Atmos. Chem. Phys. Discuss., 5, S4782–S4788, 2005
www.atmos-chem-phys.org/acpd/5/S4782/
European Geosciences Union


Atmospheric Chemistry and Physics Discussions



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

**P. Wang et al.**

Received and published: 29 December 2005

The authors would like to thank the referee for the detailed review. Below, we have replied to the comments point by point, and the corresponding changes have been incorporated in the revised version of the paper.

GENERAL COMMENTS The paper shows that AMAXDOAS can be used for quantifying SO2 emission from known sources. The emission estimates still have large uncertainties, but the authors argue that this can be improved by better knowledge on the local meteorological situation.I agree, and therefore consider it a pity that the authors didn't try or didn't succeed in retrieving the appropriate information from the local meteorological institute on wind direction, wind speed, and boundary layer characteristics (see specific comments 2 and 10). The paper is generally well written, and with

good level of detail, except for some points, which are specified below.

SPECIFIC COMMENTS

1) The applied cross sections are listed in Section 3.1. Some of them have quite low spectral resolution. The authors should explain how they dealt with the differences in resolution of the AMAXDOAS (please specify) and the laboratory cross sections. Wrong use is a potential source of errors in the SO2 slant column.

The resolution of the AMAXDOAS instrument is 0.8 nm in the UV spectral region. The GOME cross-sections used for O3 and NO2 have a significantly better spectral resolution (0.2 nm) and in addition have been deconvoluted with the well characterized slit function of the GOME instrument. The SO2 cross-section of Vandaele is a high resolution spectrum, as is the HCHO cross-section of Meller and Moortgat. All cross-sections have been convoluted with the AMAXDOAS instrument function as determined from line lamp measurements before using them in the fit. This information has now been added to the text.

2) The uncertainty on the estimate of the power plant SO2 emission is argued to be approximately 50%, dominated by the uncertainty in wind speed (2 m/s) and direction (30_). However, a few other (possibly large) error sources should also be considered: * The assumption that the SO2 is well mixed from 0 to 1.7 km is not well founded, and the error budget is very sensitive to this assumption. The horizontal distance of the location of the measurement to the power plant might not be large enough for the plume to have reached either the ground or the top of the boundary layer. A more confined plume would change the AMF considerably. For instance, if the SO2 plume at 5 km distance reached up to 1.3 km instead of 1.7 km, the AMF would decrease and the emission estimate would increase with approximately 20%.

We agree with the reviewer that the SO2 AMFs are very sensitive to the vertical SO2 profile. Our assumption that the SO2 is well mixed below 1.7 km is based on measurements from the Ultralight aircraft. These measurements were taken on the same



ACPD

5, S4782–S4788, 2005

day, at the west of the power plant. However in the revised version of the paper we used the boundary layer height from ECMWF data. According to the ECMWF data (0.5 x0.5 degree resolution), the boundary layer height at 45_N and 12.5 E at 10.5 UT is about 1 km. We recalculated the SO2 AMFs and flux, assuming SO2 is well-mixed in boundary layer. As pointed out in the text, the high values of the ground-based SO2 measurements at Scardovari taken on 26 Sep. confirm that the SO2 plume has reached the ground. We therefore believe that the assumption of a well mixed SO2 plume is reasonable in this case.

* The vertical wind profile in the plume is not discussed. Both the direction and the wind speed can have large variability. Some examples from the ECMWF analysis at 45N, 12E, 12 UT: the wind speed on 27 September 2003 was 1.7 m/s at 250 m and 10.4 m/s at 600 m, the wind direction on 26 September 2003 was 57_ (ENE to WSW) at 250 m and 295_ (WNW to ESE) at 600 m.

We agree with the reviewer that wind speed and direction change with altitude, and that these changes potentially have a large impact on the derived flux. Therefore we now used the ECMWF vertical wind profile data at 12.5 E and 45.0N, and interpolated them to the measurement time, 10.5 UT. On 26 September the wind speed and direction were quite stable, facilitating consistent results. On 27 September the (model) wind had large variations both in vertical and horizontal directions, leading to unreliable results. Detailed information can be found in the revised paper.

3) Section 4.1, page 2023, line 22: 'roughly well mixed below 1.5-1.8km is too vague. Use figure or table to quantify. Where does 1.7 km come from?

The NO2 and HCHO profiles measured by the Ultralight aircraft show that these gases are well-mixed below 1.5 km, but the measurements were taken close to Milan, at the west of Porto Tolle. As pointed out above, in the revised paper we used the ECMWF wind vertical profile data and the boundary layer height at 45N 12.5E, which is very close to the power plant Porto Tolle. According to the new data the boundary layer



ACPD

5, S4782–S4788, 2005

height is 1 km. We revised the paper with the new data.

4) Page 2024, line 5: 'The error of the fit was about 12%.' Is this the error in SO2 slant column? This seems to contradict page 2028, line 22: 'The fit error ... was between 15-50%'

The number given is the error in SO2 slant column. The reviewer is right, that the range quoted was not correct and we changed change it from 15-50% to 12 -50%.

5) Page 2024, line 16: replace 'measured' by 'detected'.

Changed as suggested

6) Page 2024, line 17-19: remove: "which indicates that ... was also similar." This is the subject of the paper and calculated later on. This handwaving argument is out of place.

Agreed and changed as requested.

7) Page 2024, line 28-29: 'should be well correlated ... much higher accuracies': give values and references.

Here, we use the spatial correlation between NO2 and SO2 slant columns to show that the SO2 plumes are in deed from the power plant. As SO2 and NO2 are emitted from the same stack we should measure both NO2 and SO2. However, the error of the NO2 slant columns is only about 2% and therefore the NO2 columns can be used to check the consistency of the SO2 data. This is now explained in more detail in the paper.

8) Page 2025, line 1-2: replace 'which is completely ... the SO2 fit.' by 'which does not overlap the SO2 fitting window.'

Changed as suggested

9) Page 2025, line 5: replace 'measured' by 'detected'.

Changed as suggested



**ACPD**

5, S4782–S4788, 2005

10) Section 4.2: see also comment 2 above. Use a simple plume model and more realistic weather conditions, e.g. obtained from a local meteorological service: vertical profiles of wind speed and direction, and other parameters that should be input to the plume model, like potential temperature. Formula (1) should then include an integral over the vertical.

We agree with the reviewer that a more detailed treatment of plume development would reduce the uncertainty in the flux estimates. In the revised paper, we used the ECMWF vertical wind profile data at 12.5 E and 45 N, with 0.5 x 0.5 degree resolution, which is the best data we could get. In the flux calculation we included the integral over the vertical wind profile in the revised paper. Incorporation of a plume model is a reasonable suggestion but seemed to be out of scope for this first case study.

11) Page 2027, line 24-27: Why not use formula (1), instead of this approximation?

We calculated the flux with formula (1). The approximation was meant to explain how to do it. As the explanation seems to be misleading, we have deleted it.

12) Section 4.3: No error estimates are given for the calculation of the SO2 pollution near Mantova. Also the vertical variability of wind direction and speed in not taken into account.

At Mantova we didn't give the SO2 flux, so no wind information was needed. The error in the SO2 slant column (13-30%) was added to the text.

13) Conclusions: "The off-axis data ... proved to be useful to determine plume altitudes". This is formulated too strong; the only thing that is concluded is that the SO2 near the city must be below 500 meter, because otherwise it would have been detected in the upward viewing directions. A lower boundary could not be given. For the power plant one could conclude that SO2 is both above and below 600 m, not that it is "well mixed in the boundary layer". It is likely that the combination of all viewing angles, together with the radiative transfer model can give more information on the vertical



**ACPD**

5, S4782–S4788, 2005

distribution. If so, this should be exploited in this paper.

A full inversion of the SO2 measurements is hampered by the relatively large uncertainty of the individual measurements and also by horizontal inhomogeneities which would have to be derived using a tomographic approach. We therefore focused on the use of zenith and nadir measurements, employing the off-axis data mainly to derive qualitative information on the vertical distribution. In response to the reviewers comment, we have re-formulated the corresponding section in the conclusions.

14) Page 2030, line 17-19: "The advantage of ... establish the emissions." This is not completely true. As suggested in comments 2 and 10, the uncertainty can probably be much reduced with a simple plume model and realistic meteorological conditions. The vertical extend of the plume will be determined by the wind speed, the distance to the source, and the turbulence.

We agree with the reviewer that the uncertainties in the vertical profiles of wind speed, wind direction and SO2 are the main error sources. Better knowledge of these parameters will reduce the uncertainties. Therefore we now use the ECMWF vertical wind profile, at 12.5 E 45N, interpolated to measurement time 10.5 UT.

15) MODIS teams are acknowledged, but I missed where the MODIS data is used in the text, except for a reference to literature.

MODIS aerosol optical thickness was used to check the aerosol settings in the AMF calculation.

16) Figure 3: The dotted line is referred to as 'SO2 fit'. It is however the measured signal after subtraction of the fit to the other absorbers. Therefore I would prefer a term like 'SO2 residual' or something similar. In the caption: " the dotted line is the measured spectrum after subtraction of the fit to the other absorbers.

The figure caption (and legend) have been changed as requested.

17) Figure 5: Include latitude and longitude of the mentioned airport.



**ACPD**

5, S4782–S4788, 2005

Changed as suggested

TECHNICAL CORRECTIONS * Formula (1) should have 'sin' instead of 'cos'. * Page 2024, line 5: 90 km instead of 100 km * Page 2027, line 25: change 'the half with' to 'the half width'.

Corrected in the paper

---

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 2017, 2005.