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► To cite this version:

M. Grutter, R. Basaldud, C. Rivera, R. Harig, W. Junkerman, et al.. SO₂ emissions from Popocatépetl volcano: emission rates and plume imaging using optical remote sensing techniques. Atmospheric Chemistry and Physics Discussions, 2008, 8 (2), pp.8119-8141. <hal-00304133>

HAL Id: hal-00304133

<https://hal.science/hal-00304133v1>

Submitted on 18 Jun 2008

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**SO₂ emissions from
Popocatépetl**

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SO₂ emissions from Popocatépetl volcano: emission rates and plume imaging using optical remote sensing techniques

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Received: 17 March 2008 – Accepted: 23 March 2008 – Published: 23 April 2008

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Published by Copernicus Publications on behalf of the European Geosciences Union.

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Abstract

Sulfur dioxide emissions from Popocatepetl volcano in central Mexico were measured during the MILAGRO field campaign in March 2006. A stationary scanning DOAS (Differential Optical Absorption Spectrometer) was used to monitor the SO₂ emissions from the volcano and the results were compared with traverses done with a COSPEC from the ground and a DOAS instrument on board an ultra-light aircraft. Daytime evolutions as well as day-to-day variation of the SO₂ emissions are reported. A value of 2.45±1.39 Gg/day of SO₂ is reported from all the daily averages obtained during the month of March 2006, with large variation in maximum and minimum daily averages of 5.97 and 0.56 Gg/day, respectively. The large short-term fluctuations in the SO₂ emissions obtained could be confirmed through 2-D visualizations of the SO₂ plume measured with a scanning imaging infrared spectrometer. This instrument, based on the passive detection of thermal radiation from the volcanic gas and analysis with FTIR spectrometry, is used for the first time for plume visualization of a specific volcanic gas. A 48-h forward trajectory analysis indicates that the volcanic plume was predominately directed towards the Puebla/Tlaxcala region (63%), followed by the Mexico City and Cuernavaca/Cuautla regions with 19 and 18% occurrences, respectively. 25% of the modeled trajectories going towards the Puebla region reached altitudes lower than 4000 m a.s.l. and all trajectories remained over this altitude for the other two regions.

1 Introduction

Volcanic emission of gases and particles can contribute significantly to the chemistry of the atmosphere, its aerosol budget and thus to the radiative forcing both in the regional and global scales. Depending on how far these emissions are transported prior to deposition, these emissions can have important environmental effects. 15–21 Tg of SO₂ are injected into the atmosphere every year due to active volcanoes worldwide (Halmer et al., 2002), either continuously by passive degassing or short-lived eruptions.

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These emissions account for approx. 7.5–10.5% of the total global sulfur emission which has as major contributors the burning of fossil fuels, oxidation of oceanic dimethyl sulfide and biomass burning. SO₂ sinks are dominated by oxidation and deposition processes and its lifetime can range from a few days to a couple of weeks, reacting mostly with OH to form H₂SO₄ or being removed by clouds and aerosols. For high water vapor producing volcanoes or low-altitude volcanoes near the coast, this reaction might occur over hours and even tens of minutes. During large eruptions, however, SO₂ can be injected to higher altitudes and form longer-lived stratospheric aerosols.

Popocatepetl (19.02° N, 98.62° W, 5465 m a.s.l.) is a high emission rate, passively degassing active volcano located 60 km south-east of Mexico City. This stratovolcano is part of the Tran-Mexican volcanic belt and after being in a repose period for nearly 70 years, it began significant fumarolic and seismic activity with a moderate eruption in December 1994 (Siebe et al., 1996). A new episode of activity began in March 1996 with pyroplastic flows and strong explosions that occurred on June 1997. Ash column reached 15 km a.s.l. and then on December 2001 another strong eruption produced lava flows reaching the timberline and ignited fires 4.5 km from the crater (Delgado-Granados et al., 2001; Macias and Siebe, 2005).

Frequent ash and gas emissions have continued irregularly since the reawakening of this volcano. SO₂ emission rate have been estimated beginning early February 2004 with several COSPEC instruments, averaging 2.2 and 3.4 Gg/d in studies performed during the 12/23/1994–01/28/1995 (Galindo et al., 1998) and 1/30/1995–06/30/1995 (Delgado-Granados et al., 2001) time periods. Other averages have been reported of 2.0 Gg/d in 1994, 1.6 Gg/d in 1995 and 15 Gg/d in 1996 with values rising occasionally to ≤50 Gg/d during 1997. Total discharge of volatile gases through November of 1996 was similar to yearly amounts released by Mount Etna (Goff et al., 1998).

The Mexican national inventory for the year 1999 reported annual emissions of 1871 and 735 Gg/y from the Popocatepetl and Colima volcanoes, respectively (INE-SEMARNAT, 2006), which can be translated into an average daily emission of 5.13 Gg/d from Popocatepetl alone. This value is comparable to 6.65 Gg, which is the

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amount of SO₂ emitted anthropogenically by the Mexico City Metropolitan Area from both point and mobile sources for an entire year, as has been officially reported for 2004 (SMA-GDF, 2006). This value, however, does not include the emissions from important industries like power plants and refineries located just outside the metropolitan area. For example, the Tula industrial complex alone, located 60 km north of Mexico City and 130 km NW of the volcano, emits around 0.43 Gg/d (158 Gg/y) as estimated from optical remote sensing measurements carried out in 2003 (de Foy et al., 2007). Thus, Popocatepetl volcano has been an important source of emissions during this past decade releasing similar or higher amounts of SO₂ than all the anthropogenic sources in the central region of Mexico.

The objective of this investigation was to study the emissions of SO₂ from Popocatepetl volcano during the MILAGRO field campaign (Fast et al., 2007) and examine its possible interaction with the Mexico City plume. The measurement of the emissions of SO₂ from Popocatepetl is important in the context of a megacity field study such as MILAGRO, where sulfate production and its radiative and chemical impacts are to be characterized in detail (Graf et al., 1997). More commonly, the monitoring of gases and their relative ratios in volcanic plumes has been sought for understanding and forecasting eruptive processes since changes in the magmatic activity is reflected in both the quantity and chemical composition of the emissions. Particular emphasis has been placed on the relative abundances of emitted gases such as HCl, HF, H₂S, SiF₄, CO₂, BrO, ClO, among others. The optical remote sensing methods employed in this investigation to characterize SO₂ both in the infrared (FTIR) and ultraviolet (DOAS) regions have been used for this purpose (Goff et al., 2001; Love et al., 1998; Oppenheimer et al., 1998). The broad possibilities of these methods as well as the ongoing work will be explained.

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2 Methodologies

Out of the many techniques available to analyze the composition of volcanic plumes, the spectroscopic remote sensing methods are preferred due to the potential hazard in approaching an active volcano for sample taking. Other advantages include the high temporal and spatial resolution which can be achieved from the different configurations and platforms in which the spectroscopic analysis is performed (Oppenheimer et al., 1998). The COSPEC (Barringer Research correlation spectrometer) has been the most widely used instrument for volcanic plume surveillance. It is configured to measure SO₂ column concentrations using sky UV radiation and estimating emissions by combining plume cross-section and wind velocity information. Differential Optical Absorption Spectrometers (DOAS) are now being implemented as more modern, light and versatile instruments. These have been deployed on the ground and from aircraft looking up to the sky and from downward-looking satellites (Afe et al., 2004; Eisinger and Burrows, 1998).

2.1 DOAS

This is a widely used technique for the continuous measurement of atmospheric gases both in active and passive configurations (Platt, 1994; Platt et al., 1979). It is based on the spectral analysis of the differential absorption by molecules in the ultraviolet and visible part of the spectrum. The broader extinction of UV light due to other processes such as dispersion by fine particles is cancelled when processed and thus not taken into account. In this investigation, dispersed light from the sun was used as the radiation source and the differential absorption of the SO₂ gas was analyzed and used to obtain differential slant columns as has been described elsewhere (Bobrowski et al., 2003; Galle et al., 2003; Lee et al., 2005).

Passive DOAS measurements were made by collecting the scattered UV light with a narrow field-of-view (< 20 mrad) telescope. This consists of a convex lens (f=100 mm), a bandpass optical filter (Hoya U330) to reduce stray light blocking visible light with

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wavelengths higher than 360 nm and a 200 μm diameter quartz optical fiber. The light is analyzed with a spectrometer (Ocean Optics, model S2000), at a resolution of <0.6 nm between 280–420 nm. This device employs a UV holographic grating and a 2048 element CCD detector. The instruments described below use their own acquisition and control interfaces, although the same spectral evaluation software DOASIS (Kraus, 2001) was used in both configurations.

2.1.1 Scanning DOAS

A scanning DOAS instrument was placed at Tlamacas station (19.06° N, 98.63° W, 4000 m a.s.l.), located on the northern flank of the volcano and 4.7 km from the crater. The instrument is equipped with a turning mirror and a housing holding a quartz window for environmental protection. A stepper motor turns the 45° mirror which is oriented so that the light coming from the quartz window is directed towards the focusing lenses and into the optical fiber. The elevation angle is scanned from a fixed position perpendicular to the direction of the plume propagation. A full scan of the plume is accomplished every 1–6 min from which an SO₂ emission rate is calculated using the slant column of every scanned position, plume height, wind direction and wind speed information. A detailed description of the instrument and spectra evaluation can be found in (Edmonds et al., 2003; Galle et al., 2003; McGonigle et al., 2003).

2.1.2 Airborne DOAS

The FZK-ENDURO Ultra-light aircraft (Junkermann, 2005) was used as the platform to perform downwind plume traverses with a portable DOAS instrument. The aircraft was stationed at Hermanos Serdán Airport (PBC at 19.16° N, 98.37° W, 2244 m a.s.l.) near Huejotzingo, Puebla. The telescope was mounted above the wing looking towards the zenith and the spectra were continuously recorded after take-off using a LabView® based interface that couples each acquisition with a longitude-latitude fix from a GPS receiver. The software is designed to automatically set the acquisition time of the

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spectrometer according to the light intensity. User defined parameters along with dark and background spectra are entered prior to each measurement along the trajectory. The traverses were planned so as to fly around the volcano with a radius of ~18 km, which is the permissible distance from the crater regulated by local aviation officials.

5 2.2 Scanning imaging infrared spectrometry

A scanning imaging infrared spectrometer for visualizing the SO₂ plume was placed at Altzomoni (19.12° N, 98.65° W, 4000 m a.s.l.) which is 11 km NNW of Popocatépetl at the flanks of Iztaccíhuatl volcano. This location allows for an adequate view of the gas plume at a safe distance from the active volcano. The system (SIGIS) (Harig et al., 2002) is based on the combination of a modified Michelson interferometer (Bruker, Opag 22), a telescope (7.5 mrad), an azimuth-elevation-scanning mirror, a video camera and a computer for control, data analysis, and display of the results. The video image is used as reference and to control the position of the scanning mirror. For visualization of gas clouds, the scanning mirror is sequentially set to all positions within the field of regard and both the video images and recorded spectra are graphically displayed on the PC. The size and the direction of the field of regard, as well as the spatial resolution (i.e. the angle between adjacent fields of view) can be varied by the operator. A complete description and specification of this system which has been successfully deployed mostly for industrial pollution monitoring, can be found in (Grutter et al., 2008; Harig and Matz, 2001; Harig et al., 2002).

In the scanning mode, a rectangular window within the video image as well as a step size (i.e. the angle between adjacent fields of view) is chosen. The instrument, capable of measuring 6 spectra/s at 4 cm⁻¹ resolution, will sequentially record a spectrum at each position such that an image of 45×30 pixels will take over 3 min to be completed. The radiation measured by the spectrometer contains the spectral signatures of the background atmosphere and the gas cloud and the atmosphere in each position in the field of regard.

The primary objective from these passive IR observations was to visualize the evo-

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lution of the SO₂ plume. This can be accomplished by analyzing the spectra and fitting the features of the expected atmospheric gases at each position. The procedure for identification of SO₂ within the field of regard follows the steps described in (Harig et al., 2002). In the first step, the spectra of the brightness temperatures of a field of view with and without the plume are calculated. In this work, the first spectrum of each row of the image is used as the background spectrum which is subtracted from the plume spectra. The reference spectra of the target gas, H₂O and other interfering gases are then fitted to the resulting spectrum using a least-squares fitting procedure. The reference spectra with different column densities are calculated by convolution of high-resolution transmittance spectra using the HITRAN database (Rothman et al., 2005) with an instrumental line function (Harig, 2004). The fitting procedure includes an approximation of the baseline. In the next step, the contributions of all fitted signatures (i.e. interferences, atmospheric species, and baseline) except the signature of the target compound are subtracted from the measured spectrum.

In order to decide if the target compound is present, the coefficient of correlation between the corrected spectrum, i.e. the result of the subtraction, and a reference spectrum is calculated. The calculation is performed for three different column densities of the target compound. In this final step, a color is assigned according to the coefficient of correlation obtained in the fitting procedure and plotted at each position on top of the video image, as presented in Fig. 4.

3 Results and discussion

Measurements of the SO₂ emissions from Popocatépetl were carried out during the month of March 2006. Apart from the scanning DOAS, which automatically measures slant column cross-sections of the plume from the Tlamacas fixed site, traverses done with COSPEC from the ground and a portable DOAS instrument on board the ultra-light aircraft were done. Calculating the emission rates with either of these techniques requires knowing the velocity at which the plume propagates at the time the measure-

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ment was done. This was derived from estimates of the wind speed at the level of the emission at ~5400 m a.s.l. The National Weather Service's National Center for Environmental Prediction (NCEP) runs a series of operational computer analyses and forecasts. One of their products is the NCEP Reanalysis data (Kalnay et al., 1996) available four times a day with $2.5^\circ \times 2.5^\circ$ horizontal resolution and that contains global meteorological fields such as the u- and v-wind components, temperature, and humidity. The NCEP provides also a more refined analysis called the North American Regional Reanalysis (NARR), with higher spatial (32 km) and time (8 times a day) resolution for the North American domain (Mesinger et al., 2006). 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 in order to examine the regional displacement for the month of March, 2006. The 48-h forward trajectories were generated for 00:00 to 21:00 UTC in three hour intervals.

Since the altitude of the volcanic plume varies and is not known with precision for every individual measurement, the 3-hourly model outputs for the 550 and 500 hPa layers, corresponding to approximate altitudes between 5100 and 5900 m a.s.l., were averaged and are shown in Fig. 1. This altitude range is thought to contain the plume above the measurement site at Tlamacas (4.7 km downwind) most of the time. Radiosonde data from the Servicio Meteorológico Nacional (SMN) in this pressure range are also plotted in Fig. 1. For consistency, the continuous NARR dataset was used for all emission calculations throughout this work.

Evidence that the emissions of Popocatépetl volcano can be influencing the particle formation in the Mexico City area has been presented elsewhere (Raga et al., 1999). In order to estimate the probability that volcanic emissions would affect neighboring urban areas during the MILAGRO field campaign, a frequency analysis was produced based on the NARR timeseries (500–550 hPa) for the month of March. A geographical division was established defining three major basins containing the largest nearby metropolitan areas: Mexico City Metropolitan Area (MCMA), Puebla/Tlaxcala and Cuautla/Cuernavaca. For that purpose, lines were drawn from the position of the

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crater in the directions 130, 270 and 353° as presented in Fig. 2, taking into account the surrounding mountains as physical barriers and major urbanized centers of the above-mentioned regions.

The corresponding wind direction ranges considered for this analysis are presented in Table 1. This table contains the percent occurrences from instantaneous wind direction datasets from NARR at the position of the crater, the final position of the forward trajectory calculated for 48 h and the radiosonde 500 hPa wind speed data which fall under this criterion. The results show that the volcanic plume was predominately directed towards the Puebla/Tlaxcala region with a 63% occurrence, followed by the Mexico City and Cuernavaca/Cuautla regions with 19 and 18%, respectively. Three dimensional plots generated for all trajectories during the month of May revealed that 25% of the modeled trajectories going towards the Puebla/Tlaxcala region reached altitudes below 4000 m a.s.l., while all the trajectories towards the other two regions remained above this altitude most of the time.

3.1 Ultra-light aircraft measurements

SO₂ emissions were calculated from selected DOAS measurements performed on board an ultra-light aircraft in order to 1) compare them with the results from the ground-based instrument and 2) determine the plume position and width during a specific event. Data is presented from only three flights since not all the flight-patterns were relevant to this investigation and the instrument performance and weather conditions were not always favorable. The result from one of these experiments, corresponding to March 18th, is graphically presented in Fig. 2. The trajectory of the flight is marked with a colored line expressed in column density (ppm·m) of SO₂, starting south east of Iztaccíhuatl volcano and ending at the Puebla airport for landing after 180 km.

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3.2 Ground-based DOAS measurements

An automated scanning DOAS was operated continuously from the Tlamacas site as described in Sect. 2a. SO₂ emissions were calculated using the NARR dataset throughout the month of March for consistency. The data has been filtered and only spectra measured under the correct conditions are included in the analysis. All the individual emission values calculated from the ground-based DOAS instrument are plotted as red dots in Fig. 3 for the month of March 2006. It is important to note that since this method requires dispersed light from the sun, only daytime values are reported. Also in this plot, the emission calculations obtained from the three traverses done from the ultra-light aircraft (blue) and two emission calculations from the COSPEC instrument are included. Ground-based traverses with COSPEC are performed routinely twice a month by CENAPRED (Centro Nacional de Prevención de Desastres) in collaboration with the Instituto de Geofísica of UNAM. The average of all measurements from the ground-based DOAS instrument, reported as 31.7 kg/s, is represented in the graph as a horizontal line.

The individual averages for every day measured during the month of March 2006 are presented in Table 2. Instantaneous emission determinations from traverses performed on selected days from the ground and from the air are also tabulated for comparison. Values obtained from the instrument on board the ultra-light aircraft agree with the daily averages from the ground based DOAS falling within their standard deviations on both days where the data is available. On the other hand, the emission calculation from the COSPEC measurements performed on the 9th and 23rd are 2.8 and 3.7 times higher than the daily average reported from the ground-based DOAS. 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.

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Two days with particularly high emission of SO₂ were the 14th and 19th, with daily averages reaching 4.42 and 5.97 Gg/d, respectively. The lowest activity was recorded on the 13, 15, 17 and 28th, all with emissions below 1 Gg/d.

3.3 Plume visualization

5 A large variability in the SO₂ emission is evident from the DOAS measurements shown above. A scanning imaging infrared spectrometer was deployed on selected days in order to visualize the plume shape and monitor its temporal evolution. The instrument, described in Sect. 2b, was placed at Alzomoni and was able to detect the SO₂ signature in the infrared spectra collected from a distance of 11 km from the crater. This
10 detection is represented in a two-dimensional image according to the coefficient of correlation R with respect to a reference spectrum of SO₂ as seen in the example shown in Fig. 4 for 16 March .

These images include the white and black video image of the volcano at the beginning of each scan and the false color image of the SO₂ plume. For clarity, only spectra
15 with R -values greater than 0.97 are plotted in order to better identify the plume shape from the background. The duration of each scan was in this particular case 3 min and 14 s, although this varies depending on the window and step sizes chosen. It is evident from these observations that the plume changes significantly from scan to scan. There are two potential reasons for this. Firstly, changes in wind speed lead to a variability of
20 the number of molecules present along the optical path if the emission rate is constant. However, the wind speed and thus also a change of the wind speed may be observed by analysis of consecutive images. The second reason is that the emission rate of gas is not constant. The presence of these “puffs” or events of higher emission can be investigated by analysis of consecutive scans.

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4 Conclusions

Knowing Popocatépetl's SO₂ emission source strength is important to assess its potential contribution to the atmospheric chemistry, aerosol formation and its radiative implications in the central region of Mexico. Optical remote sensing methods were deployed for this purpose during the MILAGRO international field campaign. An average of 2.4 ± 1.39 Mg of SO₂ were released every day to the atmosphere during the month of March 2006 as determined by a passive DOAS instrument continuously measuring from the ground and confirmed by traverses done with a similar instrument from an ultra-light aircraft. A frequency analysis of the 48-h forward trajectories calculated from the NARR model outputs from NCEP suggests that the emissions from Popocatépetl were transported towards Puebla/Tlaxcala approximately 63% of the time during the month of March 2006. At this altitude the wind direction towards the Mexico City Metropolitan Area, located only 60 km NW of the crater, accounted for only 19% of the occurrence during this period and none of these trajectories crossed below 4000 m a.s.l. The SO₂ emitted at this altitude, which is well above the planetary boundary layer, would require strong convective conditions to be directed downward and impact the metropolitan areas. This could be observed only in the case of the Puebla/Tlaxcala direction, where 25% of the calculated trajectories cross at some point below an altitude of 4000 m a.s.l. However, the particles formed as a result of these emissions are not expected to be transported that far and their interaction with the urban pollution is more likely to happen, as found in a previous study (Raga et al., 1999). It would be important to further investigate the fate of these emissions by modeling not only their trajectories, but also the chemical and physical transformations along their path.

A scanning imaging infrared spectrometer was used to visualize the dispersion of the sulfur dioxide plume and investigate the large fluctuations observed in the emissions. The thermal infrared radiation of the emitted gases was collected and used to detect the SO₂ emission band from a distance of 11 km. Two-dimensional images of the detected

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SO₂ signature were generated to determine plume shapes and monitor their evolution. These observations confirm that gas emission from the volcano is not continuous but appears rather as “puffs”. This spectroscopic technique, used for the first time for plume visualization of a specific volcanic gas using its thermal radiation, represents important

5 progress for the surveillance of volcanic activity since 1) it can operate day or night, unlike the DOAS technique, 2) does not require “perfect” blue sky conditions for gas detection, 3) can visualize shape, direction and evolution of a volcanic plume, 4) can allow for the velocity at which the plume is propagating to be determined by analyzing sequential images (this would eliminate much of the uncertainty in flux estimation) and
10 5) can be used to measure other gases and report their relative abundances. Work is in progress to determine column densities of SO₂ from the measured spectra. These results will allow for estimates of emissions from an alternative remote sensing method.

Acknowledgements. This project was partly funded by CONACyT (grant # 41531), UNAM (PA-PIIT # IN113306) and IMK-IFU. The NOVAC (Network for Observation of Volcanic and Atmospheric Change) project is acknowledged for providing the ground-based DOAS measurements
15 used in this study. A special gratitude is offered to the National Park Service and its staff (A. Lopez, A. Tagle, and J. Rodriguez) for their support. D. Baumgardner is acknowledged for his collaboration and support during this project.

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Table 1. Occurrences of the wind direction at crater height from the NARR and radiosonde (RS) datasets classified by regions during March 2006.

Region	WDR range	% Occurrence		
	Degrees	NARR	Frw Trajectory	RS-500 HPa
Mexico City Metropolitan Area	90–173	14.1	19.0	15.7
Puebla, Tlaxcala	173–310	64.1	62.9	64.7
Cuautla, Cuernavaca	310–90	21.8	18.1	19.6

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Table 2. Daily averages and standard deviation of SO₂ emissions calculated from the ground-based DOAS instrument given the number of observations. COSPEC and airborne-DOAS emission calculations are instantaneous values from single traversals.

Date	Ground-based scanning DOAS			U-Light DOAS	COSPEC
2006	No. of samples	Daily Avg.	Std. Dev.	[Gg/d]	
4-Mar	18			0.09	
8-Mar					
9-Mar	44	1.84	0.42		5.23
10-Mar	48	2.53	0.82		
11-Mar	42	3.53	2.29		
12-Mar	46	1.94	0.73		
13-Mar	28	0.99	0.30		
14-Mar	51	4.42	3.42		
15-Mar	19	0.78	0.27		
17-Mar	18	0.63	0.46		
18-Mar	28	3.41	1.85	2.71	
19-Mar	49	5.97	2.70		
20-Mar	48	2.70	0.74		
21-Mar	49	2.68	1.79	2.51	
22-Mar	38	3.96	1.94		
23-Mar	5	1.93	0.26		7.23
24-Mar	18	2.36	0.61		
25-Mar	32	2.29	1.18		
26-Mar	15	2.74	1.42		
27-Mar	32	1.05	0.38		
28-Mar	5	0.79	0.38		

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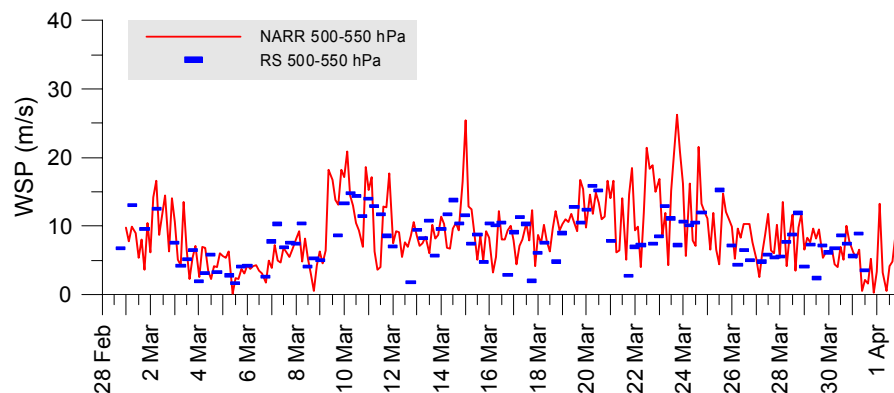


Fig. 1. 3-hourly wind speed data from NCEP (NARR) model output as well as 06:00 and 18:00 h (LST) radiosonde data (RS) from SMN for the month of March 2006. All data is averaged over the 550–500 hPa range.

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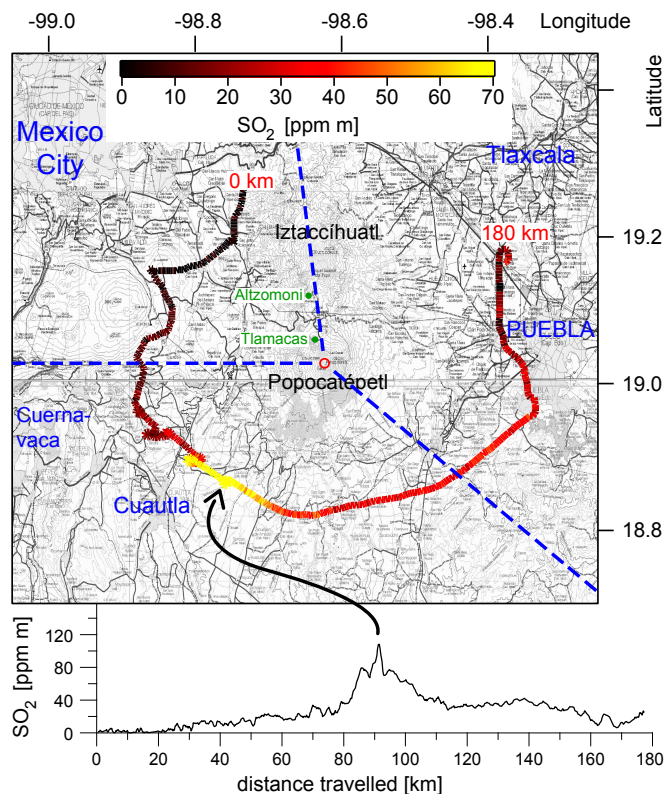


Fig. 2. Map of the region around Popocatépetl volcano showing the results from a DOAS measurement made on board an ultra-light aircraft. The color scaled line represents the slant column of SO₂ (ppm·m) measured along the path flown on 18th March 2006. Blue dashed lines separate the main regions described in Table 1.

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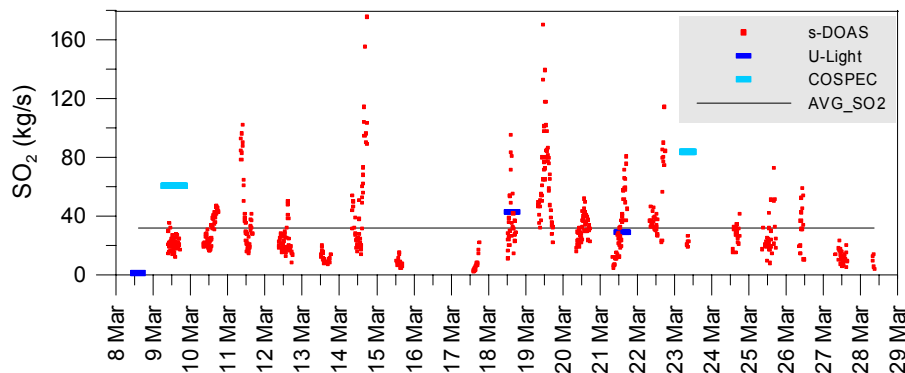


Fig. 3. Sulfur dioxide emission rates from Popocatépetl volcano calculated from the ground-based scanning DOAS measurements (red dots), from traverses done with a mobile DOAS on board an ultra-light aircraft (dark blue) and with a COSPEC instrument (light blue) in 2006.

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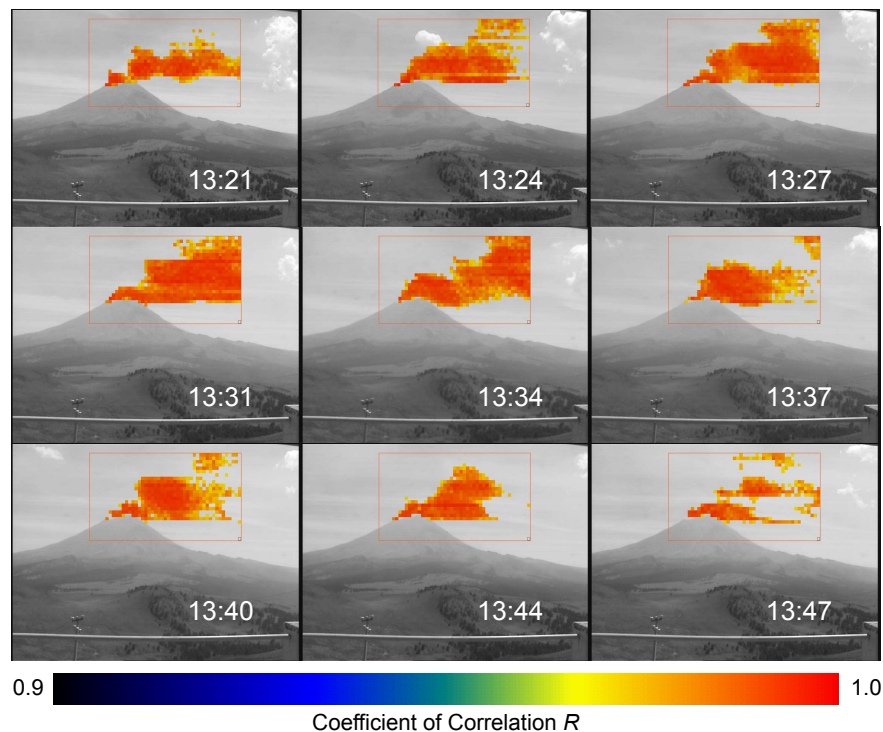


Fig. 4. SO₂ plume visualization of Popocatépetl volcano by passive infrared spectroscopy during 17th March 2006. White numbers indicate the local time.

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