

## ***Interactive comment on “Volcanic SO<sub>2</sub> and SiF<sub>4</sub> visualization and their ratio monitored using 2-D thermal emission spectroscopy” by W. Stremme et al.***

### **Anonymous Referee #1**

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The authors describe measurements of the volcano Popocatepetl, Mexico, during quiet and active periods. The measurements are recorded using a scanning FTIR instrument from the thermal emission of the atmosphere and compared to lunar measurements for validation.

The article describes an interesting method to monitor a source of gas emissions using FTIR spectroscopy during night. The authors describe two modes, a high and a low resolution mode and the retrieval of the gas contents in point and imaging geometry. They use the data to study the gas emissions before, during and after an outbreak.

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The publication is recommended for publishing after the issues addressed below have been clarified.

#### General comments

Sections 3, 4 and 5 of the supplement deal with application, validation and error analysis of the measurements. In my view this is essential in the description of the measurements presented and also a genuine research result, because a new retrieval strategy is employed. Therefore it should be part of the article, not the supplement.

All abbreviations should be explained before using them (e.g. COSPEC in the introduction). Also, instruments and methods should be appended with a reference (e.g. DOAS, SCIAMACHY in the introduction).

There are inconsistency throughout (see below) the paper which should be straightened out. These include different units and/or definitions for the same quantities. This makes reading the paper difficult, especially if the relationship between different definitions is not straight forward.

#### Specific comments:

page 5739, line 27+

The measurements are taken in the thermal infrared. Revercomb (1988) and Schreiber (1996) identified and described problems concerning the self emission of the instrument. Has this problem been taken into account and how was it dealt with?

page 5739 line 13

The sentence creates the impression, that solar absorption spectroscopy and passive spectroscopy are different methods. But solar absorption spectroscopy is a passive method (as is also explained in the next paragraph). Please change to make it consistent.

page 5740 line 10

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I would suggest to append the Italian volcano named Vulcano with a little bit of information (for example it is an volcanic island). The similarity of the name of the volcano makes one wonder if there is a misspelling or similar.

page 5741 line 11

The common unit of a column density is 'molec / area', which is also used in most of the figures. The connection between a mixing ratio and a column density is not straight forward, especially the mixing ratio is not monotone with the volume whereas the column density is.

I would understand the unit ppm m refers to the mean mixing ratio within Layer 2? In this case, I would expect it to be very sensitive to the dimensions of Layer 2.

In any case the units should be same for same quantities throughout the paper.

page 5742 line 8

Please append the geographical position with units.

page 5743 line 4 Both FASCODE and the HITRAN line-list used seem quite old. Why are those preferred to more recent ones? If the HITRAN data base in the version 2004 is used (as mentioned on page 5746), the correct citation is (Rothman, 2005).

How has the instrumental line shape be determined? Is it assumed to be triangular as mentioned in section 3.2?

page 5743 line 7

Are the baseline shifts always exceeding the spectral signatures, or are only such shifts considered which are larger then some threshold? The emission of the spectrometer does not only cause some baseline shift (Revercomb, 1988, Schreiber, 1996), has this been considered?

page 5743 eq 2

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The argument of the ln should be  $(2 h \nu^3)/(c^2 L(\nu) + 1)$

page 5743 line 20 +

I would expect a volcanic plume to contain a large amount of CO<sub>2</sub>. From the emission band at 500-600 cm<sup>-1</sup> it should be possible to estimate the temperature of the plume. Has this been considered?

page 5745 line 7

I think, estimation would be a better expression than assumption.

page 5745 line 7 pp

I do not understand the need of the effective temperature for the background layer. The emission at a certain wave number does not depend on some effective temperature but on the temperature of the emitter and its cross section.

page 5745 line 15

The description of the estimation of the temperature of the volcanic plume belongs in this section, not in the supplement

page 5745 line 17

The "natural" apodisation function of a FTIR is a boxcar function, it cannot be described by a triangular function. A triangular apodisation has to be imposed on the interferogram and is a poor choice (Harris, 1978, Davis et. al, 2001).

Is the apodisation included in the calculation of the forward model spectrum as well?

page 5745 line 20 +

"As the number of spectra was small with little computational effort, the retrieval ensures the independence of the results from the starting condition, LM-damping term and convergence criteria through the minimum number of 50 iterations."

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This argument is wrong: If the iteration has converged to some minimum in the state space, it will not matter how many additional iterations are added, the iteration stays put. In particular, independence of starting condition, LM-parameter and convergence criteria can not be ensured that way. Assume, the equation which is to be minimized has two minima, which may happen in a non-linear case, the choice of the starting argument will determine which minimum is found.

However, I suppose the authors have made sure that the iteration converges to sensible results by careful choice of the parameters. I would therefore suggest to remove this statement.

page 5746 line 7+8

How do the authors ensure that the measured SO<sub>2</sub> spectrum stems only from the volcanic layer? I would expect SO<sub>2</sub> to be present in a heavily polluted area like Mexico City.

If the amount of SO<sub>2</sub> in the foreground layer is negligible compared to the volcanic plume the authors should include some examples of the expected fractions. The same is true for CO<sub>2</sub> and other interfering gases. For example, I would expect a volcanic cloud to contain large amounts of CO<sub>2</sub>, maybe even H<sub>2</sub>O.

It may be not important in which layer it is fitted, because they are not distinguishable by pressure broadening but this should be explained and also backed with examples.

page 5746 line 12

I guess the authors mean the variability of the water vapor content and not its temperature.

page 5747 line 15

The Nyquist theorem states that the sampling frequency has to be at least twice as high as the highest frequency contained in the real signal (not in the observed signal). This

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means that the puff frequency cannot be determined using the FTIR measurements, because the sampling has already taken place. If the Nyquist theorem is violated, the frequency spectrum is wrong. It has to be ensured before the measurement, that the Nyquist theorem will not be violated, or it has to be established, that a violation of the Nyquist theorem does not matter for the study.

page 5748 eq 5

V<sub>ret</sub> does not exist in the equation (possibly V?)

page 5748 line 21

please reference the Tikhonov operator

page 5748 line 23

Is the operator D identical to the operator D<sub>1</sub>?

page 5749 line 9+, 19

Please give more details on how you arrive at the amount of the emitted gases, i.e. assumptions of the volumes considered. I would also expect an error calculation.

page 5752 line 2

Unit?

line 4

presneted -> presented

page 5754 line 3

quatified -> qualified

In the supplement:

fig 3: what is D-SO<sub>2</sub>? I would suggest to append the phrase "As mentioned" in the

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figure legend with more specific information on where to find the information.

fig 9 and fig 10

Does the retrieved SF<sub>4</sub> from lunar absorption and emission measurements agree also?

fig 11

The molecular ratio SO<sub>2</sub>/SiF<sub>4</sub> is sometimes given as SiF<sub>4</sub>/SO<sub>2</sub> (e.g. fig 9 in the main article). In order to further the readability of the article, quantities showing the same should always be defined in a consistent way, if there is no strong reason to do otherwise.

Davis, S. P.; Abrams, M. C. & Brault, J. W. Fourier transform spectroscopy Academic Press, 2001

Harris, F. J. On the Use of Windows for Harmonic Analysis with the Discrete Fourier Transform Proceedings of the IEEE, 1978, 66

Revercomb, H. E.; Buijs, H.; Howell, H. B.; Laporte, D. D.; Smith, W. L. & Stromovsky, L. A. Radiometric Calibration Of Ir Fourier-Transform Spectrometers - Solution To A Problem With The High-Resolution Interferometer Sounder Applied Optics, Optical Soc Amer, 1988, 27, 3210-3218

Rothman, L. S., et.al., The HITRAN 2004 molecular spectroscopic database Journal Of Quantitative Spectroscopy & Radiative Transfer, 2005, 96, 139-204

Schreiber, J.; Blumenstock, T. & Fischer, H. Effects of the self-emission of an IR Fourier-transform spectrometer on measured absorption spectra Applied Optics, 1996, 35, 6203-6209

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