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Interactive Comment

Interactive comment on "SO_2 emissions from Popocatépetl volcano: emission rates and plume imaging using optical remote sensing techniques" *by* M. Grutter et al.

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I would like to thank the comments from both referees (RC). Their remarks are appropriate and helpful and aim at improving the content of this manuscript. Below are the responses (AC) to the specific issues and the proposed modifications for a revised manuscript.

Responses to reviewer #2

RC: 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. AC:

More detail on the evaluation procedure of infrared spectra was included in the revised manuscript. Also, the references cited on page 8125, lines 3-4, describe the evaluation with some more depth. The following sentence was added in section 2.2: In this work, the spectral range between 1095 and 1250 cm-1 was selected for the analysis of SO2. Spectra of H2O and ozone were used as interferents in the fitting procedure. The column densities of SO2 that were used for the calculation of the reference spectra used in the fitting procedure were 800, 5000 and 18000 ppm m.

RC: 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 SO2) 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. AC In this case, water and ozone were fitted together with SO2.

RC: 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. AC: That was the reason why three different column densities are used. However, in the case of this SO2 plume, the transmission was fairly high.

RC: 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. 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. AC: In order to be more clear, the sentence was modified to: The calculation is sequentially performed for three different column densities of the target compound. In this final step, a color is assigned according to the maximum coefficient of correlation obtained in the fitting procedure and plotted at each position on top of the video image,

RC: But if different column densities are used in the correlation tests, why not plot

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the column density that best correlates with the measurement? This would give an indication of the column density of SO2 in the plume, not just a binary in plume or out of plume decision. AC: The highest correlation coefficient was plotted as stated above. In this work, no attempt to quantify the column density has been made. The goal was to visualize the plume shape. However, one of the authors is currently working on the quantification of the plume of Popocatepetl. The coefficient of correlation gives a continuous measure of the presence of the target gas. For the small column densities that are present in the diluted plume (optically thin plume), there is a high correlation between the column density and the coefficient of correlation between the measured difference spectrum and a reference spectrum with low column density. As the reviewer states, it is possible to quantify the column density by analysis of the shape of the transmission spectrum and the authors have been applying this technique in several applications (see Harig, R., Matz, G., Rusch, P.: Scanning Infrared Remote Sensing System for Identification, Visualization, and Quantification of Airborne Pollutants, SPIE Vol. 4574, 83-94, 2002.). To perform the fitting procedure with several column densities and to plot the column density of the best-fitting reference spectrum is one possible method. However, in the case of the plume of Popocatepetl that results in difficulties: In the areas without SO2 from the volcano the column density of the best-fitting reference has no meaning because the spectrum contains no signature. In this case, another quantity (such as the value of the coefficient of correlation) has to be considered in order to decide if the column density is plotted or not. Within the plume, the technique works well for high column densities (that cause a dependence of the shape of the transmission spectrum on the column density). In the limit of high transmission (small column densities) the shape is not dependent on the column density. This case is given in parts of the plume. Nevertheless, the authors agree that plotting the column density of the best-fitting reference is a possible method but in this case, as said in the text, they chose the coefficient of correlation because of the reasons stated above.

RC: 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

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authors, so one could argue that it is not necessary to describe it again here, but at least references should be given. AC: A generic reference (Stoiber et al 1983) describing the Barringer correlation spectrometer (COSPEC) in detail and its functionality was included at the beginning of the Methodology section. Since this is not the main method used in this work to quantify and characterize the SO2 volcanic plume, no subsection was dedicated to this technique.

RC: 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? AC: The sentence was completed in the revised manuscript with more information: Radiosonde data taken in Mexico City from the Servicio Meteorológico Nacional (SMN, station 76679 located at 19.4N, 99.196W, 2303 m a.s.l.) in this pressure range..

RC: One question that arises is whether or not the simulated trajectories are representative for the general situation at Popocatepetl, as only March is considered. AC: this point is addressed in the responses to referee #1

RC: 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. AC: The following sentence was removed: The data has been filtered and only spectra measured under the correct conditions are included in the analysis and replaced with this one: The emission results have been filtered so that only those data where the volcanic plume was completely crossed during a full scan of the instrument is included. In this way it is clarified that not the spectra but rather the results from complete scans are filtered for better results.

RC: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 ef-

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fects in the plume. However, this explanation seems incorrect. Lateral dispersion would increase the plume size, but at the same time reduce the SO2 concentration and therefore the measured column density, thus again yielding the correct plume cross 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 SO2 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 SO2 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. AC: We agree with the reviewers comments and the following was added in order to expand the explanation and specify what was meant with lateral dispersion.: Unfortunately, no direct comparison between the COPSPEC and DOAS instruments was done in a single transect but a previous work aiming specifically at this (Elias et al., 2006) shows that the retrieved slant columns should not differ by more than 10% from properly calibrated instruments. Thus, a poor calibration of the COSPEC instrument is not discarded but more probable causes for the observed discrepancies are explained below: It was observed from the wind trajectories calculated from NARR data, that often the wind direction changes and makes abrupt turns along the path of the plume. Although a geometrical correction is taken into account

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for the flux calculation, this considers a linear propagation between the origin of the emission and the position at which the SO2 -peak was observed. If, however, the wind direction at the position where the plume is crossed has changed and is considerably different from the one used for the correction, the cross-section and thus the flux will be overestimated. Another possibility would be for a difference in radiative transfer. It has been shown that measured SO2 slant-column densities decrease with distance to the plume due to UV scattering {Mori, 2006 #72}. If the scanning DOAS is measuring a plume close to the horizon while the zenith-looking COSPEC measurement has a shorter distance to the plume, then the flux from the DOAS would be underestimated. The geometrical factor and thus overestimation of the COSPEC flux is thought to be the most probable cause for the discrepancies. This is due to the fact that the traverses are performed at long distances (20-40 km) from the source and wind is expected to change directions as is commonly seen in the trajectories. The following paragraph was removed: Although the COSPEC measurements are within the same order-of-magnitude as the DOAS measurements, they are not completely comparable because the COSPEC data are obtained 20-40 km away from the fixed station. The larger values obtained from the COSPEC traverses can be due to lateral dispersion effects on the plume and thus longer apparent plume cross-sections, or wind speed differences compared to those near the vent.

RC: 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. AC: The original intention for the bars was to make these more visible in the figure. The figure was modified by shortening the length of the bars accordingly.

RC:The authors have gathered a fairly comprehensive dataset describing SO2 emissions from Popocatepetl volcano in March 2006. This has already been shown in the

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previous sections. Unfortunately, the conclusions from their measurements are few. In the conclusions section, mainly the measurement results already presented in the previous chapter are repeated. True conclusions are only marginally mentioned. AC: The text in the conclusions was modified as to emphasize that an important conclusion, apart from reporting the SO2 fluxes from Popocatépetl, was to argument based on the observations and the calculated trajectories, that the emitted gas is not likely impacting the metropolitan regions directly. Rather, the aerosols formed as a product of these large emissions are more likely to be deposited near the surface and interact with the urban plume. Some evidence of this has been published before (Raga et al. 1999).

AC: The reviewer comment with respect to how the different measured quantities can be integrated into one emission and propagation model, and how these measurements can contribute to learn about the atmospheric chemistry, aerosol formation, the radiative implications as well as the environmental and health impact is true and therefore the relationship is made clear in the text. However, the authors believe that it is enough for the purpose of this work to state the relationship of these issues with the output of this study and by expressing the importance to further investigate the fate of these emissions by modeling not only their trajectories, but also the chemical and physical transformations along their path.

RC: 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 SO2 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. AC: The text was modified accordingly and the benefits of using this remote sensing technique now read: 1) it can operate day or night, 2) can handle cloudy conditions for gas detection as long as the clouds are not between the instrument and the plume, 3) can visualize shape, direction and evolution of a volcanic plume, 4) can allow for the determination of the plumes velocity by analyzing sequential images (this would eliminate much of the uncertainty in flux estimation) and 5) can be used to measure other gases like HCl, HF, SiF4 (although at higher spectral resolutions) and report their relative abundances.

Responses to Reviewer #1

RC: There is a large discrepancy between COSPEC and the fixed DOAS. This is an important point to ensure continuity between the well established COSPEC and the newer DOAS and should be given a more thorough treatment. AC: this comment has been addressed (see above)

RC:The section on the plume imaging should be expanded and made a little more rigorous. How much data is available? Could more of it be presented? Is it possible to quantify how intermittent the emissions are? Is there a correlation between SO2 emissions and visible smoke? AC: The section has been expanded and more detail is available in Harig et al. 2002. More data can be shown but the authors dont think this will improve the manuscript. A separate paper is in preparation describing a more rigorous analysis of the IR spectra with the aim to quantify the SO2 column densities from newly acquired data rather than just visualize the volcanic plume.

RC: The trajectories do not add much to the analysis. They basically follow the mean flow aloft, and therefore give the same results as using a windrose. The discussion of vertical dispersion is not sufficiently rigorous at this point. I would recommend leaving this out unless you want to substantially expand it. If you do leave it out, the meteorological discussion should be expanded with wind-roses, and with climatological discussions. This could address whether the time period analyzed is representative of the longer record. On this note, I dont think it is necessary to compare NARR and Radiosonde observations - NARR assimilates this data, just refer to the literature. This would leave more space for SO2 data. AC: The instantaneous wind velocity, the results from the forward trajectories as well as the radiosonde data s summarized in Table 1.

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The % occurrences are equivalent to wind roses except for that only three specific regions, which are thought to be geographically relevant in this study, are presented. It is true that the radiosonde data is contained in the NARR output. The following sentence was added: For consistency and since the radiosonde data is assimilated by the NARR, the continuous dataset from NCEP was used for all emission calculations throughout this work.

RC: I was surprised not to see M.A. Matiella Novak et al., JVGR 170, 1-2, 76-85 in the literature review. Please include a discussion of the relationship of the present work with this paper. The number of papers on the SO2 emissions of the Popocatepetl are few enough that you can afford to cite them all. AC: The following sentence was added and the reference was included in the bibliography. In the introduction to the Methodology section: The importance of complementing satellite observations with ground-based instrumentation has been made evident, particularly when attempting to quantify volcanic emissions from space (Matiella Novak et al., 2008).

RC: How were the trajectories calculated? Where convection/terrain effects accounted for? AC: This paragraph was modified and more details on the trajectory calculations were given. The three-dimensional wind data from NARR was used to calculate the propagation velocity of the plume and the forward trajectories starting from Popocatépetl at 500 hPa using the scheme proposed by (Krishnamurti and Bounoua, 2000). The terrain effects are taken into account in the trajectory computation through the eta vertical coordinate as well as the vertical velocity field in areas where there is convection. However, no other convection effects such as washout and wet deposition were considered for the 48-h forward trajectories that were generated for 00 to 21 UTC in three hour intervals.

RC: Include some seasonal discussion of how March could or not be representative of the whole year AC: The following paragraph was added to the discussion: The March mean wind field at 500mb is representative of the boreal winter over Central Mexico. The westerly winds are dominant form the middle to upper atmosphere. During the

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summer the weaker and moister easterly winds span the lower atmosphere up to 500 hPa due to northward displacement of the trade winds. The winter pattern is modified by cold fronts (northerly winds) and the summer patterns is modulated by the local convection and mesoscale convective systems.

AC: Also, the following sentence was added in the Conclusions: The observed mean wind field at 500 hPa is typical for the month of March and is representative only for the boreal winter over Central Mexico.

AC: The unit Gg was kept in p 9121 line 29 since this corresponds to a yearly amount, as stated in the text.

AC: Where applicable, the term field of regard was changed to area of interest as suggested by the reviewer.

AC: All other technical corrections were made or addressed accordingly

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Interactive comment on Atmos. Chem. Phys. Discuss., 8, 8119, 2008.