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Regional dispersion of oxidized sulfur in Central Chile

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Abstract

Chile has a long tradition of exploiting mineral resources, particularly copper (Cu). One of the largest Cu smelters, Caletones, located some 150 km south of the country's capital, Santiago, in Central Chile, is responsible for about 0.4% of about 70 Tg S/yr oxidized sulfur (SO_x) emitted by anthropogenic sources worldwide. Santiago, a megacity with 5 million inhabitants, stands for about 5 Gg S/yr. The average meteorological conditions are unfavorable for the dispersion of pollutants in this area. All this poses risks for human health and vegetation. Also, downwind from these polluted areas there may be large-scale impacts on cloud properties and on oxidative cycles. Here, we present the first attempt to assess the regional distribution of SO_x in Central Chile using a dispersion model (MATCH) driven with data from a limited area weather forecast model (HIRLAM). Emphasis has been given to the impact of Cu smelters upon urban air quality, particularly that of Santiago. Six 1-month long periods were simulated for the years 1997, 1998 and 1999. These periods span over a broad range of typical meteorological conditions in the area including El Niño and La Niña years. Estimates of the regional dispersion and deposition patterns were calculated. The emissions from the large Cu smelters dominate the distribution of SO_x. A budget of SO_x over an area of $200 \times 200\,km^2$ around Santiago is presented. There is too low a number of monitoring stations to perform a detailed evaluation of MATCH. Nevertheless, the model reproduces consistently all the regional-scale characteristics that can be derived from the available observations.

Keywords: Oxidized sulfur; Copper smelters; Central Chile; Regional modeling

1. Introduction

Chile is facing a strong economic development. This development has consequences on the environment such as air pollution on local and regional-scales. Strong measures are required to prevent and curb these problems. Atmospheric dispersion models are one of the tools that must be developed for that purpose. The results presented here correspond to the first regional-scale modeling application in Chile. This effort was part of a cooperation project between the National Commission for the Environment (CONAMA) and the Swedish Meteorological and Hydrological Institute (SMHI) between 1999 and 2000.

Nearly 50% of the country's population is concentrated in Central Chile (See Fig. 1), mainly in Santiago (33°30′S, 70°48′W, 500 m a.s.l.). Santiago is a megacity located in a basin surrounded by the high Andes (average altitude ca. 4500 m), with 5 million inhabitants who are exposed to severe air pollution. In addition to the big cities, a number of large pollution sources exist in this part of Chile. Of particular interest are the large copper (Cu) smelters: Caletones (34°05′S, 70°27′W, 2400 m a.s.l.), Ventanas (32°44′S, 71°29′W, 10 m a.s.l.)

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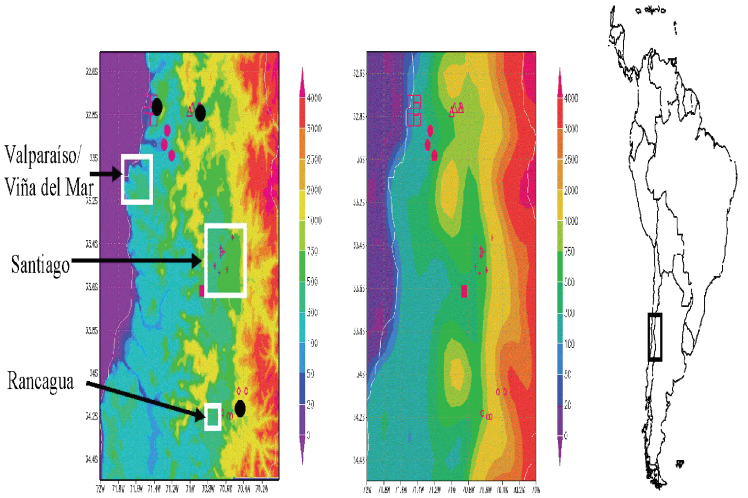

Fig. 1. South America and Central Chile (right panel) and location of the stations considered in the model evaluation as seen at 800 m horizontal resolution (left panel) and at the HIRLAM resolution (middle panel). The approximate horizontal domain considered in the MATCH model is indicated in the rightmost panel. Crosses correspond to the Santiago network, circles indicate the stations near the Caletones smelter, filled circles are for the Quillota (32°53′S, 71°15′W) network, open squares correspond to the Ventanas network, open triangles indicate the Chagres network and the filled square corresponds to the Talagante station. The main cities are highlighted with a white square. A black circle indicates the copper smelters. The color scale is the same for both colored figures and corresponds altitude levels (<0, 20, 50, 100, 300, 500, 750, 1000, 2000, 2500, 3000, >4000 m).

and Chagres (32°40′S, 70°53′, 600 m a.s.l.). These sources were responsible for an emission of 260 Gg S in 1999, which corresponds to 95% of the anthropogenic sulfur emissions in this part of Chile and about 0.4% of the global anthropogenic emissions of sulfur (e.g., Rodhe et al., 1995; IPCC, 2000).

The pollution sources in Central Chile pose environmental risks for the population (e.g., Ilabaca et al., 1999) and for agriculture (e.g., García-Huidobro et al., 2001). Most attention has been paid so far to the severe air pollution problems in Santiago. However, preliminary assessments show that other urban areas in Central Chile namely, Valparaíso (33°05′S, 71°40′W) and Viña del Mar (33°01′S, 71°33′W) some 100 km northwest of Santiago, and Rancagua (34°10′S, 70°46′W) some 150 km south of Santiago, may also suffer air pollution problems. Such problems are partly related to the Cu smelters located in the vicinity of these cities. For instance, Rancagua is some 20 km downhill from the Caletones smelter and the conurbation Valparaíso-Viña del Mar is some 30 km south of the Ventanas smelter. Also, downwind from these polluted areas there may be large-scale impacts on cloud properties (e.g., Rosenfeld, 2000) and on oxidative cycles (e.g., Dickerson et al., 1997).

Central Chile is located in a transition zone between the Pacific high in the north and the westerlies in the south (Miller, 1976). The vertical exchange of air during most part of the year is controlled by the temperature inversion due to the subsiding branch of the Hadley cell. The average meteorological conditions are unfavorable for the dispersion of air pollutants, especially during fall and winter. Often, the stagnant anticyclonic conditions are further intensified by the occurrence of coastal lows, which bring down the base of the subsidence inversion (e.g., Garreaud et al., 2001; Gallardo et al., 2002). In spring and summer, the relatively larger insolation determines an increase in the depth of the mixed layer counteracting the accumulation of pollutants. Nonetheless, actinic fluxes are also increased during spring and summer accelerating the occurrence of photochemical reactions.

Here, we present the first attempt to assess the regional distribution of oxidized sulfur (SO_x) in Central Chile using a regional scale transport/chemistry/deposition model (MATCH) coupled to a meteorological model (HIRLAM). Emphasis has been given to SO_x because it is an environmental issue of concern and because SO_x has been regularly monitored at several places over the last years, which makes it easier to bracket the model outputs for SO_x than for other pollutants. At this stage, we have focused on the long-range transport (over distances larger than 100 km) of SO_x from the Cu smelters upon urban air quality, particularly that of Santiago.

Next section briefly describes the model applied. Results are presented in Section 3, summary and conclusions in Section 4.

2. Model description and setup

To provide meteorological input data to model simulations of sulfur dispersion over Chile a three-dimensional numerical weather prediction model has been used, namely the High-Resolution Limited Area Model (HIRLAM, Källén, 1996). HIRLAM is a hydrostatic grid-point model and the resolutions used for the present study were 11 km horizontally and 31 levels in the vertical. The coordinate systems used are a rotated latitude longitude grid horizontally and a hybrid p-sigma system in the vertical. The time stepping is semi-implicit and semi-Lagrangian; a fourth-order, linear horizontal diffusion is used and the vertical diffusion is formulated through a second-order closure scheme.

HIRLAM was used as a dynamical downscaling tool, providing a more detailed representation of small-scale meteorological variations using coarse resolution forcing on the lateral boundaries. This approach has been shown to be successful for regional climate applications over Europe (Räisänen et al., 2001). HIRLAM was integrated continuously for a series of 1-month periods using lateral boundary conditions and sea surface temperature every 6 h from the ECMWF (European Centre for Medium-range Weather Forecasts) operational analysis. Model simulations for six periods were used in the present study: 15 May–15 June 1997, January and May 1998, 15 June–15 July October and 15 November–15 December 1999. The simulation periods cover a suite of meteorological conditions that affect Central Chile including extreme conditions under El Niño (1997) and La Niña (1998). The domain for the meteorological simulations extend from Northern Chile to Valdivia in Chile in the south, and in west–east direction from the Pacific to the Argentinean Atlantic coast. Comparisons between model calculated and observed meteorological variables are given in Gallardo et al. (2002).

The transport/chemistry/deposition model applied in this work is the Multiscale Atmospheric Transport and Chemistry Model (MATCH) developed at SMHI. A detailed presentation of this Eulerian transport model can be found in Robertson et al. (1999). MATCH describes the physical and chemical processes that govern emissions, atmospheric transport and dispersion, chemical transformation, wet and dry deposition of pollutants. MATCH has been applied previously to various environmental assessments, atmospheric chemistry research issues as well as in routine application for emergency response for nuclear accidents (e.g., Robertson et al., 1995; Langner et al., 1998a, b; Engardt and Holmén, 1999; Zunckel et al., 2000; Engardt, 2001).

Advection in MATCH is based on the mass conservative schemes suggested by Bott (1989). A fourth-order scheme is used in the horizontal direction and a second-order scheme in the vertical. Boundary layer processes, such as stability-dependent dry deposition flux and turbulent vertical mixing in the boundary layer is described using the surface friction velocity, surface sensible heat flux, and the boundary layer height. The dry deposition flux is proportional to the tracer's concentration and the inverse of the sum of the aerodynamic and surface resistance. The loss due to wet deposition is parameterized in terms of a scavenging coefficient and the precipitation at the surface (See Robertson et al., 1999 for details).

Two sulfur reservoirs are considered: sulfur dioxide (SO_2) and a sulfate reservoir that contains both gaseous phase sulfuric acid and sulfate aerosols. We have applied a simple sulfur only scheme in which the oxidation of SO_2 into sulfuric acid and sulfate is parameterized through a composed reaction rate, which involves separately the gas-phase oxidation through hydroxyl radical (OH) and the reactions in liquid phase (in-cloud oxidation) with ozone and hydrogen peroxide. The gas-phase oxidation rate is taken from the literature and it undergoes a diurnal and a seasonal cycle to account for the expected variations in OH. The other term is tuned so that the overall oxidation rate approximates the bulk oxidation rate proposed by Tarrasón and Iversen (1998). A complete description of the chemical scheme applied can be found in Engardt (2001). In absence of more specific information for Central Chile, deposition parameters, i.e., deposition velocities and scavenging coefficients were taken from Langner et al. (1998a).

The background values of SO_2 and sulfate were assumed to be zero. Since the SO_x emissions within the domain are very large it appears reasonable to neglect the background of SO_x. The simulations were initiated with zero fields and the outputs after 2 days of integration were considered for evaluation. MATCH was run with a horizontal resolution of $0.05^\circ \times 0.05^\circ$ (ca. $5 \times 5\,km^2$) and 15 vertical levels. The domain corresponds to an area around Santiago of $200 \times 200\,km^2$ and about 5.5 km in the vertical.

The most complete emission inventory is the one for Santiago for 1997 (CENMA, 2000a). An estimate of the emissions for 1998 and 1999 has been made considering the reduction in industrial emissions related to the introduction of natural gas (methane) instead of oil or coal combustion as energy supply in intensive industrial processes. The introduction of natural gas in industrial sources was completed in late March 1998. Therefore, the 1997 emissions were applied for January 1998 whereas for May 1998 the 1998 emissions were applied. In addition, a large point source (a molybdenum smelter, MOLYMET) in Santiago was not considered for January 1998 since this source was not operational then (C. Bustos, private communication). The May emissions do include MOLYMET. Information about emissions in other urban areas in Central Chile is scarce. Preliminary estimates have been provided by a

Table 1
Oxidized sulfur emissions considered in the model simulations for the 1997, 1998 and 1999. Unit: Gg S/yr

Sources	1997	1998	1999
Copper smelters			
Caletones	370	350	240
Ventanas	42	22	15
Chagres	8	6	5
Sub-total	420	378	260
Urban areas			
Santiago	10	5	5
Rancagua	0.5	0.5	0.5
Valparaíso-Viña del Mar	2	2	2
Others	1	1	1
Sub-total	14	9	9
Total	434	387	269

diagnostic analysis of air quality at five cities in Chile, among them Valparaíso, Viña del Mar and Rancagua. The emissions from the Cu smelters have been reported for the attainment plans enforced in the late 20th century. Table 1 indicates the annual emission used in this study. The emissions of SO_x are assumed to occur in the form of SO_2 (95%) and of sulfate (5%).

3. Results

The evaluation of the MATCH results has been limited by the fact that there is presently no regional monitoring network operating in Chile. Available observations of SO_x are largely restricted to SO_2 concentration measurements, which are reported on an hourly and monthly basis. The monitoring stations are located in the urban area of Santiago and in the surroundings of the largest Cu smelters in Central Chile (cf. Fig. 1). In addition to the data provided by these monitoring networks, monthly averaged concentrations of SO_2 retrieved in the cities of Valparaíso, Viña del Mar and Rancagua using a passive sampler technique are considered. Also, data from a monitoring campaign performed by CONAMA that provided information about sulfate in aerosol filters collected in winters 1998 (Artaxo, 1998) and 1999 (CENMA, 2000b) in Santiago are included in the analysis.

3.1. Regional distributions

According to the model simulations, SO_x is regionally dispersed over Central Chile. The model shows that there is a long-range transport of the SO_x emissions that affect the region of Chile where about 50% of the country's population lives. Fig. 2 shows the monthly averaged surface distribution of SO_2 for the six simulated periods. The regional distribution of SO_2 shows highest concentrations near the sources and positive strong gradients towards these sources. A less marked long-range transport is apparent in the rainy

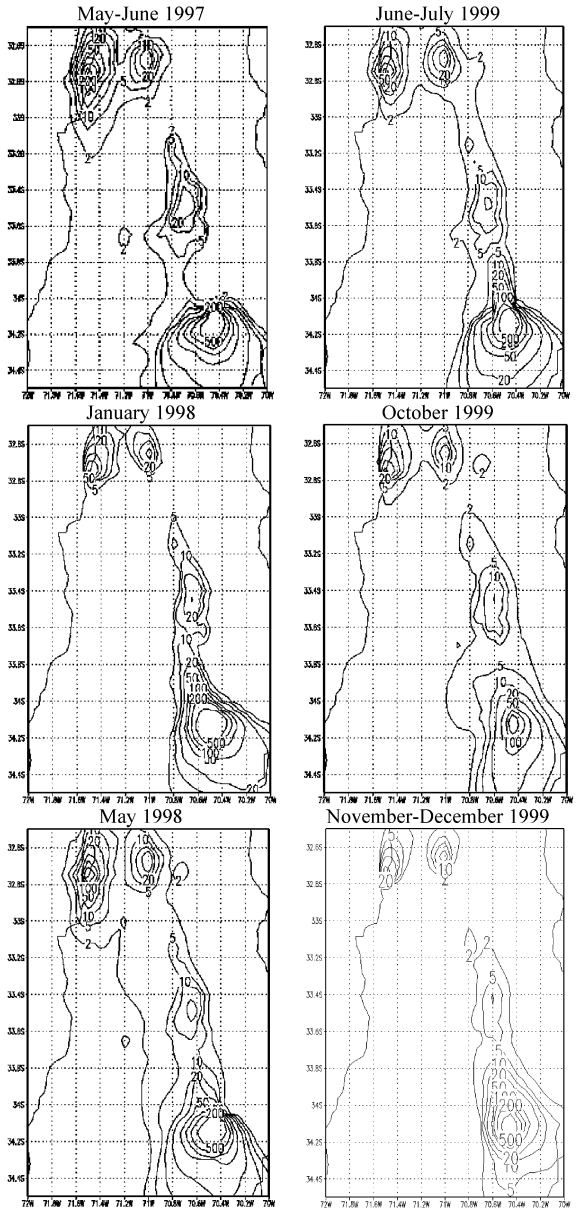

Fig. 2. Monthly averaged surface concentration of SO_2 for six different meteorological conditions. The isolines 2, 5, 10, 20, 50, 100, 200, 500 are drawn. Unit: $\mu g\,SO_2/m^{-3}$. The horizontal scale is longitude from 70°W (right) to 72°W (left) and the vertical scale is latitude from 32.5°S (top) to 34.5°S (bottom).

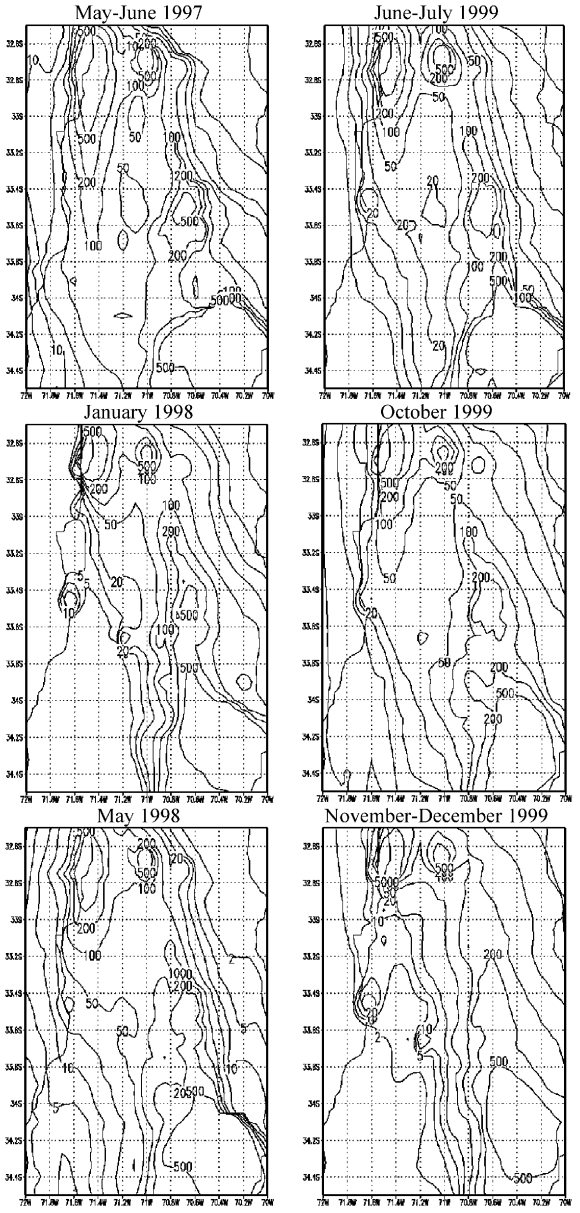

Fig. 3. Monthly accumulated total sulfur deposition for the six simulated periods. The isolines 2, 5, 20, 50, 100, 200 and 500 are drawn. Unit: mg S/m^2. The horizontal scale is longitude from 70°W (right) to 72°W (left) and the vertical scale is latitude from 32.5°S (top) to 34.5°S (bottom).

winter of 1997 because wet scavenging shortens the residence time of SO_x.

An interesting feature seen in the horizontal distribution of SO_x is the effect of changes in circulation and precipitation. In the rainy period of May–June 1997 (El Niño year) appears a north-to-south stretching related to northerly winds typical of frontal passages. This is also evident in the total sulfur deposition (See Fig. 3). During the spring and summer months the horizontal distributions show a south-to-north stretching that is related to the prevalence of southwesterly winds due to the Pacific high.

Since nearly all precipitation occurs in the winter season, wet deposition is significant only during that season and it varies markedly from year-to-year (e.g., Rutllant and Fuenzalida, 1991). Over the southwest of the domain, the model shows relatively high accumulated deposition fluxes but low monthly averaged concentrations. This suggests that there is an episodic long-range impact of the Cu smelters, particularly of Ventanas in connection with frontal passages. Unfortunately, monitoring of wet deposition is not currently available in Chile.

Table 2 shows the observed and simulated monthly averages of SO_2 surface concentrations at three selected stations. These stations are: Parque O'Higgins (EMD, 33°27′S, 70°23′W, 500 m a.s.l.) located in the urban area of Santiago; Talagante (SS4, 33°37′S, 70°51′, ca. 450 m a.s.l.), a rural site near Santiago; and Coya Club (mCY, 34°12′S, 70°34′, 1000 m a.s.l.) in the surroundings of the Caletones smelter. The model reproduces the seasonal variation observed in SO_2 concentrations with higher concentrations in wintertime than in summertime (Pairs of winter and summer periods were segregated according to the emission rates, i.e., May–June 1997 vs. January 1998; June–July 1999 vs. October and November–December 1999). However, there is a tendency to overestimate the summer and spring average concentrations in the Santiago basin. This mismatch is less noticeable in fall and winter. At mCY, nearby Caletones, there is a tendency to overestimate the average concentrations, particularly in May–June 1997 and January 1998. This overestimate of the concentrations is partly related to the coarse HIRLAM resolution (0.1°), which precludes an accurate description of surface winds and local circulations in a complex terrain area like the one around Caletones. However, systematic biases in the observations cannot be ruled out. The model roughly captures the variance of the monthly averaged SO_2 concentrations. The winter months actually show a large variance associated with synoptic disturbances such as rainfall events, characterized by cloudy skies and windy conditions, which with lower SO_2 concentrations are followed by periods of stable conditions with higher concentrations. The summer conditions are more persistent and so the variance at synoptic scale of the data is smaller than in winter (cf. Fig. 4).

In addition, the model seems able to capture a great deal of the day-to-day and week-to-week variations driven by sub-synoptic and synoptic weather patterns. Fig. 4 (upper panel) shows comparisons of simulated and measured SO_2 at EMD in downtown Santiago. Emphasis is put on comparisons with measurements at

Table 2
Observed and simulated SO_2 average concentration plus minus one standard deviation for selected stations

Period	Parque O'Higgins (EMD)		Talagante (SS4)		Coya Club de Campo (mCY)	
	Obs	Mod	Obs	Mod	Obs	Mod
May–June 1997	36±17	31±20	—	6±4 (29)	192±163 (29)	519±475 (29)
January 1998	15±6 (27)	24±7 (20)	4±1 (20)	2±2 (20)	101±160 (26)	609±415 (20)
May 1998	28±15 (30)	25±11 (30)	3±2 (30)	5±5 (30)	481±525 (30)	533±465 (30)
June–July 1999	22±13 (30)	22±7 (30)	4±3 (30)	2±1 (30)	361±411 (28)	343±259 (30)
October 1999	10±5 (30)	17±4 (30)	—	2±2 (30)	165±206 (30)	43±35 (30)
November–December 1999	11±6 (30)	17±6 (29)	—	1±1 (29)	45±90 (30)	104±172 (29)

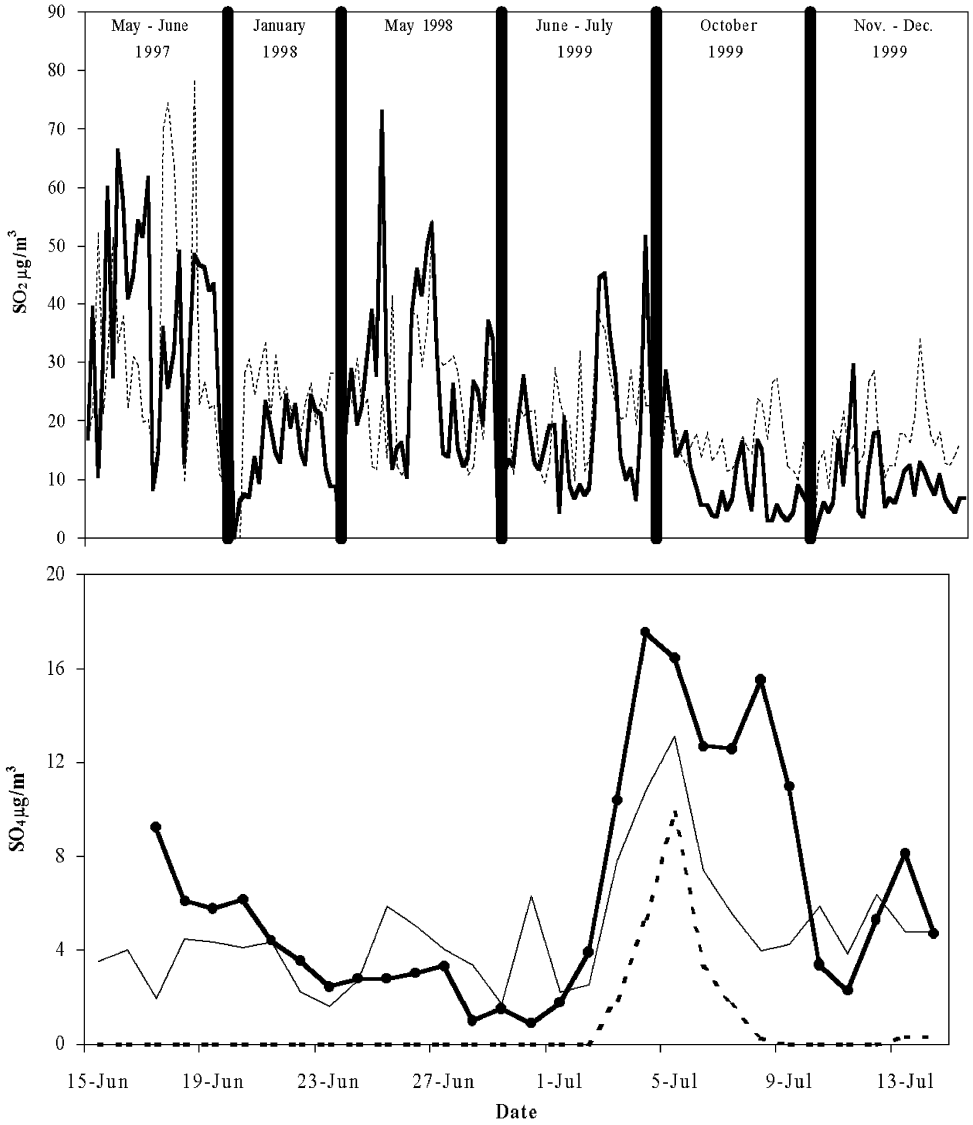

Fig. 4. The upper panel shows the simulated (dashed) and observed (full) SO_2 daily average concentration (μg SO_2/m^2) time-series at the EMD station in downtown Santiago for the six simulated periods. The lower panel presents the simulated (light) and observed (dark with marks) sulfate concentrations at station EMD in downtown Santiago between 15 June and 14 July 1999. The contribution from Caletones Cu smelter is also indicated (dashed line lower panel).

EMD since it is located in a relatively flat area of the basin, not too close to large point sources, which suggests that it is the most representative station available of the regional patterns of dispersion of SO_x. Notice that the agreement between simulations and observations is better for fall and winter (except for June–July 1997, when the meteorological simulations were less good) conditions than for summer. This may be related to the fact that in fall and winter the majority of the variations are associated with regional-scale weather patterns, whereas in summer they are strongly modulated by local wind circulations. In other words, the model seems to represent equally well the regional patterns of dispersion in winter (e.g., fronts, coastal lows, etc.) and summer (e.g., Pacific high) but this ability explains in winter more of the variance of the data than in summer when other circulations (not fully simulated by the model, i.e., driven by small-scale topography) do contribute significantly. The meteorological model has a tendency to underestimate surface wind speeds in summer, which in the case of the Santiago basin results in a less good representation of the redistribution of pollutants within the basin.

Comparisons between observations and simulations on a day-to-day or hourly basis are difficult due to the limited geographical representativity of the monitoring stations. The majority of the stations are located close to sulfur sources or in areas of complex terrain (cf. Fig. 1). Nonetheless, some regional features can be identified from the data collected at these networks. For instance, the gradient in SO_2 concentrations over 10 or more kilometers. The model generally captures these gradients for the Ventanas's and Caletones's monitoring networks as seen from the monthly averages.

Next, we evaluate the model's ability to simulate the partitioning between SO_2 and sulfate. Artaxo (1998) reports seasonal averages of the total sulfur content measured in aerosol filters for inhalable particle matter (PM_{10}, aerosol diameter $<10\,\mu m$) during winter (May–August) 1998 at several locations in Santiago and surrounding areas. Namely at EMD, Las Condes (EMM, 33°22′S, 70°31′W, ca.700 m a.s.l.) and SS4, i.e., downtown, eastwards and westwards of Santiago city, respectively. These measurements provide an upper limit for sulfate in the boundary layer at these locations during fall and winter 1998. Recall that the PM_{10} fraction typically contains soil dust and other coarse aerosols in addition to fine aerosols associated with gas-to-particle transformations. Therefore, the total sulfur content of the PM_{10} samples could contain other sulfur species than sulfate. We take, nevertheless, the available sulfur in PM_{10} data as a surrogate for sulfate (as sulfur) and we build up an "observed" partitioning, which yields an "observed" May–August 1998 average sulfur as particulate/sulfur as SO_2 ratio of 0.08, roughly consistent with the simulated May 1998 average sulfur as sulfate/sulfur as SO_2 ratio of 0.05 in downtown Santiago.

The evaluation of the simulation of the partitioning and the sulfate content is more direct for data collected on filters for completely inhalable particulate matter ($PM_{2.5}$, aerosol diameter $<2.5\,\mu m$) in winter 1999 (CENMA, 2000b). This is shown in Fig. 4 (lower panel), where the observed and simulated daily average concentrations of sulfate in downtown Santiago during one month in winter 1999. There is an overall good agreement between the simulations and the observations. The model estimated contribution of Caletones is also shown. According to the model simulations, the prevailing subsiding conditions during intensive coastal lows, e.g., early July 1999, bring down the Caletones plume, the main SO_x source in the area, over the Santiago basin. The outcome is an enhanced contribution of the Caletones smelter to the burden of SO_x in the boundary layer of the Santiago basin, especially in the form of sulfate and thus associated to fine aerosols (For details see Gallardo et al., 2002).

3.2. Relative contributions

By running the model with and without emissions from the Cu smelters, their relative contributions to the SO_x concentrations were estimated. In all the simulated periods, the emissions from the Cu smelters dominate the overall horizontal (Fig. 5) and vertical (not shown) distribution of SO_x in Central Chile. The contribution of urban sources, particularly those of Santiago, at rural sites and above the boundary layer is estimated to be $<10\%$.

It must be pointed out that similar impacts of the Cu smelters emissions can be seen in elemental analyses of particulate matter measurements for other urban areas in the region, namely Valparaíso, Viña del Mar and Rancagua (Kavouras et al., 2001). Also, in a parallel study concerning the impact of arsenic emissions, the Cu smelters were shown to be the dominating sources of airborne arsenic over Central and Northern Chile (Gidhagen et al., 2002).

These results strongly suggest that it is necessary to continue reducing the emissions from the Cu smelters in order to avoid negative impacts.

3.3. Budget calculations

Budget calculations were made over the model domain for each simulated period. The burdens and fluxes of SO_2 and sulfate were estimated for each simulated period and then averaged to give an annual estimate. Most part of the emitted sulfur (ca. 70%) is exported from the area: southward (40%), into the mountains (22%), and to the ocean (8%). Since the area studied is quite small, most part of the sulfur is found in

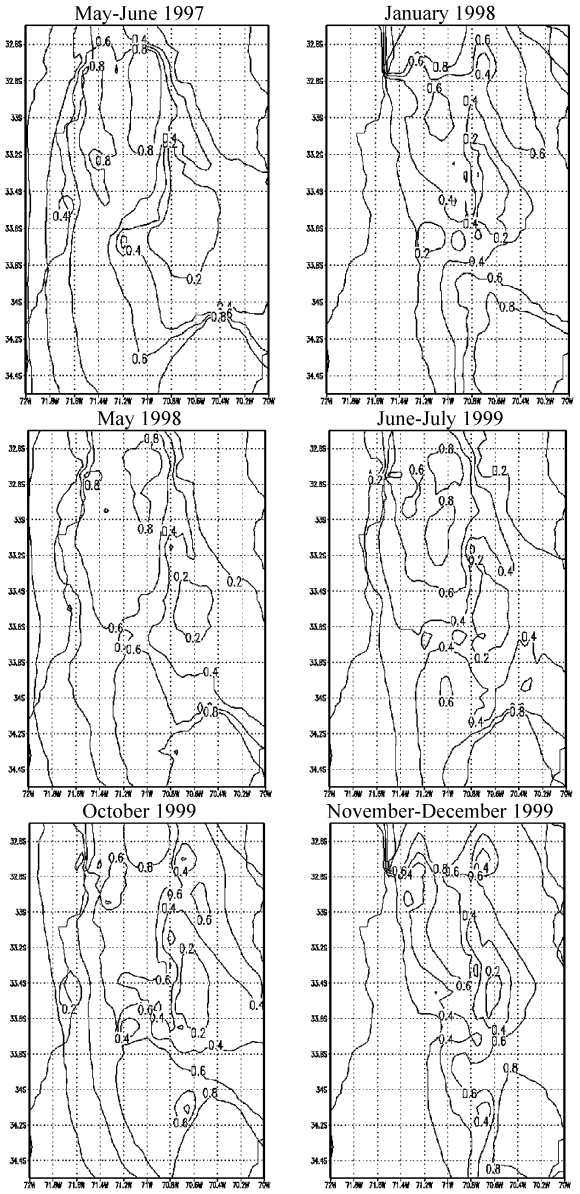

Fig. 5. Monthly averaged relative contributions (fractions) of the Cu smelters to the total surface sulfur burden, i.e. sulfur dioxide and sulfate, for the six simulated periods. The isolines are: 0.2, 0.4, 0.6, 0.8. The horizontal scale is longitude from 70°W (right) to 72°W (left) and the vertical scale is latitude from 32.5°S (top) to 34.5°S (bottom).

the SO_2 reservoir (ca. 90%) and only a small percentage of the SO_2 emission is converted into sulfate inside the domain. The most important removal mechanism is by far dry deposition. However, in rainy periods like May–June 1997, wet deposition can be as high as 40% of the total deposition. These results make it necessary to enlarge the spatial coverage of the ongoing monitoring activity and also to begin a thorough analysis of impacts in agriculture and vegetation both within the area under consideration in this study and to the south of it. In particular, they stress the necessity of installing wet-only collectors and of promoting assessments of dry deposition (e.g., Zunckel et al., 2000).

4. Summary and conclusions

A regional transport/chemistry/deposition model (MATCH) has been used to study sulfur dispersion over the central part of Chile. Six 1-month long periods were simulated for 1997, 1998 and 1999. These scenarios span over a broad range of meteorological conditions including extreme situations under El Niño and La Niña years.

On a monthly average basis the simulations reproduce the observed SO_2 concentrations as well as the variance of the data. Clear mismatches occur though for stations located in the immediate surroundings of the largest point sources, i.e., the Cu smelters, partly because the current model resolution is not enough to resolve local circulation patterns. The observed daily averaged SO_2 concentrations are well captured by the model in the Santiago basin. However, significant discrepancies occur in connection with shortcomings in the representation of the meteorological fields, e.g., mismatches in wind direction due to small-scale topography effects. The simulated sulfate concentrations are consistent with the seasonal averages of the total sulfur content measured in aerosol filters during winter 1998 in downtown Santiago. Moreover, the simulations are also in good agreement with daily averaged values observed during a campaign in winter 1999. All in all, at this stage, we cannot say that MATCH has been exhaustively validated. Nevertheless, the model reproduces consistently all the regional-scale characteristics that can be derived from available observations.

When compiling and analyzing available air quality data, it appeared clear to us that efforts should be made in terms of coordination and standardization of the ongoing monitoring activities, especially those performed by the industry. This would help in reducing costs and increasing the reliability of the collected data. Also, it appears necessary to improve the quality assurance procedures, not only thorough sampling and analysis protocols but also in terms of data reporting and interpretation. Perhaps, a common data base and an analysis and interpretation center at the regional scale would be a good way to deal with this complex matter.

The MATCH simulations show that SO_x is regionally dispersed over Central Chile, i.e., there is a long-range transport of the SO_x emissions that affect the area where about 50% of the country's population lives and where agriculture is an important activity. In all simulated

periods, the emissions from the Cu smelters dominate the overall horizontal and vertical distributions of SO_x. The urban emissions, particularly those of Santiago, only affect significantly the city surroundings. Moreover, the consistency between the simulated concentrations and the observations of both SO_2 and sulfate strongly supports the idea that on an episodic basis there is a significant contribution of the Caletones smelter to the sulfur burden in the Santiago basin.

A budget of SO_x over an area of $200 \times 200\,km^2$ around Santiago has been presented. The budget calculations show that the most important removal mechanism is by far dry deposition. However, in rainy winters wet deposition can also be important. Most part of the emitted sulfur is exported outside the model domain. In particular, a significant fraction of the emitted sulfur (ca. 40%) is exported south of the largest point source in the area, namely Caletones.

Altogether, these results point out the necessity to continue reducing the emissions from the Cu smelters in order to avoid negative impacts. Further, these results indicate that it is necessary to enlarge the spatial coverage of the ongoing monitoring activities and also to begin a thorough analysis of impacts on agriculture and vegetation. Based on model results, eight regional stations were proposed and their preliminary evaluation was initiated in 2001 as a part of a 5-yr program driven by CONAMA. Finally, future studies should assess other aspects, for instance the potential impacts of sulfate aerosols on cloud properties off the coast of Chile.

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