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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Aerosol Vertical Mass Flux Measurements During Heavy Aerosol Pollution Episodes at a Rural Site and an Urban Site in the Beijing Area of the North China Plain" *by* Renmin Yuan et al.

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Authors reply to reviewer's comments:

Dear Anonymous Referee,

Thanks for your careful review of the manuscript. We read the reviewer's comments carefully, and have responded and taken all of the reviewer's comments into consideration and revised the manuscript accordingly. My detailed responses are as follows: Comments from Anonymous Referee #2: "Quantification of the aerosol mass flux is an

important topic to understand pollutant emissions and transport over areas exposed to pollution episodes. The study utilizes an innovative large aperture scintillometer (LAS) technique to estimate the transport of aerosols over extended areas. The presented results are a valuable contribution to the understanding the emissions in urban areas and rural polluted regions."

1) "However, since the LAS technique is semi-empirical, then additional information on testing and evaluation of such measurements would help to improve confidence in results and understand the underlying uncertainties. For example, the LAS technique is capable to determine the magnitude of the flux but not the sign. In general the aerosols are very heterogeneous in space and the measured fluxes show typically large variation in magnitude including the sign. Over the polluted areas, which behave as the source, the emissions presumable overwhelmingly exceed the deposition sinks. Therefore, for example, a rough quantification of the deposition sink would allow to conclude that the sink term is indeed negligible and the flux quantified by LAS can be safely assumed to represent the upward fluxes. If available, the reference to comparison of the LAS method results with a more direct micrometeorological measurement would be very useful (if this was done in Yuan et al., 2016, please mention explicitly)."

Response: Thanks for your suggestion. We have added the statement that the sink term is indeed negligible and the flux quantified by LAS can be assumed to represent the upward fluxes. Please see L288-290. At present, we have not conducted more direct meteorological measurements to obtain aerosol fluxes, such as the use of EC methods for aerosol flux measurements. Next, we will compare the aerosol flux obtained by LAS with the aerosol flux received by the EC method.

2) "The manuscript would benefit also from better improved description/definition of the heavy pollution episodes (HPEs), how they are divided into stages of transport (transport stage TS), a cumulative stage (CS) and a removal stage (RS), and in particular what are the prevailing meteorological and aerosol emission/transport conditions during such episodes. This would help readers who are not familiar with HPE mechanisms

more easily to follow the manuscript."

Response: Based on meteorological causes of the increase or decrease in PM2.5 mass concentrations, the HPEs are divided into TSs, ASs (in the new version, CS denoted as AS by suggestion), and RSs. During the TSs, the PM2.5 is dominated by relatively strong southerly winds, which carry polluted air masses from more populated southern industrial regions (Guo et al., 2014; Zhong et al., 2018a). Before rising processes during TSs, the urban PM2.5 mass concentration of Baoding, which is typically representative of pollution conditions in the south of Beijing, was much higher than Beijing; the winds in Beijing rapidly shifted from northerly to southerly. Then the rising in PM2.5 occurred, consistently with southerly slight or gentle breezes in the BL. The southerly air mass moves more than 288 km d-1 below 500 m (estimated from the measured wind speed), which are fast enough to transport pollutants to Beijing. Such processes indicate southerly pollutant transport is primarily responsible for the rising, given the pollution transport pathway of the southwest wind belt determined by the unique geographic features of the North China Plain, with the Tai-hang Mountains and the Yan Mountains strengthening the southwest wind belt and leading the convergence of pollutant transport in Beijing (Su et al., 2004). During the ASs, PM2.5 increase is dominated by stable atmospheric stratification characteristic of southerly slight or calm winds, near-ground anomalous inversion, and moisture accumulation. When the vertical aerosols are accumulated to a certain degree, the dominant scattering aerosols will substantially back-scatter solar radiation, causing a reduction in the amount of solar radiation that reaches the surface, which creates a near-ground cooling effect through atmospheric circulation and vertical mixing(Zhong et al., 2018b). The temperature reduction induces or reinforces an inversion that further weakens turbulence diffusion and results in a lower BL height, which further worsens aerosol pollution. This condition also decreases the near-ground saturation vapor pressure and suppresses water vapor diffusion to increase the relative humidity (RH), which will further enhances aerosol hygroscopic growth and accelerates liquid-phase and heterogeneous reactions to worsen aerosol pollution (Ervens et al., 2011; Kuang et al., 2016; Pilinis et al., 1989;

Zhong et al., 2018a; Zhong et al., 2018b). This feedback effect of further worsened meteorological conditions aggravates PM2.5 pollution (Zhong et al., 2017). During the RSs, strong northwesterly winds whose velocity increases with height occur mostly. Strong northerly winds are from less populated north mountainous areas and carry unpolluted air masses to Beijing, which is favorable for pollution dispersion. We have added some descriptions about three stages in the introduction. Please L135-156 in Section introduction. Ervens, B., Turpin, B.J., Weber, R.J., Secondary organic aerosol formation in cloud droplets and aqueous particles (aqSOA): a review of laboratory, field and model studies, Atmos. Chem. Phys. 11(2011), 11069-11102. Guo, S. et al., Elucidating severe urban haze formation in China, Proc. Natl. Acad. Sci. U.S.A. 111(2014), 17373-17378. Kuang, Y., Zhao, C.S., Tao, J.C., Bian, Y.X., Ma, N., Impact of aerosol hygroscopic growth on the direct aerosol radiative effect in summer on North China Plain, Atmospheric Environment 147(2016), 224-233. Pilinis, C., Seinfeld, J.H., Grosjean, D., Water content of atmospheric aerosols, Atmos. Environ. 23(1989), 1601-1606. Su, F., Gao, Q., Zhang, Z., REN, Z.-h., YANG, X.-x., Transport pathways of pollutants from outside in atmosphere boundary layer, Res. Environ. Sci. 1(2004), 26-29. Zhong, J. et al., Feedback effects of boundary-layer meteorological factors on cumulative explosive growth of PM2.5 during winter heavy pollution episodes in Beijing from 2013 to 2016, Atmos. Chem. Phys. 18(2018a), 247-258. Zhong, J., Zhang, X., Wang, Y., Liu, C., Dong, Y., Heavy aerosol pollution episodes in winter Beijing enhanced by radiative cooling effects of aerosols, Atmos. Res. 209(2018b), 59-64. Zhong, J. et al., Relative contributions of boundary-layer meteorological factors to the explosive growth of PM 2.5 during the red-alert heavy pollution episodes in Beijing in December 2016, J. Meteorolog. Res. 31(2017), 809-819.

3) "According the author the TS is the period when the pollution over the measurement location was mainly contributed by the downwind pollution sources. But presumable also the local sources were also a significant contribution because the aerosol fluxes did not differ much in magnitude from subsequent phases. The CS (perhaps would be better to call accumulation stage?) represents the period of rapid accumulation of

pollutants and it is not evident of this occurs because of downwind transport of pollutants trapped in the atmospheric boundary layer or local emissions or both. Therefore, it is not clear if the stage differs from the TS in terms of location of emission sources or difference is made by the meteorological conditions favouring accumulation of the pollutants in the ABL. Regarding the RS, presumably the pollutant concentrations drop due to the atmospheric mixing and transport to higher levels. The other possibility is removal by scavenging or dry deposition. Dry deposition however is a slow process and also the results do not support such assumption (up-ward fluxes in Figs. 5 and 6 during the RS). "

Response: After a series of measures and actions, including air pollutant emission reduction, energy structure adjustments to decrease the dependence on fossil fuels, and other supportive policies, the emission sources in Beijing are strikingly less than the polluted southern industrial regions with large anthropogenic emissions. Therefore, the contribution of local emissions in Beijing is relatively smaller than that in the other areas such as Baoding. The TS will appear before CS. The south or southwest wind will always appear in the TS, and the concentration of PM10 in Baoding is higher than the concentration of PM10 in Beijing, which is generally maintained for one to two days. Except for the southerly or southwesterly winds for one to two days, there will be no CS in Beijing. Even if it is a southerly or southwesterly wind, if the wind speed is too small (<1ms-1), AS will not appear. Only the southerly or southwesterly wind with a wind speed higher than a specific value (>1.5ms-1), and the concentration of PM2.5 in the area to the south of Beijing is higher than that in Beijing, and then there will be CS after a small wind. Therefore, the main reason for the explosive growth of aerosol concentration during CS is that explosive growth is attributed to the horizontal transport during TS. Please L591-603.

4) "The explanation in l. 425 is confusing as if the particles are removed from the atmosphere and reduction in pollutants does not occur because of the atmospheric mixing (and upward transport of aerosols). "

Response: We modified the expression. During the RSs, strong northwesterly winds whose velocity increases with height occur mostly. Strong northerly winds are from less populated north mountainous areas and carry unpolluted air masses to Beijing, which is favorable for pollution dispersion. Please L154-157.

5) "In relation to interaction between the aerosol pollution and meteorology, the authors suggest in the abstract (and l. 498-500) that the aerosol pollution had an effect to turbulence intensity leading to further weakening of mixing and increased accumulation. Such effect is not directly evidenced by the results in the manuscript (or cannot be distinguished) and should be further supported by the literature references rather than stated as the result."

Response: Based on the results in the manuscript, the effect of the aerosol pollution cannot be drawn out to turbulence intensity leading to further weakening of mixing and increased accumulation. Based on our measurement, it can be seen that from the TS to the AS, the aerosol vertical turbulent flux decreased, but the aerosol particle concentration within surface layer increased, and it is inferred that in addition to the contribution of regional transport from upwind pollution areas during the TS, suppression of vertical turbulence mixing confining aerosols to a shallower boundary layer increased accumulation. We modified some expression. Please see L580-584.

"The manuscript would benefit also from numerous minor improvements and language editing. Please see my specific comments below."

Specific comments 1) "Line 28-29, sentence difficult to follow, please revise."

Response: The sentence means a weakened turbulence intensity and low vertical aerosol fluxes in winter and polluted areas such as GC We revised. Please see L28-29.

2) "L. 35-36, the statement is vague, see also general comments."

Response: The sentence is just a reference, not a conclusion. So the sentence is deleted.

3) “L. 60-61 “the consumption of a product” – revise phrasing”

Response: expressed as “product energy consumption”. Please Line 62-63.

4) “L. 77-79: the EC method has been used already for decades to quantify the aerosol particle number fluxes. As an example of earlier studies, see e.g. Buzorius, G., Rannik, Ü., Mäkelä, J.M., Vesala, T., Kulmala, M., 1998. Vertical Aerosol particle fluxes measured by eddy covariance technique using condensational particle counter. J. Aerosol Sci., 29, 157-171.”

Response: The manuscript was revised according to the comment. Please see line 86-87.

5) “L. 80, The EC method enables to determine the vertical turbulent flux, which can be different from total vertical transport. Also, the flux is provided by the cross-covariance (and not correlation).”

Response: “eddy correlation” modified to “eddy covariance”

6) “L. 82-83, the EC principle allows to quantify the number flux from fluctuation measurements, rephrase the sentence.”

Response: Based on EC principle, the vertical velocity fluctuations and the fluctuations in the aerosol particle number density can be measured, and the EC principle allows to quantify the number flux from fluctuation measurements. We modified the sentences. Please see Line 88-89.

7) “L. 105, The eddy correlation principles have been widely used (or something likethis, revise the sentence).”

Response: “eddy correlation” modified to “eddy covariance.”

8) “L. 126 how much the surface emissions contribute to the concentration of pollutants”

Response: At present, the question cannot be answered. We hope that with the measurement of the near-surface aerosol vertical flux, the results will help understand the accumulation of local aerosol concentrations, how much the surface emissions contribute to the level of pollutants, and how much the concentration of pollutants is attributed to upwind areas. This article does not discuss this issue for the time being. We will combine the data from the lidar and present it in a later article.

9) “L. 142-143, phrasing is not good. Rather the transport properties or the statistical aerosol transport is similar to that of scalars? In fine detail the aerosol motion can be different from the air motion and the statement is not strictly correct.”

Response: The spectral characteristics of aerosol number concentration fluctuations approximate the spectral characteristics of molecular density fluctuations. Please see Line 175-176.

10) “L. 166-167, temperature is not a passive atmospheric constituent because buoyancy affects strongly the motion of air. Also “distribution” does not seem relevant but maybe just “small particles”. Rather say that similarity of atmospheric aerosols and temperature can be assumed for the purpose.”

Response: You are very kind and helpful for the manuscript. Please see Line 206-207.

11) “L. 173 “aerosol particles are continuously dispersed in the air”, the meaning and purpose of this sentence is not clear.”

Response: Although the aerosol particles are dispersed in the air, the macroscopic behavior of the gas-particle two-phase mixture is the same as if it is perfectly continuous in structure and physical quantities, such as the mass and refractive index associated with the matter contained within a given small volume, which will be regarded as being spread continuously over that volume. We modified the manuscript. Please see Line 213-216.

12) “L. 192, Correct R_{MN}”

Response: We Modified it.

13) “L. 209-212, please provide reference and/or explanation for the relation between the high/low frequency fluctuations and the real/imaginary parts of the AERI.” Response: and can be measured by a specially made LAS (Yuan et al., 2015). After a spherical wave propagates over a distance in a turbulent atmosphere, the light intensity on the receiving end will fluctuate. When the attenuation caused by scattering and absorption along the propagation path is very weak, light intensity fluctuation depends on the variation of the real part of the AERI along the propagation path. When the attenuation caused by scattering and absorption along the propagation path is relatively strong, the light intensity fluctuation is also related to the fluctuation of the imaginary part of the AERI along the propagation path. With the spectral analysis method, the LAS light intensity fluctuations can be separated into the contributions of the real and imaginary parts of the AERI. The input of the real part of the AERI corresponds to the high frequencies, whereas the participation of the imaginary part of the AERI corresponds to the low frequencies, suggesting that the variances resulting from the real and imaginary parts are independent. Therefore, the light intensity variances induced by the real and imaginary parts can be detected separately at high frequencies and low frequencies from the LAS measurements (Yuan et al. 2015). We added the explanation. Please Line 257-271.

14) “L. 225, turbulent fluctuations of what?”

Response: Temperature

15) “L. 297, e.g. stands for “for example”, not relevant here.”

Response: Namely

16) “L. 309-310. The method for judging.. sentence difficult to follow, rephrase.”

Response: The 6-point moving average is done for the trend.

17) “L. 315, how was “mean of the adjacent difference” defined, based on the moving

average or how? Improve wording of the sentence.”

Response: The difference between adjacent moments is denoted and abbreviated as AMD. We have modified the sentences. We modified the paragraph. Please Line 373-385.

18) “L. 321-321, is the exact shape of the spectrum relevant? Or the method relies purely on the Kolmogorov’s power laws of the spectra?”

Response: The theoretical expression for the relation between light scintillation and the structure parameter are based on von Karman spectrum. It is assumed that the actual turbulence is accord with von Karman spectrum, and then the parameters are calculated. The deviation from the assumption will cause the error. We modified the sentence. Please see Line 619-620.

19) “L. 328, “heavy pollution weather conditions “is a bit weird, please rephrase”

Response: under heavy pollution weather conditions modified as during heavy pollution episodes (HPEs). Please see Line 397.

20) “L. 361, rather the wind direction varied throughout day? there was no dominant wind direction”

Response: The wind direction has diurnal variation characteristics, which are related to the sea-land breeze, valley wind and urban heat island circulation which may exist under the control of weak weather system. Please see Line 433-435.

21) “L. 368-374. The “free convection” conditions are not always easily satisfied. Free convection means that the buoyancy-driven turbulence dominates over mechanical turbulence and this is not just the unstable conditions but the free convective limit of the unstable conditions. Please clarify and evaluate the uncertainties introduced by such assumption.”

Response: At the CAMS site, local turbulence and local stability parameter measurements cannot be implemented. So we can only choose an alternative, and used the meteorological data (temperature) measured at nearby observation points, then based on the free convection assumption (using Equ. 12) the aerosol fluxes at the CAMS site were calculated. We (Yuan et al, 2015) conducted a test experiment for vertical aerosol flux in Hefei City, China, using free convection assumptions and local similarity theories to calculate aerosol fluxes. Comparison of the calculation results of the two methods shows that precarious condition, -0.15 < (z-zd) / L < 0, accounts for about 62 % of the time, and the relative difference is about 5%. Under weak unstable and stable condition, the relative error is about 15%. Although the relative error is a little significant under weak unstable stable stratification conditions, the absolute difference in flux is still small. We added some explanation. Please see L445-451.

22) "L. 421, southerly wind conditions"

Response: southerly wind

23) "Figures 3-6, the square value of the structure parameter is plotted according to label in y-axis of the relevant subplots."

Response: Regarding the naming of Cn and Cn2, I checked it. In the book of Monin and Yaglom (1975) and the book of Tatarskii (1961), there is no name for Cn and Cn2. In later books, different books have different names. For example, in Ishimaru (1978), Cn is called a structure parameter, and Cn2 is also called a structure parameter. In Andrews and Phillip (2005), Cn2 is called a structural parameter. Considering that we have always called Cn2 as a structural parameter (Yuan et al.,2015; Yuan et al. 2016), and in the application of Cn2 it generally appears in the form of Cn2, I will refer to Cn2 as a structural parameter in this manuscript. Andrews, L. C., and Phillips, R. L.: Laser beam propagation through random media, SPIE, 2005. Monin, A. S., and Yaglom, A. M.: Statistical fluid mechanics: Mechanics of turbulence, MIT Press, Cambridge, Massachusetts, 874 pp., 1975. Ishimaru, A.: Wave propagation and scattering in random media, Oxford University Press, Walton Street, Oxford OX2 6DP, 590 pp., 1978.

Tatarskii, V. I.: Wave Propagation in a Turbulent Medium, McGraw-Hill Book Company Inc., New York, 285 pp., 1961. Yuan, R., Luo, T., Sun, J., Liu, H., Fu, Y., and Wang, Z.: A new method for estimating aerosol mass flux in the urban surface layer using LAS technology, Atmospheric Measurement Techniques, 9, 1925-1937, 10.5194/amt-9-1925-2016, 2016. Yuan, R., Luo, T., Sun, J., Zeng, Z., Ge, C., and Fu, Y.: A new method for measuring the imaginary part of the atmospheric refractive index structure parameter in the urban surface layer, Atmospheric Chemistry and Physics, 15, 2521-2531, 10.5194/acp-15-2521-2015, 2015.

24) "Discussion and conclusions: how do the measured aerosol mass-fluxes compare with relevant literature values and/or earlier measurements and typical emission estimates? Please discuss this how to results contribute to understanding of pollution emissions."

Response: Compared to the results (Yuan et al. 2016) from Hefei, China, a small and medium-sized provincial capital city in East China, the measured aerosol mass-fluxes in Beijing are almost at the same amount. A series of measures and actions have been made for emission reduction in Beijing, and the primary emission is from vehicles. The difference in aerosol mass flux may be small. We added some discussion. Please Line 604-607.

Finally, the authors thank you for your constructive comments that help us to improve the clarity and the quality of the manuscript greatly. All the comments are answered and the modifications introduced in the revised manuscript correspondingly. We sincerely hope our answers can relieve doubts and give a better description of our work.

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