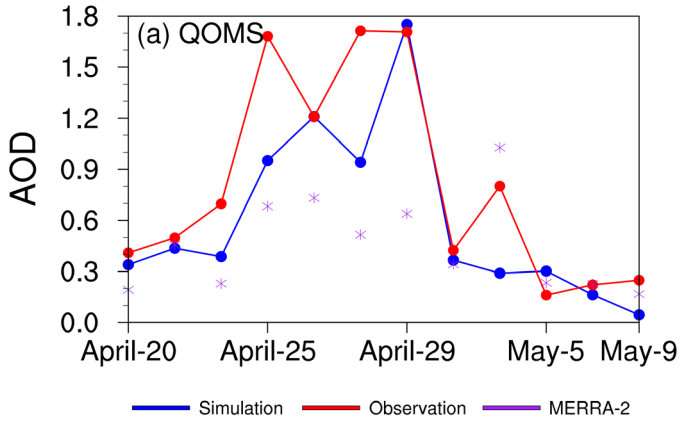

Figure A1. Inter-comparison of temporal variations in simulated BC and in-situ observed BC as well as reanalyzed BC at QOMS station for the period from April 20 to May 10, 2016.

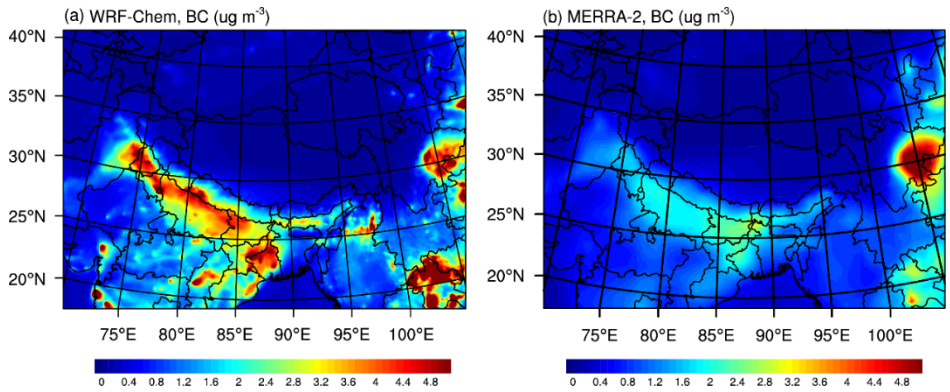

Figure S6. Spatial distributions of simulated and reanalyzed daily mean BC concentrations over the domain averaged for the period from April 20 to May 10, 2016.

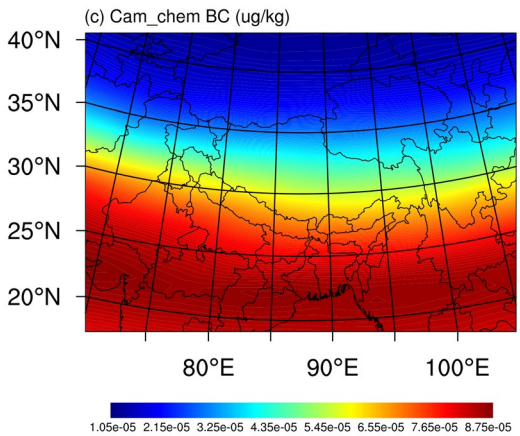

Figure A2. Spatial distribution of daily mean BC from CAM_Chem data averaged for the period from April 20 to May 10, 2016.

2. When analyzing the meteorological causes of the heavy aerosol pollution event, isotherms in the weather maps in Figure 3 are not included in the analysis, and isotherms

lead to blurring of potential heights and wind fields in weather maps, so I suggest removing them.

Response: Thank you very much for your kind remind. According to the reviewer's suggestion, we have removed the isotherms in the weather maps in Figure 3 and the replotted Figure 3 is shown as follows:

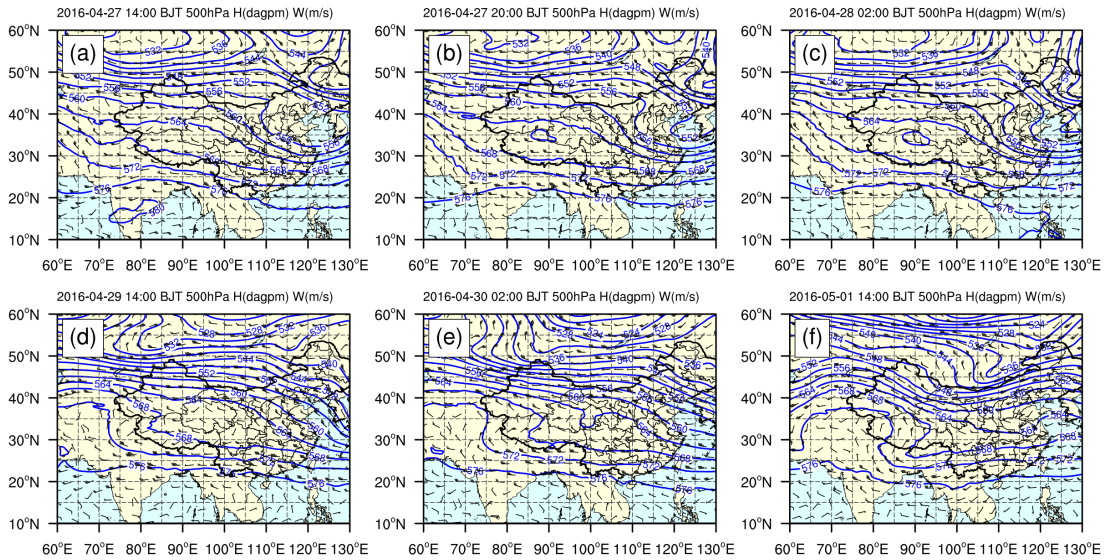

Figure 3 Weather maps at 500 hPa over the study area during the severe aerosol pollution event based on ERA-Interim reanalysis dataset. The blue lines are isopleths of geopotential height (unit: dagpm). Wind speed (unit: m/s) and direction are denoted by wind barb.

3. As the author stated in the title as well as in Figure.12, the aerosol-meteorology feedback decreased the cross-boundary transport flux of BC towards the TP. In fact, this conclusion is the result of pure model simulation because of the harsh environment, limited access for fieldwork, and the sparsity of fixed instrumental stations over the TP. So is there similar study in other places and What effect does the aerosol-meteorology feedback have on the transport flux of aerosols?

Response: Thank you very much for your good suggestion. This suggestion is somewhat similar to that made by the first reviewer. By reviewing extensive literature, it was found that there are similar studies in other places. For example, using ground-based multi-axis differential optical absorption spectroscopy (MAX-DOAS) at the Nancheng site in suburban Beijing on the southwest transport pathway of the Beijing-Tianjin-Hebei (BTH) region, Hu et at. (2022) estimated the vertical profiles of transport fluxes in the southwest-northeast direction. The results showed that the maximum net transport fluxes per unit cross-sectional area, calculated as pollutant concentration multiply by wind speed, of aerosol extinction coefficient (AEC), NO_2, SO_2 and HCHO were 0.98 $km^{-1}m\ s^{-1}$, 24, 14 and 8.0 μg $m^{-2}\ s^{-1}$ from southwest to northeast, which occurred in the 200–300 m, 100–200 m, 500–600 m and 500–600 m layers, respectively, due to much higher pollutant concentrations during southwest transport than during northeast transport in these layers. The average net column transport fluxes were 1200 $km^{-1}\ m^2\ s^{-1}$, 38, 26 and 15 mg $m^{-1}\ s^{-1}$ from southwest to northeast for AEC, NO_2, SO_2

and HCHO, respectively, in which the fluxes in the surface layer (0–100 m) accounted for only 2.3%–4.2%.

However, in terms of the influence of aerosol-meteorology feedback on transport flux of aerosols, it is found that no matter in regions with abundant observational data or in regions with sparse observational data, the influence of aerosol-meteorology feedback on the transport flux of aerosols was evaluated by means of model simulation, because sensitivity tests are involved in such studies. For instance, Huang et al. (2020) reported that long-range transport and aerosol–meteorology feedback may interact rather than act as two isolated processes as traditionally thought by investigating typical regional haze events in northern and eastern China. This interaction can then amplify transboundary air pollution transport over a distance of 1,000 km and boost long-lasting secondary haze from the North China Plain to the Yangtze River delta. The results show an amplified transboundary transport of haze by aerosol–meteorology interaction in China and suggest the importance of coordinated cross-regional emission reduction with a focus on radiatively active species like black carbon.

4. Line 949, Line 1090, Line 1176, Line 1213–1214, and Line 1227: ATMOSPHERIC CHEMISTRY AND PHYSICS --> Atmos. Chem. Phys.

Response: Thank you for your suggestion. We are very sorry for our carelessness, and "ATMOSPHERIC CHEMISTRY AND PHYSICS" has been revised as "Atmos. Chem. Phys".

5. Line 1128: JOURNAL OF GEOPHYSICAL RESEARCH-ATMOSPHERES --> Journal of Geophysical Research: Atmospheres

Response: Thank you for your suggestion. We have revised "JOURNAL OF GEOPHYSICAL RESEARCH-ATMOSPHERES" as "Journal of Geophysical Research: Atmospheres".

6. Line 1034–1035: NATURE CLIMATE CHANGE -->Nature Climate Change

Response: Thank you for your suggestion. We have revised "NATURE CLIMATE CHANGE" as "Nature Climate Change".

7. Line 1220: SCIENTIFIC REPORT--> Scientific Report

Response: Thank you for your advice. We have revised "SCIENTIFIC REPORT" as "Scientific Report".

8. Line 1221–1224: The corresponding article is quoted incorrectly and lacks the journal name.

Response: Thank you for your suggestion. The journal name has been added and the correct citation is 'Zheng, B., Zhang, Q., Zhang, Y., He, K. B., Wang, K., Zheng, G. J., Duan, F. K., Ma, Y. L., and Kimoto, T.: Heterogeneous chemistry: a mechanism missing in current models to explain secondary inorganic aerosol formation during the January 2013 haze episode in North China, Atmos. Chem. Phys., 15 (4), 2031-2049, https://doi.org/10.5194/acp-15-2031-2015, 2015.'.

Once again, special thanks to you for your good comments.
Best Regards.

Yuling Hu and Shichang Kang on behalf of all co-authors.

Bharali, C., Nair, V. S., Chutia, L., and Babu, S. S.: Modeling of the Effects of Wintertime Aerosols on Boundary Layer Properties Over the Indo Gangetic Plain, *J. Geophys. Res.: Atmos.*, *124* (7), 4141-4157, https://doi.org/https://doi.org/10.1029/2018JD029758, 2019.

Curci, G., Hogrefe, C., Bianconi, R., Im, U., Balzarini, A., Baró, R., Brunner, D., Forkel, R., Giordano, L., Hirtl, M., Honzak, L., Jiménez-Guerrero, P., Knote, C., Langer, M., Makar, P. A., Pirovano, G., Pérez, J. L., San José, R., Syrakov, D., Tuccella, P., Werhahn, J., Wolke, R., Žabkar, R., Zhang, J., and Galmarini, S.: Uncertainties of simulated aerosol optical properties induced by assumptions on aerosol physical and chemical properties: An AQMEII-2 perspective, *Atmos. Environ.*, *115*, 541-552, https://doi.org/https://doi.org/10.1016/j.atmosenv.2014.09.009, 2015.

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