

Response to comments #2:

Thanks very much for your comments, suggestions and recommendation with respect to publish this paper in AMT. Our response to all your comments are as follows.

The paper under consideration describes a useful extension of current filter radiometer approaches for the measurement of SO₂ concentrations. It fits well into the scope of AMT. I recommend publication of this work after minor revisions. Although the demonstrated method is obviously a major improvement over the linearized retrieval, I wonder about some choices made by the investigators which still seem suboptimal, perhaps the authors could comment on these items in the finalized version:

1. What is the value of assuming a Gaussian filter transmission? The filter characteristics can (and have been, as shown in figure 7) be measured with spectrometer and the transmission functions can be used as measured for calculating effective band-integrated absorptions from spectrally resolved calculations.

Response: Yes, the filter characteristics can be measured with spectrometer and the transmission functions can be used as measured for calculating effective band-integrated absorptions from spectrally resolved calculations. But before constructing an instrument, there are no filters, we have to perform some theoretical calculation, deduce the reasonable filter parameters, and then purchase filters based on the theoretical deduction. The theoretical calculation is very important because it guides us what kinds of filters should be used or purchased. In this study, the filter parameters calculation in section 3 is the theoretical deduction before constructing a instrument. Figure 7 is the actual filter transmission of purchased filters according to the theoretical parameters.

In detail:

Step 1: We want to construct an instrument that can measure industrial SO₂ emissions from ~ ppmv level to more than 10000 ppmv level. At this step we have no filters and just propose a measurement requirement.

Step 2: We theoretically deduce the filter parameters of all channels based on the range requirements, Lambert-Beer's law and some known parameters, e.g., light path, gases absorption parameters of SO₂ and all the interfering gases, etc. This step is filter parameters calculation in section 3. At this step we still have no filters, some reasonable assumptions based on previous experience have to be used. For example, backed by our experience, filter is approximated as a Gaussian function with a maximum transmission of 75%.

Step 3: We purchase all filters based on the theoretical deduction in step 2. At this step we should tell the manufacturer about what kinds of filters we want to buy. The filter parameters(center wavelengths, bandwidths and transmissions.) are based on step 2.

Step 4: We construct the instrument using all filters. Perform the real test and measurements with the optimized algorithm. To make sure the constructed instrument fulfill the proposed requirements, reliable enough to monitor SO₂ emissions in various industrial applications.

Based on above analysis, we can answer your comments briefly as:

- a). We assume a Gaussian filter transmission because we don't have filters before constructing an instrument. Gaussian assumption is backed by experience which shows that a Gaussian function form is closer to the actual transmission function of a filter.
- b). We don't use the measured filter characteristics to calculate the effective band-integrated absorptions after constructing an instrument because we have a more easier and effective method to retrieve the concentrations, i.e., using a third order polynomial to fit the concentration-absorbance relationship. We don't need to repeat the calculation task. I will answer why using ad-hoc third order polynomial fits to the optical depth is better in the following comment(see response to comment #2).

For avoiding misleading, I will add the following sentence at the beginning of section 3 in AMT version: The filter parameter and absorption characteristic of each channel should be known before constructing an instrument. We theoretically deduce the filter parameters of all channels based on the range requirements, Lambert-Beer's law and some known parameters, e.g., light path, gases absorption parameters, etc.

2. Instead of using ad-hoc third order polynomial fits to the optical depth the computational performance of even a moderate computer certainly is sufficient today for treating the whole problem numerically (convolution of measured filter transmissions with monochromatic cross-sections) using pre-calculated cross-sections for all gases involved. This numerical approach based on a physical model is a standard for the analysis of laboratory and atmospheric spectra taken with higher spectral resolution, and would probably also in the case of filter radiometer measurements result in an improved convergence behavior and an improved ability of diagnosing limiting factors of the instrumental and retrieval setup, thereby finally ensuring an optimal reconstruction of the concentrations of the various absorbers involved.

Response: In principle, two different types of optical techniques can be used for gases analysis. One is the dispersive technique, e.g., Differential optical absorption spectroscopy (DOAS) technique, Fourier transform infrared spectroscopy (FTIR) technique and Tunable diode laser absorption spectroscopy (TDLAS) technique, etc. The other one is the non-dispersive technique, e.g., Non-Dispersive Infra-Red(NDIR) technique and Non-Dispersive Ultra-Violet (NDUV) technique. In this study, NDIR technique is used. The dispersive technique can obtain the spectral absorption structure of each waveband (or channel) with a spectrometer. So the dispersive technique can retrieve the concentration of each gas by including all gases cross sections into fitting. Thus, your mentioned numerical approach based on a physical model is a standard for the analysis of laboratory and atmospheric spectra taken with higher spectral resolution. However, the non-dispersive technique don't use any spectrometer but a detector (this configuration can save the cost). So the non-dispersive technique can not obtain the spectral absorption structure of each waveband but total optical attenuations of each waveband (or channel). In order to retrieve the concentration of each gases, calibration curves have to be used, i.e., using a third order polynomial to fit the concentration-absorbance relationship (please

check the analysis in section 5.1 for details). For an example, the fitted calibration curve of SO₂(L) in this study is

$$f_{SO_2(L)}^{cal.}(\tau) = d_0 \times f_{SO_2(L)}(\tau) + j_0 \quad (1)$$

$$f_{SO_2(L)}(\tau) = 5.40386 + 94.58219 \cdot \tau + 44.59494 \cdot \tau^2 + 11.43462 \cdot \tau^3 \quad (2)$$

The nomenclature of above equations are presented in section 5.1. If the pure absorbance of this channel (this can be obtained by solving the interfering equations in Fig.3) is 0.3, substituting 0.3 into above equation (1) results in SO₂ concentration.

Basically, dispersive technique is more sensitive than non-dispersive technique. But dispersive technique is not an optimal selection for continuous emissions monitoring systems (CEMS) because of expensive production and maintenance cost, especially for multi-gases analysis. In contrast to dispersive technique, a NDIR analyzer has advantages of simple structure, wide operation spectral region and multi-gas analysis with low cost and good durability. It is an ideal tool for CEMS equipped in industrial facilities. The CEMS often exhibits high gases concentrations that stability and accuracy is more important than sensitivity.

Though the non-dispersive technique can not obtain the spectral absorption structure, it do fulfill Lambert-Beer's law, which was used to theoretically deduce the filter parameters of all channels in section 3 in this study.

3. Minor comments: It would be useful for the reader to add additional plates to figure 2, showing the absorption bands of all absorbers involved. Figure 9 indicates a systematic difference between the SO₂ retrievals based on either the strong band (low values of SO₂, bias of 2%) or the weak band (high values of SO₂, bias of 1.2%). Are there possible explanations for this finding? Figures 13 and 17 indicate that the discrepancy between the DOAS and the filter radiometer is not due to a random scatter, but periods with excellent agreement and periods during which a larger bias seems to prevail seem to alternate. Are there explanations for this finding (perhaps related with certain measurement conditions)?

a). It would be useful for the reader to add additional plates to figure 2, showing the absorption bands of all absorbers involved.

Response: In AMT version, we will add an additional plates in figure 2, showing the absorption bands of all absorbers involved.

b). Figure 9 indicates a systematic difference between the SO₂ retrievals based on either the strong band (low values of SO₂, bias of 2%) or the weak band (high values of SO₂, bias of 1.2%). Are there possible explanations for this finding?

Response: Apparently, Figure 9 indicates a systematic overestimation for all measurements. Actually, this is not real. The definitions of measurement bias and relative measurement error as shown in equation (6) indicate that the biases and relative measurement errors are absolute values, i.e., always larger than zero.

We intend to do this to show the absolute deviation level at different SO₂

concentration levels. To avoid this misleading, we will state that the biases and relative measurement errors in section 5.3 are absolute values, e.g., changing bias to absolute bias, measurement error to absolute measurement error.

c). Figures 13 and 17 indicate that the discrepancy between the DOAS and the filter radiometer is not due to a random scatter, but periods with excellent agreement and periods during which a larger bias seems to prevail seem to alternate. Are there explanations for this finding (perhaps related with certain measurement conditions)?

Response: Yes, this phenomenon related with certain measurement conditions. The volume of the sample cell in the multi-channel analyzer is larger than that in the DOAS analyzer. The reading of each analyzer are the averaged concentration of the sample in the cell. If the measurement conditions changed or in other words, the condition in the stack changed abruptly. The discrepancy would arise. We will add this analysis in the AMT version.