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***Interactive comment on “SO<sub>2</sub> emissions from Popocatepetl volcano: emission rates and plume imaging using optical remote sensing techniques” by M. Grutter et al.***

**Anonymous Referee #2**

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**General remarks**

Grutter et al. combine several optical remote sensing techniques to gather a comprehensive dataset describing the emission strength and propagation direction of emissions from Popocatepetl volcano. One of the techniques, the infrared imaging spectrometer, is novel and represents a new technology for volcanic monitoring. Because of the novel nature of this technique, a bit more care should be taken to precisely explain the evaluation. The measurements themselves are described well, and it is clear how the data was obtained. However, as these techniques are already well known, additional work should be directed towards two directions: For one, how can the different

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measured quantities be integrated into one emission and propagation model. Also, the implications of the measurements for atmospheric chemistry, aerosol formation, radiative forcing, and environmental and health impact are only marginally discussed and could be elaborated a bit more. Suggestions for changes along these lines are given below.

## Specific comments

### *Introduction*

Section 1, the introduction, is well written and gives a comprehensive overview of past work related to the field.

### *Methodologies*

In section 2, methodologies, all techniques applied in the study are described except for COSPEC. The COSPEC technique has been explained in detail by other authors, so one could argue that it is not necessary to describe it again here, but at least references should be given.

Also, the section on infrared spectrometry (2.2) is a bit unclear. On page 8126, the fitting procedure is described. The authors mention that the reference of the target gas (in this case SO<sub>2</sub>) as well as other background gases are fit to the measured differential spectrum. For one, it would be worth noting which background species were accounted for. Also, in the case of strong absorption or emission, the quality of the fit significantly depends on the column density assumed during convolution of the literature reference spectrum. The authors mention different column densities were assumed, but it is not clear if spectra with different column densities fit at the same time, or if some optimization criterion was applied to find the column densities that fit best. An iterative approach to this problem is e.g. described by (Frankenberg C, Platt U, Wagner T (2005) Iterative maximum a posteriori (IMAP)-DOAS for trace gas retrieval of strong absorbers: Model studies for CH<sub>4</sub> and CO<sub>2</sub> retrieval from near infrared spectra

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of SCIAMACHY onboard ENVISAT. Atmos Chem Phys 5:9-22)

After subtracting the background compounds, the coefficient of correlation between the remaining spectrum and the reference cross section of the target substance is calculated. Again, several column densities are used. Therefore, several correlation coefficients are obtained for every pixel. It is unclear which coefficient is then used in the plume visualization results (Fig 4). Possibly the highest coefficient could be selected. But if different column densities are used in the correlation tests, why not plot the column density that best correlates with the measurement? This would give an indication of the column density of SO<sub>2</sub> in the plume, not just a binary "in plume" or "out of plume" decision.

### *Results and discussion*

In the first part of the results section, the determination of forward trajectories starting from the volcano is described well. It is also shown, that instantaneous wind direction, forward trajectories and radiosonde data match very well. However, the authors fail to mention where the radiosonde data was collected. Near the volcano?

One question that arises is whether or not the simulated trajectories are representative for the general situation at Popocatepetl, as only March is considered.

In section 3.2., the authors mention that the ground-based DOAS data has been filtered and only measurements taken under the "correct conditions" are included. It is unclear what these conditions are, and how it was determined, which data was collected under such conditions.

The comparison with the COSPEC instrument showed flux values differing by a factor of more than 3. The authors suggest this may be caused by lateral dispersion effects in the plume. However, this explanation seems incorrect. Lateral dispersion would increase the plume size, but at the same time reduce the SO<sub>2</sub> concentration and therefore the measured column density, thus again yielding the correct plume cross

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section and therefore flux. It would be interesting to compare the utilized COSPEC instrument with the DOAS at the same location to test for consistency between the two. While this has been done in the past with some success (see e.g. Elias T, Sutton AJ, Oppenheimer C, Horton KA, Garbeil H, Tsanev V, McGonigle AJS, Williams-Jones G (2006) Comparison of COSPEC and two miniature ultraviolet spectrometer systems for SO<sub>2</sub> measurements using scattered sunlight. *Bulletin of Volcanology* 68:313-322), an instrumental problem could be the cause of the discrepancy.

Another possible explanation could be a difference in radiative transfer, as the measurement geometries of the two techniques were different. Since the COSPEC measurements were made in zenith-looking geometry, the distance to the plume for each measurement was the plume altitude. For the DOAS, the distance to the plume could be much more, depending on whether the plume was overhead or near the horizon. Mori et al. (Mori T, Mori T, Kazahaya K, Ohwada M, Hirabayashi J, Yoshikawa S (2006) Effect of UV scattering on SO<sub>2</sub> emission rate measurements. *Geophys Res Lett* 33:L17315) recently showed that the measured column density decreases with distance to the plume due to UV scattering, so DOAS measurements performed at long distances could be diluted more than the COSPEC measurements.

In Fig. 3, the aircraft measurements are drawn as a solid line covering about 12 hours, the COSPEC once even covers 16 hours. It is unclear why this should be the case, as the aircraft measurement should give almost a point measurement at a certain point in time. The COSPEC measurements could cover a certain time range, as multiple traverses could be made, but this range is likely limited to less than 16 hours.

### *Conclusions*

The authors have gathered a fairly comprehensive dataset describing SO<sub>2</sub> emissions from Popocatepetl volcano in March 2006. This has already been shown in the previous sections. Unfortunately, the conclusions from their measurements are few. In the conclusions section, mainly the measurement results already presented in the previous

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chapter are repeated. True conclusions are only marginally mentioned. The authors themselves state that the point of combining wind trajectories with SO<sub>2</sub> emission rates should be to learn about atmospheric chemistry, aerosol formation, and radiative implications in the central region of Mexico. Very little is said to this extent.

In the case of the Puebla/Tlaxcala direction, 25

One idea that was not followed up on is that the measurement of the volcanic plume close to the volcano (scanning DOAS) and further away (aircraft and COSPEC) could allow a validation of the trajectories calculated from NCEP. The combination of both could also allow an estimate of sulfur deposition between the two measurement positions.

The infrared plume imaging technique is very innovative. Its main advantage is probably the ability to measure at night, therefore allowing a 24h monitoring of SO<sub>2</sub> emissions. The fact that it does not require "perfect blue sky conditions" is true but misleading, as this is also true for DOAS and other remote sensing techniques. Most techniques can easily handle clouds above the plume but have trouble when clouds are present between the instrument and the plume. If the infrared emission instrument is superior in this respect, a specific study could be conducted to show this.

It is unclear whether the presence of "puffs" being emitted from the volcano was shown. In the results and discussion section, the authors mention the ability to measure a variation in SO<sub>2</sub> emission by examining subsequent images, extracting the wind speed from these, and then comparing the measured column densities to those expected due to variations in wind speed. It is unclear, however, whether this technique was applied to confirm the presence of "puffs". If so, details of the retrieval should be given.

### Technical corrections

p. 8120

line 26: write "...or FROM short-lived eruptions."

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p. 8121

line 5: write "For volcanic plumes with high water vapor content or low altitude volcanoes near the coast..."

line 14: write "...strong explosions that occurred in June of 1997. Ash columns reached 15 km a.s.l.. In December of 2001, another strong eruption..."

line 19: write "SO<sub>2</sub> emission rates have been..."

line 22: write "In the past, other averages have been reported: 2.0 Gg/d in 1994, 1.6 Gg/d in 1995, 15 Gg/d in 1996, and occasionally UP TO 50 Gg/d in 1997. The total discharge of volatiles..." What is meant by  $\leq 50$  Gg/d? Up to 50? Also include references for each value.

p. 8122

line 16: write "More commonly, the monitoring of gases and their relative ratios in volcanic plumes has aimed at a better understanding and forecasting of eruptive processes since changes in the magmatic activity are reflected in..."

line 21: write "Both optical remote sensing methods employed in this investigation to characterize SO<sub>2</sub> in the infrared (FTIR) and ultraviolet (DOAS) wavelength regions have been used for this purpose before (..."

line 24: write "The broad range of application of these methods..."

p. 8123

line 5: write "... can be achieved from the different measurement configurations and instrument platforms from which the..."

line 13: Here aircraft measurements are mentioned in which the zenith viewing geometry is applied. However, this is not the only possible geometry (MAX DOAS or imaging DOAS is also possible). It is suggested to write: "Possible platforms include

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ground-based measurements, aircraft, and satellites. (e.g. Afe ..."

line 15: In the Methodologies section, each applied technique has its own subchapter except COSPEC. The COSPEC technique has been explained in detail by other authors, so one could argue that it is not necessary to describe it again here, but at least references should be given.

line 16: write "DOAS is a widely..."

line 20: In atmospheric physics, the term "scattering" is usually used instead of "dispersion". Therefore, write "...scattering on air molecules and aerosol particles is cancelled during the DOAS retrieval and thus..."

line 21: write "In this investigation, scattered sunlight was used as the..."

line 27: write "...a bandpass optical filter (Hoya U330) blocking visible light with wavelengths higher than 360 nm to reduce stray light, and a..."

p. 8125

Typically, when imaging a certain area, this is referred to as "area of interest", not "field of regard". This term appears in lines 15, 16, 27, and line 3 on the next page.

line 18: add a comma "...specification of this system, which has been..."

line 24: This sentence reads "... an image of 45 x 30 pixels will take over 3 minutes to be completed". Please specify more exactly. 5 minutes? 10 minutes? Or do you mean "just over 3 minutes"?

p. 8126

line 5: "In this work" is not necessary. Begin the sentence with "The first spectrum..."

line 9: Begin the sentence with "Reference spectra with different..."

line 26: write "...light aircraft were performed. Calculating the emission rates from any of these techniques..."

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line 27: replace "velocity at which the plume propagates" with "plume velocity and direction"

p. 8127

line 1: write "... wind speed at the altitude of the volcanic emission at..."

line 2: Here, the NCEP Reanalysis according to Kalnay et al. is cited, but this data is not used in this study. Why not directly cite the NARR data: "The National Weather Service's National Center for Environmental Prediction (NCEP) runs a series of operational computer analyses and forecasts. Their North American Regional Reanalysis (NARR) products include meteorological fields such as u- and v- wind components, temperature, and humidity on a 32 km grid 8 times per day. The three-dimensional..."

line 29: write "For this purpose, lines where drawn..."

p. 8128

line 9: write "... region with an occurrence of about 63

line 10: write "... regions with approximately 19 and 18

p. 8129

line 5: specify what "correct conditions" were chosen, see Specific Comments

p. 8130

line 15: write "...in order to better separate the plume shape from the background. In this particular case, the duration of each scan was 3 min and 14 s, although the scan time generally varied depending on the scan window and step sizes chosen."

p. 8131

line 20: "that far" is too imprecise. Specify how far. See Specific Comments.

line 20: write "...their interaction with urban pollution is more likely...";

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p. 8132

line 1: "... to determine the plume shape and monitor plume evolution."

line 7: What are "perfect" blue sky conditions? See Specific Comments.

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8, S3273–S3281, 2008

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