15/09/2014

Dear Prof. Ma,

With regard to the manuscript:

Journal: ACP
Title: Evaluation of tropospheric SO2 retrieved from MAX-DOAS measurements in Xianghe, China.
Author(s): T. Wang et al.
MS No.: acp-2014-10
MS Type: Research Article
Iteration: Minor Revision

We would like to thank you for your valuable comments and suggestions on the relationship between SO_2 and aerosols in the Beijing area. Please find below our responses to them. The corresponding changes in the revised manuscript are highlighted in yellow.

Sincerely yours,

F. Hendrick (**franch@oma.be**)

Green: Editor's comment

Black: author's reply

Red: modified text in the revised manuscript

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 PM2.5 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.

Thanks for these interesting considerations which show that we have to be careful when discussing the relationship between SO_2 and aerosols from our MAX-DOAS measurements in Xianghe.

Suggestions:

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

Done.

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.

This comment has been implemented as follows in the revised manuscript:

Page 16, lines 2-5:

The sentence 'In combination to the lower SO_2 concentration, this could explain the significantly weaker correlation between anthropogenic SO_2 and aerosols obtained in J, J, A.'

has been replaced by:

'These perturbations by other aerosol sources combined to lower SO_2 emissions, shorter lifetime of SO_2 due to a more efficient oxidation, and different meteorological conditions, could likely explain the significantly weaker correlation between anthropogenic SO_2 and aerosols obtained in J, J, A.'

Page 16, line 7:

'dominate' has been replaced by 'play a significant role'.

Page 16, lines 9-12:

The sentence 'It is however important to note that co-located measurements of the chemical composition of aerosols in Xianghe would be needed to confirm our findings.'

has been replaced by

'It is however important to note that co-located measurements of the chemical composition of aerosols in Xianghe as well as additional investigations on the type and photochemical age of the air masses probed by the MAX-DOAS instrument would be needed to confirm our findings.

We have also added the Zhang et al. (2013) reference at the beginning of Sect. 3.4.

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.

Page 17, line 17: 'are the dominant aerosol sources has been replaced by 'are also important aerosol sources.'

We have also added the following sentence:

'It is however worth noting that such kind of correlation analysis should be combined to aerosol composition measurements in order to definitely conclude whether the conversion of SO_2 to sulfate is a dominant aerosol source or not.'

Given the valuable comments provided by the two anonymous referees and the Editor for improving the manuscript, we decided to acknowledge them in the 'Acknowledgements'.

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 PM2.5 in Beijing: seasonal perspective, Atmos. Chem. Phys., 13, 7053-7074, 10.5194/acp-13-7053-2013, 2013.