Dear Reviewer,

Thanks very much for your comments. We appreciate the time and effort taken to provide valuable comments on this manuscript. We have modified the manuscript accordingly. Below are our responses:

Major Comments

Comment_1: Page 8, lines 185-187: the dispersion model accuracy significantly decreases in the case of too low and too high wind speeds. The upper wind speed has been fixed to 8m/s. How do the results depend on this value? Did you make sensitivity tests before fixing this value to 8m/s?

Response:

Thanks very much for this comment. For high wind speed, or weak turbulence, the dispersion in the x direction is negligible in comparison with the advection (de Visscher, 2014). The dispersion model accuracy significantly decreases in the case of wind speeds < 1 m/s (de Visscher, 2014) and the critical wind speed is around 1.2 m/s. The high wind speeds may not affect the model accuracy, and we just limited our simulations to wind speeds < 8 m/s, because this range covers typical measurement conditions. We have revised the manuscript accordingly (lines 175-186). It is:

It should be noted that Briggs's equations are only suitable under the condition of x lower than 10 km. The dispersion in the wind direction is negligible in comparison with the advection when the wind speed is high, or for weak turbulence (de Visscher, 2014). In addition, the model accuracy significantly decreases in the case of wind speeds < 1 m/s (de Visscher, 2014). The critical wind speed for the Gaussian dispersion model is about 1.2m/s (de Visscher, 2014). For high wind speed, the effect of undetectable flux becomes very important (see e.g. results in Fig. 8). Thus for the general cases considered here measurements under high wind speed are not recommended. Only for very high emissions and close to the source (<1km), measurements for high wind speeds might be meaningful, but such situations might be rare. Since our study focuses on the general cases, we limit it to wind speeds < 8m/s, because in the range up to 8m/s the general dependencies become obvious. Therefore, the wind speed range in our simulation is between 1.2 m/s and 8 m/s. The distance in our

Comment_2: Page 9, lines 202-203: From where those reaction rate values come from? Literature source(s) should be added here.

Response:

Thanks for this suggestion. The reaction rate values are from de Visscher, 2014. We have added this literature source accordingly.

Comment_3: Page 12, lines 273-275: For low plume heights, the SCD could be assumed to be equal to the VCD. Did you determine from which plume height this assumption is not valid anymore?

Response:

Thanks very much for this very important comment. We have investigated the dependence of the AMF on altitude via 3D RTM simulations. The results indicate that for a plume height around 250m, the AMF is typically between 1.05 and 1.3. The higher values are for high Aerosol load and high SZA (here only measurements below 75 ° are considered), the lower values are for low aerosol load and low SZA. That means that for our simulations, the VCDs are not exactly the same as the measured SCDs. However, the AMFs are still smaller than for measurements of horizontally extended plumes. For layer heights below 50m, the AMF is around 1.03 and the AMF error can be neglected. We therefore add the error of the AMF to the total error calculation and also add some discussions on AMF error (lines 344-354):

VCDs are derived from SCDs applying AMF. We calculated AMFs using the Monte Carlo atmospheric radiative transfer model McArtim (Deutschmann et al., 2011). For that purpose, we calculated 3-D box-AMF for different aerosol loads and solar zenith angle (SZA). It should be noted that the application of 3-D box-AMF (in contrast to 1-D box-AMF) is important for the measurements considered in our study, because horizontal extension of the plumes perpendicular to the wind direction is rather short (compared to the average horizontal photon path lengths). Our simulations indicate that, for a plume height around 250m, the AMF is typically between 1.05 and 1.3. The higher values are for high Aerosol load and high SZA (here only measurements below 75 ° are considered), the lower values are for low aerosol load and low SZA. $\pm 10\%$. For layer heights below 50m, the AMF is around 1.03 and the AMF error can be neglected.

We also added an error analysis on the AMF error from lines 646 to 667:

4.7 AMF error

<u>AMF values depends on plume height, SZA and aerosol optical density (AOD) as shown in Figure</u> <u>16. For plume heights < 50m, the AMF is around 1.03 and its error can be neglected. For plume</u> <u>heights \leq 250m, the AMF error is about \pm 10%. Since the plume height in our study is about</u> <u>250m, the contribution from the AMF error has to be taken into account.</u>

Since VCDs are derived from SCDs by dividing the AMF, then AMF errors introduce VCD errors, which furthermore contribute to the emission flux errors. Wind speed uncertainty is the main error source when close to the source. With larger wind speed, the relative error of the wind speed becomes smaller which then also contributes less to the flux error. This indicates that the flux error that results from other error sources, such as the AMF error, have larger relative contributions under larger wind speed. Figure 17 presents R_{AMF}^2 and the total relative errors for wind speeds of 1.2 m/s and 8 m/s. From Figure 17 we could see that R_{AMF}^2 for SO₂ under the speed of 1.2 m/s is very small while it becomes larger at the speed of 8m/s, even near 0.5 when near the source. The NOx flux error, however, is less affected by the AMF error for $R_{AMF}^2 < 0.1$.



Figure 16. 3D Box-AMF dependence on plume height, SZA and aerosol optical density (AOD) for 310nm and 430nm. For the aerosols a box profile between the surface and 1km was assumed.



Figure 17. NOx and SO₂ total relative error, R_{AMF}^2 of AMF error under different wind speed (Q = 100 g/s, s=20m).

Added Reference:

Deutschmann, T.; Beirle, S.; Frieß, U.; Grzegorski, M.; Kern, C.,Kritten, L.; Platt, U.; Pra-dos-Roman, C.; Pukite, J.; Wagner, T.; Werner, B.; Pfeilsticker, K., The Monte Carlo at-mospheric radiative transfer model McArtim: introduction and validation of Jacobians and 3-D features, J. Quant. Spectrosc. Ra., 2011, 112, 1119–1137.

Vandaele, A. C., Simon P. C., Guilmot, J. M. Carleer, M., Colin, R.: SO₂ absorption cross section measurement in the UV using a Fourier transform spectrometer, J. Geophys. Res., 99, D12, https://doi.org/10.1029/94JD02187, 1994.

Vandaele, A.C., Hermans, C., Simon, P.C., Carleer, M., Colin, R., Fally, S., Mérienne, M.F., Jenouvrier, A., Coquart, B.: Measurements of the NO₂ absorption cross-section from 42 000 cm-1 to 10000 cm-1 (238–1000 nm) at 220 K and 294 K, J. Quanr. .Spectrosc. Radior. Transfer, 59,171-184, DOI: 10.1016/S0022-4073(97)00168-4, 1998.

Comment_4: Page 14, lines 308-309: Typical errors on NO₂ and SO₂ VCDs coming from previous studies are mentioned here and are used as is in the present work. These studies should be cited. Are the conditions assumed in those studies similar to the ones assumed by the authors, i.e. that SCD=~VCD and therefore the AMF error can be neglected?

Response:

Thanks for this comment. We have cited those studies accordingly and please refer to our response

to major comment_5. With respect to the AMF calculation and application, our study is partly different from previous studies. Some of previous studies measured more extended plumes, for which 'traditional' 1D-AMF calculations are appropriate. Since our study focusses on point source emissions, such 1D-simulations are not adequate. Instead, 3D-simulations taking into account the limited horizontal extent of the plume should be used. To our knowledge, such 3D-AMF simulations are applied for the first time in our study to point source emission measurements. In the revised version of the manuscript, we take the AMF value and AMF error into consideration. Please refer to our response to major comment_3.

Comment_5: Page 15, Table 4: the detection limits for NO₂ and SO₂ SCDs are assumed to be two times the corresponding retrieval errors. This should be justified.

Response:

Thanks for this comment. SCD retrieval errors are quantified by the fitting error (1 σ error), and the mean detection limit is set to 2 σ error (see e.g. Alicke et al., 2002,). We have revised the text accordingly (lines 333-343):

The SCD error can mainly be attributed to the DOAS fitting error of the SCD and the trace gas absorption cross-section error. Previous studies have indicated that the typical fit errors of NO₂ and SO₂ SCDs are $\pm (1\sim4) \times 10^{15}$ molecules cm⁻² and $\pm (1\sim6) \times 10^{15}$ molecules cm⁻², respectively (Wagner et al., 2011; Wang et al., 2014; Wu et al., 2018; Davis et al., 2019). Thus in this study, we set the fit error of NO₂ and SO₂ to be $\pm 2.5 \times 10^{15}$ molecules cm⁻² and $\pm 4 \times 10^{15}$ molecules cm⁻² (1 σ error), respectively. Here in addition, we use the 2σ values as detection limit (see e.g. Alicke et al., 2002; Platt and Stutz, 2008). The absorption cross-section errors are less than 3% for NO₂ and less than 2.4% for SO₂ (Vandaele et al., 1994, 1998). In this study, we set the total SCD error from gas absorption cross-section errors to 5% (Theys, et al., 2007) for both NO₂ and SO₂. Of course, these values are only rough estimates, but they are useful to investigate the general dependencies of the total flux error.

Added Reference:

Alicke,B., Platt, U., Stutz, J.: Impact of nitrous acid photolysis on the total hydroxyl radical budget during the Limitation of Oxidant Production/Pianura Padana Produzione di Ozono study in Milan, J. Geophys. Res., 107, NO. D22, 8196, doi:10.1029/2000JD000075, 2002.

Davis, Z. Y. W., Baray, S., McLinden, C. A., Khanbabakhani, A., Fujs, W., Csukat, C., Debosz, J. and McLaren, R.: Estimation of NOx and SO₂ emissions from Sarnia, Ontario, using a mobile MAX-DOAS (Multi-AXis Differential Optical Absorption Spectroscopy) and a NOx analyzer, Atmos. Chem. Phys., 19, 13871–13889, https://doi.org/10.5194/acp-19-13871-2019, 2019.

Wang, T., Hendrick, F., Wang, P., Tang, G., Clémer, K., Yu,H., , Fayt C., Hermans, C., Gielen, C., Müller, J.-F., Pinardi, G., Theys, N., Brenot, H., Roozendael, M. Van.: Evaluation of tropospheric SO2 retrieved from MAX-DOAS measurements in Xianghe, China, Atmos. Chem. Phys., 14, 11149–11164, doi:10.5194/acp-14-11149-2014, 2014.

Comment_6: Pages 27-28, Section 4.6: If we want to use several scans to reduce the flux error, the elapsed time between two scans at the same distance from the source is then also an important parameter. I think this point should be at least briefly discussed here.

Response:

Thanks for this comment. We have added a brief discussion at the beginning of section 4.8 (lines 670-677):

In our experiments, we only simulated a single scan of the plume by the mobile DOAS at each specific distance. In reality, we usually scan the plume cross-section several times in order to reduce the flux error. The elapsed time between two scans at the same distance from the source is then also an important parameter. The more of the elapsed time, the greater the uncertainties due to temporal variations of the flux and/or the wind fields are likely to be. Here, we assumed that the elapsed time is small and its influence can thus be neglected in our simulation. Figure 18 displays the simulation example of NOx and SO₂ flux error under different measurement times.

Technical corrections

Comment_1: The abstract is much too long. Only the most relevant findings should be reported here. **Response:** Thanks for this suggestion. We have changed accordingly:

Mobile differential optical absorption spectroscopy (mobile DOAS) has become an important tool for the quantification of emission sources, including point sources (e.g., individual power plants) and area emitters (e.g., entire cities). In this study, we focused on the error budget of mobile DOAS measurements from point sources, and we also offered recommendations for the optimum settings of such measurements via a simulation with modified Gaussian plume model. Following the analysis, we conclude that: (1) The proper sampling resolution should be between 5 m and 50 m. (2) When measuring far from the source, undetectable flux (measured SCDs are under the detection limit) resulting by wind dispersion is the main error source. The threshold for the undetectable flux can be lowered by larger integration time. When measuring close to the source, low sampling frequency results in large errors and wind field uncertainty becomes the main error source of SO₂ flux (for NOx this error also increases, but other error sources dominate). More measurements times can lower the flux error that results from wind field uncertainty. The proper wind speed for mobile DOAS measurements is between 1 m/s and 4 m/s. (3) The remaining errors by $[NOx]/[NO_2]$ ratio correction can be significant when measuring very close. To minimize the [NOx]/[NO₂] ratio correction error, we recommend minimum distances from the source, at which 5% of the NO₂ maximum reaction rate is reached and thus NOx steady-state can be assumed. (4) Our study suggests that emission rates <30 g/s for NOx and < 50 g/s for SO₂ are not recommended for mobile DOAS measurements.

Based on the model simulations our study indicates that mobile DOAS measurements are a very well suited tool to quantify point source emissions. The results of our sensitivity studies are important to make optimum use of such measurements.

Comment_2: Pages 3-4, lines 79-82: first letter of the first name should be removed in all the references that appear on these lines.

Response: Thanks for this correction. We have removed the first letter of the first name accordingly.

Comment_3: Figure 2: Point 4 in the list of dispersion simulation parameters: 'Disersion' -> 'Dispersion'

Response: Thanks for this correction. We have revised the word.

Comment_4: Page 7, line 164: Is there a reference for Pasquill and Gifford? If, yes it should be included in the list of reference.

Response:

Thanks for this suggestion. The *Pasquill and Gifford* dispersion parameters calculated using Briggs's (1973) formulas are also from de Visscher, 2014. We have cited this reference accordingly.

Comment_5: Page 8, line 186: maybe 'high' is preferable than 'strong'?

Response: Thanks for this suggestion. We have changed the word 'high' in the manuscript accordingly.

Thank you for taking care of our manuscript.

Kind regards,

Yeyuan Huang, on behalf of all authors.