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Atmospheric Measurement Techniques Discussions

Interactive comment on "Novel SO_2 spectral evaluation scheme using the 360–390 nm wavelength range" *by* N. Bobrowski et al.

N. Bobrowski et al.

nicole.bobrowski@iup.uni-heidelberg.de

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Detailed response to Andreas Richter comments :

We would like to thank Andreas Richter his helpful comments and suggestion to improve the quality and clarity of our manuscript.

For reference the original comments are always included followed by our response.

* In several places, the authors discuss the non-linearity introduced by insufficient spectral resolution of strong absorptions, making reference to work on IR absorbers. In my opinion, this is not relevant for the SO2 retrieval as none of the main absorbers (SO2 and O3) have line spectra that could lead to saturation effects. This spectral resolution dependent effect is different from the non-linearity induced by changes in light path

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under strong absorption which is a major issue in volcanic plumes in the UV.

While it is true that the problem is more severe in the IR, it can also occur in the UV (see. e.g. Volkamer et al. Atmos. Environ. 32, 3731-3747, 1998). DOAS instruments do not directly measure optical densities, they measure the light intensity convoluted with the instrumental function (see also answer to referee No. 2). For large optical densities (in case of σ times SCD is not much smaller than unity), the convolution and the exponential function are not commutative. The common DOAS approach: τ = ln(H x (exp(σ âŃĚ SCD))) = SCD (H x σ) is only valid for weak absorbers! For strong absorbers like SO2 in a volcanic plume an itarative approach like the one from Frankenberg et al. 2005 should be in theory applied. The following figure shows the SO2 absorption cross-section (A.C. Vandaele, C. Hermans, and S. Fally, "Fourier transform measurements of SO2 absorption cross sections: II. Temperature dependence in the 29000-44000 cm-1 (227-345 nm) region", J. Quant. Spectrosc. Radiat. Transfer 110, 2115-2126 (2009)) convoluted with Gaussian slit functions of 0.1 nm, 0.25 nm and 0.5 nm resolutions. Also shown are the convolutions of the absorption cross-sections in intensity space assuming 1e19 and 4e19 molec/cm2 SO2 column densities. While no effect is seen at 0.1 nm resolution, the absorption cross-section is clearly reduced in comparison to the simple convolution in optical density space for the other spectral resolutions.

In conclusion, it is true that the effect described above is significantly weaker than the influence of the strong SO2 absorption on the effective light path in the plume. Also, the approximation of a commutative convolution is fairly good above 315 nm and for high spectral resolutions. It can however not be completely ignored.

This effect was studied in more detail in Kern 2009 (section 3.4.2), and figures 3.21 and 3.22 of the thesis are reproduced below.

We added only the two citations to the article:

Kern C, Spectroscopic measurements of volcanic gas emissions in the ultra-violet

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wavelength region. Ph.D. thesis, University of Heidelberg, 318 pages, Heidelberg, 2009. http://www.ub.uni-heidelberg.de/archiv/9574

Volkamer, R., Etzkorn, T., Geyer, A., and Platt, U.: Correction of the oxygen interference with UV spectroscopic (DOAS) measurements of monocyclic aromatic hydrocarbons in the atmosphere, Atmos. Environ., 32, 3731–3747, 1998.

Fig. 3.1 - Magnitude of underestimation in standard DOAS measurements conducted at 0.8 nm optical resolution caused by a non-commutative cross-section convolution. The largest errors occur for high column densities S and short wavelengths λ, where the differential absorption bands of SO2 are strongest.

Fig. 3.2 – Magnitude of underestimation in standard DOAS measurements conducted at 0.4 nm optical resolution caused by a non-commutative cross-section convolution. Errors are slightly smaller than for 0.8 nm optical resolution (Fig. 3.1) and again occur for high column densities S and short wavelengths λ.

* The discussion of radiation dilution is important for ground-based SO2 observations where it apparently was detected as late as 2006. It would be worthwhile to point out that the same effect (reduction in sensitivity through radiation scattered into the line of sight between the observer and the plume) was already well known for a long time in the satellite community where it is discussed in the framework of airmass factors and their vertical variation (shape factors, box-AMFs).

We agree with the reviewer in this point. We didn't discussed this issue sufficciently, not mentioning also earlier studies done for ground based measurements e.g. Millan 1980 and especially not mention any of the studies done in the satellite community therefore we added now:

The following citations in the introduction:

Moffat, A.J. and Millan, M.M.: The application of optical correlation techniques to the remote sensing of SO2 plumes using skylight, Atmos. Environ., 5, 677-690, 1971.

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Millan, M.M.: Remote sensing of air pollutants. A study of some atmospheric scattering effects. Atmos. Environ. 14, 1241–1253, 1980.

Eskes, H.K., and K. F. Boersma, Averaging kernels for DOAS total-column satellite retrievals, Atmos. Chem. Phys., 3, 1285–1291, 2003

Palmer, P. I., D. J. Jacob, K. Chance, R. V. Martin, R. J. D. Spurr, T. P. Kurosu, I. Bey, R. Yantosca, A. Fiore, and Q. Li, Air mass factor formulation for spectroscopic measurements from satellites: Application to formaldehyde retrievals from the Global Ozone Monitoring Experiment, J. Geophys. Res., 106, 14,539–14,550, 2001.

Richter, A. and J.P. Burrows, Tropospheric NO2 from GOME Measurements, Adv. Space Res., 29(11),1673-1683, 2002

and changed the sentence in the introduction: "This so-called ‘radiation dilution’ effect was first observed in DOAS measurements by Mori et al. (2006), who realized that the SO2 column density (CD) retrieved from measured spectra depended on the wavelength range in which the evaluation took place." to: "This so-called ‘radiation dilution’ effect was already discussed in Moffat and Millan (1971) and modelled by Millan (1980), but in ground based DOAS measurements first observed by Mori et al. (2006)...." and additional added: "The same effect is known and has frequently been discussed in the satellite community since about a decade (e.g. Palmer et al., 2001, Richter and Burrows, 2002, Eskes et al. 2003) and appropriate suggestion how to correct it have been made."

* I think it would be good to briefly discuss the work of Yang et al. who in several papers investigated the change in SO2 absorption as a function of wavelength and how this can be used to estimate plume altitude from satellite observations and also to correct for the above mentioned wavelength dependence of airmass factor. Although the wavelength range used is limited to 330 nm, the basic concept is related to the work in this manuscript.

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We added a description to the earlier work of Yang et al. in the end of the introduction:

“Previous attempts have been made to correct for the non-linearity of SO2 absorptions as a function of wavelength (e.g. Yang et al. 2007), especially in the case of rather high SO2 concentrations. The AMF in the standard fit range (< 320nm) strongly depends on wavelength , leading to an underestimation of the regarded SO2 amount (see also figure 7). In order to account for these effects, an iterative model approach can be applied (see Yang et al. 2009, Richter et al. 2009). As such iterative model calculations are rather time intensive, switching to an fit window at longer wavelength where the response to enhanced SO2 concentrations is more linear (respectively the AMF), is a sufficient and particularly fast method to correct for the occurring non-linear effects (Yang et al. 2007)."

* The comparisons of the results from the two fitting windows shown here are in a way unfair as no attempt was made to correct for the non-linearity in the shorter wavelength window. However, such corrections can (and are) applied, e.g. in the papers by Yang et al. and also in some of my own work. With these corrections, both the spectral retrieval and the retrieved SO2 column should be of much better quality than the “naive” column assumed here.

We agree with the reviewer and added also a figure showing SO2 VCDs (derived from the long wavelength range) for the Kasatochi eruption. These VCDs are in general in good agreement with results from Richter et al., who derived VCDs from the short wavelength range, but accounted for the non-linearity effects. We also mention this now in the text in the end of section 3.2. Nevertheless, we think it is worth pointing out that in the case of high SO2 VCDs problems for the analysis in the small wavelength range arise, for which in some cases no adequate correction could be applied. In general, the retrieved SO2 VCDs from the short wavelength range will depend much more strongly on the assumed vertical profile of the SO2 concentration. In extreme cases with very high SO2 VCDs, the observation might even become completely blind for the lower part of the SO2’plume (see also answer to referee No. 2).

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* It is a pity that the discussion of the satellite data is limited to slant columns. I think that vertical columns should be shown so that readers can compare the values to results from other retrievals and get an idea of the uncertainties involved. It is interesting to note that the region of highest maxima retrieved in the new fitting window appears to be in good agreement with results from a standard fitting window using an iterative approach to correct for the non-linearities (Richter et al., 2009).

We added an additional figure (Figure 3d) showing the VCD for the long wave fit region for our example as well as the following text at the end of the section satellite measurement example: "Figure 3d shows the corresponding VCD for the long wave UV fit region. The AMF calculation was done with the Monte-Carlo radiative transfer model McArtim, assuming the SO2 layer to be between 10 and 11km (constant SO2 concentration of 1E14 molec/cm^3 within this layer), including aerosols (aerosol optical depth: 3, asymmetry parameter: 0.68, single scattering albedo: 0.9). Additionally a cloud layer extending from 5 to 6km (OD: 20, single scattering albedo: 1, asymmetry parameter: 0.85) was included. Interestingly the VCD results are in general in good agreement with results from Richter et al. (2009), who derived VCDs from the short wavelength range, but accounted for the non-linearity effects."

* The discussion of the dilution and in particular aerosol effects in the ground-based measurements is in my opinion quite sensitive to the details of the assumptions made for the plume. The balance between light path enhancement through scattering and light path reduction through absorption will depend on plume diameter, aerosol amounts and single scattering albedo, quantities which often are difficult to estimate in the field.

We again agree with the reviewer and we added more information on these aspects to the revised version of the manuscript in section 5 Model studies:

“To obtain accurate results, it is necessary to retrieve the effective radiation path length in the volcanic plume, as the measured column density can be highly dependent on

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wavelength. Only then can the true straight column density (and therefore the average concentration) be accurately assessed. It was recently shown that the wavelength-dependent air mass factor can be retrieved from the measured SO2 optical densities between 300 and 325 nm (Kern et al 2010). Furthermore, the algorithm described by Kern et al. 2010 could be expanded to include wavelengths around 380 nm, thus increasing the information content in the model inversion."

* The use of the ratio between the two retrievals as a means to estimate aerosol effects is promising but it should be kept in mind that the airmass factor varies strongly with wavelength and this also affects the retrieved slant columns in non-trivial ways if not accounted for. Therefore the results shown using single wavelengths can only be an indication but are not quantitative. Neglecting the LOS dependence of the airmass factor potentially also introduces a bias in the calculated ratio as it will be a larger effect at long wavelengths.

We agree with the reviewer and the sentences added in response to the reviewer's comment above to the model section of our manuscript should answer this comment as well.

* Somewhere in the manuscript it should also be stated that the quality of the reference spectrum for SO2 in the longer wavelength part of the spectrum introduces some uncertainty and better reference spectra (including the temperature dependence) are needed.

We included a sentence in section 4 'discussions':" Unfortunately, the relative precision and accuracy of the SO2 absorption cross-section in the long wave UV is inferior compared to the short wave UV. Further studies are required to improve this, especially for low temperatures that occur at high latitudes ."

Corrections: p 865, l28: for of the => for the

Done.

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p 880, l6: could be could => could

Done.

* Fig. 1: Lines are nearly invisible in my printout change Residuum to residual

Figure 1 was submitted now in higher resolution

* Fig. 2: Mention GOME-2 in the figure caption and give time and location

we added to the figure caption: "...the atmospheric spectrum was taken by GOME-2 (21:25UTC, center pixel coordinates 167.88W 49.69N) on the 8th of August 2008...."

References: Richter et al., poster at the EGU2009-7679, AS3.15, XY247 (http://www.doasbremen. de/posters/egu_2009_richter.pdf) Yang, K. et al., (2007), Retrieval of large volcanic SO2 columns from the Aura Ozone Monitoring Instrument: Comparison and limitations, J. Geophy. Res., 112, D24S43, doi:10.1029/2007JD008825 Yang, K., X. Liu, N. A. Krotkov, A. J. Krueger, and S. A. Carn (2009), Estimating the altitude of volcanic sulfur dioxide plumes from space borne hyper-spectral UV measurements, Geophys. Res. Lett., 36, L10803, doi:10.1029/2009GL038025

All references were added to the revised version of the manuscript.

Please also note the supplement to this comment:
http://www.atmos-meas-tech-discuss.net/3/C640/2010/amtd-3-C640-2010-supplement.zip

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Interactive comment on Atmos. Meas. Tech. Discuss., 3, 863, 2010.

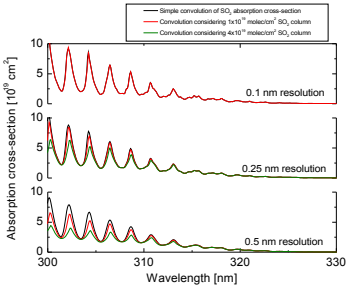

Fig. 1.

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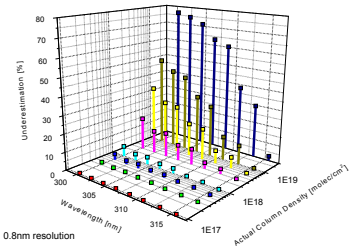

Fig. 3.1 - Magnitude of underestimation in standard DOAS measurements conducted at 0.8 nm optical resolution caused by a non-commutative cross-section convolution. The largest errors occur for high column densities S and short wavelengths λ, where the differential absorption bands of SO_2 are strongest.

Fig. 2.

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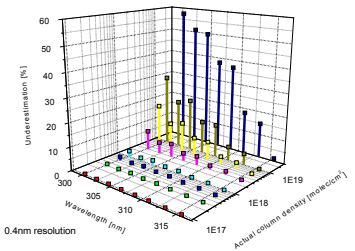

Fig. 3.2 – Magnitude of underestimation in standard DOAS measurements conducted at 0.4 nm optical resolution caused by a non-commutative cross-section convolution. Errors are slightly smaller than for 0.8 nm optical resolution (Fig. 3.1) and again occur for high column densities S and short wavelengths λ.

Fig. 3.

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