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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evaluation of tropospheric SO_2 retrieved from MAX-DOAS measurements in Xianghe, China" *by* T. Wang et al.

Anonymous Referee #1

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General Comments

The paper entitled 'Evaluation of tropospheric SO_2 retrieved from MAX-DOAS measurements in Xianghe, China' by Wang et al. presents three years of continuous SO_2 observations at a location south-east of Beijing. The emission of SO_2 is certainly one of the major environmental concerns in China, with severe consequences on public health. Therefore the general topic of the manuscript is well suited for ACP and of interest for the scientific community.

The paper is well written and the methods are clearly described. The diurnal and sea-

sonal variation of SO_2, as well as the impact of meteorology and as the year-to year variability of SO_2 abundances has been discussed in detail. However, in my opinion the potential of MAX-DOAS for the characterization of the vertical structure of the boundary layer has not been fully exploited. Apart from a very brief presentation of monthly mean vertical SO_2 profiles, only SO_2 VCDs are discussed, with the argument that the surface concentration from MAX-DOAS agrees well with measurements from an in situ SO_2 monitor, and that the SO_2 VCD is proportional to the surface concentration. From what is presented in the manuscript, one might raise the question what the advantage of MAX-DOAS measurements is since these require a very sophisticated and complex retrieval algorithm and are subject to relatively high uncertainties compared to standard SO_2 in situ monitoring instruments. One could have reached exactly the same conclusions by using only in situ SO_2 data which is readily available for many sites in China, and it does not become clear what the actual advantage of MAX-DOAS is. I would therefore appreciate if the authors would take more advantage of the capabilities of MAX-DOAS, covering the following aspects:

An important environmental concern related to SO_2 is the production of sulphuric acid and sulphate aerosols during smog conditions. MAX-DOAS measurements would be ideal to investigate the relationship between SO_2 emissions and aerosol production, since they contain information on both the aerosol extinction and the SO_2 concentration profile. It would therefore be highly desirable if aerosols retrieved from MAX-DOAS and their (potential) relation to SO_2 would be discussed.

Instead of showing only the diurnal variation of SO_2 VCDs (Fig. 13), it would be more instructive to show and discuss the diurnal variation of the vertical profile, as it also reveals the vertical extent of SO_2. Furthermore, it would be interesting to discuss the averaged aerosol extinction profiles as these would help to characterise the vertical structure of the boundary layer. This would help to investigate whether the presence of aerosols is related to the SO_2 abundance.

It has been speculated that the variability of the SO_2 VCD is partially caused by vari-

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ations in the boundary layer height, but without providing any evidence. First of all, in contrast to the surface concentration, the VCD should not vary due to a vertical dilution (except for effects arising from the reduced sensitivity at higher altitudes). Secondly, I wonder why the authors only speculate about an impact of the boundary layer height on SO_2 abundances, and do not examine the vertical structure of the boundary layer which is readily available from the MAX-DOAS SO_2 and aerosol profiles.

An important aspect of trace gas profile retrieval from MAX-DOAS measurements is the ability to constrain the light path using aerosol extinction profiles retrieved from O_4 dSCDs measured with the same instrument under exactly the same conditions. Agreement of modelled and measured O_4 dSCDs ensures that the simulated light path is compatible with the measurements, even if the aerosol profile might not exactly reflect the real atmospheric conditions. This self-consistency of the MAX-DOAS retrieval represents a great advantage. Therefore I find it hard to justify why the authors do not simply use the aerosol profile retrieved from O_4 dSCDs (converted to the wavelength of the SO_2 retrieval) directly as input for the SO_2 retrieval (which by default yields a realistic constraint for the light path), but instead use an extinction profile with predefined exponential shape and fixed layer height, scaled to the retrieved AOD. This approach will certainly introduce significant errors if the shape of the true extinction profile differs from the assumed exponential profile.

Specific Comments

6502.12: Why should the SO_2 VCD (i.e., integrated column) increase if SO_2 accumulates close to the ground? Why has this hypothesis not been tested on the basis of the retrieved SO_2 vertical profiles? From the monthly mean profile in Fig. 4, it seems that the layer height is actually highest in February which is in contradiction to this hypothesis.

6503.5ff: It is mentioned that SO_2 leads to the formation of sulphate aerosols. Why has the relationship between SO_2 and aerosols not been investigated based on the

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MAX-DOAS data?

6504.5ff: It is not true that, regarding SO_2, only little efforts have been dedicated to the retrieval and monitoring of this species from MAX-DOAS measurements. A lot has been published on SO_2 from MAX-DOAS, in particular in the framework of volcanic monitoring (e.g., Bobrowski, N., R. von Glasow, A. Aiuppa, S. Inguaggiato, I. Louban, O. W. Ibrahim, and U. Platt, Reactive halogen chemistry in volcanic plumes, J. Geophys. Res., 112, D06311, 2007, doi: 10.1029/2006JD007206). Numerous SO_2 measurements around the world are continuously performed as part of the NOVAC network (http://www.novac-project.eu, see Galle, B., M. Johansson, C. Rivera, Y. Zhang, M. Kihlman, C. Kern, T. Lehmann, U. Platt, S. Arellano and S. Hidalgo, Network for Observation of Volcanic and Atmospheric Change (NOVAC) - A global network for volcanic gas monitoring: Network layout and instrument description, J. Geophys. Res., 115, D05304, 2010, doi: 10.1029/2009JD011823). Another example are MAX-DOAS measurements of SO_2 in Korea (Lee, C., A. Richter, H. Lee, Y. J. Kim, J. P. Burrows, Y. G. Lee, and B. C. Choi, Impact of transport of sulfur dioxide from the Asian continent on the air quality over Korea during May 2005, Atmospheric Environment, 42, 1461 - 1475, 2008, doi: http://dx.doi.org/10.1016/j.atmosenv.2007.11.006).

6505.26: Please specify what you mean with 'self-calibration'.

6506.14: It is mentioned that the residual is small. But small compared to what? Either remove this statement or compare to other measurements. What is the typical error in SO_2 dSCDs?

6507.10: I do not think that any solutions are 'rejected' in the OEM method. Instead, the a priori provides additional constraints to the retrieved state vector.

6507.20: There are many different aerosol profiles in the LOWTRAN database. Which one did you choose as a priori an what are its properties?

6508.4ff: Why do you discard the retrieved aerosol profile shape and instead use an

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exponential shape for the SO_2 retrieval? As already mentioned in the general comments, this approach will lead to a less realistic simulation of the radiative transfer in the SO_2 retrieval.

6508.7: Which Ångstr öm exponent did you use for the conversion of the aerosol profile to shorter wavelengths?

6508.13: $\mathbf{S}_\epsilon$ and $\mathbf{S}_a$ are crucial parameters of the retrieval. Please specify these here instead of referring to Clémer et al.

6508.23: I am a bit confused about the RMS of the profile retrieval being specified in percent. For an ideal retrieval (no systematic errors, physically correct forward model, realistic measurement and a priori error covariances), the average RMS should equal the dimension of the measurement vector.

6509.15: In absolute numbers, the gradient of the profiles in February and November is indeed largest. However, this seems to be mainly due to the fact SO_2 amounts are highest in these months, as the layer height (in terms of something like e-folding height) appears to be very similar during all months. Why should larger surface concentrations of SO_2 necessarily lead to larger vertical gradients?

6509.19ff: Why should the SO_2 amount affect the DFS? This should not be the case as the trace gas profile retrieval usually represents a linear problem (maybe non-linear effects due to the strong absorption of SO_2 important here?). Instead, the aerosol amount should have a strong impact on the information content. It would be interesting to know if DFS correlates with AOD.

6511.11: Here it is speculated about the impact of boundary layer height on SO_2 concentrations. As already mentioned in the general comments, I wonder why the retrieved profiles of SO_2 and aerosols have not been used to confirm this hypothesis. Do you have examples where temperature inversion events result in an accumulation of SO_2 in the lower troposphere? The ratio between retrieved VCD and surface concentration

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could be used to investigate the seasonal variation of the layer height.

6512.14: This sentence should be rephrased since an inverse proportionality of the SO_2 VCD to the wind speed would imply a strict relation like $y = 1/x$. This is rather an anti-correlation.

6512.14 and 6514.20: From Fig. 11, I do not see any anti-correlation between wind speed and SO_2 VCD for eastern and south-western wind directions. Instead, this seems to be the case for north-eastern and north-western directions.

6513.13 and 6514.18: Again, the hypothesis that temperature inversions lead to increases in the SO_2 amount near the surface should be confirmed by inspecting the vertical profiles retrieved from MAX-DOAS.

Technical corrections

6503.5: remove 'Furthermore'

6503.6: '*to* a large extent'

6503.13: 'to meet the urgent demand to improve and control air quality in China'.

6506.25: I suggest to replace 'absorption by' by 'optical density of'.

6510.12 and fig. 13: I suggest to use the common abbreviations MAM, JJA, SON, DJF for the seasons.

6512.4: Delete 'For this purpose'

6513.8: Delete 'In spring and autumn'.

Fig. 11, panel (b): It is not clear to which wind speed intervals the lines refer to. Is red 0-1 m/s or 1-2 m/s? I suggest to plot panel (b) also as a wind rose, or even to merge panels (a) and (b) in a single wind rose diagram.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 6501, 2014.