P1.4 OFFSHORE TRANSPORT EPISODES OF ANTHROPOGENIC SULFUR IN NORTHERN CHILE: POTENTIAL IMPACT ON THE STRATOCUMULUS CLOUD DECK

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1 INTRODUCTION

The world's most extended and persistent stratus deck is that located under the subtropical Pacific High off the coast of Northern Chile and Southern Peru. In this region large emissions of oxidized sulfur (SO_x=sulfur dioxide (SO_2)+sulfate (SO_4)) occur, both due to anthropogenic processes (e.g., Lefhon et al, 1999), mainly copper smelting, and natural processes as biogenic emissions along the Humboldt Current system (e.g., Scholes et al, 2003), and volcanic emissions (e.g., Anders and Kasgnoc, 1998).

On the synoptic scale, it has been shown that migratory highs drifting eastwards across southern Chile ahead of mid-troposphere ridges induce subsiding easterly flow off Central Chile (Garreaud et al, 2002; Garreaud and Rutllant, 2003, Rutllant and Garreaud, 2004) and the subsequent poleward propagation of atmospheric coastally trapped disturbances (e.g., coastal lows, CLs). These conditions are crucial for determining potential fluxes of biogenic sulfur (e.g., Hormazábal et al, 2001) and offshore dispersion of pollutants, particularly anthropogenic sulfur (e.g., Gallardo et al, 2002). Aerosol-cloud-drizzle interactions in the Southeast Pacific are at present a key issue in the assessment of changes in the radiative properties of the stratus cloud deck and their simulation in numerical models (Bretherton et al., 2004).

Here we explore the plausibility of a potential anthropogenic perturbation in the stratocumulus (Sc) deck off Northern Chile due to transport of sulfur emissions from copper smelting in connection with easterly wind events. This is done by performing simulations with a chemistry-transport-deposition model under strong easterly wind conditions. The dispersion pattern of sulfur emissions is then compared with cloud droplet radii observed from satellites.

2 DATA AND METHODOLOGY

Various data sources are considered in this study, namely rawinsonde data collected daily by the Chilean Weather Service at Cerro Moreno (23.43°S, 70.43°W, 137 m.a.s.l.), ERA-40 reanalysis fields (e.g., Simmons and Gibson, 2000*), dynamically interpolated meteorological fields and sulfur emissions from copper smelters and estimates of two power plants at the coast in Northern Chile. The meteorological interpolated fields capture the regional scale circulation patterns, including synoptic variations. Also the radiatively driven circulations as those that develop at the slopes of the Andes Cordillera are well described particularly over Northern Chile (Huneeus, 2003). Sulfur emissions from copper smelters located in Northern Chile: Chuquicamata, Potrerillos, Noranda, and Paipote, are by far the dominant anthropogenic sources of oxidized sulfur in the area. The first two, responsible for more than 80% of the smelter-derived SO_2 emissions in northern Chile, are located inland at about 2900 m a.m.s.l. For satellite-derived cloud properties, we consider here the so-called level two MODIS cloud products, i.e., effective particle radius, cloud top pressure, liquid water path and cloud cover of the available data for the period July 20-August 20, 2000. These data were downloaded from the MODIS web site: http://modis-atmos.gsfc.nasa.gov/MOD06_L2/index.html

Because of the location of the two main smelters, "strong easterly wind events" were defined as those when the easterly wind component at Antofagasta exceed 5 m/s at 700 hPa.

3 RESULTS

3.1 Synoptic conditions and transport patterns

"Strong easterly wind days" (SEDs), i.e., speeds in excess of 5 m/s at 700 hPa, tend to be evenly distributed in all seasons with a slight preference for fall, with 4 to 8 SEDs per year. No discernible bias towards either extreme of the ENSO cycle appears, except for the strong La Niña in 1999 when 14 SEDs, of a total of 82 in the 14 year period, occurred. The 82 SEDs were organized into 74 "events" (SEEs), considering consecutive qualifying days as a single event.

These SEDs happen in connection with ridging in the mid-troposphere with the ridge axis just west of the study area (S-SW winds aloft), as depicted in 500 hPa geopotential height and sea-level pressure (SLP) composites (Figure 1a). The onset of anomalous easterlies are preceded by anomalous warming between 850 and 900 hPa and anomalous drying above (not shown). These circulation conditions present a

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synoptic structure typical of the coastal troughing at the onset stage of coastal lows events farther south in Chile and have been extensively documented (Garreaud et al., 2002; Garreaud and Rutllant, 2003; Rutllant and Garreaud, 2004).

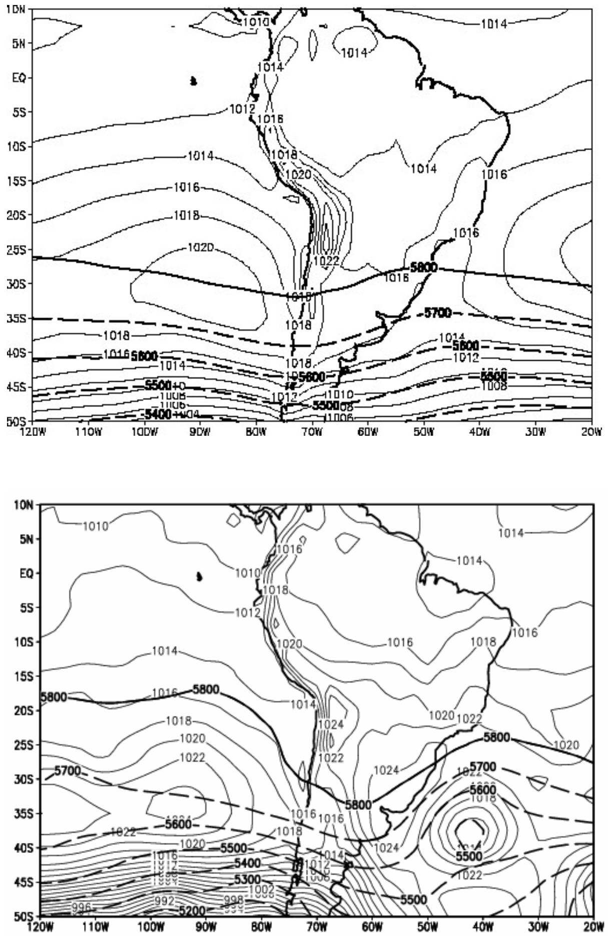

Figure 1. The upper panel (a) shows the composited surface pressure (thin lines) and 500 hPa geopotential contours (thick lines), considering 82 days of strong easterly winds at Antofagasta (23ºS). The lower panel (b) depicts the corresponding configuration for July 26th, 2000

As an example of a typical SED, we have chosen to perform a case study for the day of July 26, 2000 (Cf. next section). This day is representative of a general regional circulation pattern characterizing SEDs, as revealed by the actual 500 hPa geopotentials and SLP fields (Figure 1b), with a mid-troposphere ridge axis tilted in a NW-SE direction over Northern Chile and a surface trough between the subtropical anticyclone to the west and a migratory high east of the Andes.

3.2 Dispersion simulations

To illustrate the effects of the synoptic conditions identified above (a), we performed a numerical simulation using a transport/ chemistry/ / deposition off-line model, namely the Multi-scale Atmospheric Transport and Chemistry model (MATCH, Robertson et al, 1999). We used a similar model set-up to that applied in previous work (Olivares et al, 2002; Gallardo et al, 2002), except that the domain now spans from 30°S to 20° S and from 67°W to 75.45°W, and from the surface up to approximately 7 km height (i.e., 16 hybrid model layers). The emissions used and the deposition parameters chosen are shown in Table 1. A one month long simulation was conducted for a winter period between July 20 and August 19 2000. This period was chosen based on the availability of MODIS data.

Close to the sources and over land, SO_2 is, as expected, the prevailing form of oxidized sulfur, adding up to 80% of the total sulfur budget (not shown). On average, this fraction decreases off the coast where the partitioning is more evenly distributed between SO_2 and SO_4. This reflects the effects of oxidation processes and the lack of inputs of fresh SO_x by in situ sources.

Within the one-month period in which MODIS data were available, the SEE centered on July 26th was selected as a representative case (Cf. Figure 1). When transported off the coast on around July 26th 2000, SO_x remained more or less confined between the top of the Sc layer and up to 4 km above the surface (Cf. Figure 2). In this layer, SO_x mixing ratios increase in an order of magnitude compared with the average situation. The SO_2 transported offshore right above or below the MBL is efficiently incorporated into the Sc layer and subsequently oxidized to sulfate and wet deposited. These features are illustrated in Figure 2, where the vertical and horizontal distribution of oxidized sulfur are shown.

3.3 Satellite data

When considering the difference in cloud particle radius between days with easterly (July 20, 23, 25 and 25, 2000, 19 UTC) and westerly (July 21, 22, 24, 27, 29, 30 and August 1,2,3,4 and 5, 2000, 19 UTC) winds for the simulated period, a decrease in cloud droplet radii can be appreciated over some areas (Figure 3a). A negative anomaly in cloud droplet radii can be seen north of 28°S, and between 71ºW and 72ºW. Also, south of 25°S and west of 72°W such a decrease can be observed as well as north of 23ºS and west of 73ºW. However, those decreases in cloud droplet radii can be partly attributed to a concurrent decrease in liquid water content due to the dry easterly flow (Figure 3b). When excluding the areas where a negative liquid water anomaly is observed (Figure 3c), a non negligible portion of the domain keeps showing coherent patterns of positive SO_x burden anomalies (Figure 3d) and smaller droplet radii. Although this does not prove the anthropogenic impact over the optical properties of the status deck it provides a plausible explanation to the observed decrease in cloud droplet radius despite the increase in the liquid water at some areas.

Table 1. Emissions and deposition parameters used in the dispersion simulation.

Parameter\ Species	SO_2_S	SO_4_S
Emissions (in GgS/yr)		
Smelters		
Chuquicamata (22.32S, 68.92 W, 2850 m.a.s.l.)	100,7	5,3
Potrerillos (26.43S, 69.47W, 2850 m.a.s.l.)	40,9	2,2
Noranda (23.98S, 70.07 W, 1272 m.a.s.l)	19,0	1,0
Paipote (27.42S, 70.25 W, 540 m.a.s.l.)	12,4	0,7
Power plants		
Tocopilla (22.08ºS, 70.4ºW, 50 m.a.s.l.)	31,4	1,7
Huasco (28.5ºS, 71, 32ºW, 25 m.a.s.l.)	14,3	0,8
Dry deposition		
Over land (min/max) in cm/s	0,3/0,8	0,1/0,1
Over water in cm/s	0,5	0,05
Wet deposition (s-1· (mm· hour-1)-1)	$0,69x10^{-4}$	$2,78x10^{-4}$

4 CONCLUDING REMARKS

This study has explored the potential perturbation on the subtropical stratocumulus deck off Northern Chile by anthropogenic sulfur emissions (copper smelters) in connection with 74 strong easterly wind events (> 5 m/s at 700 hPa). within a 14 year period (1989-2002) In connection with the onset stage of coastal lows due to ridging in the middle troposphere, downslope flow over the western Andes is established (e.g., Garreaud et al, 2002). These easterly wind events appear to be quite common and evenly distributed throughout the year, reflecting the potential modification of the optical characteristics of the stratocumulus cloud deck.

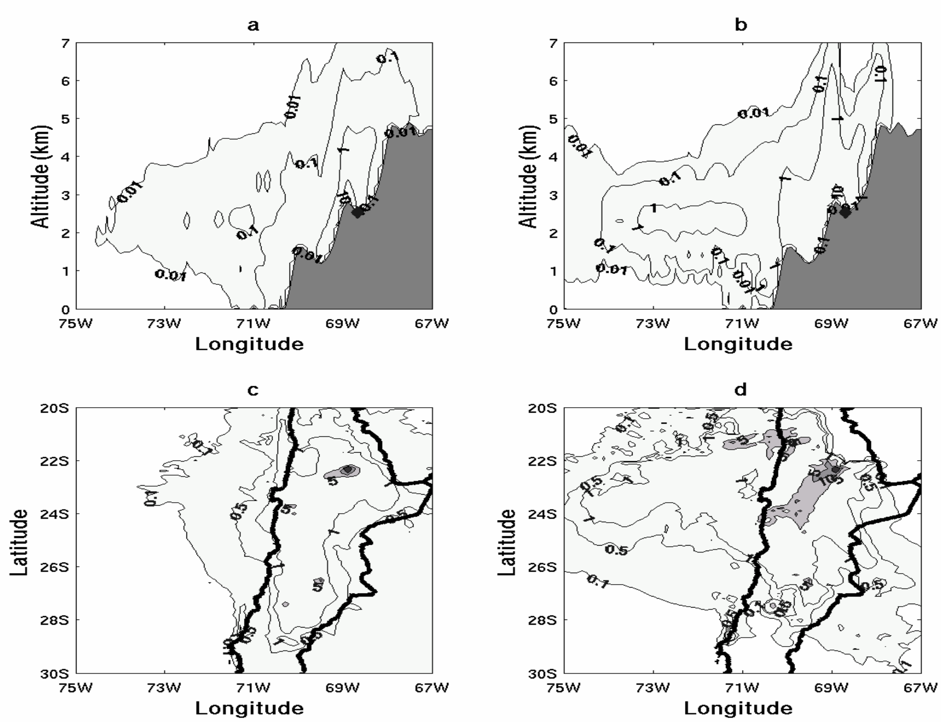

Figure 2. The upper panels (a and b) show cross-sections at latitude 23.3 ºS of mass mixing ratios of SO_x (in ppbm). To the left the monthly averaged distribution and to the right daily averages for July 26th are shown. The lower panels (c and d) show the burden, i.e., vertically integrated mass of SO_x (in mgS/m^2).

Simulations performed with a 3-D emission-transport-deposition model of an easterly wind event around July 26th 2000 show that, consequently with the synoptic configuration described above, the SO_x emitted from copper smelters located over the western slope of the Andes is transported offshore, remaining mostly within the layer between the top of the Sc and 4 km above the surface. Concomitantly, satellite retrievals indicate a decrease of cloud droplet radii despite the increase in cloud water content, which suggests a potential anthropogenic impact on the stratus deck. Although suggestive, these data and simulations do not prove nor quantify an anthropogenic perturbation of the optical properties in the stratocumulus deck off the Chilean coast. Other factors might also explain the observed changes in cloud droplet radii. For instance, biogenically produced aerosols, not only sulfur aerosols, may also induce changes, particularly considering that easterly wind events are generally associated with enhanced upwelling, near surface southerlies along the coast, also enhancing and air-sea gas exchange (e.g., Rutllant et al, 1998). Hence, in order to ascertain the validity of our hypothesis further research must be conducted.

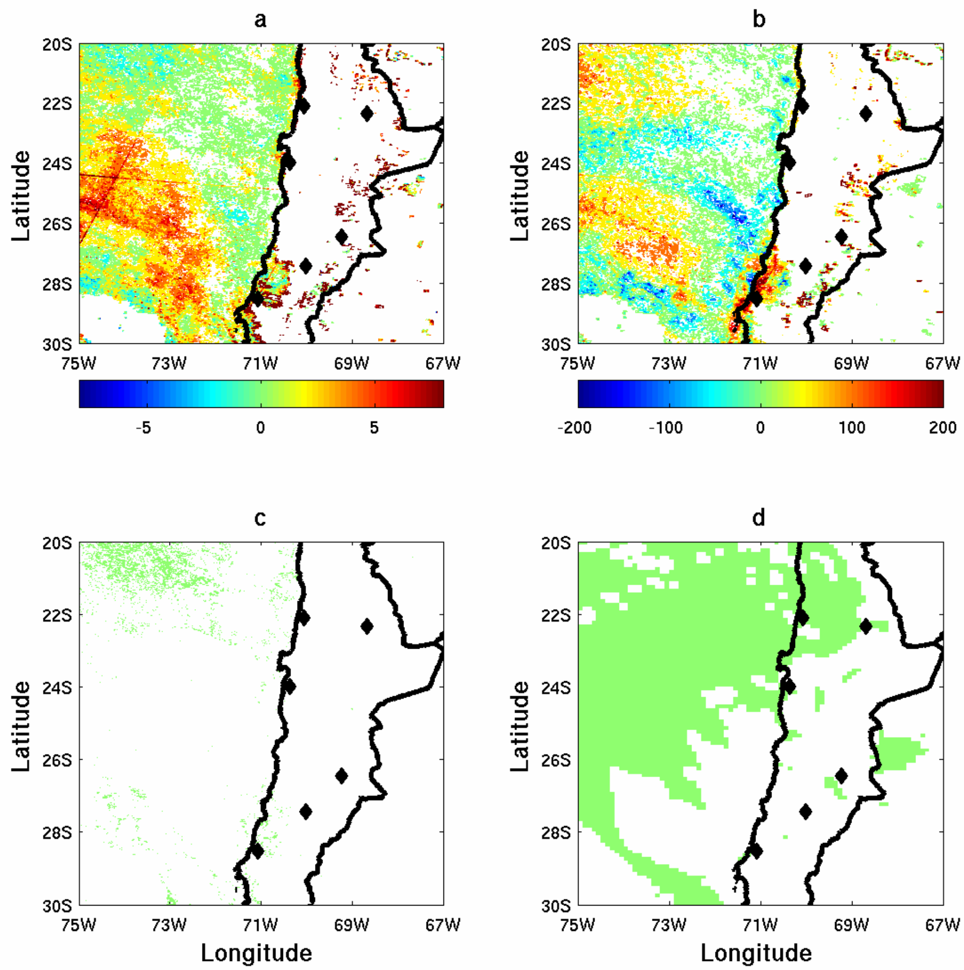

Figure 3. Difference in (a) cloud particle radii (μm) and (b) liquid water path (g/m^2) between easterly and westerly wind days. Also indicated are the corresponding patterns of (c) negative droplet radius anomalies not linked to a decrease in liquid water path, and (d) positive burden difference between easterly and westerly wind days.

Acknowledgments. This work has been developed within the framework of ECOS Sud collaboration agreement (C03U04), and under research grant FONDECYT 1020833. Support from the Multidisciplinary Program on Atmospheric and Climate Dynamics (PRODAC) from the Universidad de Chile is also acknowledged. Discussions and comments from Dr. Annica Ekman, Dr Jerome Riedi and Dr. Olivier Boucher are greatly appreciated.

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