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Interactive comment on "Airborne multi-axis DOAS measurements of tropospheric SO_2 plumes in the Po-valley, Italy" *by* P. Wang et al.

Anonymous Referee #2

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General comments

The manuscript entitled "Airborne multi-axis DOAS measurements of tropospheric SO_2 plumes in the Po-valley, Italy" by Wang et al. describes airborne multi-axis DOAS measurements of SO_2 during the FORMAT campaign in 2003. In particular, an estimate for the SO_2 emissions from a power plant as well as for the vertical column densities over a city are provided.

These are, to my knowledge, the first measurements of SO_2 by airborne MAX-DOAS, a measurement technique which offers the opportunity to determine not only information on the vertical distribution of atmospheric trace gases but also on emission rates by observing SO_2 slant column densities (SCDs) in a cross section of an emission plume. The paper therefore presents a novel concept, which addresses measurements of a trace gas with an important impact on atmospheric chemistry, air quality and - due to the potential acidification of rain droplets - the aquatic ecosystem. The analysis and interpretation of the MAX-DOAS measurements uses data from various sources obtained during the FORMAT campaign, such as SO_2 in situ measurements, satellite borne measurements of aerosols, and airborne measurements of the boundary layer height, in a synergistic way. The paper is well structured and (as far as I can judge) written in a good English. I recommend the publication of this paper in ACP after some modifications as detailed below.

The detection of SO_2 using scattered sunlight is quite challenging, in particular due to the low light intensities in the near UV (below ≈ 330nm) and, in case of observations pointing towards the ground, the low surface albedo at these wavelengths, as well as the relatively small SO_2 optical depth from industrial emissions. A detailed description of the SO_2 retrieval is therefore essential. However, I feel that the authors do not provide an appropriate discussion of the spectral analysis, including potential (systematic and random) errors, typical signal to noise ratio, and detection limits of the SO_2 retrieval. In particular, it would be interesting to provide the errors of the SO_2 SCDs for both nadir and zenith (and also the other viewing directions).

An accurate determination of airmass factors (AMFs) is crucial for the interpretation of MAX-DOAS measurements. In particular, an estimate of the aerosol extinction profile is required for the accurate modelling of the radiative transfer. The authors have demonstrated elsewhere (Wang et al., Measurements of tropospheric NO_2 with an airborne multi-axis DOAS instrument, ACP, 2005) that MAX- DOAS measurements of the oxygen dimer (O_4) provide significant information on atmospheric aerosols and clouds, and that the information on aerosols gained from O_4 measurements can serve as an input for the modelling of trace gas AMFs. The authors state that the same approach has been used for the determination of the aerosol optical depth for the SO_2 measurements

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(by using the O_4 absorption band centered around 360nm?), but do not provide any further details. It would be very informative for the reader to show and discuss a plot of the O_4 vertical column densities from different viewing directions during the flight, which would confirm the assumptions made on the aerosol properties, in particular since (as the authors state) it is possible that a higher amount of aerosols (potentially with different optical properties) is present within the exhaust plume.

Multi-axis DOAS allows to gain information on the vertical distribution of atmospheric trace gases, or at least to confirm assumptions made on the trace gas profile, by combining measurements performed along different lines of sight. A major weakness of the paper is the fact that the estimation of SO_2 emissions from the power plant is based on vertical columns from zenith sky measurements only. Although the signal to noise ratio might be best for zenith sky measurements, the problem is that these measurements are very insensitive to the partial column of SO_2 below the aircraft (as the authors mention in the discussion of the measurements at the city of Mantova). This means that even strong variations of the SO_2 concentration below the flight altitude should have only a very small impact on the SO_2 SCD measured in zenith, and this yields a large uncertainty in the derived VCDs from zenith sky measurements only. It is mentioned in the manuscript that the VCDs from other viewing directions qualitatively agree, but only zenith and nadir SO_2 VCDs are shown (Figure 8). I strongly suggest to show the VCDs derived from all available viewing directions. Although an agreement of the VCD from different lines of sight can perhaps not be expected due to the horizontal inhomogeneity of the SO_2 concentration within the exhaust plume, at least the integrated VCD along the cross section through the plume (or the respective emission rates) should agree for measurements along different lines of sight. This could confirm that the assumptions made on the vertical distribution of SO_2 (as well as on aerosols) are valid.

Specific comments

P2021, L3: It would be interesting to know the horizontal resolution of the measurements. Which horizontal distance corresponds to measurements performed in 1 min

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time intervals?

Section 3.1: The spectral analysis should be discussed in much more detail, in particular regarding the error budget for the SO_2 SCDs from different lines of sight (see general comments).

Section 3.2: I suggest to include a figure showing the O_4 VCDs from different viewing directions based on the assumptions for the aerosol scenario (see general comments).

P2022, L1: Why is a maritime aerosol used for the AMF calculations at the exhaust plume although there is a northerly wind during the measurements (which means that the air mainly comes from the continent rather than from the sea) and a significant fraction of the aerosols might be directly emitted by the power plant?

P2024, L9ff, and Figure 4: On Sept. 27, the SO_2 SCD is lower at 83° than at 97°, but one would expect the converse for geometrical reasons. Have you got any explanation for this feature?

P2024, L16: Why should the wind speed have an impact on the SO_2 SCDs measured downwind, or on the emission rates? Do you suggest that higher wind speeds cause a stronger mixing/dilution of SO_2? This would cause smaller SCDs, but would have no impact on the emission rates.

P2024, L23: Are the large variations in the background real or is this variability in SO_2 SCDs caused by random errors?

P2025, L24: It is mentioned that the plume was displaced relative to the local wind direction, and this has been attributed to the large error in measured wind direction. However, this discrepancy could also be explained by the fact that the local wind direction observed on the airplane is not necessarily equal to the average wind direction between the source and the location of the measurement.

P2026, L27ff: Although the signal to noise ratio is probably best in zenith, zenith sky measurements are very insensitive to SO_2 below the aircraft. Therefore measurements from other viewing directions should be used for the estimation of the emission flux as well (see general comments).

P2027, L7ff: It would be useful to add the VCDs from the other viewing directions to Fig. 8 to illustrate that they are in agreement with nadir and zenith VCDs. Also, flux estimates should be given for all available viewing directions (see general comments).

P2027, L24ff: Multiplying the half width of a function with its peak value does not necessarily yield the integral. Why is the integral not calculated using, for example, $\sum_i VC_i \cdot \Delta t_i$ with Δt_i being the acquisition time for the measurement of VC_i? The factor $\cos(\theta)$ ($\sin(\theta)$??) from Eq. 1 is not mentioned in the description of the flux calculation. Furthermore, it seems that the airplane was flying in a curve through the plume on 26. Sept. (Fig. 6). Do you account for the varying flight direction while crossing the plume?

P2028, L19ff: It is mentioned that the fact that SO_2 SCDs are measured relative to the background could cause a systematic error in emission rates. But isn't the increase in SO_2 relative to the background the quantity that is directly linked to the emission?

P2029, L1: The detection limit is mentioned, but it is neither defined nor quantified anywhere else in the paper. Please add this information to your 'Data Analysis' section.

P2029, L2ff: It is not mentioned which viewing direction is used for the determination of the SO_2 VCD at the city of Mantova. As for the measurements of the Porto Tolle plume (see my comments above), I would strongly suggest to show the VCDs derived from all downward viewing directions in order to provide evidence for an SO_2 layer height of 500m (or to provide a better estimate of the SO_2 layer height).

P2030, L2: It is mentioned that 'The off-axis data of the AMAXDOAS measurements proved to be useful to determine plume altitudes'. I can't see how this has been done. For the Porto Tolle measurements you assume that SO_2 is uniformly mixed in the boundary layer, with the boundary layer height determined using data from other in-

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struments. And for the SO_2 layer height above the city of Mantova you only give an upper limit for the layer height of 500m based on the fact that there is no SO_2 detected above the flight altitude. As far as I can see, lower SO_2 layer heights are also possible. As already mentioned several times above, these assumptions could be easily validated by converting the slant columns from all available viewing directions to vertical columns which should be similar if the assumptions on the vertical distribution of SO_2 (and aerosols) are realistic.

P2030, L25: It is mentioned that the SO_2 measurements '... could be improved ... by optimising the spectrometer for the SO_2 retrieval', but it is not stated how this can be done and why the instrument was not optimal during the FORMAT campaign.

Technical corrections

Please homogenise the spelling regarding U.S./U.K. English. Example: 'center' on L3 and 'centre' on L9 of P2026.

P2018, L5: Replace 'sun-light' with 'sunlight'.

P2019, L8: Replace 'air-borne' with 'airborne'.

P2019, L10: 'Remote sensing measurements of ... have been performed using TOMS .... measurements': delete second 'measurements'.

P2023, L2: According to the definition of θ as the angle between flight direction and wind direction, it should be $\sin(\theta)$ rather than $\cos(\theta)$ in Equation 1.

P2026, L20: Replace "higher altitude" with "higher altitudes".

P2028, L4: What is 'N' standing for in the unit mg Nm^{-3}?

P2029, L10: Replace 'or' with 'for'.

Figures 4 and 5: It would be useful for the reader if the locations discussed in the text (Cremona, Mantova, Porto Tolle) would be highlighted in the graphs of the SO_2 and

NO_2 time series.

Figure 6: For which viewing direction are the SO_2 SCDs shown?

Figure 9: From which viewing direction are the SO_2 VCDs derived?

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