Coastal lows and sulfur air pollution in Central Chile

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Abstract

Air pollutants in Santiago (33.5ºS, 70.8ºW, 500 m.a.s.l.), a city with 5 million inhabitants, located in a basin in Central Chile surrounded by the high Andes, frequently exceed air quality standards. This affects human health and it stresses vegetation. The most extreme winter and fall pollution events occur when the subsident regime of the Pacific high is further enhanced by coastal lows (CLs), which bring down the base of the subsidence inversion. Under these conditions, the air quality worsens significantly giving rise to acute air pollution episodes. We assess the ability of a regional transport/chemistry/deposition model (MATCH) coupled to a meteorological model (HIRLAM) to simulate the evolution of oxidized sulfur (SO_x) in connection with intensive CLs. We focus on SO_x since it is an environmental issue of concern, and the emissions and concentrations of SO_x have been regularly monitored making it easier to bracket model outputs for SO_x than for other pollutants. Furthermore, the SO_x emissions in the area are very large, i.e., about 0.4% of the global anthropogenic sources. Comparisons with observations indicate that the combination of HIRLAM and MATCH is a suitable tool for describing the regional patterns of dispersion associated with CLs. However, the low number and the limited geographical coverage of reliable air quality data preclude a complete evaluation of the model. Nevertheless, we show evidence of an enhanced contribution of the largest copper smelter in the area, i.e., Caletones, to the burden of SO_x in the Santiago basin, especially in the form of sulfate associated to fine particles (radii < 2.5 μm), during CLs. Further, we speculate that the Caletones plume may trigger or promote secondary aerosol formation during CLs in the Santiago basin.

Key words: Oxidized sulfur, $PM_{2.5}$, coastal lows, regional modeling

1. Introduction

In Central Chile, the combination of, on the one hand, meteorological conditions adverse to ventilation and, on the other hand, the existence of large emissions of pollutants to the atmosphere, determines the occurrence of high concentrations of particles and gases, which have detrimental effects on air quality and visibility (e.g., Trier and Firingueti, 1994; Rutllant and Garreaud, 1995). Maximum concentrations of particles and pollutants, mainly associated with combustion processes, occur in fall and winter months, while in spring and summer there is a maximum in ozone concentrations as the actinic fluxes increase. The most extreme winter and fall pollution events occur in connection with coastal lows (CLs).

The concentrations of pollutants in Santiago, a city with 5 million inhabitants, frequently exceed air quality standards. This affects seriously human health (e.g., Ilabaca et al., 1999). Similar risks are posed to the population of other major cities in Central Chile (e.g., Kavouras et al, 2001), namely Valparaíso (33º05'S, 71º40'W), Viña del Mar (33º01'S, 71º33'W) and Rancagua (34º10'S, 70º46'W), and in the surroundings of large pollution sources such as the copper smelters located in the area (See Figure 1). A recent risk assessment study demonstrates that there may be large impacts on vegetation and agriculture in Central Chile due to the observed high levels of ozone and sulfur dioxide (García-Huidobro, 1999; García-Huidobro et al., 2001). These problems have led authorities to enforce attainment plans for point sources, mainly copper smelters, and the Metropolitan area of Santiago.

In this work we assess the ability of a regional modeling system to simulate the evolution of high pollution episodes in connection with intensive CLs due to their significant impact on air quality in Central Chile. We chose to focus on oxidized sulfur (SO_x) since, on the one hand, it is an environmental issue of concern, and on the other hand, the emissions and atmospheric concentrations of SO_x have been regularly monitored at several places over several years making it relatively easier to bracket model outputs for SO_x than for other pollutants. Furthermore, large emissions of SO_x take place in Central Chile (269 GgS in 1999). In fact, these sources account for about 0.4 % of the 70 TgS/yr emitted worldwide by anthropogenic sources (e.g., Rodhe et al, 1995; IPCC, 2000).

In the next section we review the characteristics of CLs and of two specific episodes: one in May 1998 and another in July 1999. Section 3 describes the simulations of the selected episodes. Also, a summer case is briefly discussed in Section 3. Conclusions are summarized in Section 4.

2. Coastal lows and air quality

The occurrence of CLs is closely related to the perturbation of the mid-latitudes westerly flow by the Andes cordillera and the persistence of the Pacific high, which enables the trapping of sub-synoptic disturbances that propagate along the coast such as the warm low-level lows or CLs (For details the reader is referred to Garreaud et al, 2001 and references therein). CLs take place all through the year and rather frequently, typically once a week. This phenomenon affects a large fraction of Chile between 27ºS and 42ºS. Their effects on weather conditions and air quality have a larger impact in winter than in summer. In connection with CLs, the weather changes greatly from one day to another at the coast and inland valleys, at times over a few hours, from clear sky conditions with relatively high daytime surface temperatures to cloudy conditions with cool and moist air.

The intensity and duration of CLs varies depending on the overall synoptic configuration, the intensity of the weather systems involved and both the large- and local-scale topography. The intensity and location of the large-scale weather systems appears to be triggering factors for the onset of CLs. The large-scale topography (e.g. the Andes) seems crucial in allowing the trapping and for the strengthening of the disturbance. In addition, the local topography (e.g. east-west valleys) and the propagation velocity of the disturbance also play a role in determining the duration of the event. All these factors have consequences on the intensity and duration of the air pollution events associated with CLs as well as the strength of the emissions of pollutants and their precursors.

Ruttlant and Garreaud (1995) identify two main patterns for the CLs: type A and type BPF. Type A corresponds to the onset of a CL in Central Chile moving southward along the coast. This coastal trough appears between an enhanced Pacific high to the west of the Andes and a migratory cold high east of the Andes (See Figure 2). Type BPF is a prefrontal condition ahead of a weak and often occluded front, which slows down or becomes stationary when reaching Central Chile. Typically, CLs of type A produce more intensive air pollution episodes than those of type BPF and for that reason we focus on those of type A.

The start of the high concentration episodes associated with CLs coincides with a sharp decrease in boundary layer height (BLH) due to the establishment of easterly winds in connection with the sub-synoptic disturbance. The easterly winds are forced to subside by the high Andes giving rise to adiabatic compression and therefore to an enhancement of the subsidence inversion and clear sky conditions. Under these conditions the air quality worsens significantly giving rise to acute episodes of air pollution. The end of the episodes typically occurs in connection with humid air advection from the coast along the east-west valleys and the appearance in the Central Valley of Chile of fog conditions. The entrance of humid air from the coast is due to the reestablishment of westerly winds near the surface and a weakening of the subsidence inversion, which in turn diminishes rapidly the pollutant concentrations in the basin.

The features described above are illustrated for two intensive CLs of type A that took place around May 18th 1998 and July 5th 1999. These events resulted in increased concentrations of SO_x (i.e., about twice the monthly average) and other pollutants, particularly inhalable particles (PM_{10}, i.e., aerosol radii < 10μm) in the Santiago basin. During such episodes, increases in concentrations are observed at all monitoring networks in the region since they are triggered and modulated by a common regional weather pattern, namely a CL of type A. In fact, under such conditions common evolutions (co-variations) are found on a daily average basis, i.e., on the time-scale of changes in regional weather patterns. This is illustrated in Figure 3 where the co-variation among the time-series of SO_2 concentrations measured at three stations located in different and distant areas can be seen. The stations are: Parque O'Higgins (EMD, 33º27'S, 70º23'W, 500 m.a.s.l) in downtown Santiago, Coya Club de Campo (mCY, 34º12'S, 70º34', 1000 m.a.s.l.) near the Caletones smelter some 150 km to the south of Santiago, and Estación Sur (SUR, 32º49'S, 71º29'W, 50 m.a.s.l.) by the coast, some 100 km northwest of Santiago. Notice that large increases in daily averaged SO_2 concentrations in all time-series occur in connection with the intensive CLs observed in mid May 1998 and early July 1999. When only the Pacific high conditions (summer like) prevail the concentrations are strongly modulated by local wind circulations and no clear co-variation among the different monitoring networks can be distinguished. Nevertheless, CLs are also frequently observed in summer giving rise to co-variation patterns in Central Chile.

Figure 4 illustrates the establishment of easterly winds in the lower troposphere in connection with CLs in May 1998 and July 1999. We show the zonal wind component, temperature and humidity measured at Lo Prado (33º16', 70º 37', 1065 m.a.s.l.), a surface meteorological station located on the coastal cordillera west of Santiago. This station gives a clear signal of the establishment and retreat of easterly winds in connection with CLs due to the local topography that filters out north-south winds and amplifies the east-west wind signal. Because of this sensitivity, Lo Prado is routinely used for the forecast of pollution episodes (R. Sanguineti, M. Merino, pers. comm.). As easterly winds start to blow, the temperature rises and humidity diminishes. When the westerly winds are re-established, the temperature decreases and the air humidity goes up. Notice that according to these data, the episode in May 1998 is more clearly defined than the July episode, at least in terms of temperature and humidity. Notice that the maximum in SO_2 (Figure 4) takes place before the maximum in particulate matter, which suggests the occurrence of secondary aerosol formation during the episodes. This could possibly be related to the prevailing clear sky conditions and the accumulation of photo-oxidants and aerosols that may accelerate the oxidation rates and the gas-to-particle transformations. There may also be an effect of aerosols on the actinic fluxes and thus on the oxidative cycles (e.g., Dickerson et al, 1997). This subject goes nevertheless beyond the scope of the present work.

The evolution of temperature and wind as monitored by sounding stations by the coast some 100 km to the west of Santiago, namely Quintero (32º 28’ S, 71º19’ W, 8 m.a.s.l.) during May 1-31 1998, and Santo Domingo (33º23’S, 71º22’W, 75 m.a.s.l.) during June 15-July15 1999, respectively, is shown in Figures 5 a and b (left panels) (The sounding station at Quintero was moved to Santo Domingo in early 1999). An intensified subsidence inversion is apparent in both periods. This coincides with the establishment of weak easterly winds and decreased humidity close to the surface (not shown). A similar pattern is found at the acoustic low-level sounding (up to about 2000 m) station, namely La Platina (33º34’S, 70º22’W, 652 m.a.s.l.) in the outskirts of Santiago. This is shown in Figure 6 (left panels) for the period June-July 1999.

3. Model simulations

A combination of two numerical models was used in the present study. HIRLAM (High Resolution Limited Area Model) was used to derive meteorological fields for use in a transport/chemistry/deposition model MATCH (Multi-scale Atmospheric Transport and Chemistry model). HIRLAM is a hydrostatic grid-point model developed for short-range weather prediction and regional climate simulations (Källén et al, 1996, Räisänen et al., 2001). The resolutions used for the present study were 0.1°x0.1° (~11x11 km^2) horizontally and 31 levels in the vertical. HIRLAM was integrated continuously for one-month periods using lateral boundary conditions and sea surface temperature every six hours from the ECMWF (European Centre for Medium-range Weather Forecasts) operational analysis.

MATCH is an Eulerian regional transport/chemistry/deposition developed at SMHI. A detailed presentation of the basic 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, research studies as well as in routine application for emergency response for nuclear accidents (e.g., Robertson et al., 1995; Langner et. al., 1998 a; Langner et al, 1998 b; Engardt and Holmén, 1999; Zunckel et al, 2000; Engardt, 2001). For the present simulations two sulfur reservoirs are considered: sulfur dioxide (SO_2) and a sulfate reservoir that accounts for 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 (Engardt 2001). MATCH was run with a horizontal resolution of 0.05ºx0.05º (ca. 5x5 km^2) and 15 vertical levels. The domain corresponds to an area around Santiago of 200x200 km^2 and about 5.5 km in the vertical. Further details about the setup of MATCH for central Chile are given Olivares et al. (2002).

The pollution episodes observed around May 18th 1998 and July 5th 1999 were characterized by very stable conditions and low wind speeds in the central region of Chile. HIRLAM captures the regional-scale characteristics of these episodes. The simulations reproduce the strengthening of the subsidence inversion as shown in the right panels of Figures 5 a and b, both at the synoptic station by the coast and in the Santiago basin (Figure 6). The simulations reproduce as well the weakening of the winds, with a pronounced easterly component in the lower troposphere. The cross-sections of the simulated wind over the Santiago basin during the most intensive phases of the July 1999 CL episode indicate that easterly winds prevail in the lower part of the model domain. According to the HIRLAM simulations, the CL episode in May 1998 appears to be more intensive than the July episode since both the wind speeds and the vertical extent of the easterly winds is more pronounced in the May episode than in the July episode. This can be partly recognized in the observed evolution of temperature and humidity at Lo Prado for these periods shown in Figure 4, which suggest a more clearly defined CL episode in May 1998 than in July 1999.

Altogether, the simulations confirm the characteristic patterns described by Rutllant and Garreaud (1995), i.e., an enhancement of the subsidence inversion, the establishment of easterly winds and the reduction in BLH. The model also starts advecting humid air from the coast at the time it is actually observed. However, the humid air advected from the coast does not reach all the way into the Santiago basin resulting in too slow a change of the air mass in the Santiago basin. This may be due to the lack of detailed representation of topography and the drainage effect of the east-west valleys given the relatively coarse resolution of the meteorological model (0.1º). This contributes to overestimate the concentrations of SO_2 towards the end of the episodes. The sulfur concentrations remain high in the model until the easterly flow has ceased and the boundary layer (BL) can grow again. These features are illustrated in Figure 7, where we present the simulated time-series of BLH and those of SO_2 and relative humidity as measured and simulated in downtown Santiago. An independent estimate of BLH (Gómez, 2001), based on observed wind and temperature vertical profile at La Platina, also shown in Figure 7, is in good agreement with the model estimated BLH. Consistently with the observations at Parque O'Higgins (Cf. Figure 7), the model shows a more intensive drying of the BL in connection with easterly winds and associated subsidence in the May 1998 episode than in that of July 1999 (Cf. Figure 4). The systematic underestimate of relative humidity for these periods is mainly resulting from overestimates of the surface temperature by HIRLAM. There are several reasons for this. The scheme used for predicting soil temperature and soil moisture is simple and only one surface type was used over land. This affects the simulation of heat and moisture fluxes. Simulation of clouds is another source of error. In the present simulations HIRLAM sometimes underestimate cloudiness resulting in too high temperatures.

According to the model simulations, the prevailing subsident conditions during CLs bring down the Caletones plume, i.e., the main source of SO_x in the area, over the Santiago basin. Then whenever vertical mixing or drainage along the mountain slopes occurs air from aloft is mixed into the BL within the basin. A detailed description of exchange processes between the BL and the free troposphere under stable or very stable conditions, drainage along mountain valleys, etc., is a difficult task that may require not only higher resolution but also entirely different parameterizations than those used in this study (e.g., Seibert et al, 2000). Nevertheless, the model results indicate that during CLs there is an enhanced contribution of the Caletones smelter to the burden of SO_x in the BL of the Santiago basin.

The simulations suggest that the Caletones smelter's contribution can be as high as 50% of the SO_x concentrations and a few percent of the fine particles ($PM_{2.5}$) in the Santiago basin during CLs. In Figure 8 we show the observed and simulated daily average concentrations of SO_2 and sulfate at Parque O'Higgins in downtown Santiago (Only for the period June 15th - July 15th 1999 sulfate data are available). It also shows the contribution from Caletones as calculated by the model. We highlight two aspects from these pictures. First, the concentrations, particularly those of sulfate, both observed (June-July 1999) and simulated, increase markedly only during the CL episodes in mid May 1998 and in the beginning of July 1999. Second, the increase in SO_2 concentrations is concurrent with the increase in the Caletones contribution. In the simulations, the sulfate fraction increment occurs as the Caletones contribution to the SO_x burden in the Santiago basin increases. In the observations for June-July 1999, two relative maxima in the sulfate fraction of SO_x appear: the first one occurs, as in the simulations, at the beginning of the episode and the second towards the end of the episode. The former may be partly explained by long-range transport and entrainment from above the BL, specifically from the Caletones' plume. The second maximum in sulfate and sulfate fraction, not captured by the model, may be, as earlier stated, connected with secondary aerosol formation. Also, the increase in sulfate fraction appears to be concurrent with an increase in the observed ratio between $PM_{2.5}$ and PM_{10} (Figure 8, lower panel). These results lead us to speculate that the aerosols in the Caletones plume that entrain the Santiago basin under intensive CL conditions may trigger or further enhance the secondary aerosol (i.e., $PM_{2.5}$) formation.

So far we have discussed the impact on air quality of CLs in fall and winter. However, CLs do occur all through the year and they affect air quality, particularly SO_x, in summer as well. In fact, the inspection of SO_2 time-series in Santiago for summer conditions indicates that maxima in SO_2 concentrations take frequently place in the afternoon hours when the temperature reaches its maximum at all stations (Gómez, 2001). The ubiquitous appearance of these afternoon maxima in SO_2 suggests that they reflect long-range transport. The occurrence in connection or concurrently with the hour of highest temperature suggests that the episodic afternoon-maxima be linked with vertical mixing processes that may bring sulfur from aloft (fumigation). Such an event was observed on January 17th 1998. According to the model simulations, a plume of SO_x was effectively advected from the southeast, where Caletones is located, over the Santiago basin during January 17th in connection with a CL of type A. This CL was characterized, as those of the winter periods earlier discussed, by a strengthening of the temperature inversion and the appearance of easterly winds over the Santiago basin. Figure 9 shows the daily average horizontal distribution of SO_2 integrated in the vertical (from the surface up to ca. 5.5 km) for January 17th. Hence, the model simulations indicate that the Caletones smelter may in fact contribute to the sulfur burden in Santiago in connection with downward mixing, possibly strong vertical mixing in the afternoon hours during summer CLs. Of course, since ventilation and particularly vertical mixing are enhanced in this season, these conditions prevent the occurrence of high concentrations of SO_x and other pollutants in the BL.

4. Summary and conclusions

We have applied a regional-scale dispersion model (MATCH) coupled to a meteorological model (HIRLAM) and evaluated its ability to simulate the evolution of high pollution episodes in connection with CLs, particularly of type A due to their significant impact on air quality in Central Chile. We have focused on SO_x since it is an environmental issue of concern, and the emissions and concentrations of SO_x have been regularly monitored. However, many of the conclusions may be applicable to other pollutants. For instance, arsenic emitted from the copper smelters (Gidhagen et al, 2002).

The model is able to capture the meteorological conditions during the two major winter episodes observed in mid May 1998 and early July 1999. The modeling system reproduces the establishment of easterly winds and the reduction in BLH, which inhibit vertical mixing and dispersion resulting in an increase of the surface concentrations of SO_x and other pollutants. The end of the episodes, associated with the reestablishment of westerly winds near the surface and advection of humid air from the coast, is also captured. However, the intensity of these processes appears to be underestimated perhaps due to the lack of detailed representation of the topography and thereby of the drainage effect along the east-west valleys. This contributes to overestimate the concentrations of SO_2 towards the end of the episodes. In addition, the model simulations show that the Caletones copper smelter, the largest SO_x source in the area (240 GgS out of 269 GgS in 1999), located some 150 km south of Santiago, in connection with the occurrence of CLs contributes significantly (up to 50%) to the SO_x burden inside Santiago. The impact appears to be related to the strong subsidence associated with the configuration of intensive CLs (A type episodes). Air from aloft could be mixed into the BL within the basin via fumigation or drainage along the mountain slopes. The present model configuration precludes a detailed description of exchange processes between the BL and the free-troposphere, particularly under stable or very stable conditions and in presence of complex terrain. Nevertheless, the model consistently shows that there is a connection between the episodic impact of the copper smelters and the appearance in downtown Santiago of air masses with higher sulfate fractions and associated with secondary aerosols or fine particles (radii <2.5 μm). These findings are in good agreement with available observations.

The observations in Santiago suggest acceleration in secondary aerosol formation towards the end of the CL episodes, possibly related to increased oxidation rates due to the accumulation of photo-oxidants and aerosols under the prevailing stable and clear sky conditions during CLs. We speculate that this may be triggered or enhanced by the Caletones plume. Of course, the scarcity of data precludes any definite statement on this subject at this stage.

In summary the combination of MATCH and HIRLAM captures the main characteristics of the SO_x pollution episodes that typically occur in connection with strong CLs in Central Chile. The simulations are in good agreement with the available observations of meteorology and air quality. However, the number and the geographical coverage of reliable air quality data, preclude a more detailed and statistically based assessment. Nevertheless, the analysis made so far indicates that the model system is a suitable tool for describing the regional-scale patterns of dispersion in connection with CLs. This ability could be of use when defining measures to prevent high-pollution episodes and when designing measuring campaigns and monitoring efforts to better assess CLs and their impact on air quality.

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References

Dickerson, R., Kondragunta, S., Stenchikov, G., Civerolo, K., Doddrige, B., and Holben, B., 1997. The impact of aerosols on solar ultraviolet radiation and photochemical smog. *Science* 278, 827-830.

García-Huidobro, T., 1999. "A Risk Assessment of Potential Crop Losses Due to Air Pollution in the Central Regions of Chile". Imperial College of Science, Technology and Medicine. University of London, Centre for Environmental Technology.

García-Huidobro, T., Marshall, F., Bell, J., 2001. A risk assessment of potential crop losses due to ambient SO_2 in the central regions of Chile. *Atmos. Env*., 35, 29, 4903-4915.

Garreaud, R., Rutllant, J. and Fuenzalida, H., 2001. Coastal lows in the subtropical West Coast of South America: mean structure and evolution. *Mon. Wea. Rev*.130, 75-88.

Gidhagen, L., Schmidt-Thomé, P., Kahelin, H., Langner, J., 2000: Determinación de la línea base nacional de contenido de arsénico en material particulado respirable. Informe final. Comisión Nacional del Medio Ambiente. [Available from National Commission for the Environment, www.conama.cl, Obispo Donoso 6, Providencia, Santiago, Chile]

Gómez, 2001. Análisis de contaminantes atmosféricos en Santiago medidos por la red MACAM 2. Tesis. Departamento de Química, Facultad de Ciencias, Universidad de Chile. (In Spanish)

Ilabaca, M., Olaeta, I., Campos, E., Villaire, J., Tellez-Rojo, M., Romieu, I., 1999. Association between Levels of Fine Particulate and Emergency Visits for Pneomonia and other Respiratory Illnesses among Children in Santiago, Chile. *J. Air & Waste Manage. Assoc.*, 49, 154-163

IPCC, 2000. "Summary for Policy Makers: Emission Scenarios". Special Report of IPCC Working Group III. Intergovernmental Panel on Climate Change. [Available at www.ipcc.ch]

Kavouras, G., Koutrakis, P., Cereceda-Balic, F., Oyola, P., 2001. Source apportionment of PM10 and PM2.5 in five Chilean cities using factor analysis. *J. Air & Waste Management Asso*c., 51, 451-464.

Källén, E. (Ed.), 1996. HIRLAM documentation manual - System 2.5, pp.240. [Available from the Swedish Meteorological and Hydrological Institute, S-601 76, Norrköping, Sweden]

Robertson, L., Langner, J., and Engardt, M. 1999. An Eulerian limited-area atmospheric transport model. *J. Appl. Met*. 38, 190-210.

Rodhe, H., Langner, J., Gallardo, L., and Kjellström, E., 1995. Global scale transport of acidifying pollutants. *Water, Air and Soil Pollut.*, 85, 37-50.

Rutllant, J. and Garreaud, R., 1995. Meteorological Air Pollution Potential for Santiago, Chile: Towards an Objective Episode Forecasting. *Env. Monit. and Assess.*, 34, 223-244.

Räisänen, J., Rummukainen, M., Ullerstig, A., 2001. Downscaling of greenhouse gas induced climate change in two GCMs with the Rossby Centre regional climate model for northern Europe. *Tellus 53A*, 168-191.

Seibert, P., Beyrich, F., Gryning, S., Joffre, S., Rasmussen, A. and Tercier, P., 2000. Review and intercomparison of operational methods for the determination of the mixing height. *Atmos. Env*., 34, 1001-1027.

Trier, A. And Firingueti, L., 1994. A time series investigation of visibility in an urban atmosphere-I. *Atmos. Env.*, 28, 5, 991-996.

Figure Captions

Figure 1. South America, Central Chile and location of the air quality stations in the modeling area. The map over Central Chile shows the actual topography on an 800 m resolution. The copper smelters are indicated with white cilinders. The urban areas of Rancagua (south), Santiago (center) and Valparaíso/Viña del Mar (north) are enclosed in white squares.

Figure 2. Features of a CL of type A, which appears between a reinforced Pacific high and a migratory cold high over Argentina. The shallow cyclonic circulation gives rise to easterly winds that are forced to subside by the high Andes. This, in turn, enhances the subsidence inversion. Aloft, westerly winds prevail.

Figure 3. Normalized SO_2 concentrations as measured at EMD (continuous line), mCY (dotted line) and at SUR (dashed line). The daily average concentrations for May 1998 (upper panel) and for the period June 15^{th} – July 15^{th} 1999 (lower panel) have been normalized by the maximum of each time-series.

Figure 4. Measurements at Lo Prado station during two type A episodes in May 13-24 1998 (left panels) and July 1-11 1999 (right panels). The time-series of zonal wind speed in m/s are shown in the upper panels. Temperature in ºC (continuous line, left axis) and relative humidity in % (dashed line, right axis) are presented in the middle panels. The average concentrations of $PM_{2.5}$ (dashed, right axis) and SO_2 (continuous, left axis) in $\mu g/m^3$ measured in the Santiago monitoring network are shown in the lower panels.

Figure 5. a) Observed (left panels) and model simulated (right panels) vertical profiles of temperature (K), wind speed (m/s) and wind direction (deg) for the sounding station Quintero in May 1998. b) As in a) except for Santo Domingo in May 1998.

Figure 6. Observed (left panels) and model simulated (right panels) vertical profiles of temperature (K), wind speed (m/s) and wind direction (deg) for the acoustic sounding station at La Platina, south of Santiago in June-July 1999.

Figure 7. The upper panels show observed (dashed line) and simulated (continuous line) daily averaged SO_2 concentration in $\mu gS/m^3$ at EMD for two CL episodes in May 13-24 1998 (left panels) and July 1-11 1999 (right panels), respectively. The middle panels show the MATCH calculated BLH (continuous line) and an independent estimate by Gómez (2001) (dashed line) of BLH at EMD in m above the ground. The lower panels show simulated (continuous line) and measured (dashed line) relative humidity at EMD.

Figure 8. Simulated (continuous) and observed (dashed) SO_2 (upper panel), sulfate (middle panel) concentrations in downtown Santiago (EMD) for the periods May 1^{st} –30^{th} 1998 (left panels) and June 15^{th} to July 15^{th} 1999 (right panels). Also shown the contribution of Caletones copper smelter (dotted). The lower panel shows the ratio SO_4-S/(SO_2-S+SO_4-S) as simulated (continuous) and observed (dashed). The dotted line indicates the observed ratio between $PM_{2.5}$ and PM_{10} at EMD. Notice that the scale for the sulfate content is different for the left and right panels.

Figure 9. Vertically integrated SO_2 in ppbv, over Central Chile as simulated by the model for January 17^{th} 1998 according to the prevailing winds and considering only the emissions from the smelters.

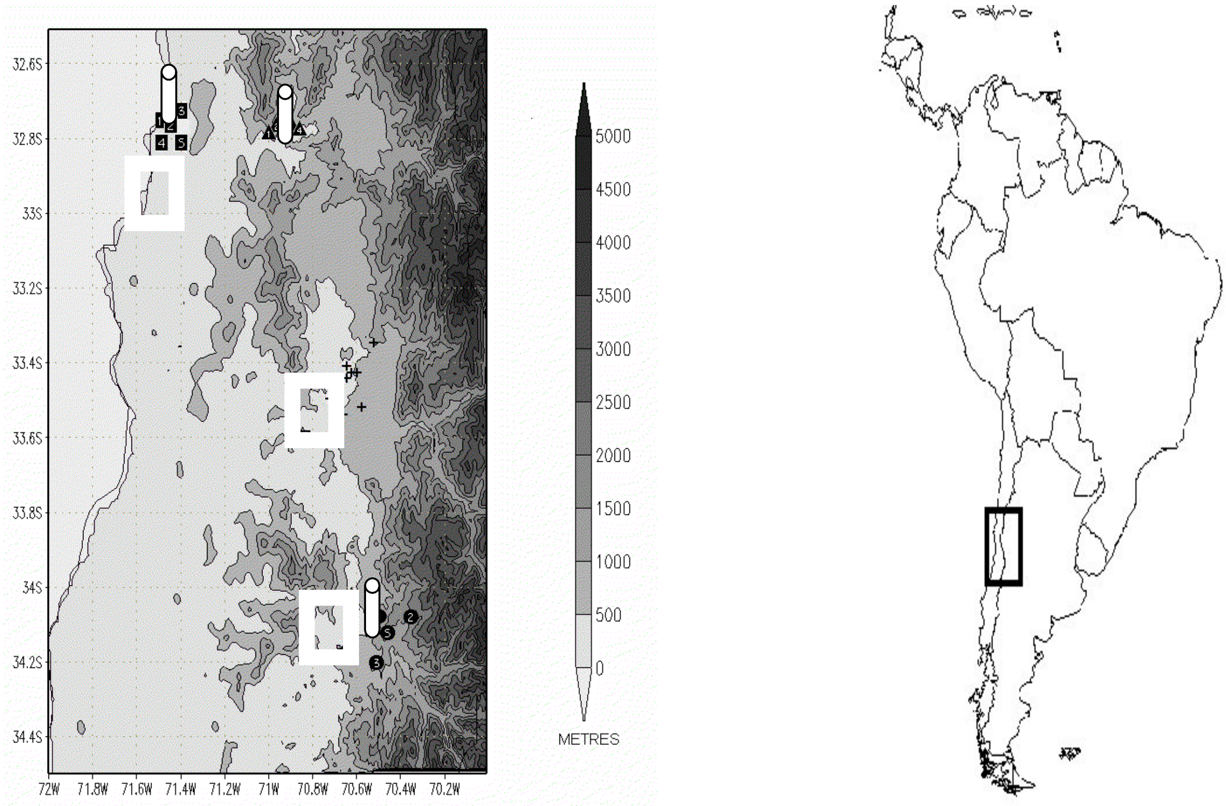

Figure 1. South America, Central Chile and location of the air quality stations in the modeling area. The map over Central Chile shows the actual topography on an 800 m resolution, every 500 m. The copper smelters are indicated with white cilinders. The urban areas of Rancagua (south), Santiago (center) and Valparaíso/Viña del Mar (north) are enclosed in white squares.

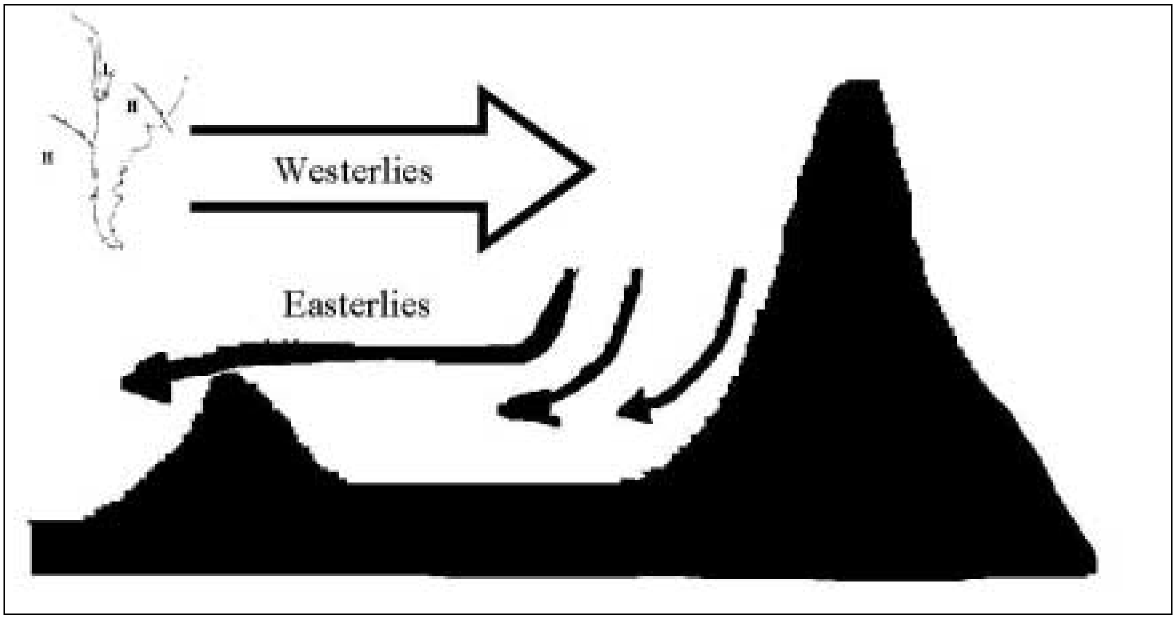

Figure 2. Features of a CL of type A, which appears between a reinforced Pacific high and a migratory cold high over Argentina. The shallow cyclonic circulation gives rise to easterly winds that are forced to subside by the high Andes. This, in turn, enhances the subsidence inversion. Aloft, westerly winds prevail.

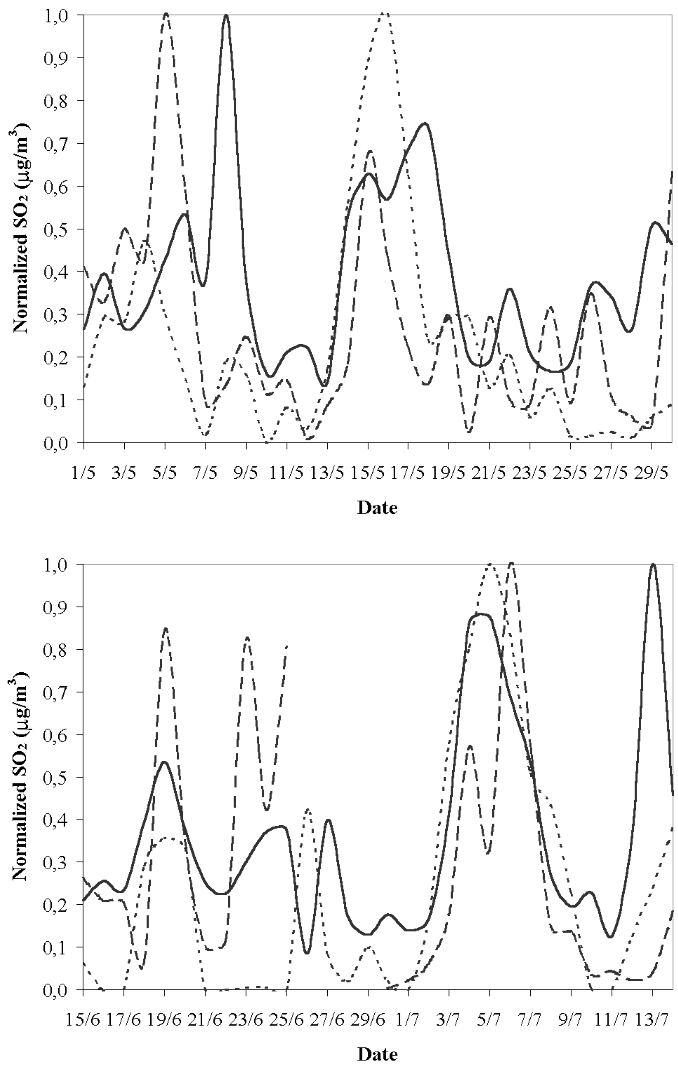

Figure 3. Normalized SO_2 concentrations as measured at EMD (continuous line), mCY (dotted line) and at SUR (dashed line). The daily average concentrations for May 1998 (upper panel) and for the period June 15^{th} – July 15^{th} 1999 (lower panel) have been normalized by the maximum of each time-series.

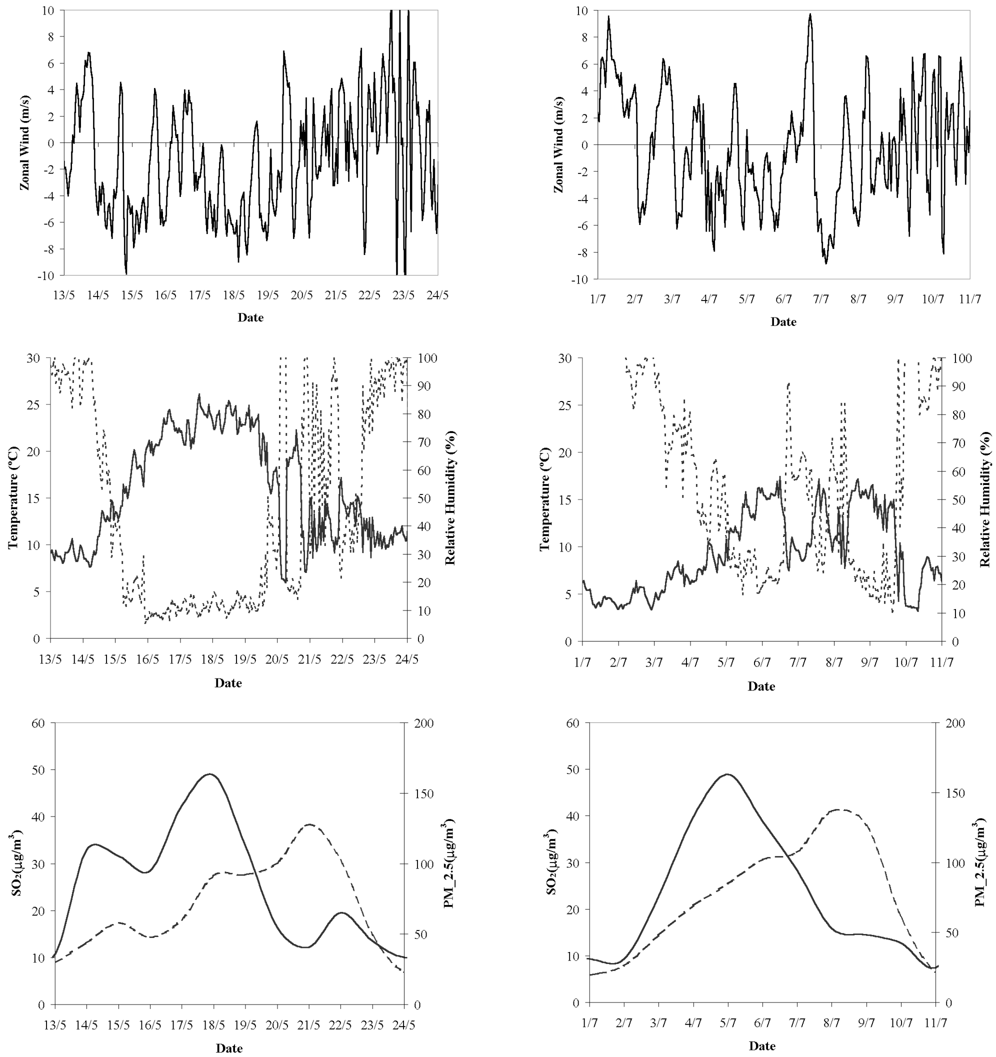

Figure 4. Measurements at Lo Prado station during two type A episodes in May 13-24 1998 (left panels) and July 1-11 1999 (right panels). The time-series of zonal wind speed in m/s are shown in the upper panels. Temperature in ºC (continuous line, left axis) and relative humidity in % (dashed line, right axis) are presented in the middle panels. The average concentrations of $PM_{2.5}$ (dashed, right axis) and SO_2 (continuous, left axis) in $\mu g/m^3$ measured in the Santiago monitoring network are shown in the lower panels.

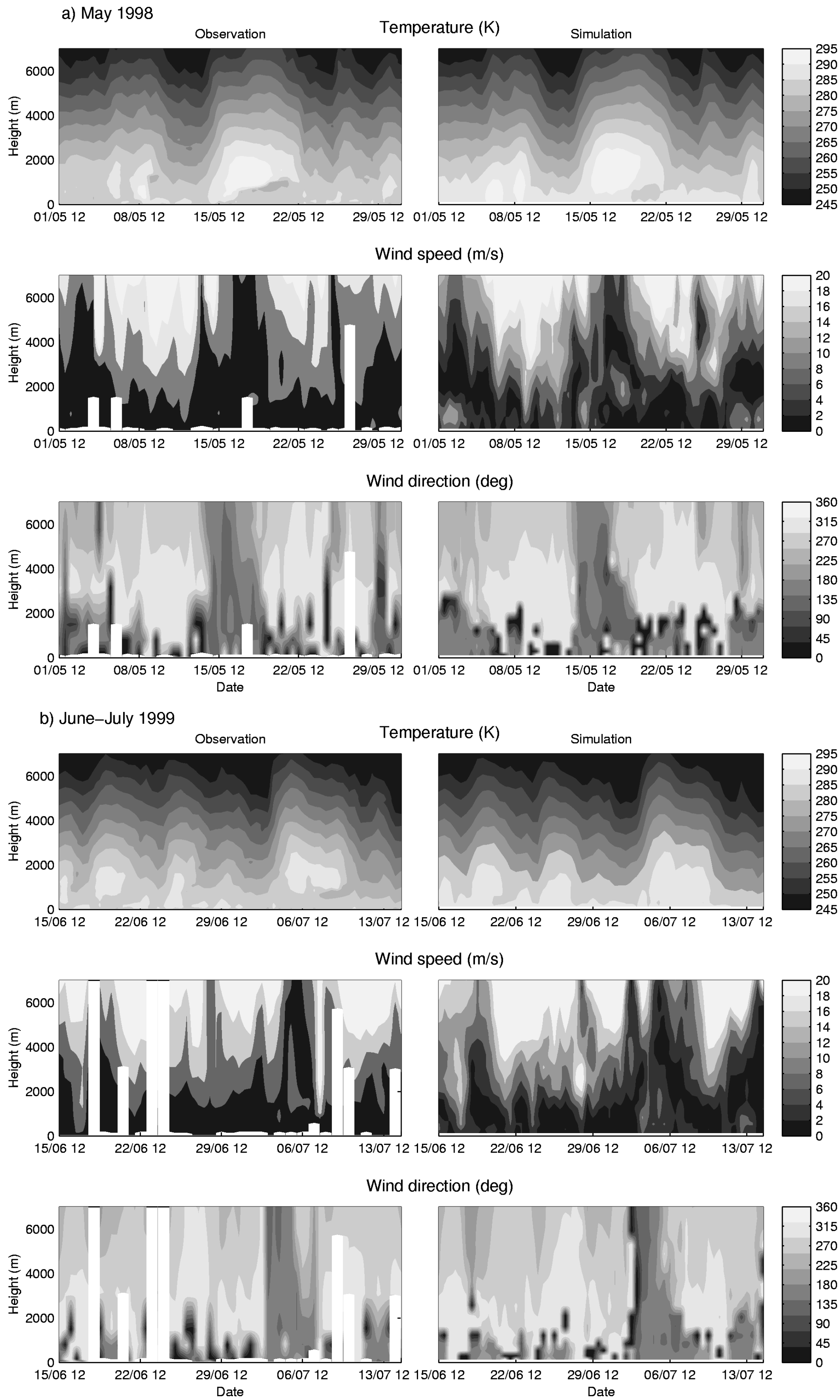

Figure 5. a) Observed (left panels) and model simulated (right panels) vertical profiles of temperature (K), wind speed (m/s) and wind direction (deg) for the sounding station Quintero in May 1998. b) As in a) except for Santo Domingo in May 1998.

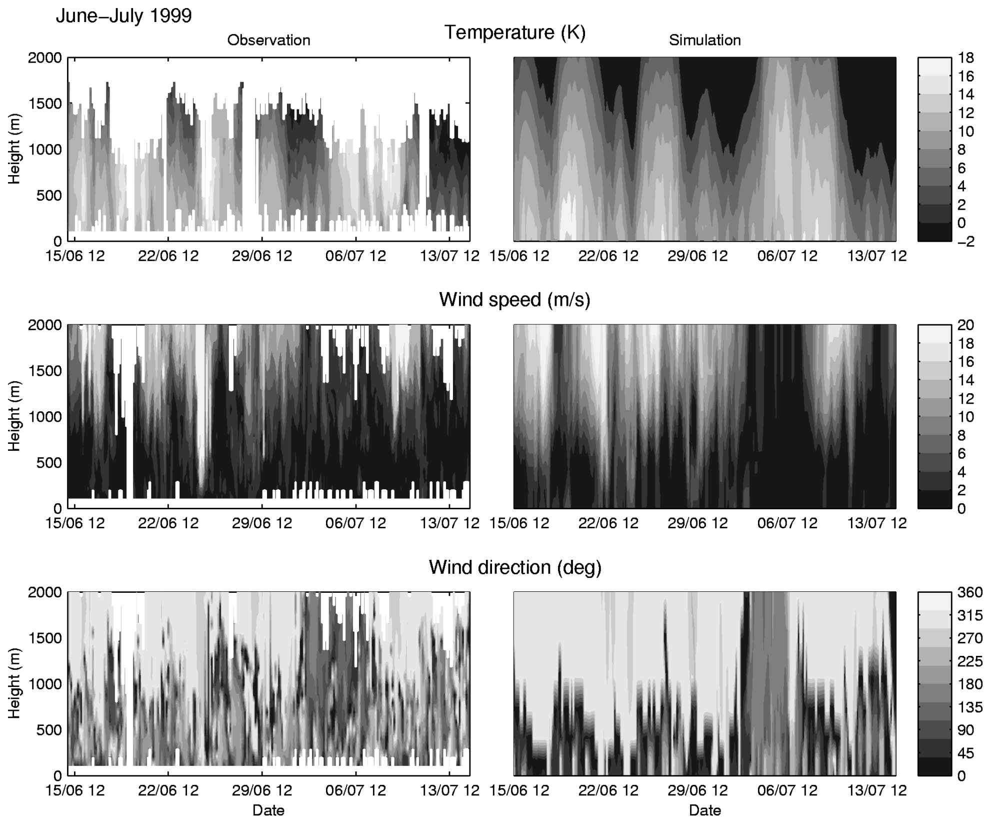

Figure 6. Observed (left panels) and model simulated (right panels) vertical profiles of temperature (ºC), wind speed (m/s) and wind direction (deg) for the acoustic sounding station at La Platina, south of Santiago in June-July 1999.

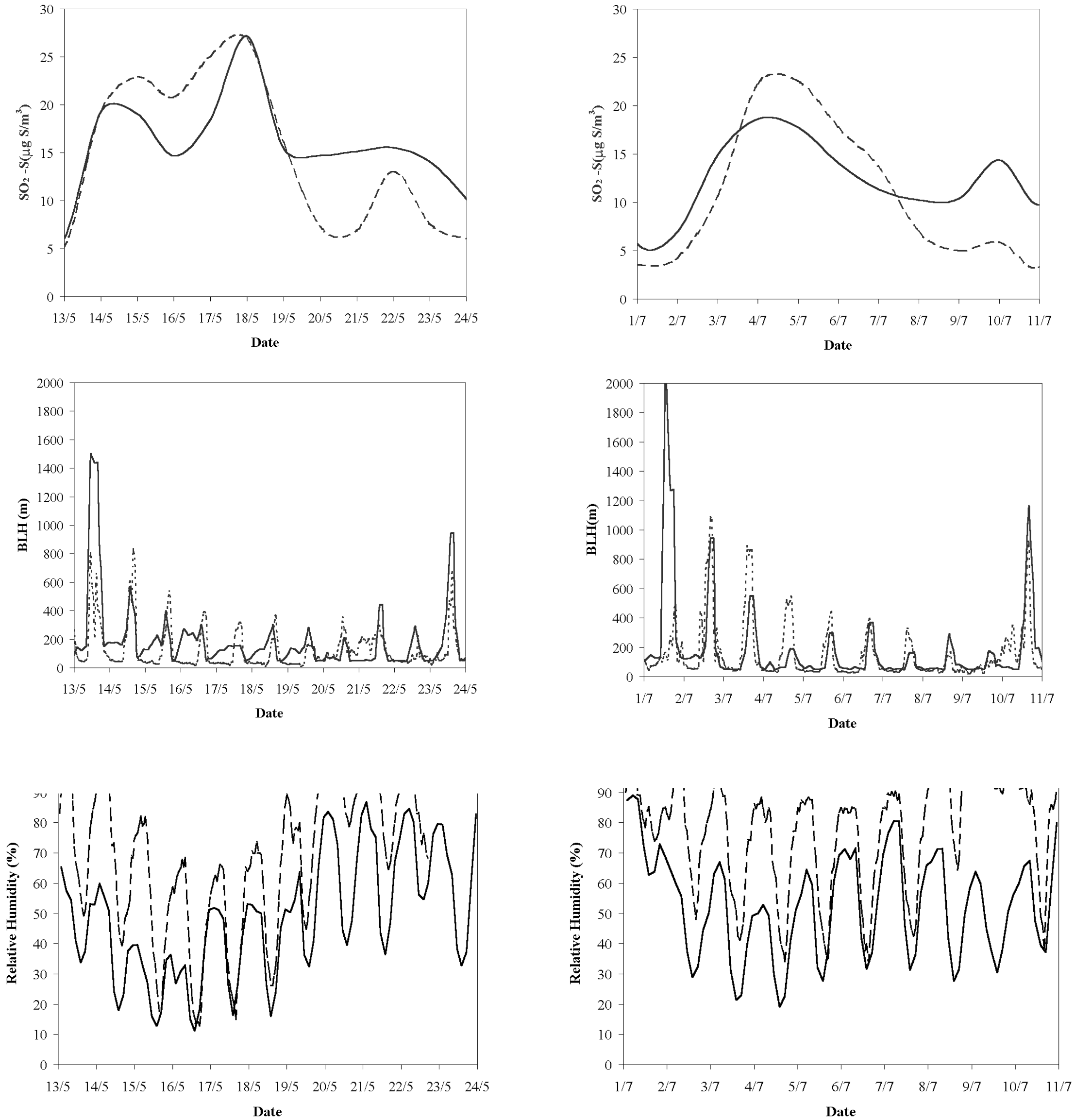

Figure 7. The upper panels show observed (dashed line) and simulated (continuous line) daily averaged SO_2 concentration in $\mu gS/m^3$ at EMD for two CL episodes in May 13-24 1998 (left panels) and July 1-11 1999 (right panels), respectively. The middle panels show the MATCH calculated BLH (continuous line) and an independent estimate by Gómez (2001) (dashed line) of BLH at EMD in m above the ground. The lower panels show simulated (continuous line) and measured (dashed line) relative humidity at EMD.

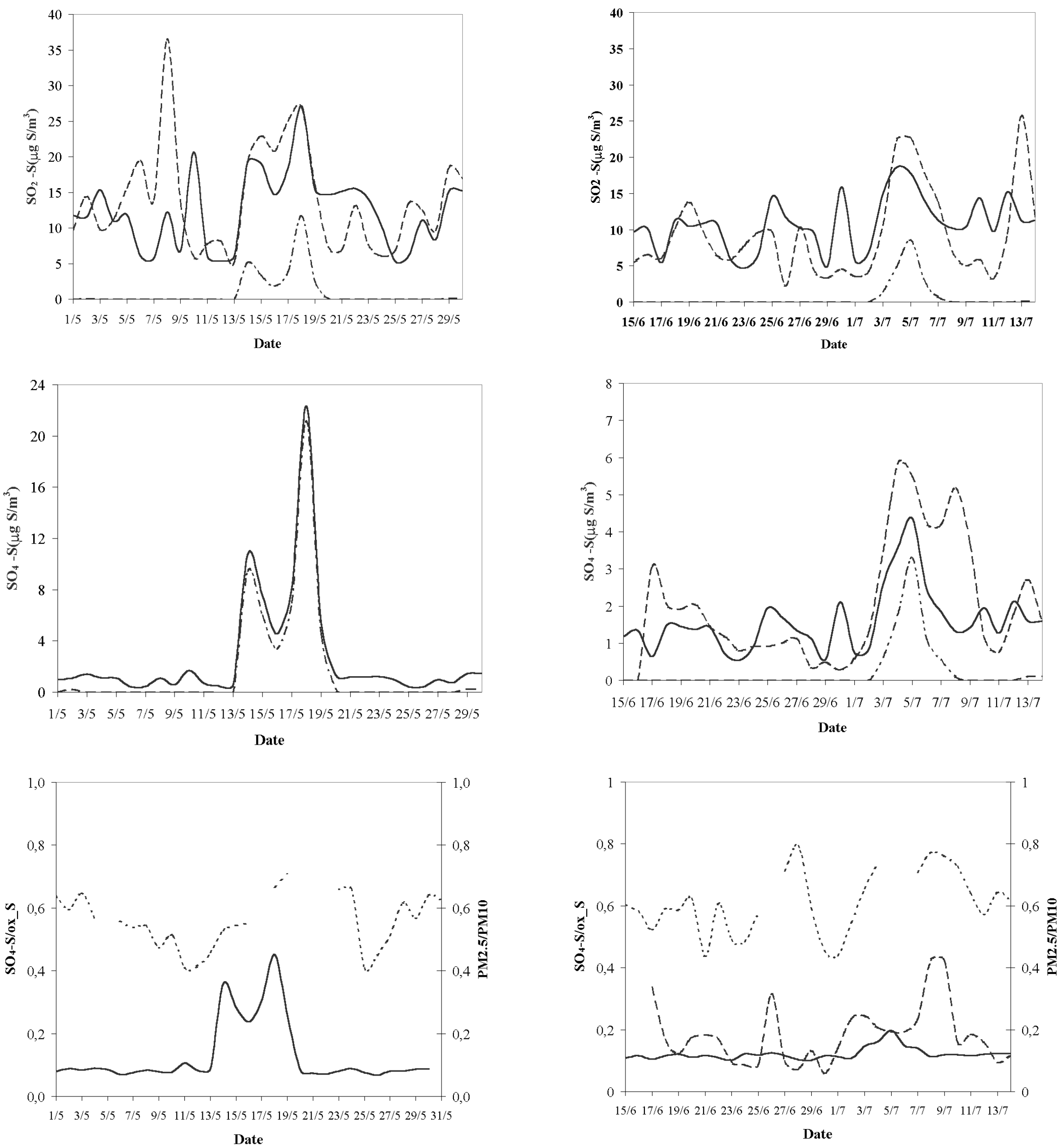

Figure 8. Simulated (continuous) and observed (dashed) SO_2 (upper panel), sulfate (middle panel) concentrations in downtown Santiago (EMD) for the periods May 1st –30th 1998 (left panels) and June 15th to July 15th 1999 (right panels). Also shown the contribution of Caletones copper smelter (dotted). The lower panel shows the ratio SO_4-S/(SO_2-S+SO_4-S) as simulated (continuous) and observed (dashed). The dotted line indicates the observed ratio between $PM_{2.5}$ and PM_{10} at EMD. Notice that the scale for the sulfate content is different for the left and right panels.

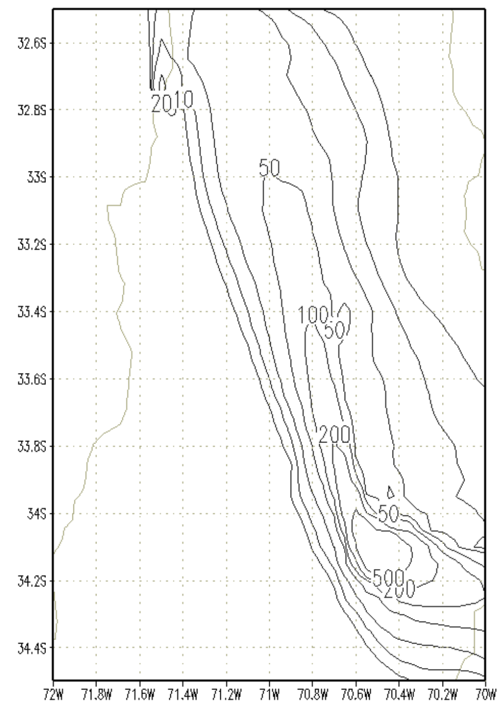

Figure 9. Vertically integrated SO_2 in ppbv, over Central Chile as simulated by the model for January 17th 1998 according to the prevailing winds and considering only the emissions from the smelters.