



Meteorological influence on the seasonal and diurnal variability of the dispersion of volcanic emissions in Nicaragua: A numerical model study

Baerbel Langmann^{a,*}, Matthias Hort^a, Thor Hansteen^b

^a Institute of Geophysics, University of Hamburg, Germany

^b IFM-GEOMAR, Kiel, Germany

ARTICLE INFO

Article history:

Received 30 September 2008

Accepted 30 January 2009

Available online 15 February 2009

Keywords:

Nicaragua

continuous volcanic degassing

atmosphere-chemistry numerical modelling

ABSTRACT

Nicaragua comprises seven historically active volcanoes (Cosigüina, San Cristobal, Telica, Cerro Negro, Momotombo, Masaya, and Concepcion), five of which are in a state of continuous degassing. Published measurements of the atmospheric dispersion of continuous emissions from Nicaraguan volcanoes, the chemical and aerosol microphysical modifications of the released gases and aerosols, and related acid deposition and impacts on the environment cover only short periods of time. We applied a three-dimensional atmosphere-chemistry/aerosol numerical model over Central America focussing on Nicaraguan volcanic emissions for month long simulation periods during the dry and wet seasons of 2003. The model is able to reproduce observed monthly precipitation and wind speed throughout the year 2003. Model results for near surface SO₂ concentrations and SO₂ dry deposition fluxes around Masaya volcano are in very good agreement with field measurements. During the dry season, oxidation of SO₂ to sulphate plays only a minor role downwind of the Nicaraguan volcanoes and over the Pacific Ocean, whereas SO₂ released from Arenal and Poas in Costa Rica is oxidised to sulphate much faster and closer to the volcanoes due to higher humidity and cloud water availability. During the wet season, more variable wind conditions lead to reduced dispersion of SO₂ over the Pacific Ocean and increased dispersion inland. The availability of liquid water in the atmosphere favours sulphate formation close to the Nicaraguan volcanoes via aqueous phase oxidation and represents another limitation for the dispersion of SO₂. Strong precipitation removes sulphate quickly from the atmosphere by wet deposition. Atmospheric SO₂ concentrations and in particular dry deposition close to the volcanoes show a pronounced diurnal cycle.

© 2009 Elsevier B.V. All rights reserved.

1. Introduction

Volcanic emissions are an important source of atmospheric gases and aerosols. The emissions consist primarily of H₂O, followed by CO₂, SO₂, HCl, H₂S and other compounds (e.g. Bardintzeff and McBirney, 2000). They play an important role in the Earth's radiation budget, in tropospheric and stratospheric chemistry, and atmospheric dynamics and can impact on terrestrial ecosystems and human health. Volcanic emissions can be released continuously by passive degassing or diffusive (soil) degassing into the troposphere. Emissions can also be released sporadically by explosive and short-lived eruptions into the stratosphere. Andres and Kasgnoc (1998) determined that 99% of volcanic SO₂ is being released continuously, while only 1% is released during sporadic eruptions. Largely due to the work following the Pinatubo eruption in 1991, there is now a better understanding of the climate impact and chemical and aerosol microphysical modifications of volcanic eruptions reaching the stratosphere (Robock, 2000). In contrast, the chemistry and climate influence of tropospheric volcanic plumes is rather poorly known. This partly stems from a wide spec-

trum of emissions and emission styles (e.g. continuous degassing, minor eruptions), high temporal variability of volcanic fluxes into the troposphere dependent on the stability of the volcano-magma system and changes in volcanic gas composition (Duffell et al., 2003; Williams-Jones et al., 2003). In addition, satellite observations of tropospheric volcanic plumes are often hampered by cloud cover.

Compared with stratosphere-reaching eruptive emissions, volcanic emissions released into the troposphere are deposited rapidly locally and regionally. Various components of volcanic emissions (including acid species and heavy metals) can be taken up by the vegetation (e.g. respiration of acid gases through stomata) and the soil (by dry and wet deposition) (e.g. Langmann and Graf, 2003). But tropospheric volcanic emissions can also have a significant atmospheric impact because such emissions are frequently released continuously for long periods of time, and because volcanoes are usually at high elevations, allowing those emissions to remain in the troposphere longer than, for example, most anthropogenic sulphur emissions. Therefore, the non-eruptive volcanic degassing of SO₂ and subsequent sulphate formation is estimated to be responsible for 24% of the global annual direct radiative forcing of sulphate aerosols at the top of the atmosphere (Graf et al., 1997) although volcanic sulphur emissions contribute only 13% to global emissions of sulphur.

* Corresponding author.

E-mail address: baerbel.langmann@zmaw.de (B. Langmann).

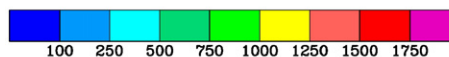
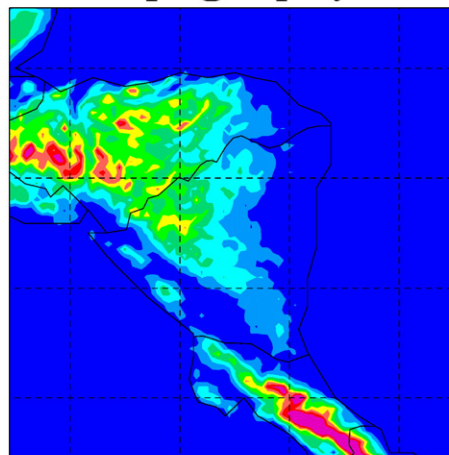
In Central America the most active volcanoes (Fig. 1) are located within the Quaternary Central American Volcanic Arc (CAVA) which is produced by subduction of the oceanic Cocos Plate beneath the southern edge of the North American Plate and the western edge of the Caribbean Plate (Simkin and Siebert, 1994). The active zone of volcanism extends 1500 km from Guatemala to northwestern Panama. Nicaragua comprises seven historically active volcanoes (Cosigüina, San Cristobal, Telica, Cerro Negro, Momotombo, Masaya, and Conception), five of which are in a state of continuous degassing. The strongest sources of volcanic gases are presently San Cristobal, Telica, Masaya and Momotombo (Mather et al., 2006; Frische et al., 2006). Masaya is one of Nicaragua's most active volcanoes, located about 25 km southwest of Managua, the capital of Nicaragua. Today Masaya as well as Telica, San Cristobal and several other volcanoes in Guatemala, El Salvador and Costa Rica emit several hundreds of tons of SO₂ per day each (Mather et al., 2006) with considerable temporal variability and sporadic ash plumes as well. During March–April 1998 and February–March 1999 mean concentrations of SO₂ measured up to 44 km downwind from Masaya volcano in Nicaragua range from ~5 to 600 µg/m³ (Delmelle et al., 2002). In May 2001, 7950 µg/m³ SO₂ maximum concentration was measured at the crater rim of Masaya volcano (Allen et al., 2002).

Until now, air sampling and optical remote sensing measurements of SO₂ have been carried out at Nicaraguan volcanoes only during restricted periods of time (Delmelle et al., 2001, 2002; Mather et al., 2006 and references therein) in order to estimate typical volcanic emissions and dispersion patterns in the atmosphere. Within the EU project NOVAC (Network for Observation of Volcanic and Atmospheric Change, <http://www.novac-project.eu>) which started in October 2005, the scanning Dual-beam Mini-DOAS (Differential Optical Absorption Spectroscopy) instrument is now used for the quantitative and continuous measurement of volcanic gas emissions during daytime with a special focus on Central America. Usually, flux measurements are carried out a few kilometres (~5–15 km) downwind a volcano. These fluxes represent the actual source fluxes from a volcano as long as no other loss or production processes occur. If, however, removal from the atmosphere, e.g. by dry deposition takes place, the actual source flux would be higher, e.g. the measured net



Fig. 1. Major volcanoes in Nicaragua (after Simkin and Siebert, 1994).

Topography



[m]

Fig. 2. REMOTE model area from 81–89° W and 9–17° N and topography [m] in 10 km resolution.

flux plus the dry deposition flux (Nadeau and William-Jones, 2008; Rodriguez et al., 2008).

In the present study we applied a regional scale atmosphere-chemistry/aerosol numerical model called REMOTE (Langmann, 2000; Langmann et al., 2008) over Central America focussing on Nicaraguan volcanic emissions for month long simulation periods during the dry and wet seasons of 2003 to investigate the meteorological influence on the seasonal and diurnal variability of the dispersion of volcanic emissions as well as the differences between source and net volcanic fluxes. To our knowledge, numerical dispersion modelling of volcanic emissions including atmospheric chemistry has not been done before in this region of the world. It contributes to an improved understanding of the atmospheric and environmental impacts of the continuous degassing from Nicaraguan volcanoes.

2. Model description and parameterisation

The regional scale three-dimensional on-line atmosphere-chemistry/aerosol model REMOTE (Regional Model with Tracer Extension, <http://www.mpimet.mpg.de/en/wissenschaft/modelle/remote.html>) (Langmann, 2000; Langmann et al., 2008) is one of the few regional climate models that determines the physical, photochemical and aerosol state of the atmosphere at every time step thus offering the possibility to consider trace species effects on climate as well (e.g. Langmann, 2007). Besides various studies on the dispersion and chemical transformation of anthropogenic and natural emissions, REMOTE has also been applied to study the dispersion of volcanic emissions in Indonesia (Pfeffer et al., 2006). The dynamical part of the model is based on the former regional weather forecast system of the German Weather Service (Majewski, 1991) which is using a hydrostatic assumption for the vertical pressure gradient. In addition to the German Weather Service physical parameterisations, those of the global climate model ECHAM-4 (Roeckner et al., 1996) have been implemented in REMOTE and are used for the current study.

REMOTE is applied with 20 vertical layers of increasing thickness between the Earth's surface and the 10 hPa pressure level (approximately 25 km) using terrain following hybrid pressure-sigma coordinates. The model domain from 81–89° W and 9–17° N covers parts of Central America, the Caribbean Sea and the Pacific Ocean with Nicaragua in the focus (Fig. 2). The domain is subdivided into 81 × 81 grid boxes of 0.1° resolution (approximately 10 km). REMOTE is

Table 1
Volcanic sulphur emissions according to Mather et al. (2006).

Volcano	Latitude (°N)	Longitude (°W)	Height (m asl)	Model height (m asl)	SO ₂ emissions (Mg/day)	Sulphate emissions (Mg/day)
San Miguel (El Salvador)	13.434	88.269	2130	1474	260	13
San Cristobal (Nicaragua)	12.702	87.004	1745	613	590	29.5
Telica (Nicaragua)	12.602	86.845	1061	334	84	4.2
Momotombo (Nicaragua)	12.422	86.540	1297	404	73	3.65
Masaya (Nicaragua)	11.984	86.161	635	551	790	39.5
Arenal (Costa Rica)	10.463	84.703	1657	980	110	5.5
Poas (Costa Rica)	10.200	84.233	2708	1856	500	25

initialised at the first time step using meteorological analysis data of the European Centre for Medium Range Weather Forecast (ECMWF), which are updated at the lateral model boundaries every 6 h. Inside the model domain, prognostic equations are solved to determine the physical and chemical state of the model atmosphere with a time step of 1 min.

SO₂ emissions from the volcanoes San Miguel, San Cristobal, Telica, Momotombo, Masaya, Arenal and Poas are prescribed as constant fluxes according to the 1972–1997 mean as published by Mather et al. (2006) (Table 1). Volcanic sulphate fluxes are assumed to make up 5% of SO₂ fluxes according to Allen et al. (2002) and are released from the volcanoes in addition (Table 1). Both are emitted into the first model layer above ground into that model grid box which is closest to the volcano location. Volcanic ash emissions are not taken into account. Due to the chosen model resolution of 10 km, the geographic topography is smoothed in the model (Fig. 2) so that the volcano height in the model is lower than in reality (see Section 3.3). Sulphate formation in the atmosphere occurs via oxidation of SO₂ in the aqueous phase and in the gas phase. Sulphate production in the aqueous phase is determined dependent on pH via oxidation by H₂O₂, O₃, methylhydrogenperoxide, peroxyacetic acid and catalysed by Fe³⁺ and Mn²⁺ (Walcek and Taylor, 1986). Cloud water pH is determined solving iteratively an ion balance which is continuously maintained (Walcek and Taylor, 1986). Photochemical production and loss is determined using the RADM II photo-chemical scheme (Stockwell et al., 1990) with 163 chemical reactions in the gas phase including a wide range of hydrocarbon degradation reactions. The diurnal cycle of photolysis rates is calculated as described by Madronich (1987) and the influence of clouds is considered according to Chang et al. (1987). The information of the spatial variability of anthropogenic, biogenic and vegetation fire emissions over Central America in 10 km resolution as chosen for the current model application is not available so that photo-oxidant precursor concentrations (e.g. NO, CO) are prescribed as constant background concentrations to enable the model to simulate reasonably the diurnal cycle of photo-oxidants like

OH, H₂O₂ which are important for sulphate formation. SO₂ and sulphate undergo transport processes (horizontal and vertical advection (Smolarkiewicz, 1983), transport in convective clouds (Tiedtke, 1989), and vertical turbulent diffusion (Mellor and Yamada, 1974)) and are removed from the atmosphere by dry and wet deposition. Dry deposition fluxes of SO₂ and sulphate are determined after Wesley (1989) and Walcek et al. (1986). Wet deposition of sulphate is computed according to Walcek and Taylor (1986) by integrating the product of the grid-averaged precipitation rate and the mean cloud water concentration which is determined from cloud base (first layer above the surface containing more than 0.001 g/kg liquid water) to cloud top (highest level exceeding an amount of 0.001 g/kg liquid water) for fair weather clouds and from the surface to cloud top for raining clouds. Scavenging efficiencies are based on Kasper-Giebl et al. (2000). The described model set-up is used for the basic model simulation which is discussed in Sections 3.1 and 3.2. Small modifications in the basic model set-up are introduced for sensitivity studies, which are presented in Section 3.3.

3. Model results and discussion

3.1. Comparison with observation data

As we apply a regional atmospheric model, we first evaluate the ability of the model to reproduce the meteorological conditions as those considerably influence modelled atmospheric trace species distributions.

The annual climate cycle in Central America is characterised by minor fluctuations in surface temperature but well defined differences in precipitation. Throughout the year, Nicaragua experiences a dry (from about December to April) and a wet season (from about May to November) (Fig. 3a). The summer rainy season is characterised by a bimodal distribution in precipitation, with maxima in June and September–October and a relative minimum during July–August (Magaña et al., 1999). This relative minimum in convective activity and precipitation is known as midsummer drought and presents a unique climatological

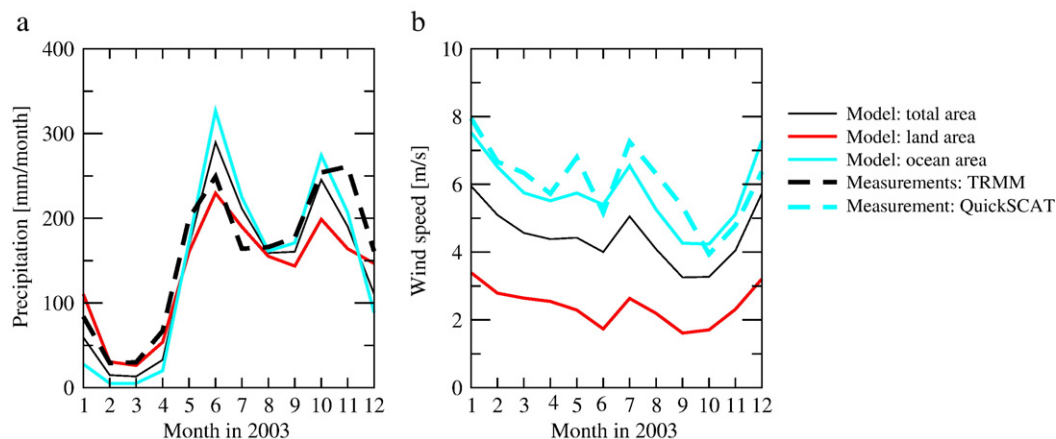


Fig. 3. a) Monthly precipitation from 81–89° W and 9–17° N and b) average 10 m wind speed from 81–89° W and 9–17° N as determined by REMOTE and measured by TRMM and QuikSCAT during 2003.

phenomenon in the tropical regions around 10–20° north or south of the equator (Magaña et al., 1999; Curtis, 2004). REMOTE is able to reproduce the above mentioned monthly variability and also the variability in magnitude of precipitation throughout the year 2003 with only a slight shifting when compared with measurements of the TRMM satellite (<http://daac.gsfc.nasa.gov/TRMM/index.shtml>) (Fig. 3a) for the area between 81–89° W and 9–17° N. During the dry season, modelled precipitation over the ocean is smaller than precipitation over land, whereas it is reverse during the wet season with a much higher land–sea contrast.

Low-level circulation in Central America is mainly affected by the northerly trades, the southerly trades and the winds blowing from the Gulf of Mexico to the Pacific through gaps in the cordillera with the Papagayo jet crossing the mountain gap in the lowlands of central Nicaragua. Strong off-shore winds occur in the lee of this mountain gap in boreal winter with a maximum in January. It weakens during the wet season, but there is a slight strengthening of the Papagayo jet during July–August which is in phase with the midsummer drought in Central America (Romero-Centeno et al., 2007). REMOTE simulated wind speed at 10 m height over ocean areas during the year 2003 reasonably reproduces this annual cycle and agrees well with 10 m wind speed obtained from SeaWinds on QuikSCAT (<http://winds.jpl.nasa.gov/missions/quikscat>) over the ocean (Fig. 3b). Modelled 10 m wind speed over continental areas is much lower, but follows the same annual cycle.

During March–April 1998 and February–March 1999 mean concentrations of SO₂ measured up to 44 km downwind from Masaya volcano in Nicaragua range from ~5 to 600 µg/m³ (about 2 to 230 ppbv) (Delmelle et al., 2002). Fig. 4a shows the dispersion of near surface SO₂ released from Masaya volcano transported to the southwest by the

prevailing North-East wind conditions and related SO₂ dry deposition (Delmelle et al., 2001, 2002). When focussing on the two lowest isolines, it is obvious that the REMOTE model results can reproduce the measured dispersion pattern and magnitude (Fig. 4b). This holds for North-East wind conditions during the dry season as well as during the midsummer drought period. The lowest isoline shown in Fig. 4b is 5 ppbv SO₂ and 5 mg SO₂/m²/day, respectively, to separate the plume of Masaya from those of the adjacent volcanoes. When wind direction shows higher variability and when cloud water availability is enhanced, volcanic SO₂ emissions from Masaya volcano are less uniformly transported into one predominant direction and do not reach so far into the Pacific Ocean. Close to the crater of Masaya volcano as well as in the Las Sierras highlands, SO₂ concentrations exceed by far European air quality standards which regulate that a threshold concentration of 125 µg SO₂/m³ may not be exceeded more than three times per calendar year (Directive 2008/50/EC, <http://eur-lex.europa.eu/LexUriServ/LexUriServ.do?uri=CELEX:32008L0050:EN:NOT>).

Altogether, the comparisons with observation data presented in this section give confidence in the ability of the REMOTE model to reasonably simulate the dispersion of volcanic emissions in Nicaragua. Unfortunately, there are only a few trace species measurements from volcanic emissions available in Nicaragua, so that a further evaluation of REMOTE model results is not possible.

3.2. Seasonal and diurnal variability of the dispersion of volcanic emissions

REMOTE monthly mean model results for the dry and wet season are illustrated in Fig. 5. For this purpose, the months of June 2003 and December 2003 have been chosen as representative for the wet and

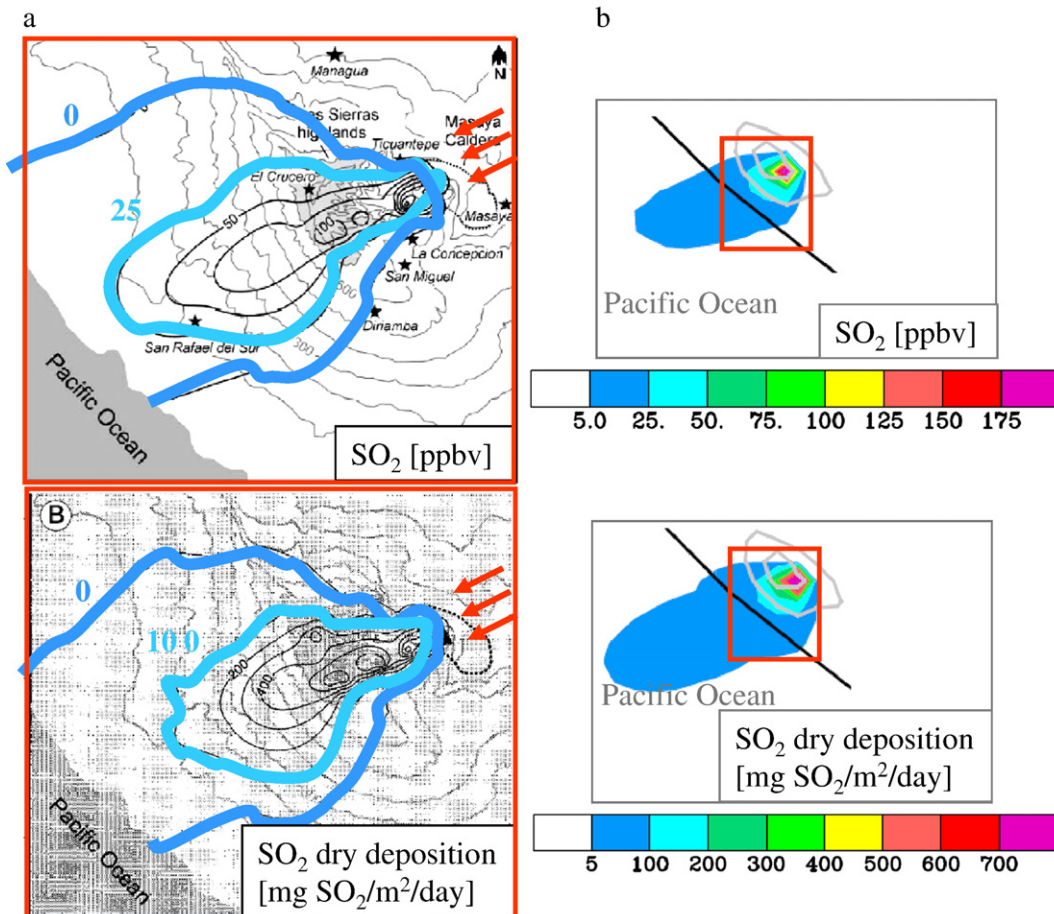


Fig. 4. a) Measured (Delmelle et al., 2001, 2002) and b) modelled (December 2003) near surface SO₂ in ppbv and SO₂ dry deposition in mg SO₂/m²/day. The measurement area is indicated as red box. The terrain height around Masaya volcano is indicated with grey isolines in 250 and 500 m height. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

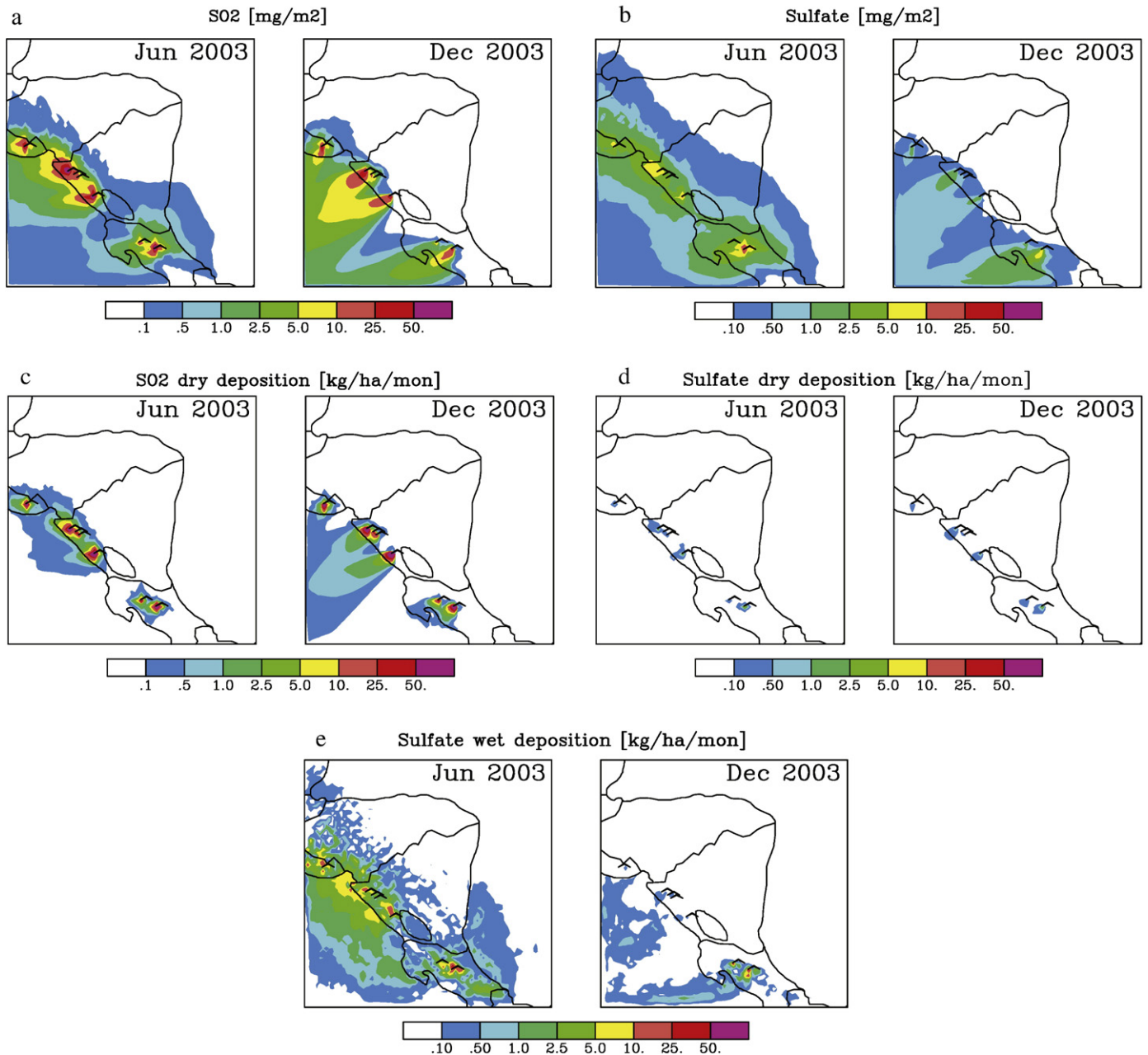


Fig. 5. REMOTE monthly mean model results during 2003 for a) SO₂ column concentration, b) sulphate column concentration, c) SO₂ dry deposition, d) sulphate dry deposition and e) sulphate wet deposition representative for the dry and wet seasons (December and June 2003, respectively). The approximate location of the volcanoes is indicated.

dry season, respectively. It should be noted, that the conditions during the midsummer drought in July 2003 are very similar to those of the dry season during December 2003. Modelled atmospheric column concentrations of SO₂ which are comparable with those measured by COSPEC or DOAS, take into account night time concentrations as well.

During the dry season with typically North-Easterly wind directions, the prevailing dispersion direction is to the South-West over the Pacific Ocean. High concentrations of SO₂ and related high dry deposition fluxes of SO₂ occur only close to the volcanoes (Fig. 6), a dilution and combination of the volcanic plumes takes place in the downwind direction over the Pacific Ocean (Fig. 4). From the two volcanoes Poas and Arenal in Costa Rica reduced SO₂ transport is visible in SO₂ column concentrations and SO₂ dry deposition fluxes compared to the volcanoes in Nicaragua, but considerably more sulphate formation

takes place close by the volcanoes and along the transport pathway. The reasons are much more humid conditions favouring sulphate formation in the aqueous phase and associated wet deposition, with wet deposition of sulphate clearly dominating over dry deposition as the major removal process.

During the wet season, more variable wind conditions lead to reduced dispersion of SO₂ over the Pacific Ocean and increased dispersion inland. The availability of liquid water in the atmosphere favours sulphate formation via aqueous phase oxidation and represents another limitation for the dispersion of SO₂. Strong precipitation removes sulphate quickly from the atmosphere by wet deposition.

Modelled time series at Masaya volcano are shown in Fig. 6 in one hour resolution starting at local midnight. Temperature in 2 m height undergoes a pronounced diurnal cycle with up to 9 K differences between day and night time temperatures. Higher night time temperatures

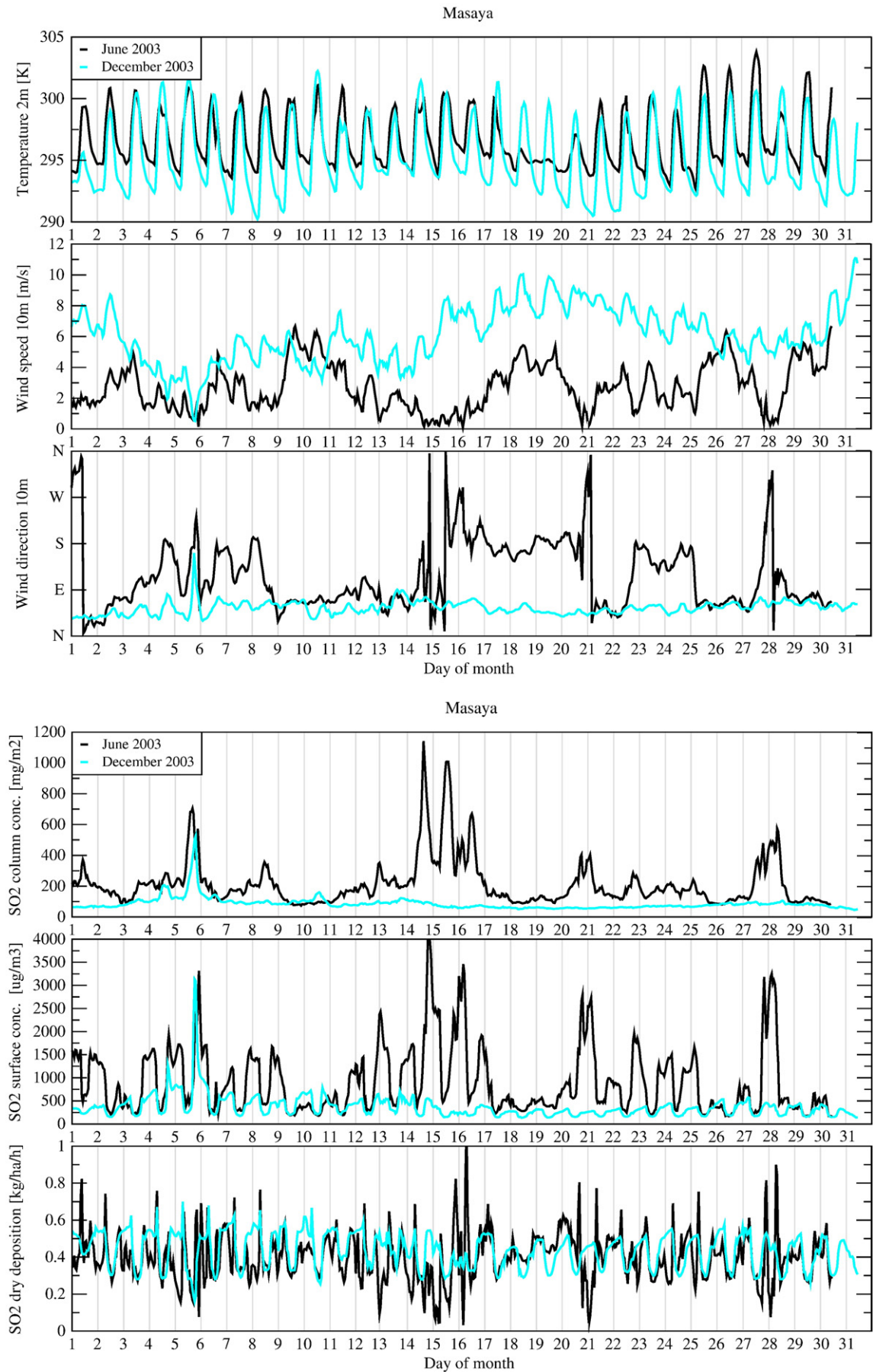


Fig. 6. REMOTE 1 h time series during June (black line) and December 2003 (light blue line) at Masaya volcano. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

during June 2003 compared to December 2003 are caused by enhanced cloud coverage during the wet season and associated storage of heat in the planetary boundary layer. Wind speed maxima in 10 m height occur during day time and go in phase with temperature maxima, in particular during the second half of December 2003 when day–night differences reach up to 4 m/s. Wind speed during December 2003 is generally higher than during June 2003. Wind direction in 10 m height is more variable during June 2003, whereas during December 2003 nearly constant North-Easterly winds are blowing. There is no pronounced diurnal cycle visible for wind direction but a slight tendency for a more easterly component in the late afternoon during December 2003.

The SO₂ column concentration at Masaya volcano is higher during June 2003 compared to December 2003 as dispersion is increased in

December 2003 due to higher wind speed. The highest concentrations are clearly correlated with low wind speeds. A continuous diurnal cycle does not occur. Looking at the SO₂ concentration in surface air, a pronounced diurnal cycle becomes visible with the minimum concentrations around noon. The maximum concentrations are reached during night time when wind speed is lower and the emissions are trapped in the night time boundary layer close to the surface thus favouring accumulation and removal by dry deposition. Dry deposition of SO₂ follows a pronounced diurnal cycle during December 2003 with a minimum during day time as the emissions are efficiently mixed within the planetary boundary layer. The diurnal cycle is less regular during June 2003. Such irregularities are caused by thin inversion layers which can develop near the

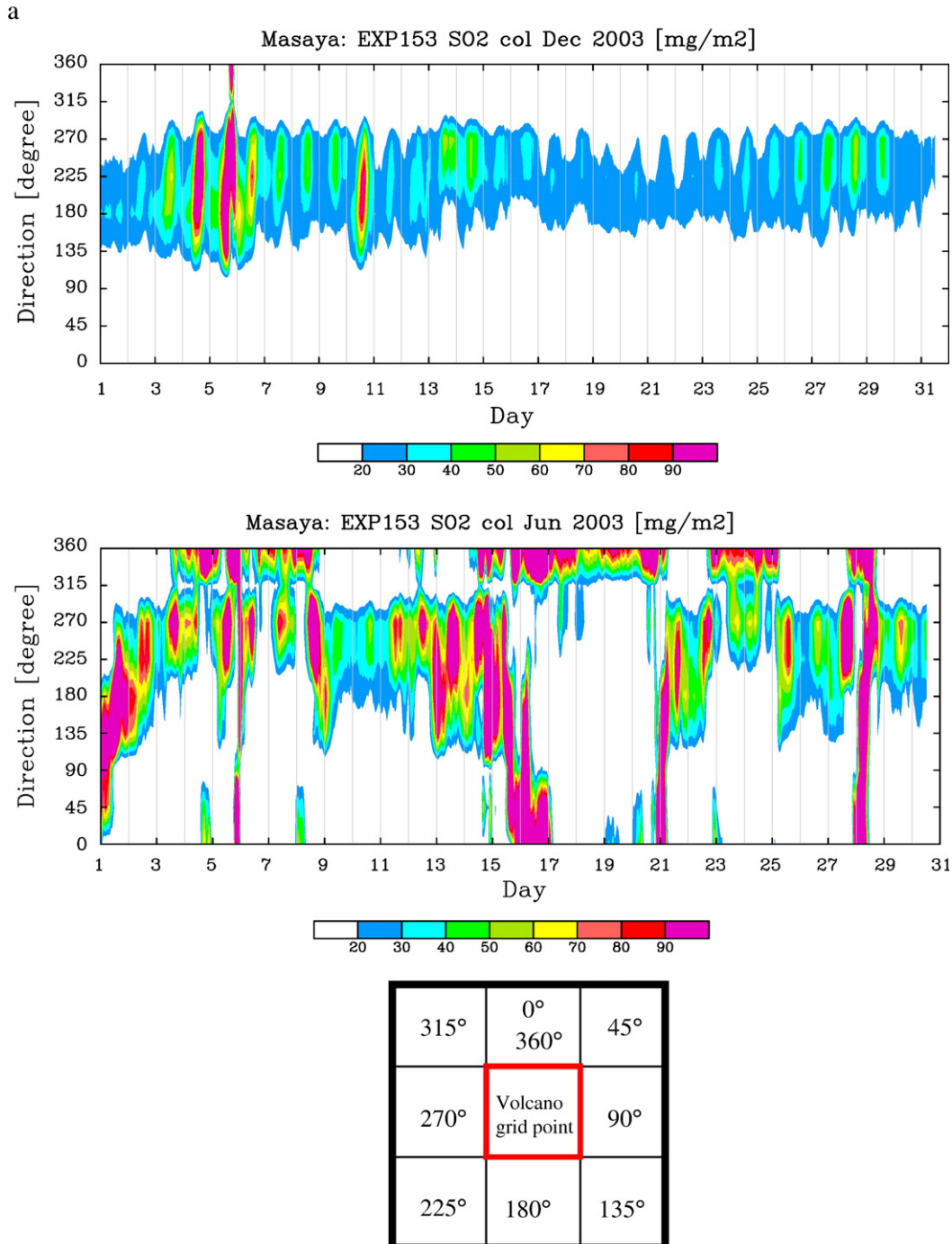


Fig. 7. REMOTE 1 h time series of a) SO₂ column concentration and b) SO₂ dry deposition during June and December 2003 around Masaya volcano.

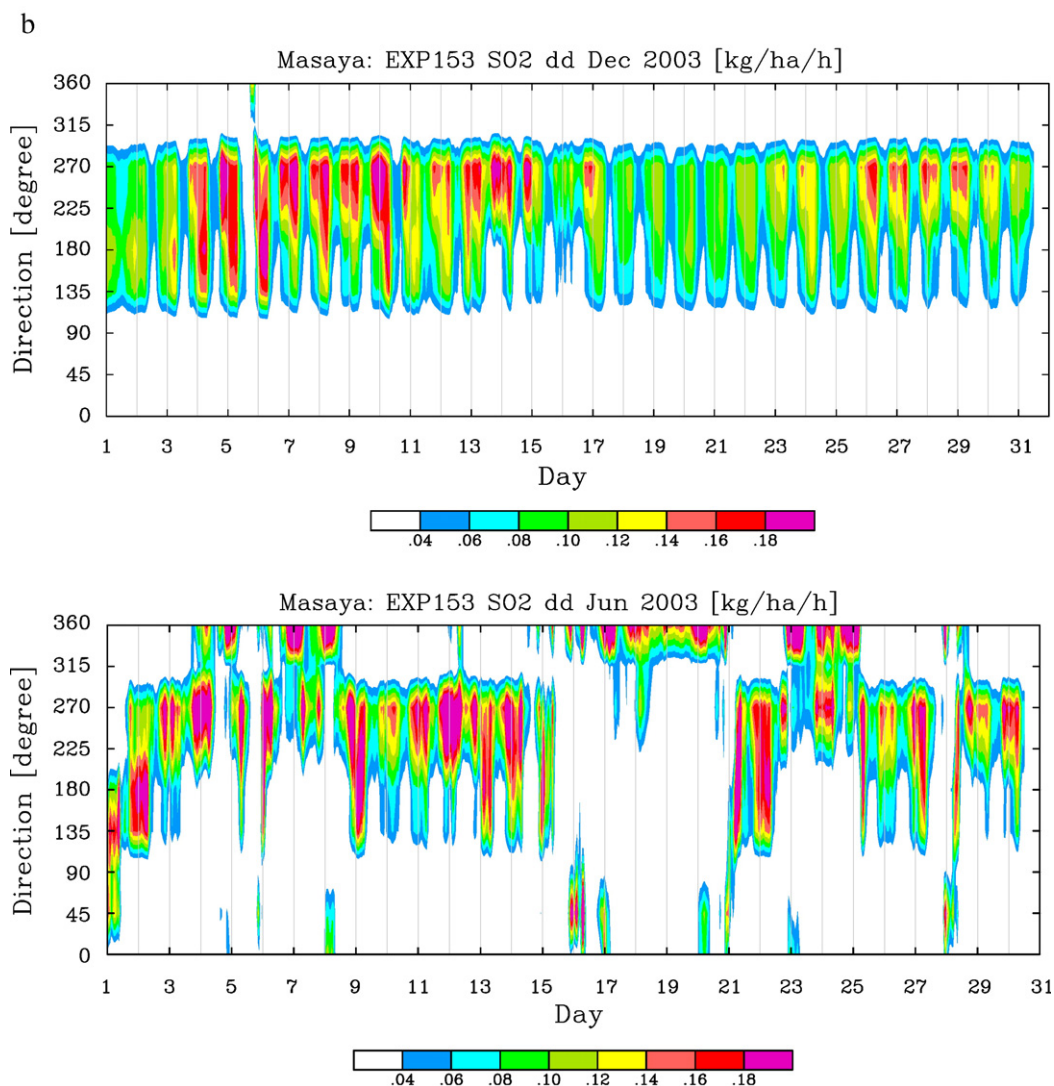


Fig. 7 (continued).

surface, thereby inhibiting the removal of atmospheric SO₂ by dry deposition.

The time series data in the vicinity of Masaya volcano reveal another diurnal cycle. Atmospheric SO₂ concentrations and dry deposition fluxes of SO₂ in the eight grid boxes around Masaya volcano are illustrated every hour in Fig. 7 by distributing the data vertically, starting at the northern neighbouring grid box and proceeding clockwise. During December 2003, the dry season, the dispersion of SO₂ around Masaya volcano follows a very regular diurnal cycle with a minimum column concentration during night and a maximum concentration around noon usually in the West–Southwest direction. The variability of the modelled SO₂ column concentration during day time is relatively small, similar to observations reported by McGonigle et al. (2004). Maximum dry deposition occurs during night, starting in the West–Southwest direction while in the early morning enhanced SO₂ dry deposition is also found in the more southern sector. Sulphate column concentration follows closely the temporal and spatial dispersion patterns of SO₂, with about 5% of the SO₂ values. This is because during the dry season primary volcanic emissions of sulphate dominate over formation processes in the atmosphere close to the volcanoes. The situation is completely different during the wet season: a very irregular dispersion of SO₂ column concentration and SO₂ dry deposition occurs due to shifting wind direction. As sulphate formation via aqueous phase oxidation and its removal from the atmosphere by wet deposition occurs also close to the

volcanoes, the dispersion patterns of SO₂ and sulphate around the volcanoes differ during the wet season.

10 km downwind of Masaya volcano, the net SO₂ flux (what can be determined from COSPEC or DOAS measurements) can be estimated from the model results by the prescribed SO₂ source flux (790 Mg/day SO₂, see Table 1) minus the dry deposition flux. When focussing on the dry season, loss of SO₂ via sulphate oxidation is negligible as indicated from model simulation results and from measurements (McGonigle et al., 2004; Nadeau and William-Jones, 2008). For the analysis of the model results, the source grid box and the surrounding eight grid boxes are taken into account. In this area, dry deposition of SO₂ represents 22% of the source flux independent of season resulting in a net SO₂ flux at Masaya volcano of only 615 Mg/day in the model simulation. Despite the associated uncertainties resulting e.g. from the chosen horizontal model resolution, this rough estimate reveals a considerable difference in the net and source fluxes of SO₂ from Masaya volcano. SO₂ flux measurements by

Table 2
Introduction of sensitivity experiments.

Abbreviation	Brief description
Flux run	Release of doubled volcanic emissions
Pulse run	Release of volcanic emissions as 1/2 h pulsed fluxes
Altitude run	Release of volcanic emissions at 'actual' volcanic height

active spectroscopy are carried out during day light hours only. Therefore, measured net and source fluxes of SO_2 at Masaya volcano during the dry season (McGonigle et al., 2004; Nadeau and William-Jones, 2008) are only slightly different, because dry deposition is relatively small during day time, it peaks during the night (Fig. 7). Dry and wet deposition fluxes of sulphate around Masaya volcano make up only 2% of the sulphate source flux during the dry season resulting in a nearly identical net and source flux of sulphate.

3.3. Sensitivity studies

The sensitivity studies introduced in this section are based on simple modifications of the assumptions in the REMOTE model to trace volcanic emissions. These simulations aim to further illuminate simulated dispersion patterns of volcanic emissions in Nicaragua. The abbreviations of the sensitivity studies and a brief description are given in Table 2.

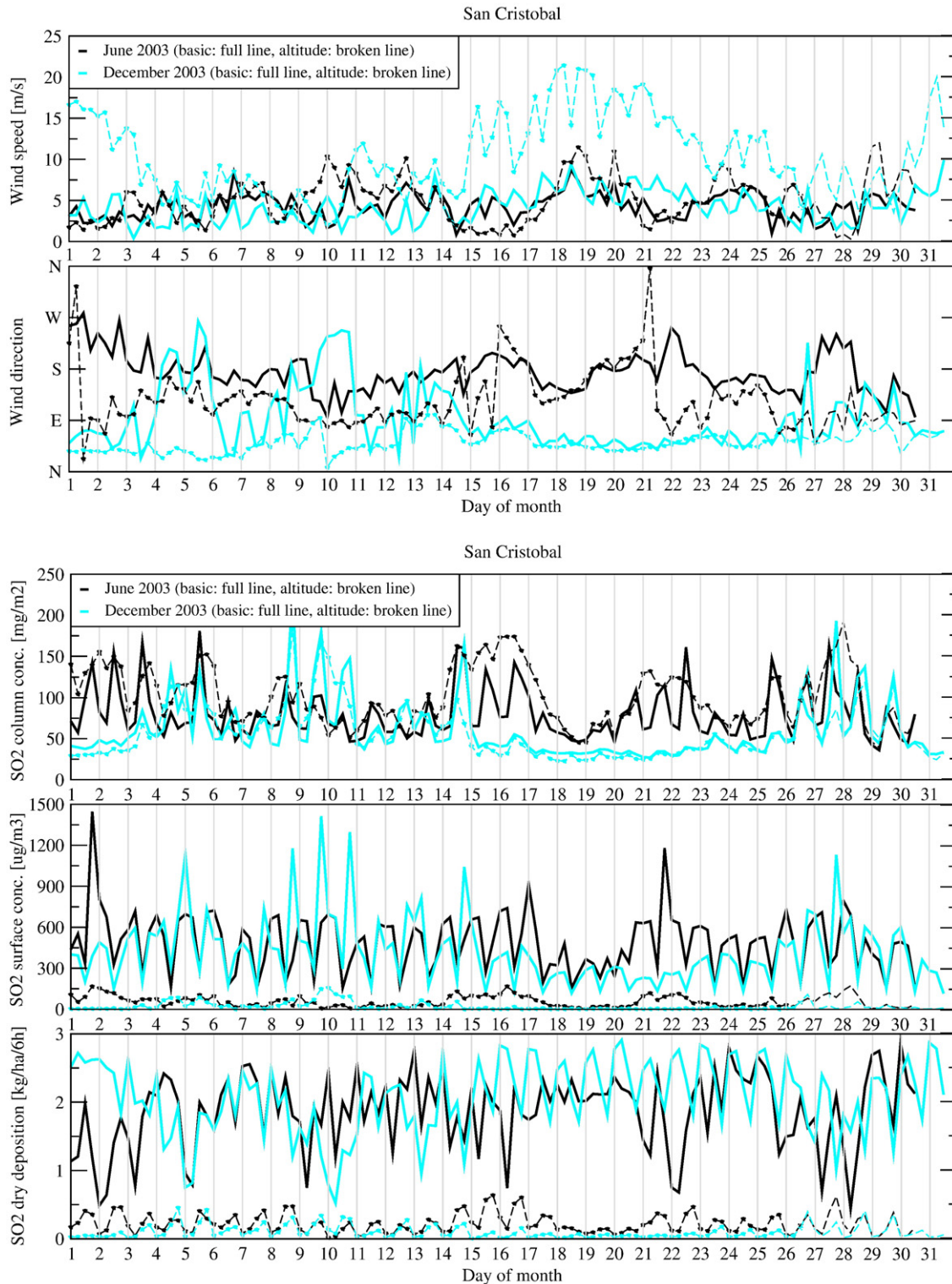


Fig. 8. REMOTE 6 h time series at San Cristobal during June (black line) and December 2003 (light blue line) for the basic model simulation (full line) and the altitude simulation (broken line). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The flux run demonstrates a strong linearity in the dispersion patterns and magnitude of the volcanic emissions: doubled volcanic emission fluxes result in approximately doubled column concentrations, surface concentration and deposition fluxes of SO₂ and sulphate. This strong linearity should be interpreted with caution due to the lack of anthropogenic, biogenic and vegetation fire emission data in appropriate spatial resolution. Therefore, photo-oxidant precursor concentrations have been prescribed (Section 2) and held constant throughout the model simulations. Under these idealised assumptions, limitations in photo-oxidant precursor and photo-oxidant concentrations are avoided and thereby explain the independency of SO₂ oxidation to sulphate on photo-oxidant availability.

The release of volcanic emissions as 1/2 h pulsed flux aims to demonstrate the influence of an accumulation of emissions in a volcanic crater during a certain time interval before the emissions are transported across the crater rim. This sensitivity study is carried out because pulsating fluxes have been observed at e.g. San Cristobal. During one 1/2 h pulse, the same amount of volcanic emissions is released as it is released continuously at every model time step in the basic model simulation during half an hour. In the monthly mean, SO₂ and sulphate concentration at the surface and throughout the atmosphere are considerably enhanced only directly at the volcanoes, slightly reduced directly around the volcano and without considerable modifications in the dispersion pattern further away. This study indicates the need to average over reasonable time periods when measuring emission fluxes close by a volcano.

In the altitude run, volcanic emissions are released into the actual volcanic height. This is done because the height of the cone-shaped volcanoes like San Cristobal is underestimated (Table 1) by the REMOTE model when using a horizontal resolution of 10 km, which leads to a smoothed topography (Fig. 2). Only the height of Masaya volcano is well captured by the model, so that there are no differences for Masaya in the basic and altitude simulation results as in both cases emissions are released into the first vertical model layer which is in about 40 m height. For the other six volcanoes, the emissions are released into the vertical model layer 4 or 5 in the altitude simulation which are between about 600 and 1000 m above the volcanoes, simulating a situation where the plume height above topography increases fast with increasing downwind distance. Due to the higher wind speeds in these altitudes, SO₂ and sulphate column concentration are slightly increased in the lee direction of the volcanoes in comparison with the basic model run, whereas near surface concentration and deposition fluxes are considerably reduced in particular close to the volcanoes. Time series at San Cristobal (Fig. 8) reveal the differences in wind speed and direction in the altitude and basic model simulation. In the altitude run, these resemble much more those at Masaya volcano (Fig. 6), but with significantly higher wind speeds at San Cristobal, both reflecting the large scale circulation. SO₂ column concentrations show a less pronounced diurnal cycle and SO₂ surface concentrations and dry deposition fluxes are much lower in the altitude run. Compared with the time series at Masaya volcano, the diurnal cycle of near surface concentration is nearly gone at San Cristobal whereas SO₂ dry deposition shows maximum values during day time compared to night time at Masaya volcano. The explanation is that during day time the concentrations are efficiently mixed within the planetary boundary layer so that even in the altitude run they can reach the surface and be partly removed from the atmosphere by dry deposition. During night time, the emissions are released above the surface inversion layer in the altitude simulation thus preventing dry deposition. Dry deposition fluxes of SO₂ around San Cristobal make up only 3% of the SO₂ source flux in the altitude simulation pointing to negligible differences in the SO₂ net and source fluxes for cone-shaped volcanoes. In summary, even in the altitude run, the volcanic emissions are still released into the planetary boundary layer (PBL). The use of the actual volcanic height leads to a more realistic reproduction of the conditions at the cone-shaped volcanoes of Nicaragua.

4. Conclusions and outlook

Numerical model simulations of the dispersion of volcanic emissions as presented here are complementary to field measurements carried out by COSPEC or DOAS, as model applications can help to interpret measurements and can also help to fill the gaps during periods where observations are not possible by providing continuous information about the diurnal and seasonal cycles of dispersion patterns of volcanic emissions. However, model results need to be carefully interpreted due to the necessary assumptions and simplifications in the model set-up. In the present study we applied the regional scale atmosphere-chemistry/aerosol numerical model REMOTE (Langmann, 2000; Langmann et al., 2008) over Central America focussing on Nicaraguan volcanic emissions for month long simulation periods during both, the dry and wet seasons of 2003 to investigate the meteorological influence on the seasonal and diurnal variability of the dispersion of volcanic emissions. For this purpose constant volcanic emissions were assumed.

During the dry season the dominant dispersion direction is to the South-West over the Pacific Ocean whereas during the wet season oxidation to sulphate and more variable winds lead to reduced dispersion of SO₂ over the Pacific Ocean and increased dispersion inland. SO₂ concentration in surface air and related dry deposition at Masaya volcano follow a pronounced diurnal cycle with maximum concentrations and fluxes during night time. As the height of Masaya volcano is well represented by the model and comparisons of near surface SO₂ concentrations and SO₂ dry deposition fluxes around Masaya volcano are in very good agreement with field measurements during the dry season, there is confidence in the ability of the REMOTE model to reasonably simulate the dispersion of the volcanic emissions from Masaya volcano, and probably of other volcanoes with similar low topographic features in a complex terrain. At Masaya volcano, the net SO₂ flux which is usually determined a few kilometres downwind the volcano differs from the actual SO₂ source flux released from the volcano due to a considerable loss of SO₂ by dry deposition along the plume transport pathway.

Generally, there is a close connection in the development of the dispersion patterns of emissions released from low level volcanoes into the PBL and the diurnal cycle of the PBL (Stull, 1988) with the PBL covering the lowest about 2 km of the atmosphere which is influenced by surface heating (cooling) and turbulence (inversion) during the day (night).

Due to the horizontal model resolution of about 10 km, the height of the other active cone-shaped volcanoes in Nicaragua is underestimated by the model. In a sensitivity simulation it is shown that the release of volcanic emissions into the actual volcanic height changes the dispersion and deposition patterns in particular close to the volcanoes thereby indicating a negligible difference in the SO₂ net and source fluxes for cone-shaped volcanoes. The application of higher resolution non-hydrostatic local models could provide a better reproduction of the volcano topography and thereby volcanic injection height, however such numerical models are still consuming huge amounts of computer time thereby limiting the length of the application periods. Further studies should also take into account the influence of volcanic plume rise on the dispersion of volcanic emissions and the influence of volcanic ash on plume development.

Acknowledgements

The authors would like to thank the German Science Foundation DFG for funding this work (Grant HO 1411/21-1) and the German Climate Computing Centre DKRZ for super computer access for model simulations. We also would like to gratefully acknowledge the helpful comments of the two reviewers of the manuscript. The availability of TRMM, QuikSCAT and ECMWF data is gratefully acknowledged. This publication is also contribution no. 163 of the Collaborative Research Center 574

“Volatiles and Hazards in Subduction Zones” at Kiel University. T. H. received additional support from the EU-funded project NOVAC.

References

- Allen, A.G., Oppenheimer, C., Fern, M., Baxter, P.J., Horrocks, L.A., Galle, B., McGonigle, A.J.S., Duffell, H.J., 2002. Primary sulfate aerosol and associated emissions from Masaya Volcano, Nicaragua. *J. Geophys. Res.* 107 (D23), 4682. doi:10.1029/2002JD002120.
- Andres, R.J., Kasgnoc, A.D., 1998. A time-averaged inventory of subaerial volcanic sulphur emissions. *J. Geophys. Res.* 103 (D19), 25251–25261.
- Bardintzeff, J.-M., McBirney, A.R., 2000. *Volcanology*, second edition. Jones and Bartlett.
- Chang, J.S., Brost, R.A., Isaksen, I.S.A., Madronich, S., Middleton, P., Stockwell, W.R., Walcek, C.J., 1987. A three-dimensional Eulerian acid deposition model: physical concepts and formulation. *J. Geophys. Res.* 92, 14681–14700.
- Curtis, S., 2004. Diurnal cycle of rainfall and surface winds and the mid-summer drought of Mexico/Central America. *Clim. Res.* 27, 1–8.
- Delmelle, P., Six, J., Bourque, C.P.A., Baxter, P.J., Garcia-Alvarez, J., Barquero, J., 2001. Dry deposition and heavy acid loading in the vicinity of Masaya Volcano, a major sulfur and chlorine source in Nicaragua. *Environ. Sci. Technol.* 35, 1289–1293.
- Delmelle, P., Stix, J., Baxter, P.J., Garcia-Alvarez, J., Barquero, J., 2002. Atmospheric dispersion, environmental effects and potential health hazards associated with the low-altitude gas plume of Masaya volcano, Nicaragua. *Bull. Volcanol.* 64, 423–434.
- Duffell, H.J., Oppenheimer, C., Pyle, D.M., Galle, B., McGonigle, A.F.S., Burton, M.R., 2003. Changes in gas composition prior to a minor explosive eruption at Masaya volcano, Nicaragua. *J. Volcanol. Geotherm. Res.* 126, 327–339.
- Frische, M., Garofalo, K., Hansteen, T.H., Borchers, R., 2006. Fluxes and origin of halogenated organic trace gases from Momotombo volcano (Nicaragua). *Geochem. Geophys. Geosyst.* 7, 5. doi:10.1029/2005GC001162.
- Graf, H.-F., Feichter, J., Langmann, B., 1997. Volcanic sulfur emissions: estimates of source strength and its contribution to the global sulfate distribution. *J. Geophys. Res.* 102, 10727–10738.
- Kasper-Giebl, A., Koch, A., Hitznerberger, R., Puxbaum, H., 2000. Scavenging efficiency of aerosol carbon and sulfate in super-cooled clouds at Mt. Sonnblick (3106 m a.s.l., Austria). *J. Atmos. Chem.* 35, 33–46.
- Langmann, B., 2000. Numerical modelling of regional scale transport and photochemistry directly together with meteorological processes. *Atmos. Environ.* 34, 3585–3598.
- Langmann, B., 2007. A model study of the smoke-haze influence on clouds and warm precipitation formation in Indonesia 1997/1998. *Atmos. Environ.* 41, 6838–6852. doi:10.1016/j.atmosenv.2007.04.050.
- Langmann, B., Graf, H.-F., 2003. Indonesian smoke aerosols from peat fires and the contribution from volcanic sulfur emissions. *Geophys. Res. Lett.* 30, 1547. doi:10.1029/2002GL016646.
- Langmann, B., Varghese, S., Marmer, E., Vignati, E., Wilson, J., Stier, P., O'Dowd, C., 2008. Aerosol distribution over Europe: a model evaluation study with detailed aerosol microphysics. *Atmos. Chem. Phys.* 8, 1591–1607.
- Madronich, S., 1987. Photodissociation in the atmosphere. I: Actinic flux and the effect of ground reflections and clouds. *J. Geophys. Res.* 92, 9740–9752.
- Majewski, D., 1991. The Europa Modell of the Deutscher Wetterdienst. Seminar Proceedings ECMWF, vol. 2, pp. 147–191.
- Magaña, V., Amador, J.A., Medina, S., 1999. The midsummer drought over Mexico and Central America. *J. Climate* 12, 1577–1588.
- Mather, T.A., Pyle, D.M., Tsanev, V.I., McGonigle, A.J.S., Oppenheimer, C., Allen, A.G., 2006. A reassessment of current volcanic emissions from the Central American arc with specific examples from Nicaragua. *J. Volcanol. Geotherm. Res.* 149, 297–311.
- McGonigle, A.J.S., Delmelle, P., Oppenheimer, O., Tsanev, V.I., Delfosse, T., Williams-Jones, G., Horton, K., Mather, T.A., 2004. SO₂ depletion in tropospheric volcanic plumes. *Geophys. Res. Lett.* 31, L13201. doi:10.1029/2004GL019990.
- Mellor, B., Yamada, T., 1974. A hierarchy of turbulence closure models for planetary boundary layers. *J. Atmos. Sci.* 31, 1791–1806.
- Nadeau, P.A., William-Jones, G., 2008. Apparent downwind depletion of volcanic SO₂ flux – lessons from Masaya Volcano, Nicaragua. *Bull. Volcanol.* 70. doi:10.1007/s00445-008-0251-9.
- Pfeffer, M.A., Langmann, B., Graf, H.-F., 2006. Atmospheric transport and deposition of Indonesian volcanic emissions. *Atmos. Chem. Phys.* 6, 2525–2537.
- Robock, A., 2000. Volcanic eruptions and climate. *Rev. Geophys.* 38, 191–219.
- Rodriguez, L.A., Watson, I.M., Edmonds, M., Ryan, G., Hards, V., Oppenheimer, C.C.M., Bluth, G.J.S., 2008. SO₂ loss rates in the plume emitted by Soufriere Hills volcano, Montserrat. *J. Volcanol. Geotherm. Res.* 173, 135–147.
- Roeckner, E., Arpe, K., Bengtsson, L., Christoph, M., Claussen, M., Dümenil, L., Esch, M., Giorgetta, M., Schlese, U., Schulzweida, U., 1996. The atmospheric general circulation model ECHAM-4: model description and simulation of present-day climate. MPI-Report No. 218. Max Planck Institute for Meteorology, Hamburg, Germany.
- Romero-Centeno, R., Zavala-Hidalgo, J., Raga, G.B., 2007. Midsummer gap winds and low-level circulation over the eastern tropical Pacific. *J. Climate* 20, 3768–3784.
- Simkin, T., Siebert, L., 1994. Smithsonian Institution, Misoula, Montana. Geoscience Press.
- Smolarkiewicz, P.K., 1983. A simple positive definite advection scheme with small implicit diffusion. *Mon. Wea. Rev.* 111, 479–486.
- Stockwell, W.R., Middleton, P., Chang, J.S., Tang, X., 1990. The second generation regional acid deposition model: chemical mechanism for regional air quality modelling. *J. Geophys. Res.* 95, 16 343–16 367.
- Stull, R.B., 1988. *An Introduction to Boundary Layer Meteorology*. Kluwer Academic Publisher.
- Tiedtke, M., 1989. A comprehensive mass flux scheme for cumulus parameterisation in large-scale models. *Mon. Wea. Rev.* 117, 1778–1800.
- Walcek, C.J., Taylor, G.R., 1986. A theoretical method for computing vertical distributions of acidity and sulfate production within cumulus clouds. *J. Atmos. Sci.* 43, 339–355.
- Walcek, C.J., Brost, R.A., Chang, J.S., Wesley, M.L., 1986. SO₂, sulfate and HNO₃ deposition velocities computed using regional landuse and meteorological data. *Atmos. Environ.* 20, 946–964.
- Wesley, M.L., 1989. Parameterization of surface resistances to gaseous dry deposition in regional-scale numerical models. *Atmos. Environ.* 23, 1293–1304.
- Williams-Jones, G., Rymer, H., Rothery, D.A., 2003. Gravity changes and passive SO₂ degassing at the Masaya caldera complex, Nicaragua. *J. Volcanol. Geotherm. Res.* 123, 137–160.