Manuscript # acp-2021-328

Responses to Referee #1

Ren et al. assess the contributions from local emissions and transport to PM_{2.5} concentrations in Chinese regions during three periods when COVID-19 affected the socioeconomic activity of the country at the beginning of 2020. In principle, the topic is interesting and relevant, but I have major reservations concerning the chosen methods and the documentation thereof as well as the interpretation and discussion of the results.

We thank the editor for all the insightful comments. Below, please see our pointby-point response (in blue) to the specific comments and suggestions and the changes that have been made to the manuscript, in effort to take into account all the comments raised here.

The finding of regionally increasing $PM_{2.5}$ during the COVID-19 period is in light of lockdowns counterintuitive and needs a clearer discussion in the text. The authors state that it is due to transport from outside of China, but the quantified 4-10% of $PM_{2.5}$ transport into a Chinese region for polluted days and even the largest 40-66% regional contributions from transport during the lockdown, when local emission should be small, are no particularly convincing evidences, especially in light of the known poor model performance for $PM_{2.5}$ indicated by the authors.

Response:

Thanks for the suggestion. The polluted days are selected for "each receptor region" in China. Therefore, the large contribution from transboundary transport is only for some specific regions in China, e.g., Southwestern China. Also, the significant impacts from South and Southeast Asian emissions have been revealed in many previous studies.

We have now revised the sentences: "In this study, the most polluted day is defined as the day with the highest daily PM_{2.5} concentration in February 2020 for each receptor region in China. The transport from outside of China only has a great impact on some specific regions in China. In Southwestern China, the relative contribution from ROW emissions, especially those from South and Southeast Asia, to the increment of PM_{2.5} concentration during the most polluted days compared with normal days is more than 50%. It is consistent with the previous studies that emissions from South and Southeast Asia have an important impact on air quality in southwest China (Yang et al., 2017a; Zhu et al., 2016, 2017). For other receptor regions in China (Northeastern China, North China Plain, Eastern China, Southern China and Central-West China), PM_{2.5} concentrations are largely contributed by local emissions during the most polluted days compared with normal days. In the future with emissions reductions for better air quality in China, decreasing air pollution should consider aerosols from both Chinese local emissions and pollutant transport from outside of China."

The results need a more compelling interpretation, making better use of the knowledge of the impact of the meteorological conditions on $PM_{2.5}$, e.g., through a discussion in light of other studies. It would be useful to have a discussion section separate from the conclusions. This would allow to fully appreciate the limits and advances of this work compared to previous studies, and draw a clear and concise conclusion from this work.

Response:

Thanks for the suggestion. We have now included such context in the discussion section as follows: "Many studies have examined the importance of meteorology on regional air quality during the COVID-19 lockdown period and emphasized that, when meteorology is unfavorable, abrupt emissions reductions cannot avoid severe air pollutions (Le et al. 2020; Sulaymon et al. 2021; Shen et al. 2021). Through model simulations, Le et al. (2020) found that abnormally high humidity promotes the heterogeneous chemistry of aerosols, which have contributed to the increase of PM2.5 by 12% in northern China during the city lockdown period. Sulaymon et al. (2021) found that significant increase in PM_{2.5} concentrations caused by unfavorable meteorological conditions in Beijing-Tianjin-Hebei region during the lockdown period based on Community Multiscale Air Quality (CMAQ) model simulations. By analyzing the observational data and model simulations, Shen et al. (2021) reported that 50% of the pollution episodes during the COVID-19 lockdown in Hubei of China were due to the stagnant meteorological conditions. Huang et al. (2020) found that the stagnant air conditions and enhanced atmospheric oxidizing capacity caused a severe haze event during the same time period. In line with previous studies, we also revealed the stagnant air condition under the anomalous high pressure system in the most polluted day over the North China Plain. In addition to the meteorological conditions, the effect of foreign transport was also raised in this study causing aerosol pollution in southwestern China during COVID-19 outbreak."

Specific comments

L. 51: "in December 2019" - give the time period of the outbreak in China Response:

As the epidemic broke out one after another in different areas, the outbreak time is a continuous time. We have now revised the text as follows: "The coronavirus disease 2019 (COVID-19) has spread worldwide since December 2019 and resulted in more than one million cases within the first four months

(Sharma et al., 2020; Dong et al., 2020)."

L. 53-54: I recommend removing "was the first country" from the sentence since it is not relevant for the scientific content, but say instead when the measures began and ended since this is indeed relevant for the interpretation of your findings.

Response:

We have now revised the sentence to reflect this: "In order to curb the virus spread among humans, measures were taken by the Chinese government on January 23, 2020 to minimize the interaction among people, including strict isolation, prohibition of large-scale private and public gatherings, restriction of private and public transportation and even lockdown of cities (Tian et al., 2020; Wang et al., 2020)."

L. 62-63: revise sentence for clarity

Response:

We have now revised the sentence: "The estimated NOx emission in eastern China was reduced by 60-70%, of which 70-80% was related to the reduced road traffic and 20-25% was from industrial enterprises shutdown during the COVID-19 lockdown period. However, severe air pollution events still occurred in East China during the COVID-19 lockdown, even though the anthropogenic emissions were greatly reduced (Huang et al., 2020)."

L. 66: "change" -> changes Response: Revised.

L. 80-83: when did the haze occur? Does your simulation reproduce this event? Response:

In the study of Huang et al. (2020), the severe air pollution events occurred on February 11, 2020. Our model reproduced the pollution event at the same time and have now included such context in the discussion as follows: "When the PM_{2.5} pollution occurred in the North China Plain on February 11, 2020, which was also reported as the polluted day in observations (Huang et al., 2020), the concentration of PM_{2.5} was 16.1 μ g m⁻³ higher than that in normal days."

L. 114-116: If there are studies already, what does your work add to the previous knowledge?

Response:

In the original text, "few" studies have focused on aerosol transport pathways and source attribution in China during the COVID-19 pandemic. Since the studies about the air quality during COVID-19 are increasing, we have emphasized our study that "Our study provides source apportionment of aerosols covering the whole China and quantifies the contribution from foreign transport for the first time in the case of COVID-19 emission reductions."

L. 146: How many simulations did you perform over what time period? Response:

By adding an additional simulation in the revised manuscript, we now have two simulations with aerosol tagging but different emission assumptions: "The anthropogenic emissions used in the baseline simulation are derived from the MEIC (Multi-resolution Emission Inventory of China) inventory (Zheng et al., 2018), referred to here as the Baseline experiment. While emissions for the other countries use the SSP (Shared Socioeconomic Pathways) 2-4.5 scenario data set under CMIP6 (the Coupled Model Intercomparison Project Phase 6). Emissions in year 2017 are used as the baseline during the simulation period considering the time limit of MEIC inventory. To better estimate the impact of restricted human activities on emission reductions owing to COVID-19 lockdown (referred to as Covid experiment), we updated China's emission inventory from January to March 2020 based on the provincial total emission reduction ratio in Huang et al. (2020). Emissions from the transportation sector are decreased by 70%. The remaining emission reduction, by excluding transport reduction from the total emission reduction, are evenly distributed to other sectors, including industry, power plant, residential, international shipping and waste treatment from January to March 2020 compared to the baseline emission in 2017. Unless otherwise specified, all the results in this study are derived from the Covid experiment."

L. 150-151: There should be an argument why emissions from SSP2-4.5 are used here, even though more recent global emission data has been created (e.g., Lamboll et al., 2020)

Response:

When we conducted the experiments, the latest global emission data has not been published. Applying the emissions from SSP2-4.5 can better compare with the simulations of CMIP6, which has been widely used in many previous studies (Lund et al. 2019; Lyakaremye et al. 2021).

L. 157-160: How were these emission estimates created? Please illustrate the results for the emissions and compare them to other new emission data. What is meant by "remaining reductions"? Response:

The emission reductions due to COVID-19 lockdown were updated based on dynamic economic and industrial activity levels, which has been applied in the previous studies (Huang et al., 2020). Emissions from the transportation sector are decreased by 70%. The remaining emission reduction, by excluding transport reduction from the total emission reduction, are evenly distributed to other sectors, including industry, power plant, residential, international shipping and waste treatment from January to March 2020 compared to the baseline emission in 2017.

L. 166: "from April 2019 to March 2020 repeatedly for six years" this needs more words to explain what you did. How did you do for instance the initialisation? What is meant by repreating the simulation for six years? Response:

The simulations are integrated for 6 years with the first five years treated as model spin-up and the last year was analyzed.

L. 169: It would be more relevant to say which weeks had the most severe lockdowns and use this information to interpret the results. Response:

The lockdown was first implemented on January 23 in Wuhan, China. Subsequently, other regions in China took measures, and the lockdown of the whole country lasted for at least three weeks varying in different regions.

L. 191: What motivates the choice of these regions? Response:

The eight source areas are divided mainly according to the geographical location and subdivided on the basis of previous studies (Yang et al., 2017a).

L. 198-201: Were these nudged simulations to MERRA-2 as well? Then say so. Otherwise, it would be useful to say a few words on the performance of MERRA-2 over China as well.

Response:

Yes. Many studies have nudged the model wind fields toward the MERRA-2 reanalysis in China (Zhuang et al., 2018; Yang et al., 2017a).

L. 212: I appreciate and encourage the open communication of uncertainties in modeling. A 50% underestimation of PM_{2.5} is large. Given your focus on PM_{2.5} in this study, how can you nevertheless trust the simulation, especially taking into account that nitrate and ammonium are known to be poorly represented in the same model (L. 217)? You revisit this point in the last paragraph of the conclusions, but I also missed guidance for the concrete implication of it there. Response:

We have now added the sentence to reflect this: "In majority of the climate models, the simulation of nitrate and ammonium aerosols are not included in the aerosol schemes, partly due to the complexity of calculation efficiency. For example, in many of the CMIP6 models, only two of them provide nitrate and ammonium mass mixing ratios. Many previous studies have evaluated the global climate models performance in reproducing aerosol concentrations (e.g., Fan et al., 2018; Shindell et al., 2013; Yang et al., 2017a,b). In general, the models can well simulate aerosols in North America and Europe but significantly underestimates aerosols in East Asia by about -36 to -58 %

compared with observations. It can lead to an underestimation of aerosols contributed by Chinese local emissions in magnitudes, but might not change the main conclusions of this study."

L. 227: State here the three time periods and motivate this choice. Response:

We now have added a description as follows. "Figure 3b presents spatial distributions of simulated mean column burden of PM_{2.5} during the three time periods ('Week 1': January 30–February 5, 'Week 2': February 6–February 12 and 'Week 3': February 13–February 19), which had the largest number of newly-diagnosed COVID-19 cases."

L. 230 - 236: Say relative to what you make the comparisons. Response:

We have now revised the sentences: "Comparing to Week 3, Week 1 and Week 2 have higher $PM_{2.5}$ loading, with values in the range of 20–40 and 20– 30 mg m⁻² in the North China Plain, Eastern China, and Southern China, while the $PM_{2.5}$ loading in Week 3 is relative lower than Week 1 and Week 2 with values ranging mostly from 10 to 20 mg m⁻²."

L. 368: It would be helpful to state the date in the text, here and/or earlier. Response:

We now have added a description as follows. "When the $PM_{2.5}$ pollution occurred in the North China Plain on February 11, 2020, which was also reported as the polluted day in observations (Huang et al., 2020), the concentration of $PM_{2.5}$ was 16.1 µg m⁻³ higher than that in normal days."

L. 273-374: 4-10% transport from outside of China on the most polluted day means that local emissions dominate. Maybe explicitly add the implication of your findings.

Response:

Thanks for the suggestion. We have now included such discussions as follows: "The transport from outside of China only has a great impact on some specific regions in China. In Southwestern China, the relative contribution from ROW emissions, especially those from South and Southeast Asia, to the increment of PM_{2.5} concentration during the most polluted days compared with normal days is more than 50%. It is consistent with the previous studies that emissions from South and Southeast Asia have an important impact on air quality in southwest China (Yang et al., 2017a; Zhu et al., 2016, 2017). For other receptor regions in China (Northeastern China, North China Plain, Eastern China, Southern China and Central-West China), PM_{2.5} concentrations are largely contributed by local emissions during the most polluted days compared with normal days. In the future with emissions reductions for better air quality in China, decreasing air pollution should consider aerosols from both

Chinese local emissions and pollutant transport from outside of China."

Arrange the order of all figures following the order of references to them in the text.

Response:

Thank you for your reminding, we have reorganized the order of figures.

Figure 1: What time period is meant here? Response:

The time period here refers to the three weeks of the study from January 30 to February 19, which had the largest number of newly-diagnosed COVID-19 cases.

Figure 2: What do the colors mean? Response:

The color is to distinguish the different weeks. We have now moved this figure to the supplement.

Table 1: State the dates of the weeks.

Response:

We have now revised the sentences: "Table 1. Fractional contributions of emissions from nine tagged source regions (vertical axis) to mean PM_{2.5} column burden in eight receptor regions (horizontal axis) during the three time periods ('Week 1': January 30–February 5, 'Week 2': February 6–February 12 and 'Week 3': February 13–February 19)."

Reference:

- Fan, T., Liu, X., Ma, P.-L., Zhang, Q., Li, Z., Jiang, Y., Zhang, F., Zhao, C., Yang, X., Wu, F., and Wang, Y.: Emission or atmospheric processes? An attempt to attribute the source of large bias of aerosols in eastern China simulated by global climate models, Atmos. Chem. Phys., 18, 1395–1417, https://doi.org/10.5194/acp-18-1395-2018, 2018.
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Responses to Referee #2

The authors investigated aerosol transport pathways in China during COVID-19. They established the source-receptor relationships among various regions of China using the CAM5 model with the capability of aerosol source tagging. The model system was developed by the same group of this paper and was evaluated in their previous studies. This work suggests that local emissions contribute largely to the severe aerosol pollution in North China Plain and Eastern China during COVID along with moderate impacts from unfavorable meteorological conditions. Overall, this paper reads well and provides interesting results, which could benefit the design of air pollution regulation strategies in China. I have two major concerns about the manuscript in its current form, which need to be resolved before it can be accepted for publication.

We thank the editor for all the insightful comments. Below, please see our pointby-point response (in blue) to the specific comments and suggestions and the changes that have been made to the manuscript, in effort to take into account all the comments raised here.

The first problem is that the CAM5 model used in this work cannot simulate nitrate and ammonium aerosols, while these compositions account for a large proportion of aerosols over China currently. Please provide detailed explanations and discussions on how this model deficiency could impact the main conclusions of this work.

Response:

Thanks for the suggestion. We have now added the following sentences in the discussion section: "In majority of the climate models, the simulation of nitrate and ammonium aerosols are not included in the aerosol schemes, partly due to the complexity of calculation efficiency. For example, in many of the CMIP6 models, only two of them provide nitrate and ammonium mass mixing ratios. Many previous studies have evaluated the global climate models in reproducing aerosol concentrations (e.g., Fan et al., 2018; Shindell et al., 2013; Yang et al., 2017a, b). In general, the models can well simulate aerosols in North America and Europe but significantly underestimates aerosols in East Asia by about -36 to -58% compared with observations. It can lead to an underestimation of aerosols contributed by Chinese local emissions in magnitudes, but might not change the main conclusions of this study."

The second problem is that the focus of this work is the aerosol source

attribution during COVID. However, the authors did not discuss much the special findings in this special period. Compared to previous literature, are there any novel results and conclusions of the contributions from local/nonlocal sources to aerosol pollutions during this period with low emission levels? And what's the implication for air pollution control policies in China, especially considering that the anthropogenic emissions will be rapidly reduced in the future?

Response:

Thanks for the suggestion. We have now included such context in the discussion section as follows: "Source tagging and apportionment is an effective way to establish aerosol source-receptor relationships, which is conducive to both scientific research and emission control strategies (Yu et al., 2012). Previous studies only focused on regional transport of aerosols, very few studies have explored the aerosol transport pathways and source attribution covering the whole China during the COVID-19 pandemic. The COVID-19 pandemic disrupted human activities and lead to abrupt reductions in anthropogenic emissions. This study first investigated the source contributions to PM_{2.5} over various regions covering the whole China during the COVID-19 pandemic. We pay attention not only to local emissions, but also to the impacts from regional and foreign transport of aerosols."

In the revised manuscript, we added an additional experiment to better reflect variations of contributions from local/nonlocal sources to aerosol pollutions during this period with low emission levels. "The anthropogenic emissions used in the baseline simulation are derived from the MEIC (Multi-resolution Emission Inventory of China) inventory (Zheng et al., 2018), referred to here as the Baseline experiment. While emissions for the other countries use the SSP (Shared Socioeconomic Pathways) 2–4.5 scenario data set under CMIP6 (the Coupled Model Intercomparison Project Phase 6). Emissions in year 2017 are used as the baseline during the simulation period considering the time limit of MEIC inventory."

"To highlight the roles of regional and foreign transport, the differences between Covid and Baseline simulations in relative contributions to PM_{2.5} burden from local, region (RCN) and foreign (ROW) emissions are given in Figure S1. During the COVID-19 period, the local and RCN emission contributions to PM_{2.5} were 1–4% lower than that in Base experiment over NCP and NEC. In Eastern China, the contribution from the local emissions decreased by 3–4% compared with Base experiment, while the contribution from ROW increased by more than 5%. In Southern China, 50–70% of the PM_{2.5} burden is contributed by emissions from ROW in Base experiment. During the COVID-19 period with low emission levels, the contribution from ROW to PM_{2.5} burden in Southern China had an increase of more than 5%. It indicates that the important role of transboundary transport needs to be considered when controlling local emissions to improve air quality in the near future."



Figure S1. Relative contributions (%) in Baseline simulation (left) and differences in relative contributions (%) between Covid and Baseline simulations (right) of local emissions (top), the emissions from the rest of China (RCN) (middle) and all sources outside China (rest of the world, ROW) (bottom) to PM_{2.5} column burden in February 2020.

Reference:

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1	Aerosol transport pathways and source attribution in China
2	during the COVID-19 outbreak
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23 Abstract

Due to the coronavirus disease 2019 (COVID-19) pandemic, human 24 activities and industrial productions were strictly restricted during January-25 March 2020 in China. Despite the fact that anthropogenic aerosol 26 emissions largely decreased, haze events still occurred. Characterization of 27 aerosol transport pathways and attribution of aerosol sources from specific 28 regions are beneficial to the air quality and pandemic control strategies. 29 This study establishes source-receptor relationships in various regions 30 of covering the whole China during the COVID-19 outbreak based on the 31 Community Atmosphere Model version 5 with Explicit Aerosol Source 32 Tagging (CAM5-EAST). Our analysis shows that PM_{2.5} burden over the 33 34 North China Plain between January 30 and February 19 is largelymostly contributed by local emissions (40–66%). For other regions in China, PM_{2.5} 35 36 burden is largely contributed from non-local sources. During the most polluted days of COVID-19 outbreak, local emissions within North China 37 Plain and Eastern China, respectively, contribute 66% and 87% to the 38 increase in surface PM_{2.5} concentrations. This is associated with the 39 anomalous mid-tropospheric high pressure at the location of climatological 40 East Asia trough and the consequently weakened winds in the lower 41 troposphere, leading to the local aerosol accumulation. The emissions 42 outside China, especially those from South and Southeast Asia, contribute 43 over 50% to the increase in PM2.5 concentration in Southwestern China 44

45 through transboundary transport during the <u>most_polluted day</u>. As the 46 reduction in emissions in the near future, aerosols from long-range 47 transport together with unfavorable meteorological conditions are 48 increasingly important to regional air quality and need to be taken into 49 account in clean air plans.

50 **1. Introduction**

The coronavirus disease 2019 (COVID-19) had an outbreak in China 51 inhas spread worldwide since December 2019. It has and resulted in more 52 than one million cases within the first four months worldwide (Sharma et 53 al., 2020; Dong et al., 2020). In order to curb the virus spread among 54 humans, China was the first country to take dramatic measures were taken 55 by the Chinese government on January 23, 2020 to minimize the 56 interaction among people, including strict isolation, prohibition of large-57 scale private and public gatherings, restriction of private and public 58 transportation and even lockdown of cities (Tian et al., 2020; Wang et al., 59 2020).-The estimated NOx emission in eastern China was reduced by 60-60 61 70%, of which 70-80% was related to the reduced road traffic and 20-25% was from industrial enterprises shutdown during the COVID-19 lockdown 62 period (Huang et al., 2020). However, severe air pollution events still 63 occurred in East China during the COVID-19 lockdown. It is of great 64 concern that why severe air pollution was not avoided by decreasing, even 65 though the anthropogenic emissions were greatly reduced (Huang et al., 66 2020). The unprecedented large-scale restrictions resulting from the 67 COVID-19 epidemic provide an opportunity to research the relationship 68 between dramatic anthropogenic emission reductions and air quality 69 changechanges (e.g., Bao et al., 2020; Li et al., 2020; Wang et al., 2020). 70 Bao et al. (2020) reported that, during the COVID-19 lockdown period, the 71

72 air quality index (AQI) and the $PM_{2.5}$ (particulate matter less than 2.5 μ m in diameter) concentration were decreased by 7.8% and 5.9%, respectively, 73 on average in 44 cities in northern China, mainly due to travel restrictions. 74 By applying the WRF-CAMx model together with air quality monitoring 75 data, Li et al. (2020) revealed that although primary particle emissions were 76 reduced by 15%-61% during the COVID-19 lockdown over the Yangtze 77 River Delta Region, the daily mean concentration of PM2.5 was still 78 relatively high, reaching up to 79 μ g m⁻³. Wang et al. (2020) found that the 79 relative reduction in PM_{2.5} precursors was twice as much as the reduction 80 in PM_{2.5} concentration, in part due to the unfavorable meteorological 81 conditions during the COVID-19 outbreak in China that led to the 82 83 formation of the heavy haze. Huang et al. (2020) and Le et al. (2020) reported that stagnant air conditions, high atmospheric humidity, and 84 enhanced atmospheric oxidizing capacity led to a severe haze event in 85 northern China during the COVID-19 pandemic. 86

Aerosols are main air pollutants that play important roles in the atmosphere due to their adverse effects on air quality, visibility (Vautard et al., 2009; Watson, 2002), human health (Lelieveld et al., 2019; Heft-Neal et al., 2018), the Earth's energy balance, and regional and global climate (Ramanathan et al., 2001; Anderson et al., 2003; Wang et al., 2020; Smith et al., 2020). With the rapid development in recent decades, China has experienced severe air pollutions that damage human health and cause

regional climate change (Chai et al., 2014; Liao et al., 2015; Fan et al., 94 2020). In order to control air pollution, the Chinese government issued and 95 implemented the Air Pollution Prevention and Control Action Plan in 2013 96 (China State Council, 2013). Although emissions in China have decreased 97 significantly in recent years (Zheng et al., 2018), aerosols transported from 98 other source regions could add on top of local emissions (Yang et al., 2017a, 99 2018a; Ren et al., 2020). Therefore, it is important to understand the 100 relative effects of local emissions and regional transport on aerosols in 101 China. 102

Source tagging and apportionment is an effective way to establish 103 aerosol source-receptor relationships, which is conducive to both scientific 104 105 research and emission control strategies (Yu et al., 2012). By applying the Particulate Source Apportionment Technology in CAMx model, Xue et al. 106 (2014) found that the contributions of regional transport to annual average 107 PM_{2.5} concentrations in Hainan, Shanghai, Jiangsu, Zhejiang, Jilin and 108 Jiangxi provinces of China are more than 45%. By adding a chemical tracer 109 into the WRF model, Wang et al. (2016) studied the sources of black carbon 110 (BC) aerosol in Beijing and reported that about half of BC in Beijing came 111 from the central North China Plain. Liu et al. (2017) applied WRF-Chem 112 model and showed that Foshan, Guangzhou and Dongguan, respectively, 113 with relatively high emissions contributed 14%, 13% and 10% to the 114 regional mean PM_{2.5} concentration in the Pearl River Delta. 115

Currently, manyprevious studies only focused on regional transport of 116 aerosols, very few studies have investigated the impact of reduced human 117 activity on regional air quality, as a result of the COVID-19 outbreak. Few 118 studies have focused on explored the aerosol transport pathways and source 119 attribution incovering the whole China during the COVID-19 pandemic. In 120 this study, the global aerosol-climate model CAM5 (Community 121 Atmosphere Model, version 5) equipped with an Explicit Aerosol Source 122 Tagging (CAM5-EAST) is employed to quantify source-receptor 123 relationships and transport pathways of aerosols during the COVID-19 124 outbreak in China. We also provide model evaluations of PM_{2.5} 125 concentrations against observations made during the COVID-19 outbreak. 126 127 With the aerosol source tagging technique, source region contributions to PM_{2.5} column burden over various receptor regions and transport pathways 128 in China are analyzed. The source contributions to the changes in near-129 surface PM_{2.5} in the most polluted days compared to the monthly means 130 during February 2020 are also quantified. This paperOur study provides 131 source apportionment of aerosols incovering the whole China during 132 theand quantifies the contribution from foreign transport for the first time 133 in the case of COVID-19 emission reductions, which is beneficial to the 134 investigation of policy implications for future air pollution control. 135

136 **2. Methods**

137 **2.1 Model description and experimental setup**

The CAM5 model is applied to estimate the $PM_{2.5}$ changes during the 138 139 COVID-19 period.-In CAM5, which is the atmospheric component of the earth system model CESM (Community Earth System Model, Hurrell et 140 al., 2013). In this study, major aerosol species including sulfate, BC, 141 primary organic matter (POM), secondary organic aerosol (SOA), sea salt, 142 and mineral dust, are represented by three lognormal size modes (i.e., 143 Aitken, accumulation, and coarse modes) of the modal aerosol module 144 (MAM3) (Liu et al., 2012). The detailed aerosol representation in CAM5 145 was provided in Liu et al. (2012) and Wang et al. (2013). The aerosol 146 mixing states consider both internal mixed (within a same mode) and 147 external mixed (between modes). On top of the default CAM5, additional 148 modifications that improve the representation of aerosol wet scavenging 149 and convective transport (Wang et al., 2013) are also included in the model 150 version used for this study. 151

In this study, simulations were conducted with a horizontal resolution 152 of $1.9^{\circ} \times 2.5^{\circ}$ and 30 vertical layers up to 3.6 hPa in year 2020. 153 Anthropogenic The anthropogenic emissions used in Chinathe baseline 154 simulation are derived from the MEIC (Multi-resolution Emission 155 Inventory of China) inventory (Zheng et al., 2018). while), referred to here 156 as the Baseline experiment. While emissions for the other countries use the 157 SSP (Shared Socioeconomic Pathways) 2-4.5 scenario data set under 158 CMIP6 (the Coupled Model Intercomparison Project Phase 6). Emissions 159

in year 2017 are used as the baseline during the simulation period 160 considering the time limit of MEIC inventory. To better estimate the impact 161 of restricted human activities on emission reductions owing to COVID-19 162 lockdown, (referred to as Covid experiment), we updated China's emission 163 inventory from January to March 2020 based on the provincial total 164 emission reduction ratio in Huang et al. (2020). Emissions from the 165 transportation sector are decreased by 70% and the%. The remaining 166 reductions emission reduction, by excluding transport reduction from the 167 total emission reduction, are evenly distributed to other sectors, including 168 industry, power plant, residential, international shipping and waste 169 treatment from January to March 2020 compared to the baseline emission 170 171 in 2017. Unless otherwise specified, all the results in this study are derived from the Covid experiment. 172

The sea surface temperature, sea ice concentrations, solar radiation and 173 greenhouse gas concentrations are fixed at present-day climatological 174 levels. To capture the large-scale atmospheric circulations during the 175 COVID-19, we nudge the model wind fields toward the MERRA-2 176 (Modern-Era Retrospective Analysis for Research and Applications, 177 version 2) reanalysis (Gelaro et al., 2017) from April 2019 to March 2020 178 repeatedly for six years. Only model results from the last year are used to 179 represent year 2020- with the first five years as model spin-up. In this study, 180 we analyze the transport pathways and source attribution of aerosols during 181

the three weeks that had the largest number of newly-diagnosed COVID19 cases (Fig. <u>2S1</u>, hereafter referred to as the 'Week 1': January 30–
February 5, 'Week 2': February 6–February 12 and 'Week 3': February 13–
February 19), when unexpected hazardous air pollution events also
occurred during this time period (<u>Huang et al., 2020;</u> Le et al., 2020).

187 **2.2 Explicit aerosol source tagging and source regions**

To examine the source apportionment of aerosols in China, the Explicit 188 Aerosol Source Tagging (EAST) technique was implemented in CAM5, 189 which has been utilized in many aerosol source attribution studies (e.g., 190 Wang et al., 2014; Yang et al., 2017a, b, 2018a, b, c, 2020; Ren et al., 2020). 191 Different from the emission sensitivity method that assumes a linear 192 193 response to emission perturbation and the traditional backward trajectory method, aerosols from each tagged region or sector are calculated 194 independently in EAST within one single simulation. Without relying on a 195 set of model simulations with emission perturbations or assuming constant 196 decaying rate, EAST is more accurate and time-saving than the source 197 apportionment method mentioned above. In addition to the sulfate, BC and 198 POM species that were tagged in previous studies (e.g., Yang et al., 2020), 199 SOA and precursor gas are now also tagged in the EAST. These types of 200 aerosols from independent source regions and sectors can be explicitly 201 tagged and tracked simultaneously. In this study, focusing on the aerosols 202 in China during the COVID-19 outbreak period, the domestic aerosol and 203

precursor emissions are divided into<u>from</u> eight geographical source regions
(Fig. 1), including Northeastern China (NEC), North China Plain (NCP),
Eastern China (ESC), Southern China (STC), Central-West China (CWC),
Southwestern China (SWC), Northwestern China (NWC) and the
Himalayas and Tibetan Plateau (HTP), and the rest of the world (ROW)
emissions), are tagged separately.

210 **3. Model evaluation**

Many previous studies have assessed the spatial distribution and 211 seasonal to decadal variations in aerosol concentrations in China and 212 worldwide simulated by CAM5 with the observations (e.g., Wang et al., 213 2013; Yang et al., 2017a,b, 2018b,c, 2020). In order to evaluate the model's 214 215 performance in simulating aerosols during the COVID-19 outbreak period in China, the surface concentrations of PM2.5, estimated as the sum of 216 sulfate, BC, POM and SOA for model results, during the analyzed time 217 periods are compared with measurements from the China National 218 Environmental Monitoring Center (CNEMC), as shown in Fig. 3a2a. The 219 model reasonably reproduces the overall spatial distribution of near-220 surface PM_{2.5} concentrations during the three time periods, with high 221 values in the North China Plain and low values in western China. However, 222 as reported in many CAM5 model studies (e.g., Yang et al., 2017a,b), the 223 model underestimates the PM_{2.5} concentrations with normalized mean 224 biases (NMB) of -55%~-49%, compared to the available site observations 225

(Fig. S1S2). The discrepancies are related to coarse-resolution model sampling bias relative to the observational sites, uncertainties in aerosol emissions, wet removal, and gas-particle exchange. In addition, the model version used in this study is not able to simulate nitrate and ammonium aerosols, which are also the main components of $PM_{2.5}$ (Kong et al., 2020; Xu et al., 2019).

The long-distance transport of aerosols mainly occurs in the upper 232 troposphere rather than near the surface (Hadley et al., 2007; Zhang et al., 233 2015). Aerosols are lifted from the atmospheric boundary layer of the 234 emission source regions to the free troposphere and then undergo the 235 transboundary and intercontinental transport effectively driven by the 236 237 upper tropospheric circulations. Therefore, it is helpful to analyze the relative contributions of local and non-local sources by focusing on the 238 column burden of aerosols. Figure 3b2b presents spatial distributions of 239 simulated mean column burden of PM_{2.5} during the three time periods-240 ('Week 1': January 30–February 5, 'Week 2': February 6–February 12 and 241 'Week 3': February 13-February 19), which had the largest number of 242 newly-diagnosed COVID-19 cases. The contrast in column burden does 243 not differ significantly from that of near-surface concentrations. Among the 244 three time periodsComparing to Week 3, Week 1 and Week 2 have higher 245 $PM_{2.5}$ loading, with values in the range of 20–40 and 20–30 mg m⁻² in the 246 North China Plain, Eastern China, and Southern China, while the PM_{2.5} 247

loading in Week 3 is relative lower with <u>than Week 1 and Week 2 with</u> values ranging mostly from 10 to 20 mg m⁻². Note that the column burden of PM_{2.5} in South and Southeast Asia is higher than 20 mg m⁻² in three time periods and reaches up to 50 mg m⁻² in Week 2, which potentially influences aerosol concentrations in China through transboundary transport.

254 **4. Transport Pathways**

The explicit aerosol tagging technique can clearly identify the transport 255 pathways of aerosols moving from their source regions to their destination. 256 Figure 43 shows the spatial distribution of mean column burden of 257 simulated PM_{2.5} originating from the six tagged source regions in central 258 259 and eastern China and outside of China during the three time periods. Aerosols and/or precursor gases emitted from the various regions follow 260 quite different transport pathways determined by their source locations, 261 meteorological conditions, emission injection height, and physical and 262 chemical characteristics of aerosol species. Aerosols from Northeastern 263 China are transported southeastward by the northwesterly winds (Fig. 1b). 264 From the North China Plain, aerosols can be transported either southward 265 reaching Eastern, Southern and Southwestern China during Week 1 or 266 across east coast of China to the oceanic region during Week 2-3. Aerosols 267 originating from Eastern China move straight to Southwestern and 268 Southern China during Week 1-2, while they also entered the North China 269

Plain during Week 2-3. Aerosols emitted from Southern China and Central-West China have no obvious transport due to their relatively weak emissions. In additional to the local impact, emissions from Southwestern China affect mostly the Southern China and Eastern China. Air parcels with high levels of $PM_{2.5}$ from South and Southeast Asia moved into Southwestern, Southern and Eastern China and even the North China Plain during the three time periods.

The vertical distributions of PM_{2.5} emitted from six major tagged 277 source regions are shown in Figs. <u>S2S3</u> and <u>S3S4</u>. The PM_{2.5} has much 278 higher concentrations in the lower troposphere and decreases with 279 increasing height. During Week 1-2, owing to the presence of high PM_{2.5} 280 281 loadings, a stronger vertical mixing and transport brought more PM_{2.5} to the upper troposphere compared to that during Week 3. High 282 concentrations of PM_{2.5} originating from the North China Plain extended 283 southeastward by strong northwesterly winds. Weak winds over Eastern 284 China led to accumulations of PM_{2.5} within this region, which is consistent 285 with the findings in Yang et al. (2017a). Strong southwesterly winds in the 286 south of Southwestern China and weak winds in the north of this region 287 produced convergences and updrafts that lift aerosols up to 700 hPa. 288

Considering that the emissions outside China contribute greatly to PM_{2.5} concentrations in Southwestern China through transboundary transport (Yang et al., 2017a) and aerosols from East Asia can be

transported to the North Pacific and even North America (Yu et al., 2008; 292 Yang et al., 2018c), it is of great importance to study the inflow and outflow 293 of $PM_{2,5}$ across the boundaries of China. Figures 54 and 65 show the 294 vertical distribution of PM_{2.5} concentrations resulting from emissions 295 within and outside China over 29°N, 88°E and 21°N around the south 296 boundaries (cross-sections (CS) 1-3 in Fig. 1a) and 123° E around the east 297 boundary (CS 4 in Fig. 1a) of the mainland of China. Over the southern 298 border, PM_{2.5} concentrations are more influenced by transboundary 299 transport of aerosols from ROW than those originating from domestic 300 emissions. The high concentrations of PM_{2.5} from South and Southeast 301 Asia are lifted into the free atmosphere of the Tibetan Plateau and Yun-Gui 302 303 Plateau, and then transported to Southern and Southwestern China by southwesterly winds. Over the North China Plain and Eastern China, 304 northwesterly winds at 35-45° N and southwesterly winds at 25-35° N 305 cause aerosols to accumulate in the lower atmosphere and then export 306 across east border of China below 700 hPa. 307

5. Source apportionment of PM_{2.5} in China during the COVID-19

309

5.1 Source contributions to PM_{2.5} burden

Figure 76 shows the simulated relative contributions in percentage to PM_{2.5} column burden from local source emissions, regional transport from the untagged regions of China (rest of China, RCN) and rest of the world (ROW). Over the North China Plain, where emissions are relatively high,

PM_{2.5} column burden is dominated by local emissions during the three time 314 periods. In contrast, regions with relative low emissions are mainly 315 affected by nonlocal sources, especially by foreign contributions. 316 Emissions from the ROW contribute a large amount to PM_{2.5} burden over 317 Northeastern, Southern, Central-West, Southwestern, Northwestern China 318 and the Tibetan Plateau. PM2.5 burden in Eastern China is greatly 319 contributed by the sources from RCN, especially in Week 1 when regional 320 transport of PM_{2.5} from the North China Plain is relatively strong (Fig. 321 322 S3S4).

Table 1 summarizes the contributions of tagged source regions to the 323 PM_{2.5} burden over different receptor regions in China. In Northeastern 324 325 China, 36%-43% of the PM_{2.5} column burden comes from local emissions, while a larger portion (39%-54%) is contributed by emissions from ROW 326 during the three time periods. The impacts of nonlocal sources within 327 China on PM_{2.5} burden are relatively low in Northeastern China during 328 Week 1 with the contribution of less than 5%, while RCN is responsible for 329 23% and 25% during Week 2 and Week 3, respectively. 330

In the North China Plain, the majority of the $PM_{2.5}$ burden is attributed to local emissions in all cases, with local contributions in a range of 40– 66%. Emissions from the North China Plain also produce a widespread impact on $PM_{2.5}$ over its neighboring regions. The sources from North China Plain account for 14–33% of the $PM_{2.5}$ burden in Eastern China and 7-23% in Southern China during the three time periods.

In Eastern China, local emissions account for 27-40% of PM_{2.5} column 337 burden, while ROW contributes 20-45%. Southern China and Central-338 West China have 13-18% and 25-31% of local source contributions, 339 respectively, whereas 37–64% are due to emissions from outside China in 340 these two regions. In Southwestern China, 15-18% of the PM_{2.5} burden 341 originates from local emissions and 7-24% is from RCN. ROW emissions 342 play important roles in affecting PM_{2.5} burden over this region, with 343 relative contributions in a range of 59–78% during the three time periods, 344 which is associated with the transboundary transport by southwesterly 345 winds. PM_{2.5} burden over the Northwestern China and Himalayas and 346 347 Tibetan Plateau with relatively low local emissions are strongly influenced by nonlocal sources, where more than 70% of the PM_{2.5} burden originates 348 from emissions outside China. 349

5.2 Aerosol source attribution during polluted days

In spite of the large reductions in emissions, severe air pollution events still occurred in China during the COVID-19 lockdown. Source attribution of $PM_{2.5}$ during polluted days in China has policy implications for future air pollution control. In Beijing, capital of China over the North China Plain, a serious haze event happened from February 11 to 13, 2020 during the COVID-19 outbreak period according to observations released by CNEMC. CAM5-EAST reproduced the polluted day on February 11 over the North China Plain. In this study, the most polluted day is defined as the day with the highest daily $PM_{2.5}$ concentration in February 2020 for each receptor region in China. Figure <u>87</u> presents the composite differences in near-surface $PM_{2.5}$ concentrations and 850 hPa wind fields between the <u>most polluted daysday</u> and normal days (all days in February 2020) for each receptor region. The local and nonlocal source contributions to the $PM_{2.5}$ differences are summarized in Fig. <u>98</u>.

Unexpectedly, near-surface PM_{2.5} concentrations in the North China 365 Plain and Eastern China experienced remarkable increases during the same 366 most polluted daysday of COVID-19 lockdown. The simulated PM_{2.5} 367 concentrations increased, with the largest increases of more than 20 μ g m⁻³ 368 in the North China Plain and Eastern China, 10 µg m⁻³ maximum increase 369 in the Southwestern China and 5 μ g m⁻³ in the Northeastern, Southern and 370 Central-West China, during the most polluted days compared to the normal 371 days. 372

The increase in near-surface $PM_{2.5}$ concentrations during the most polluted day over Northeastern China is largely influenced by the local emissions, which contribute to a regional averaged concentration increase of 1.1 µg m⁻³. This is mainly due to the accumulation of local aerosols under the weakened prevailing northwesterly winds over this region.

When the PM_{2.5} pollution occurred in the North China Plain_{$\overline{5}$} on February 11, 2020, which was also reported as the polluted day in

observations (Huang et al., 2020), the concentration of PM_{2.5} was 16.1 µg 380 m⁻³ higher than that in normal days. The contribution from local emissions 381 accounts for 66% of the averaged increase, which was related to the 382 stagnant air condition (i.e., weakened lower tropospheric winds) resulting 383 from the anomalous mid-tropospheric high pressure located at the 384 385 climatological location of the East Asia trough (Fig. <u>\$4\$5</u>). Sources from Eastern China also explain 4.3 μ g m⁻³ (27%) of the total increase over the 386 North China Plain. 387

During the most polluted day in Eastern China (the same day as the 388 most polluted day in North China Plain), the regional averaged increase in 389 concentration of $PM_{2.5}$ concentrations is was 16 µg m⁻³ higher than that in 390 391 normal days, which is primarily contributed by the local emissions. While the contribution from the North China Plain decreased in the most polluted 392 day, the anomalous southerly winds brought more aerosols from Southern 393 China and ROW into Eastern China, contributing to 4% and 10% aerosol 394 increase, respectively. 395

Owing to the enhanced northerly winds, emissions from the North China Plain and Eastern China contribute 33% and 39% of the increase, respectively, in $PM_{2.5}$ concentration over Southern China. The most polluted day in Central-West China is mostly caused by local emissions (65% of the total increase).

401 When Southwestern China was under the polluted condition, $PM_{2.5}$

concentration was increased by 2.1 μ g m⁻³. Emissions from ROW, 402 especially those from South and Southeast Asia, are of great significance 403 to the increase of PM2.5 concentrations due to the enhanced southwesterly 404 winds over this region. The relative contribution from ROW emissions is 405 more than 50% over Southwestern China during the most polluted day. It 406 highlights that the important role of transboundary transport needs to be 407 considered when controlling local emissions to improve air quality in the 408 near future. 409

410

411 **6. Conclusions and discussions**

The COVID-19 pandemic disrupted human activities and lead to abrupt 412 413 reductions in anthropogenic emissions. This study first investigated the source contributions to PM_{2.5} over various regions covering the whole 414 China during the COVID-19 pandemic. We pay attention not only to local 415 emissions, but also to the impacts from regional and foreign transport of 416 aerosols. An explicit aerosol source tagging is implemented in the 417 Community Atmosphere Model version 5 (CAM5-EAST) to examine the 418 aerosol transport pathways and source attribution of PM_{2.5} in China during 419 the first few weeks of the COVID-19 outbreak (Week 1: January 30-420 February 5, Week 2: February 6-February 12 and Week 3: February 13-421 February 19). The contributions of emissions to $PM_{2.5}$ originating from 422 eight source regions in the mainland of China, including Northeastern 423

China, North China Plain, Eastern China, Southern China, Central-West 424 China, Southwestern China, Northwestern China and Himalayas and 425 Tibetan Plateau, and sources outside China (ROW) to near-surface 426 concentrations, column burdens, transport pathways of PM_{2.5}, and haze 427 formation in different receptor regions in China are quantified in this study. 428 Aerosols emitted from the North China Plain, where the air quality is 429 often poor, can be transported through Eastern China and reach 430 Southwestern China during the three time periods. Similarly, aerosols from 431

Eastern China move straight to Southern China and Southwestern China
during Week 1 and Week 2, and a significant portion can also enter the
North China Plain during Week 2 and Week 3.

Across the southern boundary of the mainland of China, high concentrations of $PM_{2.5}$ from South and Southeast Asia are lifted into the free atmosphere and then transported to Southern and Southwestern China. While $PM_{2.5}$ from the North China Plain and Eastern China can also be brought out of China via westerly winds, mostly below 700 hPa.

PM_{2.5} in China is affected not only by local emissions but also by longrange transport of pollutants from distant source regions. Over the North China Plain, 40–66% of the PM_{2.5} burden is attributed to local emissions during the COVID-19 outbreak. They also impact PM_{2.5} in neighboring regions, accounting for 14–33% of the PM_{2.5} burden in Eastern China and 7–23% in Southern China during the three time periods. Northeastern

China has 36%-43% of local source contributions to its PM_{2.5} column 446 burden, while 39%-54% is contributed by emissions from ROW during the 447 three time periods. In Eastern China, local emissions explain 27-40% of 448 PM_{2.5} burden, while ROW contributes 20–45%. In Southwestern China, 449 59-78% of the PM_{2.5} burden is contributed by emissions from ROW. Over 450 the Northwestern China and Himalayas and Tibetan Plateau, ROW 451 emissions have a great contribution of more than 70% to the PM_{2.5} column 452 burden. 453

454 In this study, the most polluted day is defined as the day with the highest daily PM_{2.5} concentration in February 2020 for each receptor 455 region in China. The transport from outside of China only has a great 456 impact on some specific regions in China. In Southwestern China, the 457 relative contribution from ROW emissions, especially those from South 458 and Southeast Asia, to the increment of PM_{2.5} concentration during the 459 most polluted days compared with normal days is more than 50%. It is 460 consistent with the previous studies that emissions from South and 461 Southeast Asia have an important impact on air quality in southwest China 462 (Yang et al., 2017a; Zhu et al., 2016, 2017). For other receptor regions in 463 China (Northeastern China, North China Plain, Eastern China, Southern 464 China and Central-West China), PM_{2.5} concentrations are largely 465 contributed by local emissions during the most polluted days compared 466 with normal days. In the future with emissions reductions for better air 467

468 <u>quality in China, decreasing air pollution should consider aerosols from</u>
469 both Chinese local emissions and pollutant transport from outside of China.

Despite the large reductions in emissions, near-surface $PM_{2.5}$ 470 concentrations in the North China Plain and Eastern China increased a lot 471 during the most polluted days of COVID-19 lockdown (with the highest 472 daily PM_{2.5} concentration in February 2020), with the largest increases of 473 more than 20 μ g m⁻³. In addition to local emissions, regional transport of 474 pollutants is also an important factor that causes haze events in China. The 475 increases in PM_{2.5} concentrations during the most polluted days over the 476 North China Plain and Eastern China are largely influenced by the stagnant 477 air condition resulting from the anomalous high pressure system and 478 479 weakening of winds, which lead to a reduced ventilation and aerosol accumulation in the North China Plain, together with an increase in aerosol 480 inflow from regional transport. During the most polluted day in 481 Southwestern China, ROW contributes over 50% of the PM_{2.5} 482 concentration increase, with enhanced southwesterly winds that drive 483 pollution transport from South and Southeast Asia. It indicates that regional 484 transport and unfavorable meteorology need to be taken into consideration 485 when controlling local emissions to improve air quality in the near future. 486

487 <u>To highlight the roles of regional and foreign transport, the differences</u>
 488 <u>between Covid and Baseline simulations in relative contributions to PM_{2.5}</u>
 489 <u>burden from local, region (RCN) and foreign (ROW) emissions are given</u>
in Figure S6. During the COVID-19 period, the local and RCN emission 490 contributions to $PM_{2.5}$ were 1–4% lower than that in Base experiment over 491 NCP and NEC. In Eastern China, the contribution from the local emissions 492 decreased by 3-4% compared with Base experiment, while the 493 contribution from ROW increased by more than 5%. In Southern China, 494 50–70% of the $PM_{2.5}$ burden is contributed by emissions from ROW in 495 Base experiment. During the COVID-19 period with low emission levels, 496 the contribution from ROW to PM_{2.5} burden in Southern China had an 497 increase of more than 5%. It indicates that the important role of 498 transboundary transport needs to be considered when controlling local 499 emissions to improve air quality in the near future. 500

501 Many studies have examined the importance of meteorology on regional air quality during the COVID-19 lockdown period and 502 emphasized that, when meteorology is unfavorable, abrupt emissions 503 reductions cannot avoid severe air pollutions (Le et al. 2020; Sulaymon et 504 al. 2021; Shen et al. 2021). Through model simulations, Le et al. (2020) 505 found that abnormally high humidity promotes the heterogeneous 506 chemistry of aerosols, which have contributed to the increase of PM_{25} by 507 12% in northern China during the city lockdown period. Sulaymon et al. 508 (2021) found that significant increase in $PM_{2.5}$ concentrations caused by 509 unfavorable meteorological conditions in Beijing-Tianjin-Hebei region 510 during the lockdown period based on Community Multiscale Air Quality 511

(CMAQ) model simulations. By analyzing the observational data and 512 model simulations, Shen et al. (2021) reported that 50% of the pollution 513 episodes during the COVID-19 lockdown in Hubei of China were due to 514 the stagnant meteorological conditions. Huang et al. (2020) found that the 515 stagnant air conditions and enhanced atmospheric oxidizing capacity 516 caused a severe haze event during the same time period. In line with 517 previous studies, we also revealed the stagnant air condition under the 518 anomalous high pressure system in the most polluted day over the North 519 China Plain. In addition to the meteorological conditions, the effect of 520 foreign transport was also raised in this study causing aerosol pollution in 521 southwestern China during COVID-19 outbreak. 522

There are a few uncertainties in this study. The CAM5 model has low 523 biases in reproducing the near-surface PM_{2.5} concentrations in China, 524 compared to observations, in part due to the incapability of simulating 525 some aerosol components of PM_{2.5} (e.g., ammonium and nitrate), excessive 526 aerosol wet removal during the long-range transport (Wang et al., 2013), 527 and uncertainties in observations. In majority of the climate models, the 528 simulation of nitrate and ammonium aerosols are not included in the 529 aerosol schemes, partly due to the complexity of calculation efficiency. For 530 example, in many of the CMIP6 models, only two of them provide nitrate 531 and ammonium mass mixing ratios. Many previous studies have evaluated 532 global climate models performance in reproducing aerosol the 533

concentrations (e.g., Fan et al., 2018; Shindell et al., 2013; Yang et al., 534 2017a,b). In general, the models can well simulate aerosols in North 535 America and Europe but significantly underestimates aerosols in East Asia 536 by about -36 to -58 % compared with observations. It can lead to an 537 underestimation of aerosols contributed by Chinese local emissions in 538 magnitudes, but might not change the main conclusions of this study. 539 Uncertainties in the estimate of emission reductions in different source 540 regions during the COVID-19 pandemic can also introduce uncertainties 541 to our results. During the COVID-19 lockdown, greenhouse gas emissions 542 also decreased (Le Quéré et al., 2020), but the effect of greenhouse gas 543 reduction on meteorology that potentially influence aerosol distributions 544 was not taken into consideration. Nevertheless, this study is the first 545 attempt to provide source apportionment of aerosols incovering the whole 546 China during the COVID-19 outbreak, which is beneficial to the 547 investigation of policy implications for future air pollution control. 548

The CAM5 model is available 550 at http://www.cesm.ucar.edu/models/cesm1.2/ (last access: 25 October 20203 551 August 2021). CAM5-EAST model code and results can be made available 552 upon request. The surface PM_{2.5} observations are from the China National 553 Environmental Monitoring Center (CNEMC, http://www.cnemc.cn, last 554 access: 25 October 20203 August 2021) 555

- 556 *Competing interests.*
- 557 The authors declare that they have no conflict of interest.
- 558 Author contribution.
- 559 YY and LR designed the research; YY performed the model simulations;
- 560 LR analyzed the data. All authors discussed the results and wrote the paper.
- 561 Acknowledgments.

This study was supported by the National Key Research and Development Program of China (grant 2020YFA0607803<u>and 2019YFA0606800</u>) and the National Natural Science Foundation of China (grant 41975159). HW acknowledges the support by the U.S. Department of Energy (DOE), Office of Science, Office of Biological and Environmental Research (BER). The Pacific Northwest National Laboratory (PNNL) is operated for DOE by the Battelle Memorial Institute under contract DE-AC05-76RL01830.

569 **Reference**

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Table 1. Fractional contributions of emissions from nine tagged source regions (vertical axis) to mean $PM_{2.5}$ column burden in eight receptor regions (horizontal axis) during the three time periods₇ ('Week 1': January 30–February 5, 'Week 2': February 6– February 12 and 'Week 3': February 13–February 19).

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Figure 1. (a) Tagged source regions (NEC: Northeastern China, NCP: North China 841 Plain, ESC: Eastern China, STC: Southern China, CWC: Central-West China, SWC: 842 Southwestern China, NWC: Northwestern China, HTP: Himalayas and Tibetan Plateau, 843 ROW: rest of the world) and (b) mean wind field (units: m s⁻¹, vectors) at 850 hPa 844 845 during the time period of interest.three weeks of the study from January 30 to February 19, which had the largest number of newly-diagnosed COVID-19 cases. Lines in (a) 846 mark the cross-sections (CS) defined to study the transport of aerosols to and from 847 848 China.

- 849
- 850



PM_{2.5} concentrations (μg m⁻³) and (b) PM_{2.5} column burden (mg m⁻²) during January
30–February 5 (Week 1), February 6–February 12 (Week 2) and February 13–February
19 (Week 3).



PM_{2.5} column burden (mg m⁻²)

Figure 43. Spatial distribution of $PM_{2.5}$ column burden (mg m⁻²) originating from the six major source regions in China (NEC, NCP, ESC, STC, CWC and SWC) and sources outside China (ROW) during the three time periods.



Figure 54. Vertical distributions of $PM_{2.5}$ concentrations (µg m⁻³), originating from emissions outside China (i.e., ROW sources), across the latitudinal and/or longitudinal extents marked in Fig.1, respectively, during the three time periods.



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Figure 65. Vertical distributions of $PM_{2.5}$ concentrations (µg m⁻³), originating from domestic emissions in China, across the latitudinal and/or longitudinal extents marked in Fig.1, respectively, during the three time periods. The values along CS 1 and CS 2 are negligibly small.

(a) Local contribution





Figure 76. Relative contributions (%) of (a) local emissions, (b) the emissions from the
rest of China (RCN) and (c) all sources outside China (rest of the world, ROW) to PM_{2.5}
column burden during the three time periods.



Figure 87. Composite differences in winds at 850 hPa (m s⁻¹) and near-surface PM_{2.5}
concentrations (μg m⁻³) between the most polluted and normal days in February 2020.
The most polluted day is defined as the day with the highest daily PM_{2.5} concentration
in February 2020 in each receptor region in China.



Figure 98. Composite differences in near-surface $PM_{2.5}$ concentrations (µg m⁻³) averaged over receptor regions (marked on the horizontal axis) in China between the most polluted and normal days in February 2020 originating from individual source regions (corresponding color bars in each column).