

Dear Dr. Hendrick,

With regards to your revised manuscript, the referee did not give a response until now although he had expressed a willing to review your manuscript during the last-round review process. It seems to me that most of the issues raised by this referee are addressed in author response as well as in the revised manuscript. Thanks to the two referees, your manuscript has improved a lot after discussions and revisions. Regarding to the issue on the aerosol sources indicated by a correlation between SO₂ and extinction coefficient, I have additional comments/suggestions for you to consider.

Please do not hesitate to contact me if you have any questions.

Best regards,

Jianzhong Ma

Comments:

It is well known that SO₂ is an important gas precursor of sulfate aerosols. While a plume with high SO₂ (e.g. originally from a power plant) transports to the observation site, the amount of SO₂ should become less and less while being oxidized to form sulfate through either gas- or liquid- phase chemistry. At the same time, the amount of sulfate aerosols should increase in relative to SO₂ assuming that no strong wet remove processes occur. Note that aerosols are also primarily emitted along with SO₂ from the same sources. A strong correlation between SO₂ and aerosol extinction coefficient is most likely to be found in relatively fresh plumes, e.g. from the power plants, and the later (aerosol extinction coefficient) can be attributed to both primary (e.g. BC and OC) and secondary (sulfate) aerosols. In photo-chemically aged air masses, however, the correlation might not be so significant since SO₂ could be very deficient due to its conversion to sulfate while the concentration of aerosols might increase relatively due to the formation of sulfate. Therefore, a weaker correlation between SO₂ and aerosol extinction coefficient in summer than in winter may not necessarily indicate that sulfate is less important in summer considering that SO₂ should be oxidized more

efficiently in summer. Previous studies have indicated that for urban atmosphere over Beijing, the concentration of sulfate aerosols in PM_{2.5} is typically higher in summer than in winter (e.g. see Table 3 of Ma et al. (2012) and Table 1 of Zhang et al. (2013)). This argument does not mean that further measurements of aerosol composition in Xianghe are not necessary, but the photochemical age and associated evolution processes should be taken into account when doing a correlation analysis for different air masses.

Suggestions:

P2, L6-10: The sentence “while other sources dominate in summer” should be removed.

Sect. 3.4 (p15-16): In addition to the perturbation from other aerosols, lower emissions and shorter lifetime of SO₂ in summer, as well as different meteorological conditions, are likely to result in a weaker correlation between SO₂ and aerosol extinction coefficient. Such effect of air mass ages as well as air mass types on the correlation analytical results needs to be mentioned.

P17, L13-17: It should be pointed out that such kind of correlation analysis is not sufficient to conclude whether the conversion of SO₂ to sulfate is a dominant aerosol source or not.

References

Ma, J. Z., Xu, X. B., Zhao, C. S., and Yan, P.: A review of atmospheric chemistry research in China: Photochemical smog, haze pollution, and gas - aerosol interactions, *Adv. Atmos. Sci.*, 29, 1006-1026, 10.1007/s00376-012-1188-7, 2012.

Zhang, R., Jing, J., Tao, J., Hsu, S. C., Wang, G., Cao, J., Lee, C. S. L., Zhu, L., Chen, Z., Zhao, Y., and Shen, Z.: Chemical characterization and source apportionment of PM_{2.5} in Beijing: seasonal perspective, *Atmos. Chem. Phys.*, 13, 7053-7074, 10.5194/acp-13-7053-2013, 2013.