

01/08/2014

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: Correction

Please find below our responses to both referees comments. They also contain the changes implemented in the revised version of the manuscript. The latter are highlighted in yellow in the new manuscript.

Sincerely yours,

F. Hendrick (franch@oma.be)

Response to Anonymous Referee #1

First, we would like to thank Anonymous Referee #1 for his/her helpful comments.

Please find our replies below. We used the following color code:

Green: Referee's comment

Black: author's reply

Red: modified text in the revised manuscript

General Comments

The paper entitled 'Evaluation of tropospheric SO₂ retrieved from MAX-DOAS measurements in Xianghe, China' by Wang et al. presents three years of continuous SO₂ observations at a location south-east of Beijing. The emission of SO₂ 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 seasonal variation of SO₂, as well as the impact of meteorology and as the year-to-year variability of SO₂ 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 SO₂ profiles, only SO₂ VCDs are discussed, with the argument that the surface concentration from MAX-DOAS agrees well with measurements from an in situ SO₂ monitor, and that the SO₂ 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 SO₂ in situ monitoring instruments. One could have reached exactly the same conclusions by using only in situ SO₂ 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:

***Referee's comment #1:** An important environmental concern related to SO₂ is the production of sulphuric acid and sulphate aerosols during smog conditions. MAX-DOAS measurements would be ideal to investigate the relationship between SO₂ emissions and aerosol production, since they contain information on both the aerosol extinction and the SO₂ concentration profile. It would therefore be highly desirable if aerosols retrieved from MAX-DOAS and their (potential) relation to SO₂ would be discussed.*

Author's reply: We agree Referee #1 on the fact that our MAX-DOAS observations should allow to investigate the relation between SO₂ and aerosols. In the revised manuscript, we have included a new Section (3.4) on the relationship between SO₂ and aerosols. This relationship has been investigated through a correlation study of SO₂ VCD and surface concentration versus AOD and surface extinction coefficient, respectively. Here is the new Sect. 3.4:

Fig. 16 shows monthly scatter plots of the SO₂ concentration versus aerosol extinction coefficient retrieved in the 0-200m layer for the March 2010 – February 2013 period. A strong correlation (correlation coefficients in the 0.6-0.9 range) is obtained in JFM and OND while a significantly lower correlation is observed in late spring/summer with correlation coefficients around 0.3 in JJA. Similar features are found from the scatter plots of SO₂ VCD versus AOD (not shown here). The marked seasonality of the correlation between SO₂ and aerosols is further illustrated in Fig. 17 where monthly correlation coefficients for both surface concentration and integrated column are reported. The positive correlation (>0.2) observed throughout the year indicates that in most cases, high pollution events in Xianghe are associated with enhanced SO₂ and aerosol levels (Chan and Yao, 2008; Li et al., 2007).

The higher correlation coefficients obtained in winter (>0.6) suggest that anthropogenic SO_2 plays a more significant role in the aerosols formation during this period of the year due to its larger concentration and lower temperatures favoring the formation of sulfates (Lin et al., 2012). In late spring/summer, the Beijing area is also strongly influenced by other sources of aerosols, especially particles emitted from massive agricultural fires in the surrounding region (Xia et al., 2013) as well as dust particles transported from the Kumutage and Taklimakan deserts in western China and from the Mongolian deserts (Yu et al., 2009). In combination to the lower SO_2 concentration, this could explain the significantly weaker correlation between anthropogenic SO_2 and aerosols obtained in JJA. However, measurements of the chemical composition of aerosols in Xianghe would be needed to further support our findings.

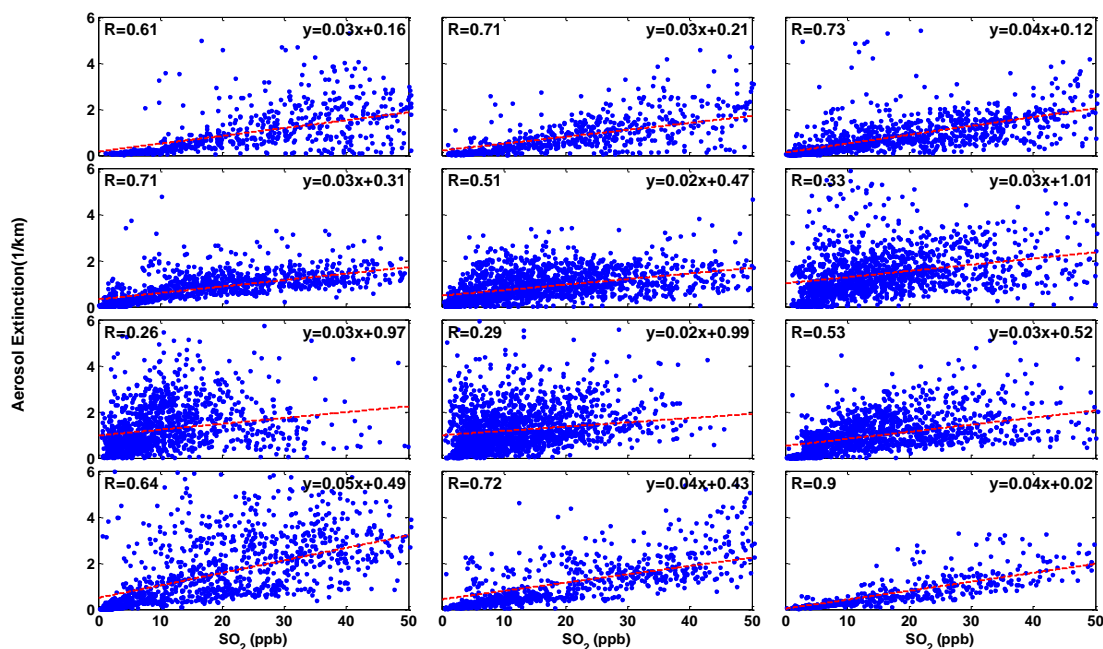


Figure 16: Scatter plots of aerosol extinction coefficient versus SO_2 concentration in the 0-200m layer for months 1-12 of the March 2010 – February 2013 period (first row is for JFM, second row for AMJ, third row for JAS, and fourth plots for OND). The data points correspond to the different MAX-DOAS scans. The red line denotes the linear least-squares fit to the data.

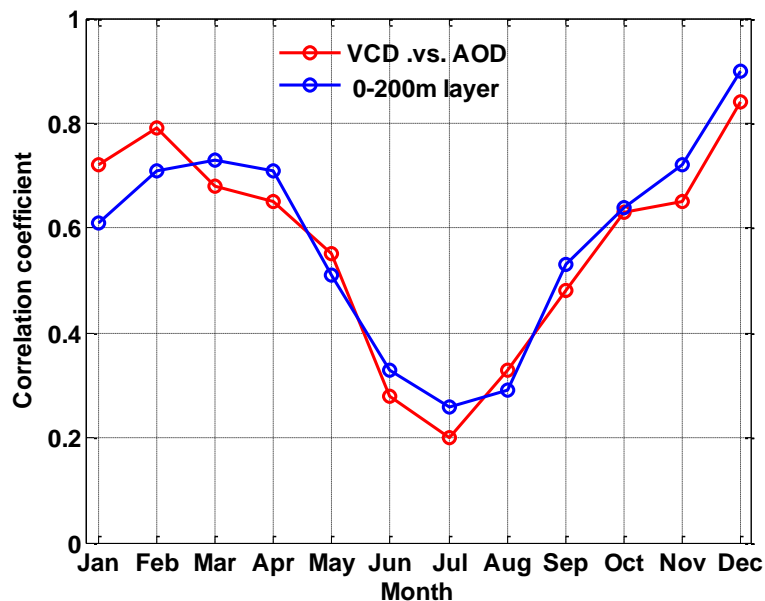


Figure 17: Seasonal variation of the correlation coefficient between SO_2 and aerosols over the March 2010-February 2013 period. The red curve corresponds to VCD versus AOD and the blue curve to SO_2 concentration versus aerosol extinction coefficient in the 0-200m layer.

The abstract and conclusions have been also modified accordingly.

Referee's comment #2: 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 characterize the vertical structure of the boundary layer. This would help to investigate whether the presence of aerosols is related to the SO_2 abundance.

Author's reply: With a maximum DFS around 2 on average for both SO_2 and aerosol retrievals, we think that the information content is not high enough to quantitatively investigate the vertical extent of both SO_2 and aerosols. With such DFS, the shape of the retrieved profiles can be strongly influenced by the a priori profile shape and for this reason the retrieved profiles are only a first-order estimates of the true profiles (see Vlemmix et al., 2011 and 2014). Therefore, investigating the vertical structure of the boundary layer from these profiles can lead to erroneous conclusions unless the retrieved boundary layer heights are verified through comparisons with correlative measurements (e.g. from a ceilometer) and/or model calculations, which is beyond the scope of the present study. For these reasons, we decided that the discussion in the revised manuscript should remain mainly focused on the VCDs and surface concentrations. The combination of both quantities is the main advantage of the MAX-DOAS

technique and this combination is certainly not possible from in-situ measurements. Moreover, in-situ monitors need to be calibrated on a regular basis, which is not the case for MAX-DOAS spectrometers.

Regarding the relationship between SO₂ and aerosols, it is now investigated through a correlation study of SO₂ VCD and surface concentration versus AOD and surface extinction coefficient, respectively (see above our reply to general comment #1).

Referee's comment #3: It has been speculated that the variability of the SO₂ VCD is partially caused by variations 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 SO₂ abundances, and do not examine the vertical structure of the boundary layer which is readily available from the MAX-DOAS SO₂ and aerosol profiles.

Author's reply: We agree on the fact that (1) VCD should not vary due to vertical transport resulting from the variation of the boundary layer height, in contrast to the surface concentration, and (2) the way it is discussed in the manuscript can be confusing.

In order to address this comment, we have proceeded to the following changes in the revised manuscript:

- (1) Page 6502, lines 11-13 (page 1, lines 20-22 in the revised manuscript): ‘This can be explained by the larger emissions in winter due to the domestic heating and more favorable meteorological conditions for the accumulation of SO₂ close to the ground during this period.’ is replaced by ‘This can be explained by the larger emissions in winter due to the domestic heating and, in case of surface concentration, by more favorable meteorological conditions for the accumulation of SO₂ close to the ground during this period.’
- (2) Page 6511, lines 11-13 (page 12, lines 15-17 in the revised manuscript): ‘In addition, the reduced atmospheric boundary layer height and frequent temperature inversion events result in an accumulation of SO₂ in the lower troposphere (Meng et al., 2009).’ is replaced by ‘In addition, the reduced atmospheric boundary layer height and frequent temperature inversion events result in larger surface concentrations due to an accumulation of SO₂ in the lower troposphere (Meng et al., 2009).’
- (3) Page 6513, lines 13-15 (page 14, lines 16-17 in the revised manuscript): ‘This can be explained

by more frequent temperature-inversion events and a strengthened diurnal variation of emission sources during this period (Meng et al., 2009)’ is replaced by ‘This can be explained a strengthened diurnal variation of emission sources during this period (Meng et al., 2009)’

- (4) Page 6514, lines 15-19 (page 16, lines 16-17 in the revised manuscript): ‘The high levels of SO₂ during the cold season are further enhanced by the weakness of the wet deposition mechanism and the frequent temperature-inversion events occurring during this period, favoring the accumulation of SO₂ in the atmospheric layers close to the ground.’ is replaced by ‘The high levels of SO₂ during the cold season are further enhanced by the weakness of the wet deposition mechanism and, in case of surface concentration, by the frequent temperature-inversion events occurring during this period, favoring the accumulation of SO₂ in the atmospheric layers close to the ground.’

Regarding the second point (impact of the boundary layer height), see above our reply to general comment #2.

Referee’s comment #4: 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 O₄ dSCDs measured with the same instrument under exactly the same conditions. Agreement of modeled and measured O₄ 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 O₄ dSCDs (converted to the wavelength of the SO₂ retrieval) directly as input for the SO₂ 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.

Author’s reply: In the revised version of the paper, we have applied the Angström exponent approximation directly to the retrieved extinction profiles. More details on how this approximation is applied are now also given (see below our reply to specific comment 6508.7).

Specific Comments

Referee's comment (6502.12): Why should the SO₂ VCD (i.e., integrated column) increase if SO₂ accumulates close to the ground? Why has this hypothesis not been tested on the basis of the retrieved SO₂ 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.

Author's reply: See above our reply to general comment #3.

Referee's comment (6503.5ff): It is mentioned that SO₂ leads to the formation of sulfate aerosols. Why has the relationship between SO₂ and aerosols not been investigated based on the MAX-DOAS data?

Author's reply: Thank for the useful suggestion. The relationship between SO₂ and aerosol is investigated in the revised version of the manuscript (see our reply to the first general comment).

*Referee's comment (6504.5ff): It is not true that, regarding SO₂, only little efforts have been dedicated to the retrieval and monitoring of this species from MAX-DOAS measurements. A lot has been published on SO₂ 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 SO₂ 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 SO₂ 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>).*

Author's reply: A lot of work has been done about MAX-DOAS measurements of volcanic SO₂, but so far, only a few studies deal with MAX-DOAS SO₂ observations in polluted area. This point is discussed in more details in the revised manuscript and the above references are now included (see page 3, lines 18-23).

Referee's comment (6505.26): Please specify what you mean with 'self-calibration'.

Author's reply: 'self-calibration' is now specified in the revised manuscript (see page 5, lines 11-17):

(MAX-)DOAS is recognized as a “self-calibrating” technique because differential absorptions are measured and therefore the impact of possible instrumental degradations can be largely removed by using appropriate reference spectra. In contrast, in-situ instruments need to be optically and/or chemically calibrated on a regular basis, especially when performing long-term measurements. For tropospheric studies, a zenith spectrum is frequently chosen as reference, in this way also removing the contribution of the stratosphere in off-axis DSCDs.

Referee’s comment (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 SO₂ dSCDs?

Author’s reply: We agree that the sentences ‘We see that the residual is small, ranging from -2×10^{-3} to 2×10^{-3} , which indicates a limited retrieval error. In this illustrative case, the retrieved SO₂ DSCD is 7.27×10^{16} molec·cm⁻².’ suffer from a lack of clarity and can be misleading. We have reformulated this part as follows:

‘We see that fitting residuals range in between -2×10^{-3} and 2×10^{-3} , corresponding to a root-mean-squares (RMS) of 9×10^{-4} , which appears to be small in comparison to the SO₂ differential structures represented in the lowest panel of the figure. The typical fitting uncertainty on SO₂ DSCDs is of about $1-6 \times 10^{15}$ molec·cm⁻² (~less than 10%), and for the case illustrated here corresponds to 2%.’

Referee’s comment (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.

Author’s reply: We agree that solutions are not formally rejected in the OEM but constraining the retrieval with a priori information leads indirectly to the rejection of unrealistic solutions. So, ‘...to reject unrealistic solutions...’ is replaced by ‘...to indirectly reject unrealistic solutions...’ (see page 7, line 22 in the revised manuscript).

Referee’s comment (6507.20): There are many different aerosol profiles in the LOWTRAN database. Which one did you choose as a priori and what are its properties?

Author’s reply: The sentence on line 20-22 page 6507 is not correct. Actually, we did not use a fixed aerosol extinction profile taken from the LOWTRAN climatology as a priori for our aerosol retrievals. What we used is an exponentially decreasing profile corresponding to an AOD of 0.2 and a scaling height of 0.5km. This is corrected in the revised manuscript (see page 8, lines 6-16).

Referee's comment (6508.4ff): Why do you discard the retrieved aerosol profile shape and instead use an exponential shape for the SO₂ retrieval? As already mentioned in the general comments, this approach will lead to a less realistic simulation of the radiative transfer in the SO₂ retrieval.

Author's reply: This point is already discussed above. See above our replies to general comment #4 and specific comment 6508.7 below.

Referee's comment (6508.7): Which Ångström exponent did you use for the conversion of the aerosol profile to shorter wavelengths?

Author's reply: The application of the Ångström exponent approximation is discussed into more details in the revised manuscript. In AERONET database, 5 different Ångström exponents are available: 340-440nm, 380-500nm, 440-675nm, 440-870nm, 500-870nm. The 340-440 nm exponent, which is closest to the SO₂ fitting interval (305-317.5nm) has been used in a first approximation. It is now applied directly to the retrieved aerosol extinction profiles instead to the AODs (and then assuming exponentially decreasing extinction profiles). The corresponding mean scaling factor for converting aerosol extinction profiles from 360 to 313 nm is of 1.16 ± 0.06 . SO₂ vertical profiles have been retrieved with these new aerosol extinction profiles and all Figures and Table 2 in the revised manuscript have been updated with these new aerosol and SO₂ data sets. Results and findings remain similar to those obtain with the previous data set, except the retrieved profile shape in spring and fall which now displays a maximum in the 200-400m layer instead of in the first layer. The discussion on the profiles is modified accordingly in the revised manuscript (see page 10, lines 18-20).

We have proceeded to the following text changes for addressing this comment (see page 8, line 16 up to page 9, line 1):

The sentences 'Since the DOAS fitting intervals are different for SO₂ and aerosols, the aerosol extinction profiles utilized as input for the calculation of SO₂ weighting functions have been derived by converting the AODs retrieved in the 338-370 nm wavelength range to the 305-317.5 nm interval using the Ångström formula (Cachorro et al., 2000), and assuming an exponentially decreasing profile shape with a SH of 0.5 km (see Eq. 2).'

have been replaced by

'Since the DOAS fitting intervals are different for SO₂ and aerosols, the aerosol extinction profiles utilized as input for the calculation of SO₂ weighting functions have been derived by directly

converting the aerosol profiles retrieved in the 338-370 nm wavelength range to the 305-317.5 nm interval using the Ångström exponents (Cachorro et al., 2000) retrieved from collocated CIMEL/AERONET sunphotometer measurements (Holben et al., 1998; see <http://aeronet.gsfc.nasa.gov>):

$$\text{Extinction}(z, 313 \text{ nm}) = \text{Extinction}(z, 360 \text{ nm}) \times (313/360)^{-\alpha} \quad (3)$$

where z is the altitude and α is the Ångström exponent.

The 340-440 nm exponents are used in a first approximation since values for a wavelength range closer to the SO₂ fitting interval (305-317.5 nm) are not available so far. The corresponding mean scaling factor for the March 2010 – February 2013 period is of 1.16 ± 0.06 .

Referee's comment (6508.13): S_ε and S_a are crucial parameters of the retrieval. Please specify these here instead of referring to Clémer et al.

Author's reply: We specified the following details for S_ε and S_a in the revised manuscript (see page 9, lines 6-15): . S_ε and S_a matrices are similar as in Clémer et al. (2010) and Hendrick et al. (2014). S_ε is a diagonal matrix, with variances equal to the square of the DOAS fitting error. For S_a, the diagonal element corresponding to the lowest layer, S_a (1,1), is set equal to the square of a scaling factor β times the maximum partial VCD (AOD) of the profiles. Here β=0.4 for SO₂ and 0.2 for aerosol. The other diagonal elements decrease linearly with altitude down to 0.2×S_a(1,1). The off-diagonal terms in S_a, were set using Gaussian functions as follows:

$$S_a(i, j) = \sqrt{S_a(i, i)S_a(j, j) \exp(-\ln(2)\left(\frac{z_i - z_j}{\gamma}\right)^2)} \quad (4)$$

where z_i and z_j are the altitudes of i^{th} and j^{th} levels, respectively. The correlation length is set to 0.1 km for SO₂ and 0.05km for aerosol in order to optimize the DFS.

The retrieval altitude grid is also the same as in Clémer et al. (2010) and Hendrick et al. (2014), i.e. ten layers of 200 m thickness between 0 and 2 km, two layers of 500 m between 2 and 3 km and 1 layer between 3 and 4 km.

Referee's comment (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.

Author's reply: For each scan, we calculate a 'standard' RMS expressed in $\text{molec}\cdot\text{cm}^{-2}$ and then we divide it by the mean DSCD of the scan to get a relative RMS expressed in percent, which is used to reject 'bad' scans. This is clarified in the revised manuscript (see page 9, lines 23-24).

Referee's comment (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?

Author's reply: We agree that larger surface concentrations do not necessarily lead to larger vertical gradients but this is what we obtain here. The sentence of 'which is due to the difference in SO_2 concentration near ground for the different months.' is replaced by 'This is mainly due to the fact that the SO_2 emissions are the highest in February and November.' (see page 10, lines 21-22 in the revised manuscript).

Referee's comment (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.

Author's reply: Our experience with optimal estimation shows that profile retrieval from low trace gas amounts generally associated with larger uncertainties is more difficult (no convergence or spurious oscillations in the retrieved profiles with low DFS). The low DFS for low trace gas amounts could be also related to the way the \mathbf{S}_a matrix is constructed (see above reply to the 6508.13 comment): low trace gas amounts lead to low a priori profiles and therefore to low values in the \mathbf{S}_a and averaging kernel matrices. It is known that high AODs make the forward modeling more difficult and therefore should affect more significantly the trace gas retrievals but from our experience, we know also that high AODs do not necessarily lead to low trace gas DFS.

Referee's comment (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 could be used to investigate the seasonal variation of the layer height.

Author's reply: See above our reply to General Comment #3. Moreover, since we don't have the observed vertical profiles of temperature corresponding to our MAX-DOAS measurements, we are not able to show examples of temperature inversion events. So the discussion on this subject remains speculative in the revised manuscript.

Referee's comment (6512.14): This sentence should be rephrased since an inverse proportionality of the SO₂ VCD to the wind speed would imply a strict relation like $y = 1/x$. This is rather an anti-correlation.

Author's reply: Corrected.

Referee's comment (6512.14 and 6514.20): From Fig. 11, I do not see any anti-correlation between wind speed and SO₂ VCD for eastern and south-western wind directions. Instead, this seems to be the case for north-eastern and north-western directions.

Author's reply: We agree that the discussion about the impact of wind speed and direction is a bit confusing. Panel (b) of Fig. 11 (actually Fig. 13 in the revised manuscript) is now plotted also as a wind rose (see last technical correction here below) and the text is reformulated as follows (see page 13, lines 14-23):

It can be seen that the amount of SO₂ is strongly dependent on the wind direction (Fig. 13a): high VCDs are prominent when the winds blow from the east, because Tangshan, a heavy industrial city releasing large amounts of SO₂, is situated to the east of Xianghe (see Fig. 1); in contrast, the north-west direction corresponds to a minimum in SO₂ VCD, since it is a mountain area, characterized by much less emissions than in Xianghe. The wind therefore contributes significantly to the dispersion of the pollutants, as expected. Regarding the dependence of the SO₂ VCD on wind speed, Fig. 13(b) shows that the VCD is almost constant with wind speed for the E and SW, which means no good dispersion happens with the wind from these directions, since high-emission industrial areas and Tangshan are located to the southwest and east of Xianghe, respectively. In contrast, an anti-correlation is observed for NE/NNE, NW, and SE, which means the wind from these directions corresponding to less polluted areas can efficiently disperse pollutants. In addition, the SO₂ content at Xianghe is more sensitive to the emission sources in Tangshan (E) than in Beijing (WNW), which is consistent with the fact that Beijing has taken regulatory actions to reduce air pollution through traffic-control measures

and the closure of heavy polluting industries initiated before the 2008 Olympic Games (Yu et al., 2010).

The abstract and conclusions have been also modified accordingly.

Referee's comment (6513.13 and 6514.18): Again, the hypothesis that temperature inversions lead to increases in the SO₂ amount near the surface should be confirmed by inspecting the vertical profiles retrieved from MAX-DOAS.

Author's reply: See above our reply to general comment #3 and specific comment 6511.11.

Technical corrections

Referee's comment (6503.5): remove 'Furthermore'

Author's reply: Corrected.

Referee's comment (6503.6): 'to a large extent'

Author's reply: Corrected.

Referee's comment (6503.13): 'to meet the urgent demand to improve and control air quality in China'.

Author's reply: Corrected.

Referee's comment (6506.25): I suggest to replace 'absorption by' by 'optical density of'.

Author's reply: Corrected.

Referee's comment (6510.12 and fig. 13): I suggest to use the common abbreviations MAM, JJA, SON, DJF for the seasons.

Author's reply: Corrected.

Referee's comment (6512.4): Delete 'For this purpose'

Author's reply: Corrected.

Referee's comment (6513.8): Delete 'In spring and autumn'.

Author's reply: Corrected.

Referee's comment (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.

Author's reply: The legend '1m/s' means the average in the 0-2m/s interval, '2m/s' the average in the 1-3m/s interval, and so on. This is corrected in the new Fig. 11 (actually Fig. 13 in the revised manuscript) where panel (b) is now also plotted as a wind rose (see below), as suggested by Referee #1.

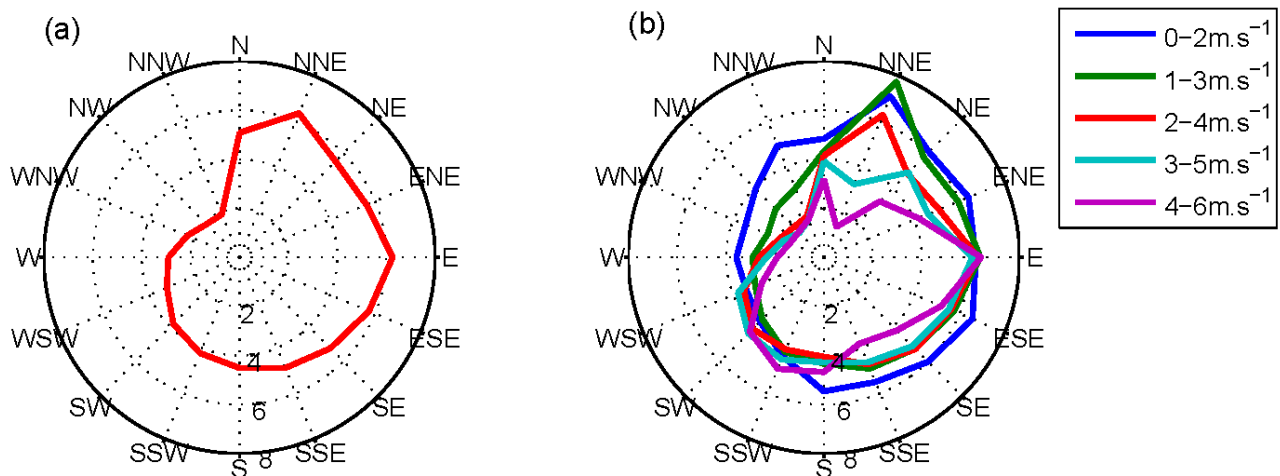


Figure 13: (a) Wind rose showing the SO₂ VCD (10^{16} molec.cm⁻²) as a function of the wind direction (average for all wind speed). (b) Dependence of SO₂ VCD (10^{16} molec.cm⁻²) on wind direction for different wind speeds.

References:

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Response to Anonymous Referee #2

First, we would like to thank Anonymous Referee #2 for his/her helpful comments.

Please find our replies below. We used the following color code:

Green: referee's comment

Black: author's reply

Red: modified text in the revised manuscript

The study of "Evaluation of tropospheric SO₂ retrieved from MAX-DOAS measurements in Xianghe, China" by T.Wang et al. presents the seasonal and diurnal variability of SO₂ in the boundary layer of Xianghe, China retrieved by ground based MAX-DOAS. The interpretation of the results is aided by independent in-situ SO₂ and meteorological measurements. The three years of MAX-DOAS measurements in the urban atmosphere of Xianghe, China, demonstrate the importance of this technique for air quality purposes and as indicator of boundary layer in-homogeneity. The manuscript is well written, however, it does not show an evident/noticeable novelty from the three continuous years of MAX-DOAS measurements, as they are not fully exploited. I suggest the publication of this manuscript after considering changes/improvements according to the comments below:

***Referee's comment #1:** According with the authors, the reliability of the SO₂ retrieval vertical profile is demonstrated with the comparison of the near surface concentration retrieval profiles with in-situ and independent SO₂ measurements. However, the sensitivity of air masses is quite distinctive for both methods. In-situ measurements detect air mass close to the instrument and might be able to detect localized air mass while moving close to the site. On the other hand, MAX-DOAS measures/averages the air mass over a long distance. From my point of view, this comparison is actually important in terms of air mass homogeneity in the boundary layer rather than validation process. I would recommend to use backward trajectories at different altitudes to identify air masses, especially aloft. This could help in a deep explanation of the vertical profile extent which is missing in the manuscript.*

Author's reply: To our opinion, the good agreement between MAX-DOAS and in-situ SO₂ measurements means that there is no major horizontal representativeness issues in these comparisons, probably because Xianghe is a sub-urban site not so much affected by local emissions, in contrast to the Beijing City Centre. Moreover, as suggested by Referee #2, we have performed backward trajectory calculations using the HYSPLIT model (<https://ready.arl.noaa.gov/HYSPLIT.php>) for the 9 days for

which MAX-DOAS and in-situ data have been compared. Fig. A shows that in most cases, the air masses corresponding to the three selected altitude levels (100m, 500m, and 1000m) have similar origins and trajectories, coming mostly from the north (pointing direction of our instrument), indicating that in-situ and MAX-DOAS instruments are more likely detecting similar air masses. We have decided to not discuss this point in the revised manuscript.

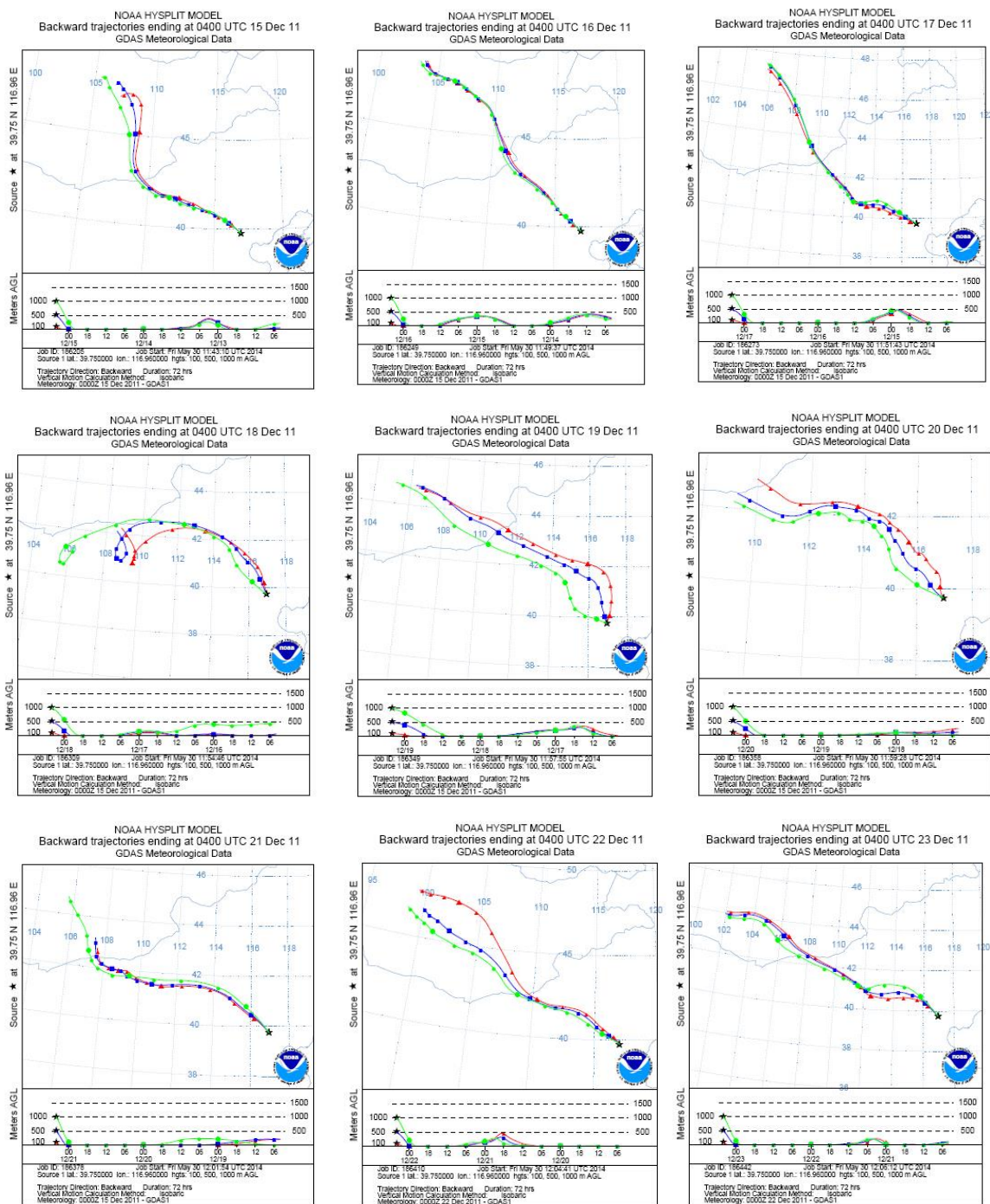


Figure A: Backward trajectories for the 15-23 December 2011 period (upper plots are for 15-17, middle plots are for 18-20, lower plots are for 21-23 December). The selected altitude levels are 100m (red), 500m (blue), and 1000m (green).

Referee's comment #2: One important advantage of MAX-DOAS over other techniques, is the capability of measuring several species simultaneously. In the present manuscript, solely results of SO₂ are shown, even though other species can be retrieved, such as NO₂, and aerosol extinction profiles. Undoubtedly the manuscript would improve if results of NO₂ and aerosol extinction (which are actually retrieved in the first step approach) are shown. The ratio of SO₂/NO₂ can be used, for example, as a metric to understand in more detail the emission level and atmospheric transport in the boundary layer. In the current manuscript meteorological conditions and qualitative seasonal domestic heating are used in order to know emission sources, however the metric SO₂/NO₂ ratio could be used adequately to know industry or power plant SO₂ episodes, not only at the surface but also in the vertical profile inside the boundary layer. On the other hand, the correlation of SO₂ and aerosol extinction would be important as an indication of SO₂ conversion and aerosol production.

Author's reply: We agree with Referee #2 on the fact that the SO₂/NO₂ ratio is an important parameter for investigating the emission sources. This parameter has been investigated in several papers (e.g., Li et al., 2010; Chai et al., 2014). However, we think that looking at particular emission episodes from e.g. power plants and possible transport of SO₂ and NO₂ to Xianghe is beyond the scope of the present study, as it will require an important modelling part. The aim of the present paper is to describe this unique three-year data set of MAX-DOAS SO₂ observations and give reasonable explanations to what we observe. Once published, this data set could then be used for more detailed/dedicated studies like the one suggested by Referee #2.

Regarding point 2, we have included a new Section (3.4) on the relationship between SO₂ and aerosols in the revised manuscript. This relationship has been investigated through a correlation study of SO₂ VCD and surface concentration versus AOD and surface extinction coefficient, respectively. Here is the new Sect. 3.4:

Fig. 16 shows monthly scatter plots of the SO₂ concentration versus aerosol extinction coefficient retrieved in the 0-200m layer for the March 2010 – February 2013 period. A strong correlation (correlation coefficients in the 0.6-0.9 range) is obtained in JFM and OND while a significantly lower correlation is observed in late spring/summer with correlation coefficients around 0.3 in JJA. Similar

features are found from the scatter plots of SO_2 VCD versus AOD (not shown here). The marked seasonality of the correlation between SO_2 and aerosols is further illustrated in Fig. 17 where monthly correlation coefficients for both surface concentration and integrated column are reported. The positive correlation (>0.2) observed throughout the year indicates that in most cases, high pollution events in Xianghe are associated with enhanced SO_2 and aerosol levels (Chan and Yao, 2008; Li et al., 2007). The higher correlation coefficients obtained in winter (>0.6) suggest that anthropogenic SO_2 plays a more significant role in the aerosols formation during this period of the year due to its larger concentration and lower temperatures favoring the formation of sulfates (Lin et al., 2012). In late spring/summer, the Beijing area is more influenced by other sources of aerosols, especially particles emitted from massive agricultural fires in the surrounding region (Xia et al., 2013) as well as dust particles transported from the Kumutage and Taklimakan deserts in western China and from the Mongolian deserts (Yu et al., 2009). In combination to the lower SO_2 concentration, this could explain the significantly weaker correlation between anthropogenic SO_2 and aerosols obtained in JJA. However, measurements of the chemical composition of aerosols in Xianghe would be needed to further support our findings.

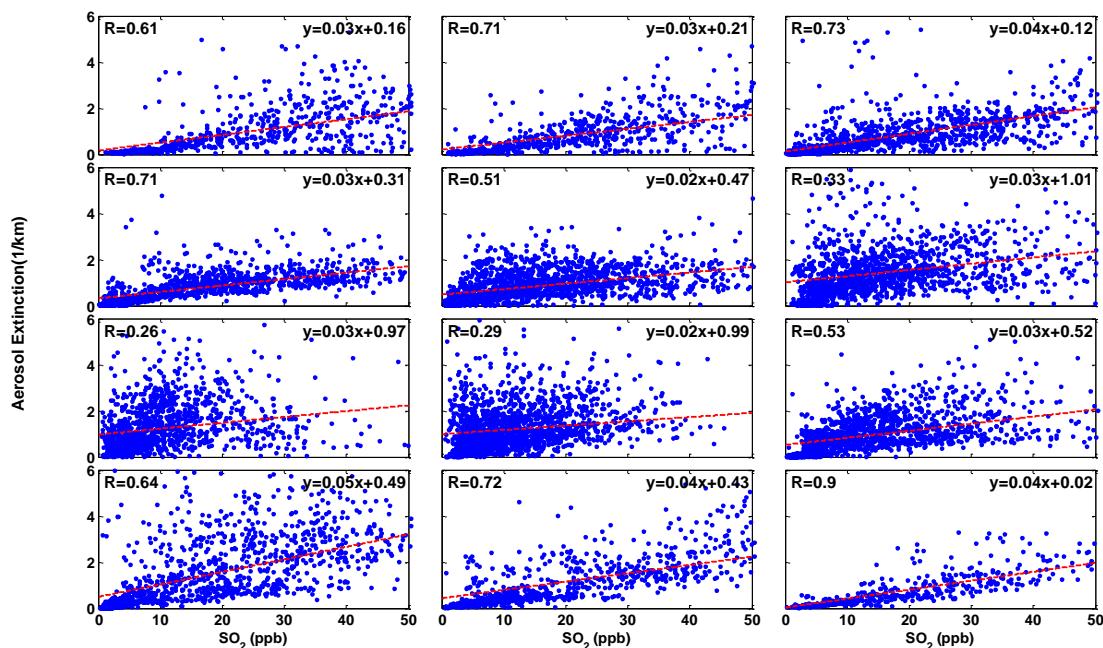


Figure 16: Scatter plots of aerosol extinction coefficient versus SO_2 concentration in the 0-200m layer for months 1-12 of the March 2010 – February 2013 period (first row is for JFM, second row for AMJ, third row for JAS, and fourth plots for OND). The data points correspond to the different MAX-DOAS scans. The red line denotes the linear least-squares fit to the data.

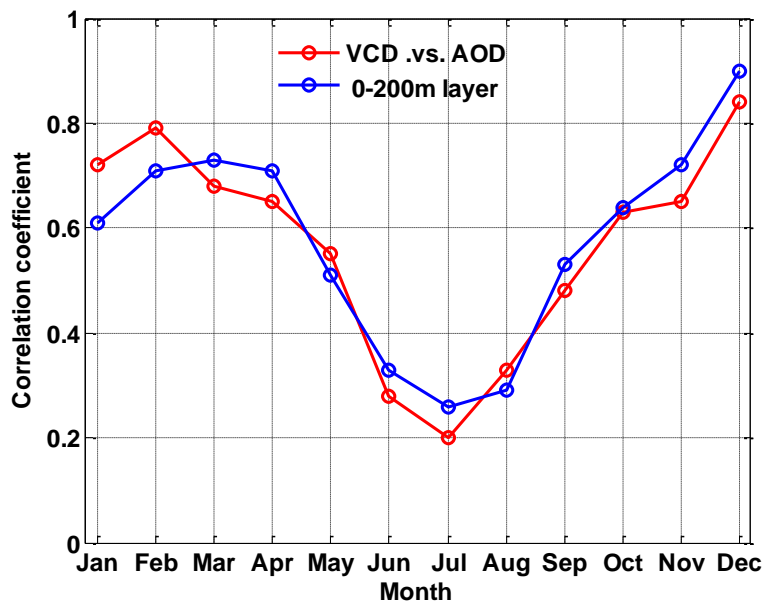


Figure 17: Seasonal variation of the correlation coefficient between SO_2 and aerosols over the March 2010-February 2013 period. The red curve corresponds to VCD versus AOD and the blue curve to SO_2 concentration versus aerosol extinction coefficient in the 0-200m layer.

The abstract and conclusions have been also modified accordingly.

Referee's comment #3: As pointed out in the introduction and in the conclusion, these three years of measurements are quite important for tropospheric SO_2 satellite validation/comparison. Have you thought in incorporating existing tropospheric SO_2 VCDs obtained with satellites and compare with your data?. Throughout the manuscript the main results, being the annual and diurnal cycles, are shown in terms of tropospheric SO_2 VCDs. Incorporating SO_2 VCD comparisons with satellite retrievals would improve the quality of the paper.

Author's reply: We have done comparisons between satellite (OMI, GOME-2, and IASI) and MAX-DOAS SO_2 VCDs, but these will be part of a separate paper about the description of the BIRA-IASB satellite SO_2 products (OMI, GOME-2) and their validation (Theys et al., in preparation, 2014). As an illustration, the comparison with OMI is shown in Fig. B. We see that calculating the SO_2 AMF for OMI by using MAX-DOAS vertical profiles significantly improves the agreement between both data sets.

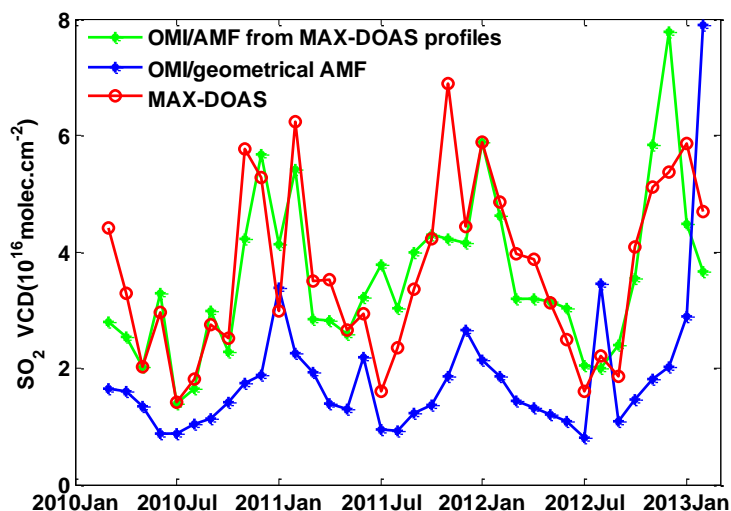


Figure B: Monthly-averaged SO₂ VCD of MAX-DOAS (red line), OMI/geometrical AMF (blue) and OMI/AMF from MAX-DOAS profiles (green) from March 2010 to February 2013.

Specific Comments

6505. DOAS analysis

Referee's comment: As pointed out in the introduction, SO₂ retrieval by MAX-DOAS have seldom been conducted in places far away from punctual sources such as volcanoes and/or industry. As far as I know, SO₂ retrieval represent a challenge due to absorption by stratospheric O₃ at the same wavelengths (< 325nm). In the DOAS analysis section it is mentioned that sensitivity tests were performed in order to choose the DOAS settings. Therefore, it would be valuable to know what kind of sensitivity tests were performed and applied in this work. I would recommend a detailed explanation and provide these results. This can be part of the supplementary information. Besides O₃ interference and DOAS analysis, more instrumentation details might be necessary For example, were filters used in the spectrometer system? It is known that instrument artifacts might lead to a bias due to noise in the spectral features.

Author's reply: The reliability and stability of the SO₂ DOAS analysis has been investigated through sensitivity tests on several key parameters, such as wavelength interval, choice of absorption cross sections, polynomial order, intensity offset terms. In the revised manuscript, we present the results of the sensitivity tests on the fitting window selection and discuss in more detail the ozone fitting. Here is the new text on DOAS settings (see page 5 line 18 -> page 6 line 17):

SO₂ fitting windows ranging between 303 and 325 nm have generally been used in previous studies (Bobrowski and Platt, 2007; Lee et al., 2008; Galle et al., 2010; Irie et al., 2011). At wavelengths shorter than 303 nm, the limiting factor is the strong ozone absorption which interferes with SO₂, leading to lower signal to noise ratio. At wavelengths longer than 325 nm, the SO₂ differential absorption signal becomes too weak. In order to identify the wavelength interval which minimizes both random and systematic uncertainties on SO₂ retrieval, 6 wavelength intervals have been investigated. The results of these sensitivity tests for two example days are presented in Figs. 2 and 3. On the first day (1st October 2011), the SO₂ content is minimum and stable in time. On the second day (4th October 2011), large variations of the SO₂ content occur, so the ability of the different intervals to give consistent and stable values can be verified. As can be seen, the 305-317.5 nm interval provides the lowest fitting errors throughout the day and the smallest dependence on the solar zenith angle (SZA) for both days. Due to the larger absorption and therefore interference by O₃ at large SZAs, it has been decided to exclude measurements taken at SZAs larger than 75°. For these tests, the following spectral signatures have been included: SO₂, O₃, NO₂, and the Ring effect (Grainger and Ring, 1962; Chance and Spurr, 1997). Daily zenith-sky radiance spectra recorded around local noon have been selected as reference. To account for the temperature dependence of the ozone absorption, cross sections at 2 different temperatures (223°K and 243°K) were used according to Van Roozendaal et al. (2006). A fifth-order polynomial is applied to fit the low-frequency spectral structure due to Rayleigh and Mie scattering and instrumental effects. Attempts to further adjust these settings, e.g. by adding BrO cross-section or by including additional ozone correction terms according to Puķīte et al. (2010) were not successful (less stable retrievals with larger noise on the SO₂ DSCDs).

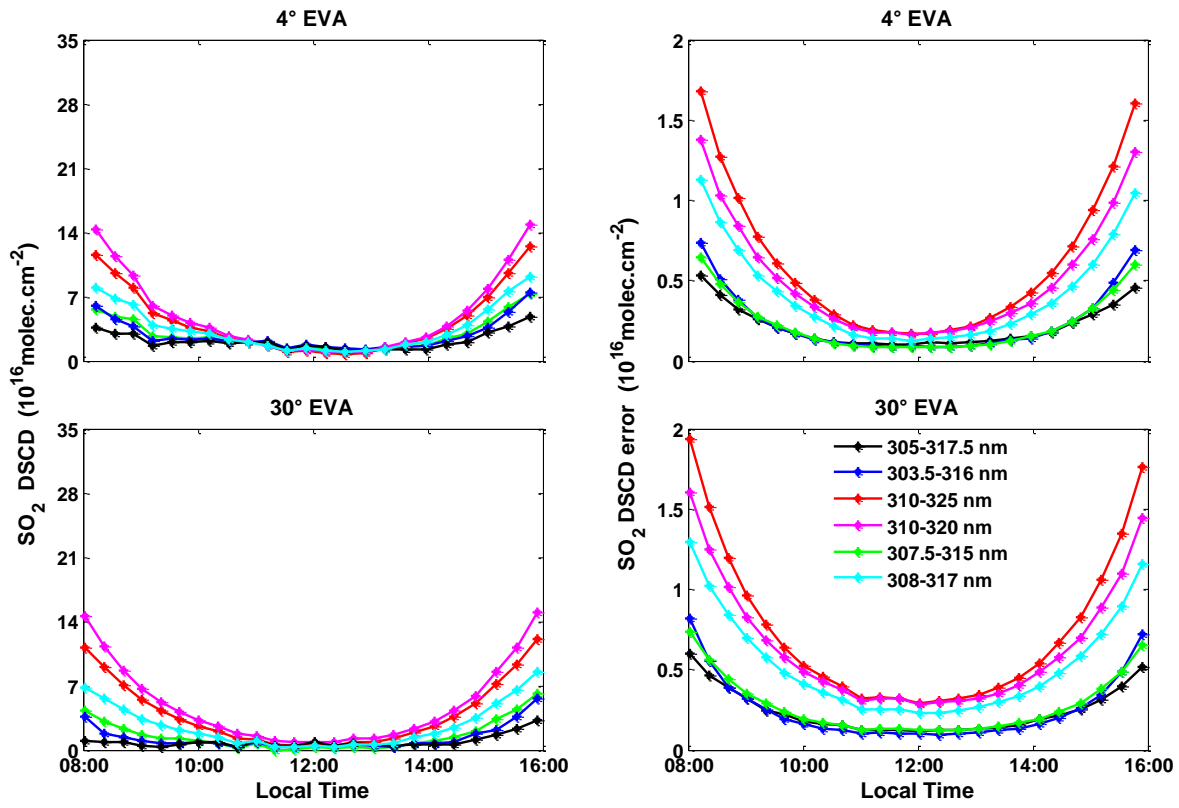


Figure 2: SO₂ DSCDs (1st column) and corresponding fitting uncertainties (2nd column) retrieved at 4° (upper plots), 30° (lower plots) elevation for different wavelength intervals on 1st October 2011.

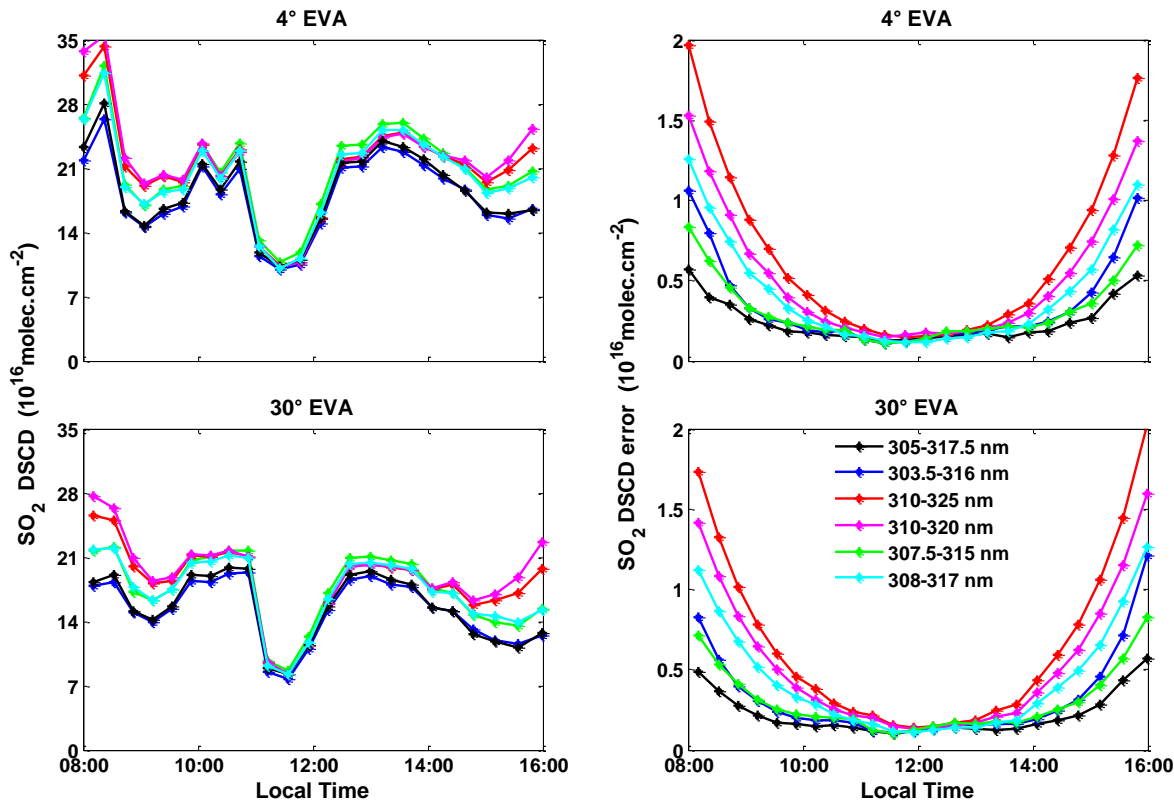


Figure 3: Same as Figure 2, but for 4th October, 2011.

Regarding the second point (instrumental set-up), we used for the UV channel a band-pass filter (Hoya U340) centered at 340 nm in order to avoid stray light from the visible range.

Referee's comment: In the same section it is mentioned that the residual achieved in the fitting example is small, ranging from -2×10^{-3} to 2×10^{-3} . Please provide more information about the quality of the DOAS analysis. Please explain why a value of 2×10^{-3} residual error is small? What is considered a good fit residual, and RMS, etc. Since this is the first SO_2 measurements of the MAX-DOAS what would be the detection limit of the MAX-DOAS (or please include a reference where this is mentioned).

Author's reply: We agree that the sentences ‘We see that the residual is small, ranging from -2×10^{-3} to 2×10^{-3} , which indicates a limited retrieval error. In this illustrative case, the retrieved SO_2 DSCD is $7.27 \times 10^{16} \text{ molec} \cdot \text{cm}^{-2}$.’ suffer from a lack of clarity and can be misleading. We have reformulated this part as follows including a discussion about the detection limit:

‘We see that fitting residuals range in between -2×10^{-3} and 2×10^{-3} , corresponding to a root-mean-squares (RMS) of 9×10^{-4} , which appears to be small in comparison to the SO_2 differential structures presented in the lowest panel of the figure. The typical fitting uncertainty on SO_2 DSCDs is of about 1-

6×10^{15} molec·cm⁻² (less than 10%), and for the case illustrated here, corresponds to 2%. For near-noon conditions, the detection limit on the SO₂ DSCD can be conservatively estimated as 3 times the one-sigma uncertainty on the slant column, which means approximately 3×10^{15} molec·cm⁻². This detection limit is similar for the vertical columns estimated using the geometrical approximation at 30° elevation (see Sect. 2.3). Vertical columns derived from the full inversion generally have a smaller detection limit, owing to the gain in sensitivity obtained when including near horizontal viewing measurements.’

6506. Profile Retrieval

Referee’s comment: It is mentioned that aerosol extinction and SO₂ vertical profiles are obtained by means of a non-linear approach. Usually this is the case for strong absorbers such as aerosols. Is SO₂ considered a strong absorber? could you apply a linear inversion and save time in the analysis?

Author’s reply: Actually, both the linear and the non-linear iterative approaches have been implemented in our profiling algorithm. For strong absorbers like O₄, the non-linear iterative approach is used. In case of weak absorbers like NO₂, HCHO, SO₂, the linear method is selected. This is corrected in the revised manuscript (see page 7, lines 12-15).

Referee’s comment: The retrieval approach is based in a two-step approach. First, the aerosol extinction is retrieved at different wavelengths and then is extrapolated to a shorter wavelength using just the AOD, the Angstrom formula, and an exponential profile shape. The determination of the aerosol extinction based in the O₄ has been demonstrated before, but it is not well explained how and why the AOD, the Angstrom formula, and the exponential decrease profile were used here. In order to know the spectral dependence of the aerosol extinction and/or AOD you might need at least two wavelengths. Please describe what wavelengths you used in this step. Also, explain why the AOD was used with an exponential profile shape instead of applying the aerosol angstrom exponent approximation to the aerosol extinction profile?

Author’s reply: The application of the Ångström exponent approximation is discussed into more details in the revised manuscript. In AERONET database, 5 different Ångström exponents are available: 340-440nm, 380-500nm, 440-675nm, 440-870nm, 500-870nm. The 340-440 nm exponent, which is closest to the SO₂ fitting interval (305-317.5nm) has been used in a first approximation. It is now applied directly to the retrieved aerosol extinction profiles instead to the AODs (and then assuming exponentially decreasing extinction profiles). The corresponding mean scaling factor for converting aerosol extinction profiles from 360 to 313 nm is of 1.16 ± 0.06 . SO₂ vertical profiles have been retrieved with these new aerosol extinction profiles and all figures and Table 1 in the revised

manuscript have been updated with these new aerosol and SO₂ data sets. Results and findings remain similar to those obtain with the previous data set, except the retrieved profile shape in spring and fall which now displays a maximum in the 200-400m layer instead of in the first layer. The discussion on the profiles is modified accordingly in the revised manuscript (see page 10, lines 18-20).

We have proceeded to the following text changes for addressing this comment: (see page 8, line 16 -> page 9, line 1):

The sentences ‘Since the DOAS fitting intervals are different for SO₂ and aerosols, the aerosol extinction profiles utilized as input for the calculation of SO₂ weighting functions have been derived by converting the AODs retrieved in the 338-370 nm wavelength range to the 305-317.5 nm interval using the Ångström formula (Cachorro et al., 2000), and assuming an exponentially decreasing profile shape with a SH of 0.5 km (see Eq. 2).’

have been replaced by

‘Since the DOAS fitting intervals are different for SO₂ and aerosols, the aerosol extinction profiles utilized as input for the calculation of SO₂ weighting functions have been derived by directly converting the aerosol profiles retrieved in the 338-370 nm wavelength range to the 305-317.5 nm interval using the Ångström exponents (Cachorro et al., 2000) retrieved from collocated CIMEL/AERONET sunphotometer measurements (Holben et al., 1998; see <http://aeronet.gsfc.nasa.gov>):

$$\text{Extinction}(z, 313 \text{ nm}) = \text{Extinction}(z, 360 \text{ nm}) \times (313/360)^{-\alpha} \quad (3)$$

where z is the altitude and α is the Ångström exponent.

The 340-440 nm exponents are used in a first approximation since values for a wavelength range closer to the SO₂ fitting interval (305-317.5 nm) are not available so far. The corresponding mean scaling factor for the March 2010 – February 2013 period is of 1.16 ± 0.06

Referee’s comment (6510):

I suggest to change “A very good agreement is found between both data sets, indicating the good overall reliability and the robustness of our MAXDOAS retrievals” according with the comments above.

Author’s reply: For the reasons given above (see our reply to the first general comment), we have decided to keep this sentence as is.

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