Dear Editors and Referees:

Thank you very much for your constructive suggestions and helpful comments for improving our manuscript acp-2019-758. We have accordingly made the careful revisions. Revised portions are highlighted in the revised manuscript. In the following we quoted each review question in the square brackets and added our response after each paragraph.

Responses to Referee #1

[Yu et al investigated the impacts of regional transport to the heavy haze pollution in January 2016 over Wuhan, a city located over the Yangtze River Middle Basin in the central part of China. This study characterized unique "non-stagnant" conditions (e.g., high winds, no inversion layers) associated with extreme high levels of $PM_{2.5}$ concentrations (e.g., strong correlation between $PM_{2.5}$ concentrations greater 150 $\mu g\ m^{-3}$ and wind speed), which differed significantly from traditional haze pollutions with low near-surface winds and inversion layers found in the literatures. The authors employed both observational and modeling analyses to prove the importance of the contribution of regional transport to the excessive $PM_{2.5}$ concentrations over Wuhan. This is an interesting study to demonstrate the complexity and challenge of the severe haze pollution over central-eastern China during wintertime, with research scope aligned with topics suitable for ACP. However, the current format of the manuscript is not accepted, due to ambiguous structure of the manuscript, lack of detailed descriptions of observational and modeling methods, concerns of technical methodology as well as numerous grammar errors and typos over the entire manuscript. A major revision is needed for this manuscript before further consideration of publication in ACP. My comments for the manuscript are shown as follows.]

Response 1: Many thanks for the encouraging comments and constructive suggestions on our manuscript acp-2019-758. Accordingly, we have restructured the manuscript with detailed descriptions of observational and modeling methods, concerns of technical methodology as well as corrected the grammar errors and typos over the entire manuscript (please find them in the following responses and the highlighted revisions in the revised manuscript).

[Major Comments

1. Research Methodology and Results/Discussions for the paper are not clear I have difficulty in following the paper's research methodology/results. The authors mix the research methodology and results in the same section. I highly recommend that the authors should re-organize the structure of the paper. The descriptions of observational data from various sites and FLEXTPART-WRF (Sect. 3.2.1 and Sect. 3.2.2) should be placed in Section 2. And Results and Discussions, including the analysis of the observational data and modeling study, should be placed in Section 3.]

Response 2: Following the referee's suggestions, we have re-organized the structure of the paper. In the revised manuscript, the descriptions of observational data from various sites and FLEXTPART-WRF (Sect. 3.2.1 and Sect. 3.2.2) are placed in Section 2. And Results and Discussions, including the analysis of the observational data and modeling study, are placed in Section 3.

[2. The descriptions of the data used in this study are not adequate and needed to be expanded to provide a more detailed and rigorous documentation.

We don't know the spatial locations of the observational sites for $PM_{2.5}$ measurements, especially ten sites over Wuhan, which need to be presented. A spatial map of WRF modeling domain, with $PM_{2.5}$ measurement sites inserted, will be very helpful. Moreover, what is the measurement technique used for $PM_{2.5}$? What is the measured frequency/quality data control method, and measurement uncertainty associated with $PM_{2.5}$ concentrations and other meteorological *parameters for each site? How do you represent Wuhan's hourly $PM_{2.5}$ concentrations out of the ten measurement sites? And how do you calculate the correlation coefficients between $PM_{2.5}$ concentrations and wind speed/temperature over Wuhan in January 2016 out of ten measured sites?]*

Response 3: According to the referee's suggestions, we have added the spatial locations of the ten observational sites for $PM_{2.5}$ measurements over Wuhan in the supplemental (Fig. s1). Besides, the $PM_{2.5}$ data used in this study were collected from the national air quality monitoring network operated by the Ministry of ecology and environmental protection of China, The mass volume concentrations of surface $PM_{2.5}$ are operationally hourly observed with the instrument of the Thermo Fisher Scientifi. The observation data are under quality control based on the China's national standard of air quality observation before released by the Ministry of ecology and environmental protection of China. The source of the data has been added in the revised manuscript. At last, the surface $PM_{2.5}$ concentrations averaged over 10 observation sites in Wuhan are used to calculate the correlation coefficients with the changing meteorological drivers (wind speed/temperature etc.) over Wuhan in January 2016 to investigate the local meteorological influences on hourly changes of surface $PM_{2.5}$ concentrations in Wuhan.

[3. In terms of quantification of regional transport contributions for PM2.5 over Wuhan, the authors have utilized FLEXPART-WRF model. However, I have concerns about the convolution of FLEXPART-WRF residence time with the PM2.5 bottom-up emission fluxes from MEIC. Firstly, what is the definition of residence time here? Is it the PM2.5 lifetime? With Lagrangian method, it will result in a Jacobian matrix (footprint), in unit of mass per volume per unit flux. It is helpful for the authors to mathematically derive the residence time for particles out of FLEXPART, the product of the residence time and the bottom-up emission flux, and ultimately the regional transport contribution rate in the "Research Methodology" Section. The authors should insert the unit for each variable out of FLEXPART modeling. Meanwhile, please help the readers about the purpose of the WRF model here. Further, FLEXPART does not consider chemistry and deposition

in the model, the only part it accounts for is the transport, driven by reanalysis data. PM2.5 contains a significant portion of secondary organic and inorganic aerosols, which come from important and complex physiochemical processes in the atmosphere. How this methodology (FLEXPART-WRF) is proven robustness to quantify the regional transport contribution? What is the uncertainty range here?]

Response 4: Thanks for the comments. In the revised manuscript, we have clarified the quantification of regional transport contributions with utilizing the model FLEXPART-WRF in the revised manuscript as followings:

In the model FLEXPART-WRF, the trajectory of a large number of particles released from a source is simulated with consideration of the processes of tracer transport, turbulent diffusion, wet and dry depositions in the atmosphere. With Lagrangian method, it could result in a Jacobian matrix (footprint), in unit of mass per volume per unit flux. Stohl et al. (2005) mathematically derived the residence time for particles out of FLEXPART. Generally, in the backward trajectory of FLEXPART modeling, a large number of particles is released at a receptor and transported backward in time. Then the residence time (not the lifetime) of all particles, normalized by the total number of released particles, is determined on a uniform grid. In this study for the receptor of Wuhan, the residence time for a thickness of 100 m above the surface was calculated and considered the ‘‘footprint’’ (in unit of s). By multiplying the residence time with the air pollutant emission flux in the respective grid cell (in unit of μg m^{-2} s^{-1}) calculated from the Multi-resolution Emission Inventory of year 2016 for China (MEIC, http://www.meicmodel.org/), the emission source contribution (in μg m^{-2}) from this grid cell to the receptor could be estimated (Stohl, 2003; Stohl et al., 2005;Ding er al.,2009), yielding a so-called potential source contribution map, which is the geographical distribution of the regional transport contribution rates (%) of the emission source grid cell to $PM_{2.5}$ pollution at the receptor of Wuhan (Fig. 9).

A need for further multiscale modeling and analysis has encouraged new developments in FLEXPART-WRF, a FLEXPART version that works with the Weather Research and Forecasting (WRF) mesoscale meteorological model (Brioude et. al., 2013). For the refined simulation of air pollutant sources and transport, FLEXPART modeling driven by mesoscale meteorology from WRF modeling has been widely used to investigate the potential sources of air pollutants in consideration of air pollution change.

In this study, the $PM_{2.5}$ contributions of regional transport to air pollution in the downwind receptor region could be approximately estimated based on the product of the residence time of air particles during regional transport simulated by FLEXPART-WRF, and the $PM_{2.5}$ emission flux over the source grid in Central and Eastern China. The potential source contribution is estimated based on transport alone, ignoring chemical and removal processes. We also understand that the physical and chemical processes such as complex deposition and chemical conversion for the formation of secondary particles are not introduced in the FLEXPART-WRF emulation, which could represent the basic features of contribution and patterns of regional $PM_{2.5}$ transport over central and eastern China, when limited to the primary $PM_{2.5}$ particles highlighted in this study. Considering less precipitation in the winter monsoon season, how this methodology (FLEXPART-WRF) is proven robustness to quantify the regional transport contribution with the uncertainty range here could mostly rely on a portion of secondary organic and inorganic aerosols, which are resulted from important and complex physiochemical processes in the atmosphere.

References

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Brioude, J., Arnold, D., Stohl, A., Cassiani, M., Morton, D., Seibert, P., Angevine, W., Evan, S., Dingwell, A., Fast, J. D., Easter, R. C., Pisso, I., Burkhart, J., and Wotawa, G.: The Lagrangian particle dispersion model FLEXPART-WRF version 3.1, Geoscientific Model Development, 6, 1889-1904, https://doi.org/10.5194/gmd-6-1889-2013, 2013.

Minor Comments

[1. Line 48: The order of the references is messed up, which should follow the order of the first letter of the first author for each reference alphabetically, and should be "An et al., 2019; Fuzzi et al., 2015; Nel, 2005" for this case. Please check the entire manuscript.]

Response 5: We have corrected all the similar errors in the revised references.

[2. Line 50: The definition of PM2.5 "particulate matter with an aerodynamical diameter equal to or less than 2.5 micrometers".]

Response 6: It has been revised.

[3. Line 99: change "humid environment. (see Fig. 1b)" to "humid environment (see Fig.1b)". There are so many similar typos across the entire manuscript. Please CHECK!]

Response 7: We have corrected all the similar errors in the revised manuscript.

[4. Line 101: The associated temporal variations of $PM_{2.5}$ concentrations for the study period out

of ten sites in Wuhan are strongly recommended to be plotted and placed in the Supplemental.]

Response 8: According to the referee's suggestions, we have added the temporal variations of $PM_{2.5}$ concentrations for the study period out of ten sites in Wuhan in the supplemental file (Fig. s2 and s3).

[5. Line 107: Change "obviously" to "obvious".]

Response 9: It has been revised, "obviously" has been changed to "obvious".

[6. Line 124: "heavy $PM_{2.5}$ pollution the over central-eastern China" should be revised as "heavy PM2.5 pollution over the central-eastern China".]

Response 10: It has been revised.

[7. Line 128: The number and unit should be separated (75 μg m^{-3}). Similar changes should be applied for the entire manuscript.]

Response 11: All the similar errors have been corrected in the revised manuscript.

[8. Line 146: "at same day." should be changed to 'at the same day,".]

Response 12: "at same day." has changed to 'at the same day".

[9. Lines 147-Line 149: The authors use "am" and "a.m." interchangeable. Please be consistent for the entire manuscript. Similar for "pm" and "p.m.".]

Response 13: All the similar errors have been corrected in the revised manuscript.

[10. Lines 161-165: Grammar error here. Please re-write this sentence. And what is the logical relationship between this sentence and the previous one? Do you try to demonstrate the reasons for this result? If so, probably it is better to begin the sentence with "There are several reasons associated with this result. Firstly, ……".]

Response 14: We are so sorry for the grammar error here. Following the referee's suggestion, we have re-written the sentence as follows:

There are several reasons associated with this result. Firstly, the lower near-surface wind speed could alter the concentrations of air pollutants with a weaker advection of cold air, in conjunction with strong subsidence and stable atmospheric stratification, easily producing a stagnation area in the lower troposphere with resulting in regional pollutant accumulations for the development of haze events.

[11. Line 165: what is "CEC" here?]

Response 15: The CEC stands for Central-eastern China; and it has been revised in the manuscript.

[12. Lines 165-170: There are many typos and grammar errors in this sentence. And I am confused by this sentence as well, which looks very odd to me. Is this your statement or conclusion? Several references to support your statement will be necessary.]

Response 16: We are so sorry for the typos and grammar error here. Following the referee's suggestion, we have modified the sentence with adding the relevant references to support our statement as follows:

Secondly, in the presence of high soil moisture, strong surface evaporation could increase the near-surface relative humidity, which is also conducive to hygroscopic growth of participles for haze formation (Dawson et al., 2014; Xu et al. 2016). High air temperature and strong solar radiation could enhance chemical conversions for the formation of secondary aerosols in the atmosphere (He et al., 2012; Huang et al., 2014). Furthermore, precipitation could alter the emissions, and depositions of air pollutants (Dawson et al., 2007; Cheng et al. 2016).

References

Cheng, X., Zhao, T., Gong, S., Xu, X., Han, Y., Yin, Y., Tang, L., He, H., and He, J.: Implications of East Asian summer and winter monsoons for interannual aerosol variations over central-eastern China, Atmospheric Environment, 129, 218-228, https://doi.org/10.1016/j.atmosenv.2016.01.037, 2016.

Dawson, J., Adams, P., and Pandis, S.: Sensitivity of $PM_{2.5}$ to climate in the Eastern US: a modeling case study, Atmospheric chemistry and physics, 7, 4295-4309, https://doi.org/10.5194/acp-7-4295-2007, 2007.

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Xu, X., Zhao, T., Liu, F., Gong, S. L., Kristovich, D., Lu, C., Guo, Y., Cheng, X., Wang, Y., and Ding, G.: Climate modulation of the Tibetan Plateau on haze in China, Atmospheric Chemistry and Physics, 16, 1365-1375, https://doi.org/10.5194/acp-16-1365-2016, 2016.

[13. Lines 184-185: There should be spaces between references, which should be "(Miao et al., 2018; Xu et al., 2016b). There are many cases (e.g., Line 187, 254, 263 and etc) like this. Please check over the entire manuscript.]

Response 17: We have corrected all the similar errors in the revised manuscript.

[14. Line 210: "the stagnation meteorological conditions" should be revised as"meteorological conditions of the stagnation".]

Response 18: It has been revised as suggested by the referee.

[15. Lines 233-234: References relevant to secondary organic and inorganic aerosols study over Wuhan?]

Response 19: Following the referee's suggestion, we have modified the sentence with adding the references relevant to secondary organic and inorganic aerosols study over Wuhan as follows:

The meteorological drivers of air quality change are complicated by a series of physical and chemical processes in the atmosphere especially the formation of secondary air pollutants with strong hygroscopic growth in the humid air environment overlying the dense water network (see Fig. 1b) in the YRMB region (Cheng et al., 2014, He et al., 2012, Huang et al., 2014),

References

Cheng, H., Gong, W., Wang, Z., Zhang, F., Wang, X., Lv, X., Liu, J., Fu, X., and Zhang, G.: Ionic composition of submicron particles ($PM_{1.0}$) during the long-lasting haze period in January 2013 in Wuhan, central China, Journal of Environmental Sciences, 26, 810-817, https://doi.org/10.1016/s1001-0742(13)60503-3, 2014.

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Huang, R. J., Zhang, Y., Bozzetti, C., Ho, K. F., Cao, J. J., Han, Y., Daellenbach, K. R., Slowik, J. G., Platt, S. M., Canonaco, F., Zotter, P., Wolf, R., Pieber, S. M., Bruns, E. A., Crippa, M., Ciarelli, G., Piazzalunga, A., Schwikowski, M., Abbaszade, G., Schnelle-Kreis, J., Zimmermann, R., An, Z., Szidat, S., Baltensperger, U., El Haddad, I., and Prevot, A. S.: High secondary aerosol contribution to particulate pollution during haze events in China, Nature, 514, 218-222, https://doi.org/10.1038/nature13774, 2014.

[16. Line 276: Change "relatively" to "relative".]

Response 20: It has been changed.

[17. Lines 296-299: First of all, there are grammar errors in this sentence (e.g., ...by winter monsoonal winds the from Tongling and Hefei to Wuhan (...). Second of all, the site numbers of Tongling and Hefei are 6 and 5 respectively, as indicated by legend of Figure 6a?)]

Response 21: In the revised manuscript, the grammar errors have been corrected, and the site numbers have been modified as indicated by legend of Figure 6a.

[18. Lines 311-313: It seems that this sentence belongs to the beginning of Section 3.2.]

Response 22: Yes, this sentence (Lines 311-313) has been moved to the beginning of Section 3.2.

[19. Lines 331-333: I recommend that the authors make a plot associated with the modeling domains, which demonstrates the regions with the coarse and finer horizontal resolutions (refer to my major comment #2).]

Response 23: We have added the modeling domains with the coarse and finer horizontal resolutions in the supplemental file (Fig.s4).

[20. Lines 341-342: I have concerns about the release of the number of particles in FLEXPART-WRF. Firstly, for particles from FLEXPART, it is not PM2.5 particles, it is just particles to represent the air parcels. Secondly, can you double check that the model release 50,000 particles per hour? From my understanding, for each hourly mean PM2.5 observation at the receptor list, the release of particles in the 48-h backward trajectory simulation in FLEXTPART just happens in the first hour, with the rest of the time tracking the routes/transport of the particles over the simulation domain?]

Response 24: Yes. Many thanks for the kind review. We have carefully checked our model configuration, and accordingly corrected the errors in the revised manuscript as follows:

For particles from FLEXPART, it is not $PM_{2.5}$ particles, it is just particles to represent the air parcels, and the release of particles in the 48-h backward trajectory simulation in FLEXTPART just happens in the first hour, with the rest of the time tracking the routes/transport of the particles over the simulation domain.

[21. Line 374: Change "Eq (1)" to "Eq. (1)".]

Response 25: It has been corrected.

[22. Lines 634-637: For "K km^{-1}", it should be "K km^{-1}".]

Response 26: It has been revised.

[23. Lines 640-645: There are many typos for Figure 1. For Y-axis title in Figure 1a, it should be "Latitude". Moreover, both units of X-axis and Y-axis in Figure 1a are missing. In Line 643, "YPD" is a typo. And where is the description of PRD here?]

Response 27：Many thanks for the careful review of referee. We are sorry for the typos, which have been corrected in the revised manuscript.

[24. Lines 663-664: The solid line for heavy $PM_{2.5}$ pollution and the dash line for clean air period are missing in the caption for Figure 5.]

Responses 28： We have modified the caption for Figure 5.

[25. Lines 679-680: Why there are no "comma" among "P1 P2 and P3". I suggest changing the caption of the last part of the caption of Figure 7 as ".... pollution periods of P1 (upper panel), P2 (middle panel) and P3 (lower panel), respectively, in January 2016".]

Response 29: We are sorry for the typos. We have add a"comma" between P1 and P2 as well as modified the caption for Figure 7.

Responses to Referee #2

[General comments.

In this manuscript, the authors present an observational analysis to characterize the unique features of meteorological conditions that account for the heavy air pollution events in Wu Han, a metropolis in the Yangtze River Middle Basin, China. and then use a Lagrange particle dispersion model to quantify the percentage contribution of regional transport to such heavy pollution events. They found that $PM_{2.5}$ concentrations show a positive correlation with wind speeds and no stable atmospheric boundary conditions are required to support the accumulation of air pollutants when 24-hr average $PM_{2.5}$ concentrations are higher than 150.0 $\mu g \cdot m^{-3}$. Regional transport driven by strong wind speed contributed more than 65% increase in surface $PM_{2.5}$ concentrations during the development of air pollution events in this region. The study represents a great interest to air quality community given the unique features which are very different from those presented in the textbooks. This version is improved to some extent as compared to the first submission. Part of my comments have been addressed but not all. Especially, the manuscript structure is not re-organized as suggested, a lot of grammar errors or typos need to be corrected throughout the manuscript. In addition, I have several major concerns with the authors' arguments during their analyses and discussion. Thus, a major revision is still required before it is accepted for publication.]

Response 1: Many thanks for the encouraging comments and constructive suggestions on our manuscript acp-2019-758. According to the suggestions of referee, we have re-organized the manuscript structure with detailed descriptions of observational and modeling methods, concerns of technical methodology as well as corrected the grammar errors and typos over the entire manuscript (please find them in the following responses and the highlighted revisions in the revised manuscript).

[Major comments

1. It is strongly recommended to re-organize the structure of the manuscript. Both Methodology and Results/Discussion parts are mixed together in the current version. For instance, it is suggested to move "Model Description (Section 3.2.1)", "Model Configuration (Section 3.2.2)", and the way of calculating "contribution rates" (Lines 360-375 in Section 3.3) to a new section like "Data and methods" (say Section 2 in the new version), and then move part of current Section 2.1, Sections 2.2 and 2.3 to Section 3 like "Results and Discussion" in the revised or new version something like that.]

Response 2: Following the referee's suggestions, we have re-organized the structure of the paper. In the revised manuscript, the descriptions of observational data from various sites and FLEXTPART-WRF (Sect. 3.2.1,Sect. 3.2.2 and the way of calculating "contribution rates" in Section 3.3) are placed in Section 2. And Results and Discussions, including the analysis of the observational data and modeling study, are placed in Section 3 *"Results and Discussion"* .

[2. The East Asian winter Monsoon was mentioned at least 10 times throughout the manuscript to highlight its importance in driving the regional transport during development of heavy pollution events observed in Wuhan. As we know, the East Asian Monsoon represents a seasonal mean behavior and its temporal scale is much longer than that of air pollution events which usually have a scale of one to several day(s) but not longer than one week according to the authors' argument. The authors need show some scientific evidences to support their arguments on how the East Asian winter Monsoon can drive the regional transport which may lead to the development of heavy pollution events. Otherwise the readers may get confused when they read Fig.9b in where the regional transport was from East China other than North China. My suggestion is to limit the emphasis of the East Asian winter Monsoon in this study.]

Response 3: We totally agree with the referee's comments and suggestions. Following the them to correct this misunderstanding on the East Asian Monsoons, we have changed "the East Asian monsoon" to "the cold air activity of East Asian winter monsoon over central-eastern China" to limit the emphasis of the East Asian winter Monsoon in this study (please see the highlighted revisions in the revised manuscript.

[3. Estimate of percentage contribution of regional transport to the heavy pollution events in the YRMB region is one of the major works proposed by this study. As described in Eq.1 and 2, simulation of residence time of $PM_{2.5}$ is critical to do such calculations. Please define residence time. How does the FLEXPART simulate the residence time? A little bit more details are helpful for our readers to understand the percentage contribution of regional transports to the three different episodes.]

Response 4: Thanks for the comments. In the revised manuscript, we have clarified the quantification of regional transport contributions with utilizing the model FLEXPART-WRF in the revised manuscript as followings:

In the model FLEXPART-WRF, the trajectory of a large number of particles released from a source is simulated with consideration of the processes of tracer transport, turbulent diffusion, wet and dry depositions in the atmosphere. With Lagrangian method, it could result in a Jacobian matrix (footprint), in unit of mass per volume per unit flux. Stohl et al. (2005) mathematically derived the residence time for particles out of FLEXPART. Generally, in the backward trajectory of FLEXPART modeling, a large number of particles is released at a receptor and transported backward in time. Then the residence time (not the lifetime) of all particles, normalized by the total number of released particles, is determined on a uniform grid. In this study for the receptor of Wuhan, the residence time for a thickness of 100 m above the surface was calculated and considered the ''footprint'' (in unit of s). By multiplying the residence time with the air pollutant emission flux in the respective grid cell (in unit of μg m^{-2} s^{-1}) calculated from the Multi-resolution Emission Inventory of year 2016 for China (MEIC, http://www.meicmodel.org/), the emission source contribution (in μg m^{-2}) from this grid cell to the receptor could be estimated (Stohl, 2003; Stohl et al., 2005;Ding er al.,2009), yielding a so-called potential source contribution map, which is the geographical distribution of the regional transport contribution rates (%) of the emission source grid cell to $PM_{2.5}$ pollution at the receptor of Wuhan (Fig. 9).

References

Stohl, A., Forster, C., Eckhardt, S., Spichtinger, N., Huntrieser, H., Heland, J., Schlager, H., Wilhelm, S., Arnold, F., and Cooper, O.: A backward modeling study of intercontinental pollution transport using aircraft measurements, Journal of Geophysical Research: Atmospheres, 108, https://doi.org/10.1029/2002jd002862, 2003..

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Ding, A., Wang, T., Xue, L., Gao, J., Stohl, A., Lei, H., Jin, D., Ren, Y., Wang, X., and Wei, X.: Transport of north China air pollution by midlatitude cyclones: Case study of aircraft measurements in summer 2007, Journal of Geophysical Research: Atmospheres, 114, https://doi.org/doi:10.1029/2008JD011023, 2009.

[4. Lines 323-329: I assume that the FLEXPART simulations were driven by the WRF outputs rather than ECMWF or NCEP reanalysis data. If this is the case, please make clarification and delete lines 323-325.]

Response 5: Following the referee's suggestion, we have made clarification with deleting lines 323-325 in the revised manuscript as follows:

In this study on the fine and multiscale modeling of air pollutant sources and regional transport, FLEXPART was coupled offline with the Weather Research and Forecasting Model (WRF) to effectively devise the combined model FLEXPART-WRF.

[5. Fig.5b: We can see that the heavy air pollution events had stronger winds within the 1-km

layer but weaker winds above the 1-km layer. Does this mean that regional transport are mainly limited to the 1-km layer. Some discussions on this will be helpful.]

Response 6: Thanks for the referee's comment. We have accordingly added the following discussions in the revised manuscript:

Compared to the clean air period, the heavy air pollution events had stronger winds within the 1-km layer but weaker winds above the 1-km layer (Fig. 5b), indicating that regional transport of $PM_{2.5}$ was mainly limited to the 1-km layer, especially between 0.25 km and 0.8 km. These vertical structure of horizontal wind could conduce the downward mixing of the regionally transported air pollutants and produce the near-surface accumulations of air pollutants over the YRMB area with elevated ambient $PM_{2.5}$ concentrations, thus contributing to a heavy air pollution.

[6. Writing needs a heavy edit work. There are a lot of grammar errors or typos and many sentences need further improvement. Some of examples include "obviously differences (L107)", "relative high (L109)", "suffering under significant (L133)", "has significantly influence (L162)", "relatively to (L276)", "a horizontally resolution (L344)", etc. I am not going to list all of them since there are many.]

Response 7: Many thanks for the referee's careful review. We are so sorry for the grammar errors or typos, which have been corrected in the revised manuscript.

By the way, this revised manuscript was edited by Elsevier Language Editing Services to improve the English language.

Minor comments:

[1. L19: central China → Central China.]

Response 8: It has been changed.

[2. L20: I am not sure "excessive" is appropriate in this manuscript.]

Response 9: The "excessive $PM_{2.5}$ concentrations" has been changed to "hourly $PM_{2.5}$ concentrations" in the revised manuscript.

[3. L30: I did check "List of regions of China" at Wikipedia at https://en.wikipedia.org/wiki/List_of_regions_of_China, and didn't find "central-eastern China". So "Central China" should be better and sufficient.]

Response 10: I am sorry for the geographical misleading. In the revised manuscript, we have changed *"central-eastern China" to "Central and Eastern China"* covering the major anthropogenic pollutant sources over the vast flatlands from the eastern edges of the Tibetan Plateau and the Loess Plateau to China's Pacific coast for the regional transport of air pollutants toward to YRMB.

[4. L33: FLEXPART-WRF or WRF-FLEAPART? I would suggest the latter since it is WRF-driven FLEXPART. In addition, please define any abbreviated terms at its first appearance. Please check similar issue for other abbreviations throughout the manuscript.]

Response 10: We totally agree with the referee's comments. However, the Lagrangian particle dispersion model FLEXPART-WRF was developed by Brioude et al. (2013), therefore, we adopted the model name FLEXPART-WRF in the manuscript.

All the abbreviated terms were defined at its first appearance in the revised manuscript.

Refereces

Brioude, J., Arnold, D., Stohl, A., Cassiani, M., Morton, D., Seibert, P., Angevine, W., Evan, S., Dingwell, A., Fast, J. D., Easter, R. C., Pisso, I., Burkhart, J., and Wotawa, G.: The Lagrangian particle dispersion model FLEXPART-WRF version 3.1, Geoscientific Model Development, 6, 1889-1904, https://doi.org/10.5194/gmd-6-1889-2013, 2013.

[5. L155-157: Please define these abbreviations at their first appearances.]

Response 11: We have defined these abbreviations at their first appearances in the revised manuscript.

[6. L251: change "the atmospheric stability in the boundary layer" to "the stability of the atmospheric boundary layer"?]

Response 12: It has been changed.

[7. L272: are you sure "it is in Section 3.1"?]

Response 13: It is "3. Regional transport of PM2.5 in heavy air pollution periods".

[8. L342: Please change to (30.61°N, 114.42°E).]

Response 14: It has been changed.

[9. L232-235: I feel a "jump" when I read this sentence.]

Response 15: We have modified this sentence in the revised manuscript as follows:

The meteorological drivers of air quality change are complicated by a series of physical and chemical processes in the atmosphere especially the formation of secondary air pollutants with strong hygroscopic growth in the humid air environment overlying the dense water network (see Fig. 1b) in the YRMB region (Cheng et al., 2014, He et al., 2012, Huang et al., 2014),

References

Cheng, H., Gong, W., Wang, Z., Zhang, F., Wang, X., Lv, X., Liu, J., Fu, X., and Zhang, G.: Ionic composition of submicron particles ($PM_{1.0}$) during the long-lasting haze period in January 2013 in Wuhan, central China, Journal of Environmental Sciences, 26, 810-817, https://doi.org/10.1016/s1001-0742(13)60503-3, 2014.

He, K., Zhao, Q., Ma, Y., Duan, F., Yang, F., Shi, Z., and Chen, G.: Spatial and seasonal variability of $PM_{2.5}$ acidity at two Chinese megacities: insights into the formation of secondary inorganic aerosols, Atmospheric Chemistry and Physics, 12, 1377-1395, https://doi.org/10.5194/acp-12-1377-2012, 2012.

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[10. L352: "The simulated meteorology" → "The simulated meteorological fields".]

Response 16: It has been changed.

[11. L373-374: Change "by calculation of the $PM_{2.5}$ contribution rates with Eq (1)" to "by using the $PM_{2.5}$ contribution rates calculated with Eq.1" something like that.]

Response 17: Thanks for careful editing. We have been accordingly changed in the revised manuscript.

[12. L309-313: I do not think this paragraph is necessary since it does not provide any useful information. Similar issue can be found in other places of the manuscript.]

Response 18: Following the referee's suggestion, we have deleted the unnecessary sentences and paragraphs.

[13. L338-339: What are the horizontal resolutions of the NCEP reanalysis data?]

Response 19: *the horizontal resolutions of the NCEP reanalysis data* is 1°×1°, which has been added in the revised manuscript.

[14. L419-423: Does this paragraph represent any significant findings or conclusions obtained from this study? I am not sure this paragraph is really needed here.]

Response 20: We have accepted the suggestion of referee and deleted this paragraph in the revised manuscript.

[15. L424-426: We know this already and I don't think you need iterate this sentence here. It does not provide any more useful information to me.]

Response 21: Following the suggestion of referee, we have deleted the sentence (L424-426) and modified the paragraph as follows:

This study of environmental and meteorological observations in the YRMB region revealed a unique "non-stagnant" meteorological condition of the boundary layer characterized by strong wind, no inversion layer and a more unstable structure in the atmospheric boundary layer associated with heavy air pollution periods with excessive $PM_{2.5}$ concentrations in the YRMB region, which facilitates understanding of the air pollutant source-receptor relationship of regional air pollutant transport. The study represents a great interest to air quality community given the unique features of air pollution meteorology which are very different from those "stagnant" meteorological conditions presented in the textbooks.

[16. L625-629: Please define WS, T, P, and RH in the description of Table 1 and Table 2.

Response 22: WS, T, P, and RH have been defined with wind speed, air temperature, air pressure and relative humidity respectively in the revised manuscript.

[17. Fig.1b: The font size of those cities shown in Fig.1b is too small. Is it possible to add the locations of 10 sites presented on Page 5 at Lines 101-103 in this plot?]

Response 23: We have added the locations of 10 sites in Wuhan in the supplemental file (Fig. s1).

[18. Fig.9: I believe that the values of the percentage contribution rates are not correct.]

Response 24: Thanks for the comments. We have confirmed that the values of the percentage contribution rates in the Fig.9 are correct. Fig.9 is composed by 151 × 161 grid points, and the total contribution rate of all grid points is 100%.

Heavy air pollution with the unique "non-stagnant" atmospheric boundary layer in the Yangtze River Middle Basin aggravated by regional transport of $PM_{2.5}$ over China

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Abstract: Regional transport of air pollutants controlled by both emission sources and meteorological factors results in a complex source-receptor relationship of air pollution change.

Wuhan, a metropolis in the Yangtze River Middle Basin (YRMB) of ~~c~~Central China experienced heavy air pollution characterized by ~~excessive~~hourly $PM_{2.5}$ concentrations reaching 471.1 μg m^{-3} in January 2016. In order to investigate the regional transport of $PM_{2.5}$ over China and the meteorological impact on wintertime air pollution in the YRMB area, observational meteorological and other relevant environmental data from January 2016 were analyzed. Our analysis presented the noteworthy cases of heavy $PM_{2.5}$ pollution in the YRMB area with the unique "non-stagnant" meteorological conditions of strong northerly winds, no temperature inversion and additional unstable structures in the atmospheric boundary layer. This unique set of conditions differed from the stagnant meteorological conditions characterized by near-surface weak winds, air temperature inversion, and stable structure in the boundary layer observed in heavy air pollution over most regions in China. The regional transport of $PM_{2.5}$ over C~~c~~entral and ~~-~~E~~e~~astern China (CEC) aggravated $PM_{2.5}$ levels present in the YRMB area, thus demonstrating the source-receptor relationship between the originating air pollution regions in ~~central-eastern~~~~Central-eastern China~~CEC and the receiving YRMB regions. Furthermore, a backward trajectory simulation using FLEXPART-WRF to integrate the air pollutant emission inventory over China was used to explore the patterns of regional transport of $PM_{2.5}$ governed by the strong northerly winds in the cold air activity of the East Asian winter monsoon over ~~central-eastern~~~~Central-eastern China~~CEC, which contributes markedly to the heavy $PM_{2.5}$ pollution in the YRMB area. It was estimated that the regional transport of $PM_{2.5}$ of non-local air pollutant emissions could contribute more than 65% of the $PM_{2.5}$ concentrations to the heavy air pollution in the YRMB region during the study period, revealing the importance of the regional transport of air pollutants over ~~central-eastern~~~~Central-eastern~~ China in the formation of heavy air pollution over the YRMB region.

Key words: $PM_{2.5}$ pollution; Yangtze River Middle Basin; meteorological condition; regional transport; FLEXPART-WRF

1. Introduction

~~Air~~Haze pollution events could result in serious environmental problems with adverse influence on traffic, human health, climate change and other significant aspects (An et al., 2019; Fuzzi et al., 2015; ~~An et al., 2019;~~ Nel, 2005). Based on the observations in China, there is a well-established association between haze pollution and high concentrations of $PM_{2.5}$ (particulate matter with an aerodynamical diameter equal to or less than 2.5 μm). Air pollution levels are highly dependent on emissions of air pollutants and changes in meteorology (An et al., 2019; Tie et al., 2017; Xu et al., 2016b; ~~An et al., 2019;~~ Xu et al., 2016a). The accumulation, maintenance and dissipation of haze pollution events are generally determined by meteorological changes (Zhang et al., 2014; Zhang et al., 2015~~Kan et al., 2012~~), among which the boundary layer structures play the most important role (Zhao et al., 2015). Meteorological conditions of stagnation characterized by near-surface low winds, high humidity and stable boundary layer could govern the periodic variations of haze pollution, which present as typical wintertime air pollution in ~~central-eastern~~Central-eastern China (Huang et al., 2018; Xu et al., 2016b; Zhang et al., 2014~~; Huang et al., 2018~~). Four major regions exhibiting haze pollution with high $PM_{2.5}$ concentrations and overall poor air quality are centered over North China Plain (NCP), Yangtze River Delta (YRD) in East China, Pearl River Delta (PRD) in South China and Sichuan Basin

(SCB) in Southwest China (Cheng et al., 2008; Zhang et al., 2012; Deng et al., 2011; Qiao et al., 2019; Tie et al., 2017; Wang et al., 2016; Zhang et al., 2012;).

The source-receptor relationship describes the impacts of emissions from an upwind source region to pollutant concentrations or deposition at a downwind receptor location. Regional transport of source-receptor air pollutants is generally complicated by two types of – factors: emission and meteorology. The emission factor includes the emission source strength, chemical transformation and production; the meteorological factor determines the transport pathway from the source to receptor regions, exchanges between boundary layer and free troposphere, the removal processes occurring over the source and receptor regions as well as along the transport pathways. Regional transport of air pollutants with the source-receptor relationship is an important issue in our understanding of changes in air quality. Driven by atmospheric circulation, the regional transport of $PM_{2.5}$ from source regions can deteriorate air quality in the downwind receptor regions, leading to the regional haze pollution observed in a large area over Ccentral-eastern China –(Chang et al., 2018; Wang et al., 2014; He et al., 2017; Chen et al., 2017b; He et al., 2017; Hu et al., 2018; Jiang et al., 2015; Wang et al., 2014;). The Yangtze River Middle Basin (YRMB) in central China is geographically surrounded by four major haze pollution regions in all directions with NCP to the north, the YRD to the east, the PRD to the south and the SCB to the west (Fig.1–a). Due to this specialized location of the YRMB as a regional air pollutant transport hub with subbasin topography (see Fig. 1b), the regional transport of air pollutants driven by the cold air activity of East Asian winter monsoonal winds in central-easternCentral- and Eeastern China could develop a source-receptor relationship between major haze pollution regions (NCP, YRD, etc.) in central-easternCentral-eastern China and the downwind YRMB region. However, there are unresolved questions regarding the meteorological processes involved in the regional transport of air pollutants~~–~~ and the pattern of regional transport with contribution to the air quality changes observed in the YRMB.

Wuhan, a metropolis located in the YRMB, has confronted the problems associated with urban air pollution, especially heavy $PM_{2.5}$ pollution events that occur in the winter (~~Zhong et al., 2014;~~ Gong et al., 2015;~~ Tan et al., 2015;~~ Xu et al., 2017~~; Tan et al., 2015 Zhong et al., 2014~~). Local emissions of air pollutants from urban transportation, industrial exhaust and bio-combustion play an important role in YRMB urban air pollution (Acciai et al., 2017; Zhang et al., 2015). Many observational and modeling studies on air pollution in this urban area have been conducted (Wu et al., 2018; Zheng et al., 2019~~; Wu et al., 2018~~). However, regional transport routes of $PM_{2.5}$ from ~~central-eastern~~Central-eastern China and its contribution to air pollution over the YRMB are still poorly understood, especially in relation to heavy air pollution episodes in the YRMB area. This study selected the Wuhan area as a representative area within the YRMB for investigation of the meteorological conditions of air pollution events in January 2016 and the contribution of regional transport of $PM_{2.5}$ to heavy air pollution over the YRMB region.

2. Data and methods~~Observational analysis~~

2.1 Data

Wuhan, the capital of Hubei province, is located across the Yangtze River, where its surrounding water network attributed with a humid environment~~.~~ (see Fig. 1b). In order to analyze the air quality change, the hourly concentrations of air pollutants including $PM_{2.5}$ in January 2016 were collected from the national air quality monitoring network operated by the Ministry of ecology and environmental protection of Chinasites (http://www.mee.gov.cn/) over central-easternCentral-e and Eastern China, including ten observational sites in Wuhan-. These ten sites include nine urban sites in residential and industrial zones as well as one suburban site within , The mass volume concentrations of surface $PM_{2.5}$ are operationally hourly observed with the instrument of the Thermo Fisher Scientifi. The observation data are under quality control based on the China's national standard of air quality observation before released by the Ministry of ecology and environmental protection of China. The concentrations of air pollutants were distributed spatially in less difference over the suburban and urban sites with the similar patterns and peaks of hourly changes during the heavy pollution events, demonstrating the regional heavy air pollution in a large area of the YRMB region with the contribution of regional transport from central-easternCentral-eEastern China, while the obviously differences in air pollutant concentrations were measured with the relative high and low $PM_{2.5}$ concentrations respectively at –urban sites and suburban site during the clean air period, reflecting the important influence of high air pollutant emission over urban area on local air quality.

The $PM_{2.5}$ concentrations averaged over the ten observational sites were used to characterize the variations of air pollution in January 2016 over this urban area within the YRMB. The surface $PM_{2.5}$ concentrations averaged over 10 observation sites in Wuhan are used to calculate the correlation coefficients with the changing meteorological drivers (wind speed/temperature etc.) over Wuhan in January 2016 to investigate the local meteorological influences on hourly changes of surface $PM_{2.5}$ concentrations in Wuhan.

The meteorological data of surface observation and air sounding in Wuhan and other observatories in Central-eEastern China were obtained from the China Meteorological Data

Sharing Network (http://data.cma.cn/). Meteorological data selected for this study included horizontal visibility, air temperature, relative humidity, air pressure, and wind speed and direction with temporal resolutions of 3 h for surface observation and 12 h for sounding observation in order to analyze the variations of the meteorological conditions in the atmospheric boundary layer in January 2016.

The ERA (ECMWF ReAnalysis) -Interim reanalysis data of meteorology from the ECMWF (European Centre for Medium-Range Weather Forecasts) (https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/) were applied to explore the cold air activity of East Asian winter monsoonal winds in January 2016 and their anomalies during heavy $PM_{2.5}$ pollution ~~the~~ over the ~~e~~ ~~entral-eastern~~Central-eastern China.

2.2 FLEXPART-WRF model

2.2.1 Model description

The Flexible Particle dispersion (FLEXPART) model (Stohl, 2003) is a Lagrange particle diffusion model developed by the Norwegian Institute for Air Research (NIAR). In this model, the trajectory of a large number of particles released from a source is simulated with consideration of the processes of tracer transport, turbulent diffusion, and wet and dry depositions in the atmosphere (Brioude et al., 2013). Applying backward trajectory simulation can determine the distribution of potential source regions that may have an impact on a target point or receptor region (Chen et al., 2017a; Chen et al., 2017b; Seibert and Frank, 2003;Zhai et al., 2016).

Initially, FLEXPART could be driven by the global reanalysis meteorological data obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF) or the National

Centers of Environmental Prediction (NCEP). In this study on the fine and multiscale modeling of air pollutant sources and regional transport, FLEXPART was coupled offline with the Weather Research and Forecasting Model (WRF) to effectively devise the combined model FLEXPART-WRF (Fast and Easter, 2006; Brioude et al., 2013), which has been widely used to investigate the potential sources of air pollutants in consideration of environmental change (An et al., 2014; De Foy et al., 2011; Sauvage et al., 2017; Stohl, 2003).

2.2.2 WRF Modeling configuration and validation

The WRF model was configured with two nested domains. The coarse domain covered the entirety of Asia with a 30 km×30 km horizontal resolution, and the nested fine domain included most of China and surrounding regions with a 10 km×10 km horizontal resolution. The physical parameterizations used in WRF were selected with the Morrison microphysics scheme (Morrison, 2009), the Rapid Radiative Transfer Model (RRTM) scheme for long and short wave radiation (Mlawer et al., 1997), the Yonsei University (YSU) boundary layer scheme (Hong, 2006), Grell 3D cumulus parameterization, and the Noah land surface scheme (Grell et al., 2005). Driven with the reanalysis meteorological data in the horizontal resolutions of 1°×1° obtained from NCEP for initial and boundary meteorological conditions, the WRF simulation ran 12 h each time with the first 6 h simulations constituting spin-up time.

The WRF-simulated meteorological fields, which included wind speed, air temperature, relative humidity and surface pressure, were compared with observations at five sites (Wuhan, Changsha, Hefei, Zhengzhou and Nanchang) over Central-eastern China. The correlation coefficients and normalized standardized deviations were calculated and are shown in Figure 8

(Taylor, 2001). Based on the results with correlation coefficients passing the significance level of 0.001 and low normalized standardized deviations (Fig. 8), it was confirmed that WRF-modeled meteorology that is consistent with observations could be used to drive the FLEXPART backward trajectory simulation in this study.

2.3 Estimating contribution of regional transport of $PM_{2.5}$ to heavy pollution

In the model FLEXPART-WRF, the trajectory of a large number of particles released from a source is simulated with consideration of the processes of tracer transport, turbulent diffusion, wet and dry depositions in the atmosphere. With Lagrangian method, it could result in a Jacobian matrix (footprint), in unit of mass per volume per unit flux. Stohl et al. (2005) mathematically derived the residence time for particles out of FLEXPART. Generally, in the backward trajectory of FLEXPART modeling, a large number of particles is released at a receptor and transported backward in time. Then the residence time (not the lifetime) of all particles, normalized by the total number of released particles, is determined on a uniform grid. In this study for the receptor of Wuhan, the residence time for a thickness of 100 m above the surface was calculated and considered the ‘‘footprint’’ (in unit of s). By multiplying the residence time with the air pollutant emission flux in the respective grid cell (in unit of μg m^{-2} s^{-1}) calculated from the Multi-resolution Emission Inventory of year 2016 for China (MEIC, http://www.meicmodel.org/), the emission source contribution (in μg m^{-2}) from this grid cell to the receptor could be estimated (Stohl, 2003; Stohl et al., 2005;Ding er al.,2009)

In this study, the FLEXPART-WRF simulation was conducted for the 48-hr backward trajectory with a release of 50,000 particles at first hour in Wuhan (30.61 ° N, 114.42 ° E) for

January 2016. The 48-hr backward trajectory simulation results were output with the residence time of $PM_{2.5}$ particles in a horizontally resolution of 0.1°×0.1°. The FLEXPART simulations of $PM_{2.5}$ particle residence time over the 48-hr backward trajectory pathways were multiplied with the regional primary $PM_{2.5}$ emission fluxes to quantify the contribution of regional transport of $PM_{2.5}$ to air quality change in the YRMB region with identifying the patterns of regional transport of $PM_{2.5}$ over Central and Eastern China. The primary $PM_{2.5}$ emission data of 2016 obtained from the Multi-resolution Emission Inventory for China (MEIC, http://www.meicmodel.org/) were selected for use as the regional $PM_{2.5}$ emission fluxes in this study.

Based on the FLEXPART-WRF backward trajectory simulation, the upstream sources of $PM_{2.5}$ emissions for heavy air pollution in Wuhan could be identified. The contribution rates $\boldsymbol{rate_{i,j}}$ of regional transport of $PM_{2.5}$ from the upstream sources to air pollution in the downstream receptor region of YRMB were calculated by Eq.(1), and the total contribution $\boldsymbol{R}$ of regional transport from the non-local emission sources are estimated by Eq. (2) (Chen et al., 2017b).

$$rate_{i,j} = \frac{E_{i,j} \times r_{i,j}}{\sum_{1,1}^{N,S} E_{i,j} \times r_{i,j}} \qquad (1)$$

$$R = \sum_{(N_1,S_1)}^{(N_2,S_2)} rate_{i,j} \qquad (2)$$

where the subscripts $\boldsymbol{i}$ and $\mathbf{j}$ represent a grid location; $\mathbf{r_{i,j}}$ represents the residence time of $PM_{2.5}$ particles simulated by FLEXPART-WRF; and, $E_{i,j}$ represents the $PM_{2.5}$ emission flux over the grid. The first grid location (N_1, S_1) and the last grid location (N_2, S_2) over the non-local emission sources and the local area of Wuhan were determined respectively by the regional transport of $PM_{2.5}$ pathways and the YRMB region as simulated by FLEXPART-WRF.

3. Results and Discussion

3~~2~~.1~~2~~ Variations in $PM_{2.5}$ concentrations and meteorology in January, 2016

Based on the National Ambient Air Quality Standards of China released by the Ministry of Ecology and Environment of China in 2012 (http://www.mee.gov.cn/), light and heavy air pollution levels of $PM_{2.5}$ are categorized by daily average $PM_{2.5}$ concentration exceeding 75 ~~μg m^{-3}~~ and 150 μgm^{-3} in ambient air, respectively. The daily variations of $PM_{2.5}$ concentrations over January 2016 in Wuhan are illustrated in Figure 2a. The average monthly $PM_{2.5}$ concentration reached 105.8μg m^{-3}. The national secondary standard was exceeded on 27 days with daily $PM_{2.5}$ concentrations exceeding 75 μg m^{-3} during the entire month of January 2016 in Wuhan, indicating that this urban area in the YRMB was suffering under significant $PM_{2.5}$ pollution during this period. As shown in Figure 2a, a 21-day prolonged air pollution event resulted from high levels of daily $PM_{2.5}$ concentrations (>75 μg m^{-3}) over the period of January 1 to 21. During this 21-day period of air pollution, three notably heavy air pollution events occurred on January 4, 10-12 and 18 with excessive daily $PM_{2.5}$ concentrations (>150 μg m^{-3}); these events are marked as P1, P2 and P3 in Figure 2. Based on the observation in January 2016, we found the interesting phenomenon of an apparent 7-day cycle of heavy air pollution in January 2016, reflecting an important modulation of meteorological oscillation in the East Asian winter monsoon affecting air pollution concentrations observed over the YRMB region (Xu et al., 2016a). A period analysis on long-term observation data of air quality could provide more information on air pollution oscillations with meteorological drivers.

Figure 2b presents the hourly changes of $PM_{2.5}$ concentrations for the three heavy air pollution events P1, P2 and P3. The heavy pollution event P1 on January 4 started at 11:00 a.m. (local time is used for all events) and ended at 11:00 p.m. at the same day. with an observed $PM_{2.5}$ concentration peak of 471.1 μg m^{-3}. The event P2 occurred from 10:00 p.m. on January 10 to 00:00 a.m. on January 12 with a duration of 26 h and two peaks in $PM_{2.5}$ concentrations of 231.4 μg m^{-3} and 210.6 μg m^{-3}. The event P3 was observed between 7:00 p.m. on January 17 and 2:00 p.m. on January 18 with an explosive growth rate of 42.9 μg m^{-3} h^{-1} in $PM_{2.5}$ concentrations. Those three heavy $PM_{2.5}$ pollution episodes over the YRMB region were characterized by short durations of less than 26 h from rapid accumulation to fast dissipation.

Using the environmental and meteorological data observed in Wuhan in January 2016, the effects of the meteorological conditions on $PM_{2.5}$ concentrations in the YRMB region were statistically analyzed in regards to hourly variations of surface $PM_{2.5}$ concentrations, near-surface wind speed (WS) and wind direction (WD), as well as surface air temperature (T), air pressure (P) and relative humidity (RH) (Fig. 3). Among the observed hourly changes in $PM_{2.5}$ concentrations and meteorological elements shown in Figure 3, the obvious positive correlations to surface air temperature and relative humidity, as well as a pronounced negative correlation to surface air pressure and a weak positive correlation to near-surface wind speed were found with the change of $PM_{2.5}$ concentrations in January 2016 (Table 1). ~~T~~There are several reasons associated with this result. Firstly, the lower near-surface wind speed could alter the concentrations of air pollutants with a weaker advection of cold air, in conjunction with strong subsidence and stable atmospheric stratification, easily producing a stagnation area in the lower troposphere with resulting in regional pollutant accumulations for the development of haze events. Secondly~~he near-surface wind speed associated with East Asian monsoons has significantly~~

~~influence concentrations of air pollutants mainly by the changes in weak advection of cold air, in conjunction with strong subsidence and stable atmospheric stratification, can easily produce a stagnation area in the lower troposphere resulting in regional pollutant accumulations, which are favorable for the development of CEC haze~~ in the ~~Central-eastern China events. In addition, i~~, in the presence of high soil moisture, strong surface evaporation could increase the near-surface relative humidity, which is also conducive to hygroscopic growth of participles for haze formation (Dawson et al., 2014; Xu et al. 2016). High air temperature and strong solar radiation could enhance chemical conversions for the formation of secondary aerosols in the atmosphere (He et al., 2012; Huang et al., 2014). Furthermore, precipitation could alter the emissions, and depositions of air pollutants (Dawson et al., 2007; Cheng et al. 2016). T~~n the presence of high soil moisture, strong surface evaporation results in increases in the near-surface relative humidity, which is also conducive to hygroscopic growth of participles for haze formation; high air temperature and strong solar radiation could enhance chemical reactions and conversions for the formation of secondary aerosols in the atmosphere, precipitation could alter the emissions, and depositions of air pollutants. T~~hese observations could reflect the special influences of meteorological factors (winds, air temperature, humidity, precipitation ~~etc~~etc.) on physical and chemical processes in the ambient atmosphere, in particular that of wind driving air pollutant transport and affecting air quality change in the YRMB region.

When we focused on the changes leading to excessive $PM_{2.5}$ levels during these heavy air pollution events, it is noteworthy that all three heavy pollution episodes P1, P2 and P3 were accompanied with strong near-surface wind speeds in the northerly direction, as well as evident turning points in prevailing conditions leading to falling surface air temperatures and increasing surface air pressure (noted as a rectangle with red dashed lines in Fig. 3). The conditions observed during these three heavy pollution episodes reflect the typical meteorological characteristics of cold front activity over the East Asian monsoon region. The southward advance of a cold front could drive the regional transport of air pollutants over ~~central-eastern~~Central-eastern China (Kang et al., 2019). Climatologically, a strong northerly wind, low air temperature and high air pressure are typical features of an incursion of cold air during East Asian winter monsoon season in ~~central-eastern~~Central-eastern China, which could disperse air pollutants and improve air quality in the NCP region (Miao et al., 2018; Xu et al., 2016b). Compared to the meteorological conditions for stagnation with weak winds observed for heavy air pollution events in the major air pollution regions of ~~central-eastern~~Central-eastern China (Ding et al., 2017; Huang et al., 2018~~; Ding et al., 2017~~), meteorological conditions with strong near-surface wind were anomalously accompanied with the intensification of $PM_{2.5}$ during heavy air pollution periods over the study area in the YRMB in January 2016 (Fig. 3). This could imply the importance of regional air pollutant transport in worsening air quality over the YRMB, driven by the strong northerly winds of the East Asian winter monsoon over China.

3.2~~.3~~ A unique "non-stagnation" meteorological condition for heavy $PM_{2.5}$ pollution

To further investigate the connection of meteorological elements in the near-surface layer with changes in air quality affected by $PM_{2.5}$ concentrations in the YRMB region, we carried out a more detailed correlation analysis of $PM_{2.5}$ concentrations in Wuhan with near-surface wind speed and air temperature and three different levels of $PM_{2.5}$ concentrations: clean air environment ($PM_{2.5}$<75 μg m^{-3}), light air pollution (75 μg m^{-3} ≤ $PM_{2.5}$ <150 μg m^{-3}) and heavy air pollution

($PM_{2.5}$ ≥150 μg m^{-3}) periods (Table 2). As seen in Table 2, the surface $PM_{2.5}$ concentrations were positively correlated with air temperature, as well as negatively correlated with wind speeds during the periods of clean air environment and light air pollution. It should be emphasized here that a significantly negative correlation (R=-0.19) of $PM_{2.5}$ concentrations with near-surface wind speeds for the light air pollution period could indicate that weak winds are favorable for local $PM_{2.5}$ accumulation, reflecting an important effect of local air pollutant emissions on light air pollution periods over the YRMB area. In January 2016, the overall wind speed of Wuhan was weak with a monthly mean value of 2.0 m s^{-1}, which could prove beneficial to maintaining the high $PM_{2.5}$ levels in the prolonged air pollution event experienced during January 2016. However, a significantly positive correlation (R=0.41) existed between excessive $PM_{2.5}$ concentrations ($PM_{2.5}$ >150 μg m^{-3}) and strong near-surface wind speeds during the heavy air pollution period, which was inconsistent with ~~the stagnation~~ meteorological conditions of the stagnation observed in the near-surface layer with weak winds associated with heavy air pollution in eastern China (Cao et al., 2012;~~ Zhang et al., 2016~~). The meteorology and environment conditions in the YRMB region indicate the close association of heavy air pollution periods with the intensification of regional transport of air pollutants driven by strong winds (Fig. 3, Table 2) reflecting a key role of regional air pollutant transport in the development of the YRMB's heavy air pollution periods.

In order to clearly illustrate the impact of wind speed and direction on the $PM_{2.5}$ concentrations associated with the regional transport of upwind air pollutants, Figure 4 presents the relation of hourly changes in surface $PM_{2.5}$ concentrations (in color contours) to near-surface wind speed (in radius of round) and direction (in angles of round) in Wuhan during January 2016. As can be seen in Figure 4, strong northerly winds of the East Asian winter monsoon accompanied extremely high

$PM_{2.5}$ concentrations (>150 μg m^{-3}) during heavy air pollution periods, including the northeast gale that exceeded 5 m·s^{-1} during the extreme heavy pollution period with excessive high $PM_{2.5}$ concentrations (>300 μg m^{-3}) over the YRMB region. These results reveal a unique meteorological condition of “non-stagnation” with strong winds during events of heavy air pollution over YRMB area. Conversely, the observed $PM_{2.5}$ concentrations ranging between 75 and 150 μg m^{-3} for light air pollution periods generally corresponded with low wind speed (<2 m s^{-1}) in the YRMB region (Fig. 4); therefore, it is the meteorological condition for stagnation characterized by weak winds involved in the accumulation of local air pollutants that is responsible for the YRMB’s light air pollution periods. Meteorological impacts on air quality could include not only the stagnation condition with weak winds and stable boundary layer, but also air temperature, ~~humidity, precipitation~~humidity, precipitation, atmospheric radiation etc. in close connection with atmospheric physical and chemical processes. The Meteorological drivers of air quality change are complicated by a series of physical and chemical processes in the atmosphere especially the formation of secondary air pollutants with strong hygroscopic growth in the humid air environment overlying the dense water network (see Fig. 1b) in the YRMB region (Cheng et al., 2014, He et al., 2012, Huang et al., 2014).~~Therefore, meteorological~~Therefore, meteorological ~~drivers of air quality change are complicated by a series of physical and chemical processes in the atmosphere, especially the formation of secondary air pollutants in the humid air environment overlying the dense water network in the YRMB region (see Fig. 1b), thus pointing out the need for further comprehensive study.~~

As shown in Figure 2a, the heavy pollution periods with the daily average $PM_{2.5}$ concentrations exceeding 150 μg m^{-3} in ambient air, respectively occurred on January 4, 10-12 and

18, and the clean air periods with the daily average $PM_{2.5}$ concentrations below 75 μg m^{-3} occurred on January 22 and 24-27, 2016, in the YRMB region. The air sounding data of Wuhan were used to compare the structures of the atmospheric boundary layer of the heavy air pollution and clean air periods. Figure 5 presents the vertical profiles of air temperature, wind velocity and potential temperature averaged for the heavy $PM_{2.5}$ pollution and clean air periods in January 2016. It can be clearly seen that the inversion layer of air temperature did not exist during the heavy pollution periods, but a near-surface inversion layer appeared at the height of about 200 m during the clean air periods (Fig. 5a). Compared to the clean air period, the heavy air pollution events had stronger winds within the 1-km layer but weaker winds above the 1-km layer (Fig. 5b), indicating that regional transport of $PM_{2.5}$ was mainly limited to the 1-km layer, especially between 0.25 km and 0.8 km. These vertical structure of horizontal wind could conduce the downward mixing of the regionally transported air pollutants and produce the near-surface accumulations of air pollutants over the YRMB area with elevated ambient $PM_{2.5}$ concentrations, thus contributing to a heavy air pollution.

The comparison of vertical profiles of horizontal wind velocity experienced during the clean air periods further revealed the stronger wind speed observed in the heavy air pollution period below a height of 850 m located in the atmospheric boundary layer exhibiting the vertical structure similar to a low-level jet stream (Fig. 5b); these conditions could conduce the downward mixing of the regionally transported air pollutants and produce a local near-surface accumulation in the YRMB area with elevated ambient $PM_{2.5}$ concentrations, thus contributing to a heavy air pollution.

To characterize the stability of the atmospheric boundary layer the atmospheric stability in the boundary layer, the vertical profiles of potential air temperature (θ) were calculated with air temperature and pressure (Fig. 5c). The vertical change rate of θ was used to quantify the static stability of the boundary layer in this study (Oke, 2002; Sheng et al., 2003). A lower vertical change rate of θ generally indicates a decreasing stability or increasing instability of the boundary layer. The averaged static stability values of the near-surface layer below a height of 200 m during the heavy pollution and clean air periods were approximately 4.4 K~~·~~ km^{-1} and 13.2 K km^{-1}, respectively (Table 3). This obvious decrease in stability of the boundary layer from clean air to heavy pollution periods reflects an anomalous tendency for instability in the boundary layer during heavy pollution periods in the YRMB region during January 2016.

The meteorological conditions of stagnation characterized by weak wind, temperature inversion and a stable vertical structure of the atmospheric boundary layer is generally accepted as the typical meteorological drivers for heavy air pollution (An et al., 2019; Ding et al., 2017). Nevertheless, this study of environmental and meteorological observations in the YRMB region has revealed a unique meteorological condition of "non-stagnation" in the atmospheric boundary layer during heavy air pollution periods characterized by strong wind, lack of an inversion layer and a more unstable structure of the atmospheric boundary layer; these conditions are generally regarded as the typical pattern of atmospheric circulation that facilitates the regional transport of air pollutants from upstream source to downwind receptor regions. Regional transport of $PM_{2.5}$ associated with the source-receptor relationship between the air pollution regions in ~~central-eastern~~Central-eastern China and the YRMB was investigated based on the observational analysis described in Sect~~.~~ 3.~~1~~.

3. Regional transport of $PM_{2.5}$ in heavy air pollution periods

3.13 Changes of $PM_{2.5}$ and winds observed in central-easternCentral- and Eeastern China

The monthly averages of observed $PM_{2.5}$ concentrations and the anomalies of wind speed averaged in three heavy air pollution periods relatively to the monthly mean wind speed in January 2016 over central-easternCentral-eastern China are shown in Figure 6. In January 2016, a large area of central-easternCentral-eastern China experienced air pollution with high levels of $PM_{2.5}$ (>75 μg m^{-3}), especially serious in the NCP region and the Fenhe-Weihe Plain in central China (Fig. 6a). As seen in Figure 6, the YRMB region (Site 1, Wuhan) was situated in the downwind southern edge of an observed air pollution area located over central-easternCentral-eastern China, where the northerly winds of the East Asian winter monsoon prevail climatologically in January (Ding, 1994). It is notable that the anomalously stronger northerly winds were observed over the upstream region in central-easternCentral-eastern China during three periods of wintertime heavy $PM_{2.5}$ pollution in the YRMB region (Fig. 6b). Driven by the strong northerly winter monsoonal winds (Fig. 6b), the regional transport of air pollutants from the source regions in central-easternCentral-eastern China could largely contribute to wintertime heavy air pollution periods in the downwind receptor region of YRMB.

In order to explore the connection of regional transport of $PM_{2.5}$ over central-easternCentral-eastern China to three events of heavy air pollution in the YRMB region, six observational sites were selected from the northwestern, northern and northeastern upwind areas located over central-easternCentral-eastern China (Fig. 6a) to represent the temporal $PM_{2.5}$

and wind variations along the different routes of regional transport of $PM_{2.5}$ with the southward incursion of stronger northerly winds of East Asian monsoon across ~~central-eastern~~Central-eastern China (Fig. 7). —The southeastward movement of heavy $PM_{2.5}$ pollution driven by stronger northerly winds from Luoyang and Xinyang to Wuhan (Sites 3, 2, and 1 in Fig. 6) presents a northwestern route of regional transport of $PM_{2.5}$ for the heavy air pollution period P1 in the YRMB (see upper panels of Fig. 7). The westward ~~southwestward~~ advance of $PM_{2.5}$ peaks governed by ~~winter monsoonal~~eastern winds ~~the~~ from Tongling and Hefei to Wuhan (Sites ~~5~~6, ~~6~~5, and 1 in Fig. 6). Regional transport of $PM_{2.5}$ across Eastern China to the YRMB exert~~ed~~ed a significant impact on the heavy air pollution period P2 (see middle panels of Fig. 7). ~~aggravated by regional transport of $PM_{2.5}$ across Eastern China to the YRMB region (see middle panels of Fig. 7).~~ A northern pathway of regional transport of $PM_{2.5}$ connected Zhengzhou and Xinyang to Wuhan (Sites 4, 2, and 1 in Fig. 6) during the YRMB's heavy air pollution period P3 with anomalously strong northerly winds (see Fig. 6b and lower panels of Fig. 7). It is noteworthy in Fig. 7 that the heavy $PM_{2.5}$ pollution periods at the upstream sites Hefei, Tongling, Luoyang, Xinyang and Zhengzhou (Fig. 6a) were generally dispelled by strong northerly winds, while strong northerly winds could trigger the periods of heavy $PM_{2.5}$ pollution in the YRMB region (Wuhan, Fig. 6), and such inverse effects of strong winds on heavy air pollution in the source and receptor regions reflect an important role of regional air pollutant transport in worsening air pollution in the YRMB's receptor region.

The regional transport over ~~central-eastern~~Central and -E~~e~~astern China associated with the source-receptor relationship directing heavy $PM_{2.5}$ pollution to the YRMB region was revealed with observational analysis. Backward trajectory modeling with FLEXPART-WRF was used to further confirm the patterns of regional transport of $PM_{2.5}$ over central-easternCentral-eastern China and estimate the resulting contribution to heavy air pollution in the YRMB region, as described in the following Sects.

3.2 FLEXPART-WRF model

3.2.1 Model description

The Flexible Particle dispersion (FLEXPART) model (Stohl, 2003) is a Lagrange particle diffusion model developed by the Norwegian Institute for Air Research (NIAR). In this model, the trajectory of a large number of particles released from a source is simulated with consideration of the processes of tracer transport, turbulent diffusion, and wet and dry depositions in the atmosphere (Brioude et al., 2013). Applying backward trajectory simulation can determine the distribution of potential source regions that may have an impact on a target point or receptor region (Seibert and Frank, 2003;Zhai et al., 2016;Chen et al., 2017a; Chen et al., 2017b; Seibert and Frank, 2003;Zhai et al., 2016).

Initially, FLEXPART could be driven by the global reanalysis meteorological data obtained from the European Centre for Medium-Range Weather Forecasts (ECMWF) or the National Centers of Environmental Prediction (NCEP). For the refined simulation of air pollutant sources and transport, FLEXPART was coupled offline with the Weather Research and Forecasting Model (WRF) to effectively devise the combined model FLEXPART-WRF (Fast and Easter, 2006), which *has been widely used to investigate the potential sources of air pollutants in consideration of* environmental change (An et al., 2014; De Foy et al., 2011;Stohl, 2003;De Foy et al., 2011; An et al., 2014; Sauvage et al., 2017; Stohl, 2003).

3.2.2 Model configuration

The WRF model was configured with two nested domains. The coarse domain covered the entirety of Asia with a 30 km×30 km horizontal resolution, and the nested fine domain included most of China and surrounding regions with a 10 km×10 km horizontal resolution. The physical parameterizations used in WRF were selected with the Morrison microphysics scheme (Morrison, 2009), the Rapid Radiative Transfer Model (RRTM) scheme for long and short wave radiation (Mlawer et al., 1997), the Yonsei University (YSU) boundary layer scheme (Hong, 2006), Grell 3D cumulus parameterization, and the Noah land surface scheme (Grell et al., 2005). Driven with the reanalysis meteorological data obtained from NCEP for initial and boundary meteorological conditions, the WRF simulation ran 12 h each time with the first 6 h simulations constituting spin-up time.

The FLEXPART-WRF simulation was conducted for the 48-hr backward trajectory with a release of 50,000 $PM_{2.5}$ particles per at first hour in Wuhan (30.61°N, 114.42°E) for January 2016. The 48-hr backward trajectory simulation results were output with the residence time of $PM_{2.5}$ particles in a horizontally resolution of 0.1°×0.1°. The FLEXPART simulations of $PM_{2.5}$ particle residence time over the 48-hr backward trajectory pathways were multiplied with the regional primary $PM_{2.5}$ emission fluxes to quantify the contribution of regional transport of $PM_{2.5}$ to air quality change in the YRMB region with identifying the patterns of regional transport of $PM_{2.5}$ over$_5$ over central-easternCentral-eastern China. The primary $PM_{2.5}$ emission data of 2016 obtained from the Multi-resolution Emission Inventory for China (MEIC, http://www.meicmodel.org/) were selected for use as the regional $PM_{2.5}$ emission fluxes in this study.

3.2.3 Validation of modeling results

The simulated meteorological fieldssimulated meteorology, which included wind speed, air temperature, relative humidity and surface pressure, were compared with observations at five sites (Wuhan, Changsha, Hefei, Zhengzhou and Nanchang) over central-easternCentral-eastern China. The correlation coefficients and normalized standardized deviations were calculated and are shown in Figure 8 (Taylor, 2001). Based on the results with correlation coefficients passing the significance level of 0.001 and low normalized standardized deviations (Fig. 8), it was confirmed that WRF-modeled meteorology that is consistent with observations could be used to drive the FLEXPART backward trajectory simulation in this study.

3.34 Contribution of regional transport of $PM_{2.5}$ to heavy pollution

Based on the FLEXPART-WRF backward trajectory simulation, the upstream sources of $PM_{2.5}$ emissions for heavy air pollution in Wuhan could be identified. The contribution rates ***rate*$_{i,j}$** of regional transport of $PM_{2.5}$ from the upstream sources to air pollution in the downstream receptor region of YRMB were calculated by Eq. (1), and the total contribution ***R*** of regional transport from the non-local emission sources are estimated by Eq. (2) (Chen et al., 2017b).

$$rate_{i,j} = \frac{E_{i,j} \times r_{i,j}}{\sum_{1,1}^{N,S} E_{i,j} \times r_{i,j}} \qquad (1)$$

$$R = \sum_{(N_1,S_1)}^{(N_2,S_2)} rate_{i,j} \qquad (2)$$

where the subscripts *i* and **j** represent a grid location; $\mathbf{r}_{i,j}$ represents the residence time of $PM_{2.5}$ particles simulated by FLEXPART-WRF; and, $E_{i,j}$ represents the $PM_{2.5}$ emission flux over the grid. The first grid location (N_1, S_1) and the last grid location (N_2, S_2) over the non-local emission

~~sources and the local area of Wuhan were determined respectively by the regional transport of $PM_{2.5}$ pathways and the YRMB region as simulated by FLEXPART-WRF.~~

In this study for the receptor of Wuhan, the $PM_{2.5}$ contributions of regional transport to air pollution in the downwind receptor region could be approximately estimated based on the product of the residence time of air particles during regional transport simulated by FLEXPART-WRF, and the $PM_{2.5}$ emission flux over the source grid in Central and Eastern China, yielding a so-called potential source contribution map, which is the geographical distribution of the regional transport contribution rates (%) of the emission source grid cell to $PM_{2.5}$ pollution at the receptor of Wuhan (Fig. 9).

The non-local emission sources that affected $PM_{2.5}$ concentrations during three heavy pollution periods through regional transport to the YRMB region were quantified by using the $PM_{2.5}$ contribution rates calculated with Eq.(1)~~by calculation of the $PM_{2.5}$ contribution rates with Eq (1)~~. Combining the distribution of high $PM_{2.5}$ contribution rates with the prevailing winds experienced during the three heavy $PM_{2.5}$ pollution periods, the spatial distribution of the major pathways of regional transport of $PM_{2.5}$ over ~~central-eastern~~Central- and Eeastern China could be recognized as shown in Figure 9. During the heavy air pollution period P1 in the YRMB region, the regional transport of air pollutants was centered along a northwestern route from the Fenhe-Weihe Plain in central China and a northeastern route from the YRD region (Fig. 9a). The YRD emission sources of air pollutants in East China exerted an important impact on the heavy air pollution period P2 through regional transport of $PM_{2.5}$ cross East China to the YRMB region along the north side of Yangtze River (Fig. 9b). Two major regional transport pathways of $PM_{2.5}$ indicated by the spatial distribution of high contribution rates of $PM_{2.5}$ from the NCP and YRD regions respectively to the elevated $PM_{2.5}$ concentrations during the YRMB's heavy air pollution period P3 (Fig. 9c). Governed by the northerly winds of the East Asian winter monsoon, the regional transport of air pollutants from the ~~central-eastern~~Central-eastern air pollutant emission source regions in China provided a significant contribution to the wintertime heavy $PM_{2.5}$ pollution observed in the YRMB region (Figs. 6-7), which was confirmed by the results of the FLEXPART-WRF backward trajectory simulation utilized in this study.

In this study, the $PM_{2.5}$ contributions of regional transport to air pollution in the downwind receptor region could be approximately estimated based on the product of the residence time of $PM_{2.5}$ particles during regional transport simulated by FLEXPART-WRF, and the $PM_{2.5}$ emission flux over the source grid. The $PM_{2.5}$ contributions of regional transport over ~~central-eastern~~Central-eastern China to $PM_{2.5}$ concentrations during three heavy $PM_{2.5}$ pollution periods P1, P2 and P3 in the YRMB region were estimated using Eq. (2) with resulting high contribution rates of 68.1%, 60.9%and 65.3%, respectively (Table 4), revealing the significant contribution of regional transport of $PM_{2.5}$ over ~~central-eastern~~Central-eastern China to the enhancement of $PM_{2.5}$ levels in the YRMB area during wintertime heavy air pollution periods.

It should be pointed out that the potential source contribution is estimated based on transport alone, ignoring chemical and removal processes. We also understand that the physical and chemical processes such as complex deposition and chemical conversion for the formation of secondary particles are not introduced in the FLEXPART-WRF emulation, which could represent the basic features of contribution and patterns of regional $PM_{2.5}$ transport over central and eastern China, when limited to the primary $PM_{2.5}$ particles highlighted in this study.

Normally people rely on 3-D numerical models with process analysis capability such as integrated process rates (IPRs) to quantify the contributions of regional transport to the occurrence of air pollution episodes. The simulations with a Lagrange particle dispersion model FLEXPART-WRF are utilized to calculate the percentage contribution of regional transport with identifying the transport pathway in this study. The major uncertainty of this method for such calculation as compared to other methods like IPRs is ~~that the~~that the physical and chemical processes such as wet-deposition and chemical conversion for the formation of secondary particles are not introduced in the FLEXPART-WRF simulation, Considering less precipitation in the winter monsoon season, how this methodology (FLEXPART-WRF) is proven robustness to quantify the regional transport contribution with the uncertainty range here could mostly rely on a portion of secondary organic and inorganic aerosols, which are resulted from important and complex physiochemical processes in the atmosphere.

4. Conclusions

This study investigated the ambient $PM_{2.5}$ variations over Wuhan, a typical urban YRMB region in ~~central-eastern~~Central-~~eastern~~ China in January 2016 through analysis of observational data of environment and meteorology, as well as via FLEXPART-WRF simulation to explore 1) the meteorological processes involved in the regional transport of air pollutants and 2) regional transport patterns of $PM_{2.5}$ with the contribution to the air pollution in the YRMB region. Based on observation and simulation studies on the meteorological conditions of air pollution events in January 2016 ~~and regional~~and regional transport of $PM_{2.5}$ to heavy air pollution over the YRMB region, it is revealed heavy air pollution with the unique "non-stagnant" atmospheric boundary layer in the YRMB region aggravated by regional transport of $PM_{2.5}$ over central and eastern

China.

The study of the effects of meteorology and regional transport of $PM_{2.5}$ on— heavy air pollution werewas focused on three heavy $PM_{2.5}$ pollution periods in January 2016. The heavy pollution episodes observed with the peak of $PM_{2.5}$ concentrations exceeding 471 μg m^{-3} over the YRMB region were characterized by a short duration of less than 26 hrhr. from rapid outbreak to fast dissipation.

This study of environmental and meteorological observations in the YRMB region revealed a unique "non-stagnant" meteorological condition of the boundary layer characterized by strong wind, no inversion layer and a more unstable structure in the atmospheric boundary layer associated with heavy air pollution periods with excessive $PM_{2.5}$ concentrations in the YRMB region, which facilitates understanding of the air pollutant source-receptor relationship of regional air pollutant transport. The study represents a great interest to air quality community given the unique features of air pollution meteorology which are very different from those "stagnant" meteorological conditions presented in the textbooks.

The "stagnation" meteorological condition in the boundary layer characterized by weak wind, air temperature inversion and a stable vertical structure of the atmospheric boundary layer is currently accepted as a typical meteorological driver for heavy air pollution. Conversely, this study of environmental and meteorological observations in the YRMB region revealed a unique "non-stagnation" meteorological condition of the boundary layer characterized by strong wind, no inversion layer and a more unstable structure in the atmospheric boundary layer associated with heavy air pollution periods with excessive $PM_{2.5}$ concentrations in the YRMB region, which facilitates understanding of the air pollutant source-receptor relationship of regional air pollutant

~~transport.~~

Although the emissions and local accumulation of air pollutants in the YRMB could lead to the formation of light air pollution, in regards to $PM_{2.5}$, over the YRMB region, the regional transport of $PM_{2.5}$ from ~~Central-eastern~~ emission source regions in China contributed significantly to 65% of the exceedances of $PM_{2.5}$ concentrations during wintertime heavy air pollution periods in the downwind YRMB region in January 2016, as governed by the strong northerly winds of the East Asian winter monsoon.

Based on the variations of air quality and meteorology in a typical urban YRMB region in January 2016, this study revealed a unique “non-stagnant” meteorological condition for the development of heavy air pollution in the YRMB region with strong contributions of regional transport of $PM_{2.5}$ over China. These conditions and contributions can be investigated further with climate analyses of long-term observations and a more comprehensive modeling of air quality and meteorology.

Data availability: Data used in this paper can be provided by Chao Yu (ychao012@foxmail.com) upon request.

Author contributions: CY, TZ and YB conducted the study design. XY, LZ and SK provided the observational data. LZ assisted with data processing. CY wrote the manuscript with the help of TZ and XY. YB, SK, JH, CC, YY, GM, MW and JC were involved in the scientific interpretation and discussion. All of the authors provided commentary on the paper.

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Table 1. Correlation coefficients between hourly $PM_{2.5}$ concentrations and near-surface meteorological elements WS (wind speed), T (air temperature) , P (air pressure) and RH (relative humidity) in ~~over~~ Wuhan in January 2016.

Correlation coefficients	WS	T	P	RH
$PM_{2.5}$	0.10	0.31	-0.47	0.20

Table 2. Correlation coefficients of $PM_{2.5}$ concentrations with wind speed and air temperature in different air quality levels during the study period.

Air quality	$PM_{2.5}$ levels	Number of samples	WS	T

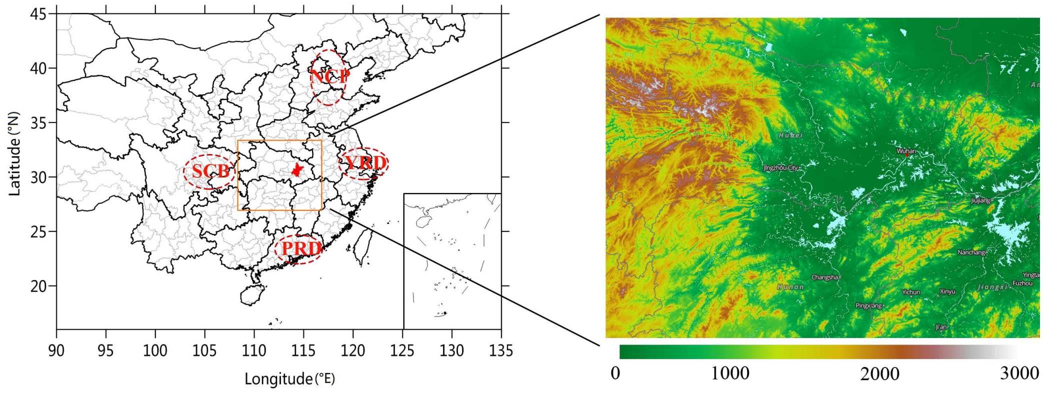
Clean	$PM_{2.5}<75\mu g\cdot m^{-3}$	73	-0.20	0.56
Light pollution	$75\mu g\cdot m^{-3}\leq PM_{2.5}<150\mu g\cdot m^{-3}$	135	-0.19	0.15
Heavy pollution	$PM_{2.5}\geq 150\mu g\cdot m^{-3}$	37	0.41	-0.08

Table 3. Atmospheric static stability below heights of 200 m in the boundary layer during heavy pollution and clean air periods with the anomalies relative to the average over January, 2016 in Wuhan.

Period	heavy pollution period ($K\cdot km^{-1}$)	clean air period ($K\cdot km^{-1}$)	monthly average ($K\cdot km^{-1}$)
Static stability	4.4	13.2	8.6
Anomalies of stability	-4.2	4.6	-

Table 4. The relative contributions of regional transport over ~~central-eastern~~Central-eastern China to three $PM_{2.5}$ heavy pollution periods P1, P2 and P3 in the YRMB with the local contributions.

Contribution rates	P1	P2	P3	Averages
Regional transport	68.1%	60.9%	65.3%	65.1%
Local contribution	31.9%	39.1%	34.7%	34.9%



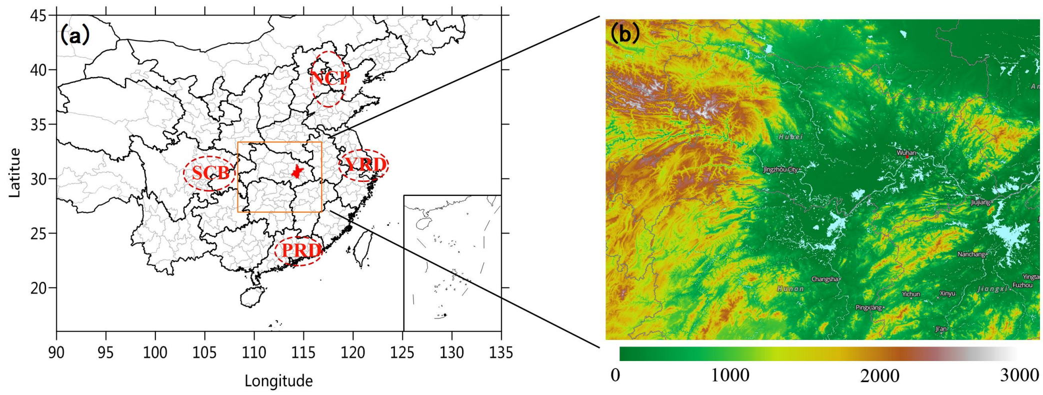


Fig. 1. (a) Distribution of the Yangtze River Middle Basin (orange rectangle) with the location of Wuhan (red area) and the major haze pollution regions of NCP, YRPD, PRD and SCB in central-easternCentral and -Eeastern China as well as (b) the YRMB region with terrain height (color contours, m in a.s.l.), the rivers and lake network (blue areas),downloaded from https://worldview.earthdata.nasa.gov.

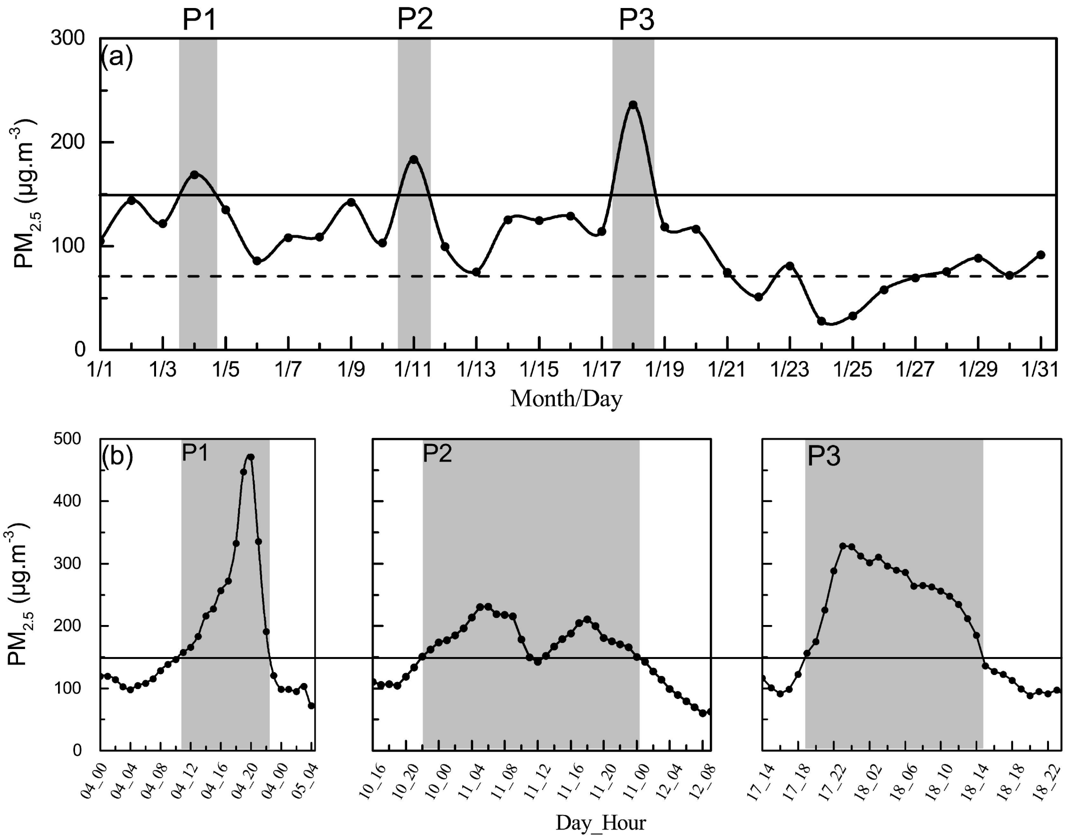


Fig. 2. (a) The daily changes of surface $PM_{2.5}$ concentrations in Wuhan in January 2016 with $PM_{2.5}$ concentrations exceeding 75 μg·m^{-3} (dash line) and 150 μg·m^{-3} (solid lines), respectively, for light and heavy haze pollution, and (b) the hourly variation of surface $PM_{2.5}$ concentrations in three heavy air pollution events P1, P2 and P3 with excessive $PM_{2.5}$ levels (>150 μg m^{-3}) marked by the shaded areas.

Fig. 3. Hourly variations of meteorological elements and $PM_{2.5}$ concentrations in Wuhan in January 2016 with heavy air pollution periods marked with the columns in red dash lines and $PM_{2.5}$ concentrations exceeding 150 $\mu g \cdot m^{-3}$ (solid line).

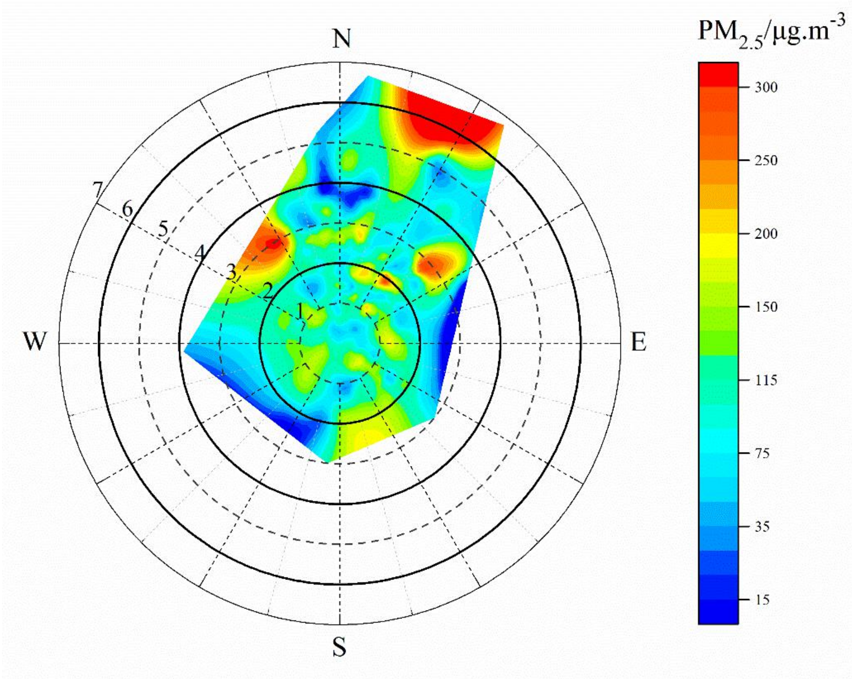


Fig. 4. A polar plot of hourly variations in wind speed (round radius, units is m·s^{-1}) and direction (angles) to surface $PM_{2.5}$ concentrations (color contours, units is μg·m^{-3}) in Wuhan in January, 2016.

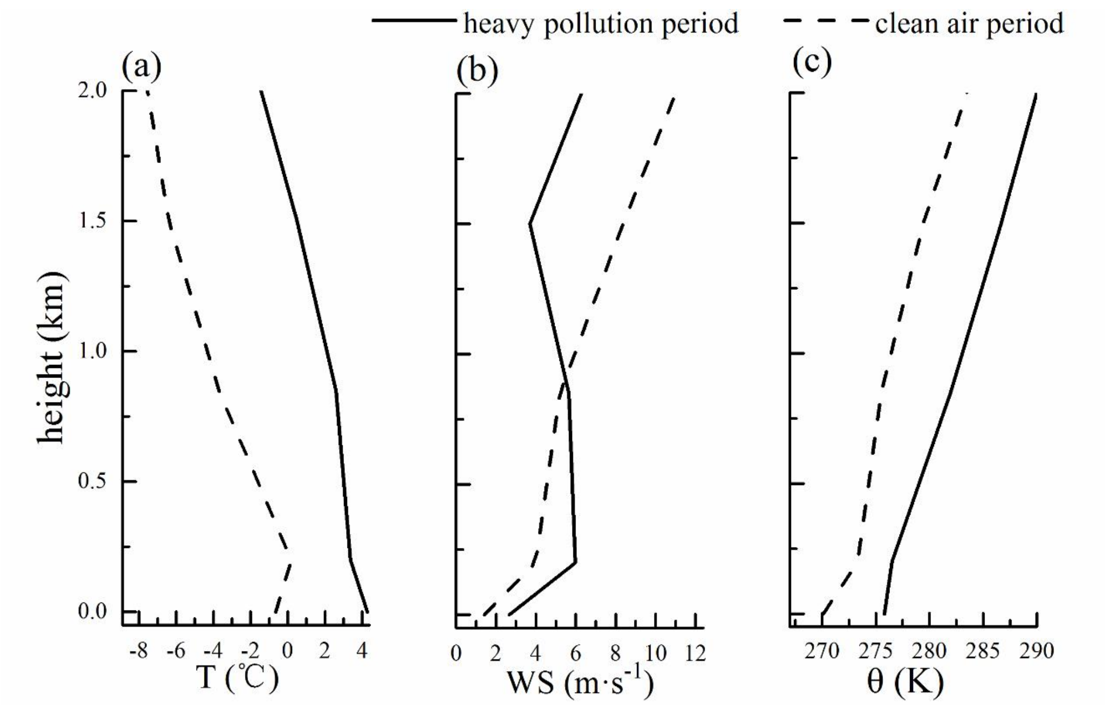


Fig. 5. Vertical profiles of (a) air temperature, (b) wind velocity and (c) potential temperature

averaged in heavy $PM_{2.5}$ pollution (solid line) and clean air (dash line) periods over Wuhan during January 2016.

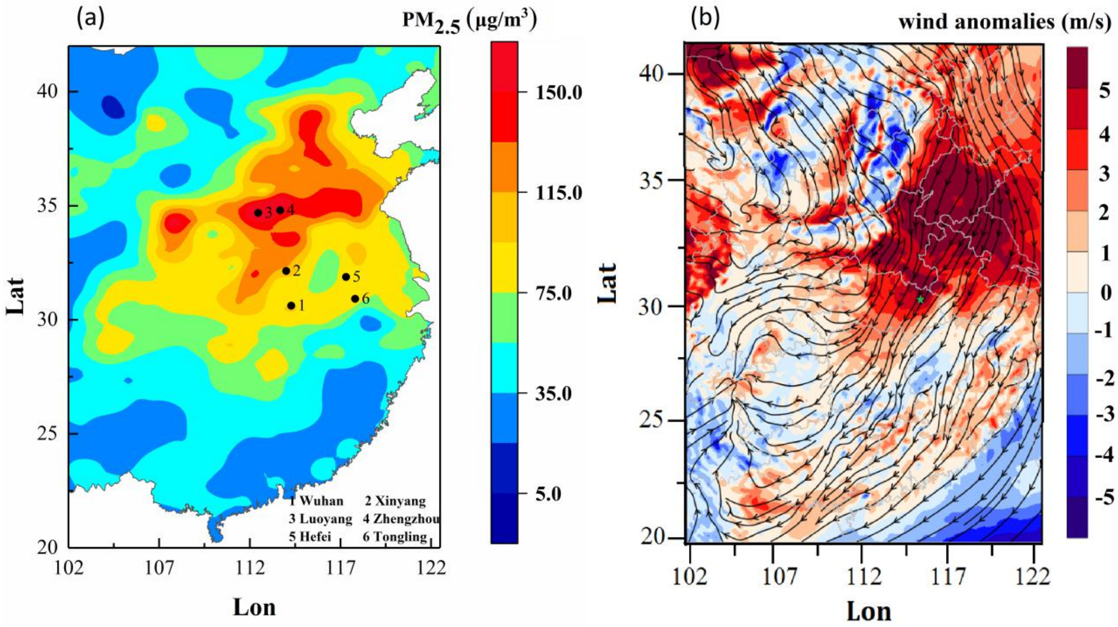
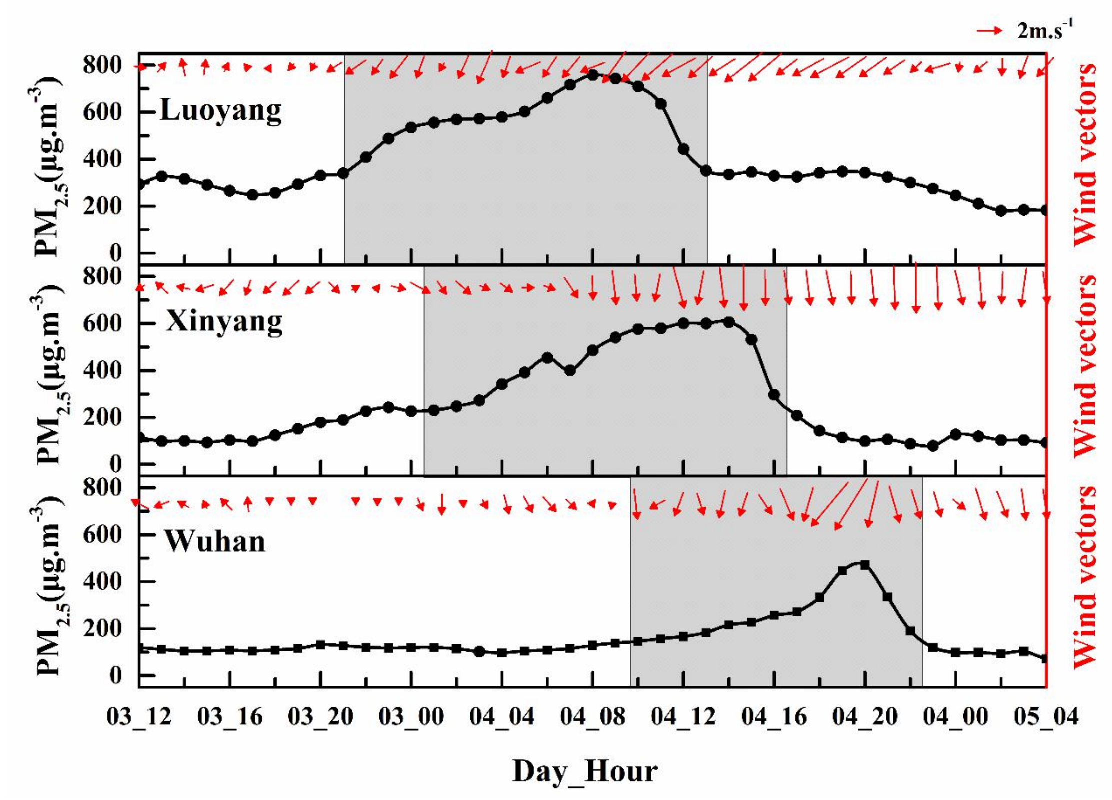
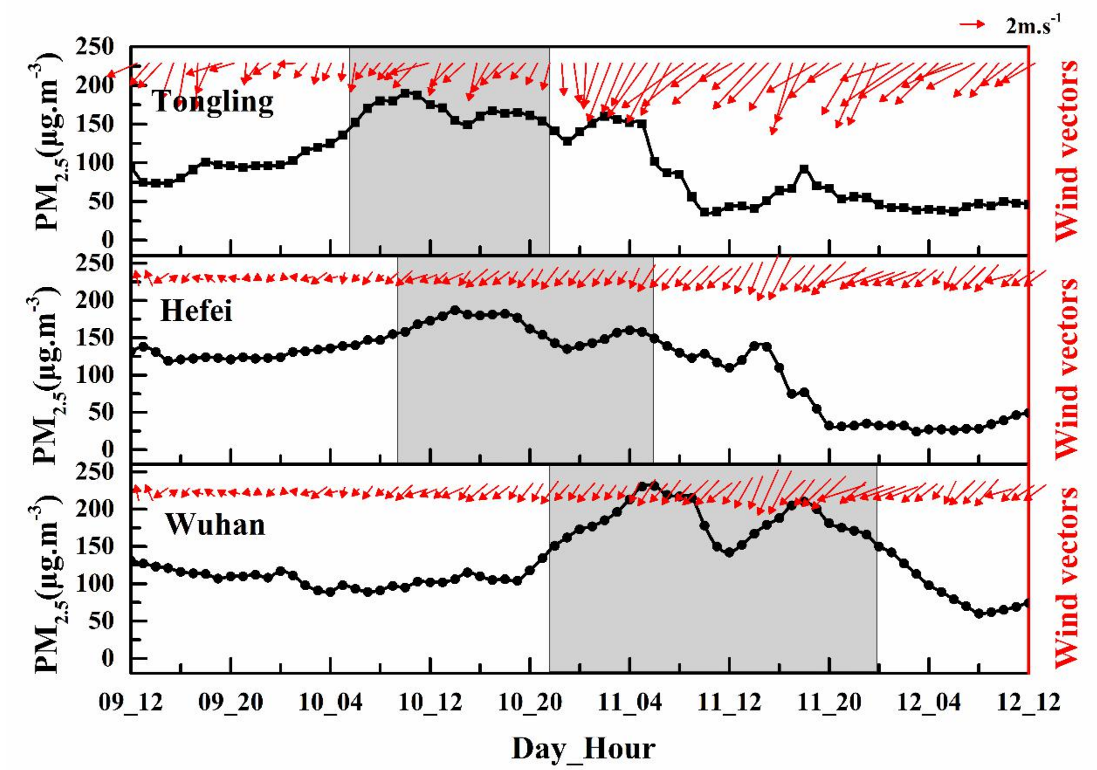


Fig. 6 Distribution of (a) monthly averages of surface $PM_{2.5}$ concentrations observed in January 2016 over ~~central-eastern~~Central-eastern regions in mainland China with the locations of six sites 1. Wuhan, 2. Xinyang, 3. Luoyang, 4. Zhengzhou, 5. Hefei and 6.Tongling as well as (b) the anomalies (color contours) of 200m wind speeds averaged during three heavy air pollution periods relatively to the monthly wind averages (streamlines) in January 2016 over ~~central-eastern~~Central-eastern China with the location of Wuhan (a light blue star).


2m.s-1
Luoyang
Xinyang
Wuhan
PM2.5(μg.m-3)
Wind vectors
800
600
400
200
0
03_12
03_16
03_20
03_00
04_04
04_08
04_12
04_16
04_20
04_00
05_04
Day_Hour


2m.s-1
Tongling
Hefei
Wuhan
PM2.5(μg.m-3)
Wind vectors
250
200
150
100
50
0
09_12
09_20
10_04
10_12
10_20
11_04
11_12
11_20
12_04
12_12
Day_Hour

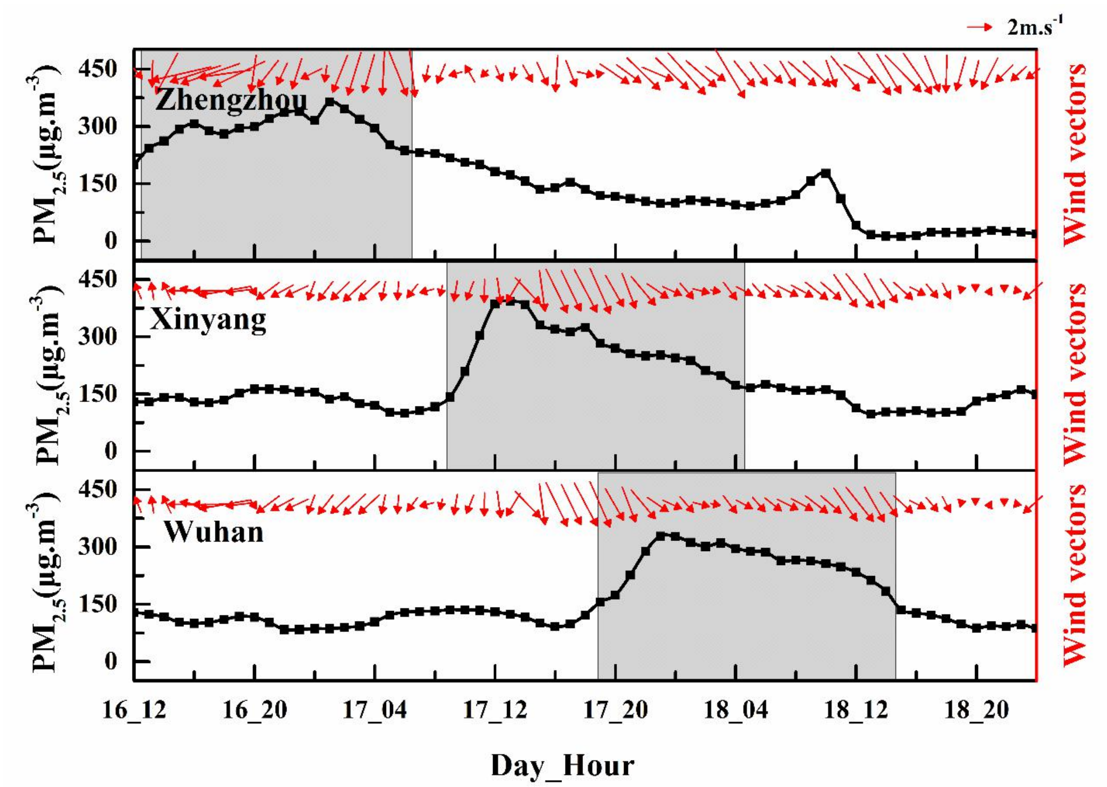


Fig. 7. Temporal changes of $PM_{2.5}$ concentrations (dot lines) and near-surface winds (vectors) observed at five upstream sites (Fig. 6) and Wuhan with shifts of $PM_{2.5}$ peaks (marked with shaded areas) to the YRMB's heavy $PM_{2.5}$ pollution periods <u>P1 (upper panel), P2 (middle panel) and P3 (lower panel), respectively, in January 2016</u>~~P1 P2 and P3 (respectively in upper, middle and lower panels) in January 2016~~.

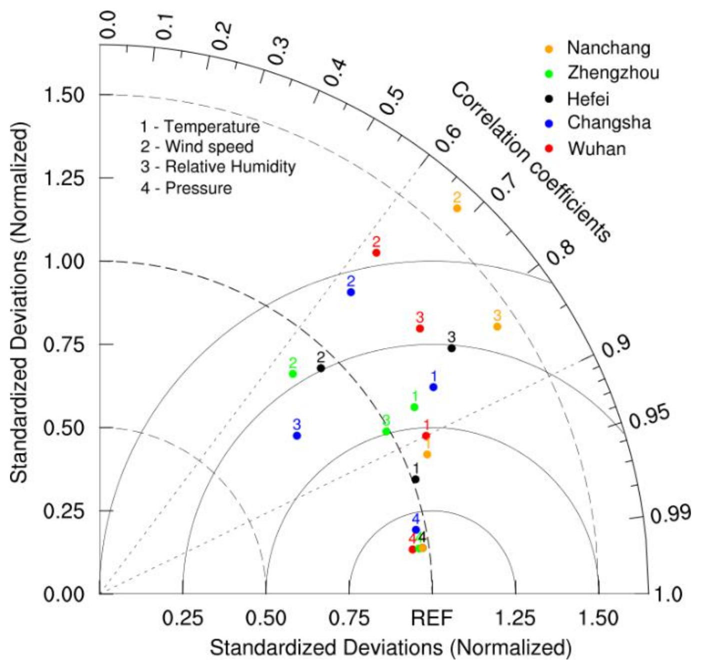


Fig. 8. Taylor plots with the normalized standard deviations and correlation coefficients between simulated and observed meteorological fields. The radian of the sector represents the correlation coefficient, the solid line indicates the ratio of standard deviation between simulations and observations, the distance from the marker to "REF" reflect the normalized root-mean-square error (NRMSE).

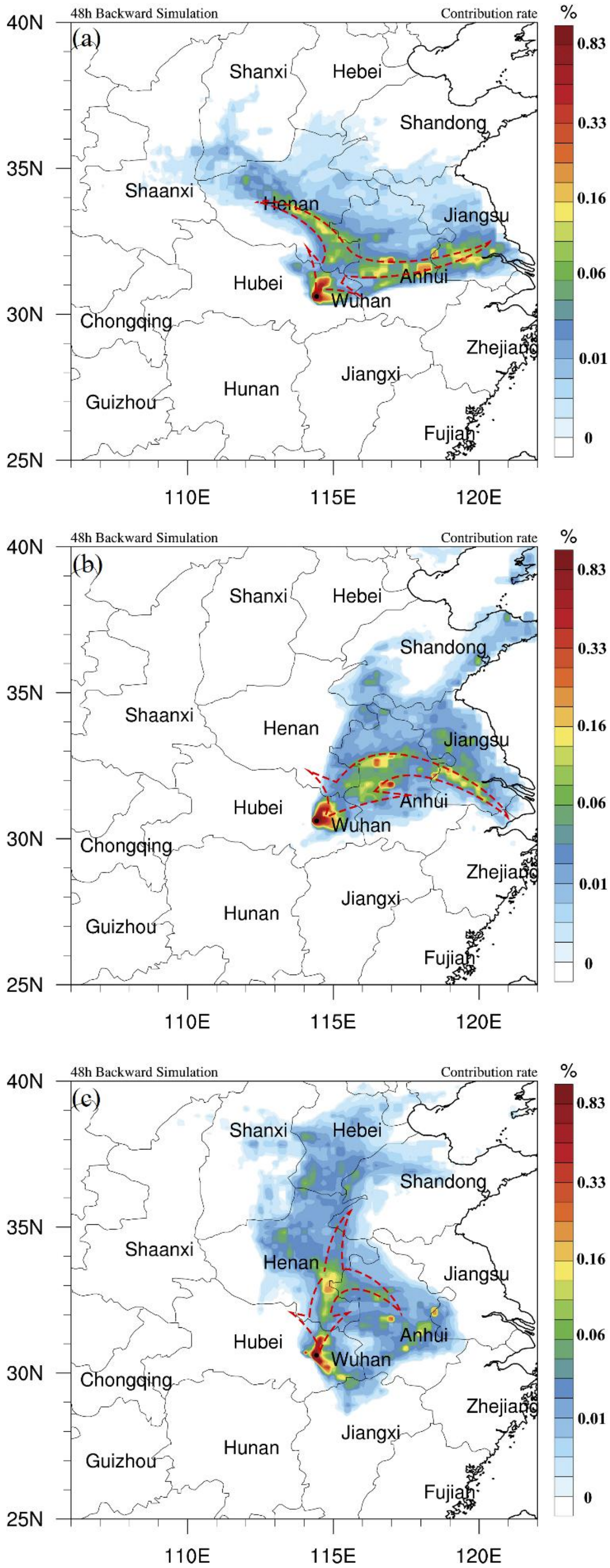


Fig. 9. Spatial distribution of contribution rates (color contours) to $PM_{2.5}$ concentrations in Wuhan

with the major pathways of regional transport over ~~central-eastern~~Central-eastern China (dash arrows) for (a) heavy pollution periods P1, (b) P2 and (c) P3 in January, 2016 simulated by the model FLEXPART-WRF.