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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.

Anonymous Referee #2

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General comments

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.

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).

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. 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). 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). 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.

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.

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

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

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.

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).

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

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

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

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.

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.

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

12. L. 192, Correct R_{MN}

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.

14. L. 225, turbulent fluctuations of what?

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

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

17. L. 315, how was “mean of the adjacent difference” defined, based on the moving average or how? Improve wording of the sentence.

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?

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

20. L. 361, rather the wind direction varied throughout day?

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.

22. L. 421, southerly wind conditions

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

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.

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