# Analysis of Aerosol Particles and Coarse Particulate Matter Concentrations in Chillán, Chile, 2001–2003

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## ABSTRACT

Daily particle samples were collected in Chillán, Chile, at six urban locations from September 1, 2001, through September 30, 2003. Aerosol samples were collected using monitors equipped with a Sierra Andersen 246-b cyclone inlet on Teflon filters. Average concentrations of coarse particulate matter ($PM_{10}$) for the 2001–2003 period ranged from 43.4 $\mu g/m^3$ to 81.8 $\mu g/m^3$ across the six sites. Annual $PM_{10}$ concentration levels exceeded the European Union concentration limits. Mean $PM_{10}$ levels during the cold season (April through September) were more than twice as high as those observed in the warm season (October through March). Average contributions to $PM_{10}$ from organic matter, soil dust, nitrate ($NO_3^-$), elemental carbon, ammonium ($NH_4^+$), and sulfate ($SO_4^{2-}$) were 31%, 27%, 11%, 8%, 7%, and 5%, respectively. The chemical analyses indicated that carbonaceous substances were the most abundant components of $PM_{10}$ in cold months, whereas crustal material was the most abundant component of $PM_{10}$ during warm months. Higher concentration levels were observed in the downtown area suggesting a clear anthropogenic origin, whereas in the rural sites the source was mainly natural, such as resuspended soil dust associated with traffic on unpaved roads and agricultural activities.

### IMPLICATIONS

This paper examines $PM_{10}$ particle measurements in Chillán, Chile, from 2001 to 2003. Air pollution studies in Chile have been principally performed in the capital city of Santiago, which is one of the most polluted cities in South America. The current results indicate that respirable particulate air pollution in Chillán is also high, exceeding both U.S. and European particle standards. Particle levels are particularly high during the cold season, most likely because of the influences of meteorology and anthropogenic sources. Chillán is a city of ~170,000 inhabitants with a rapidly growing population. This city needs to develop its research and pollution prevention programs to avoid developing the environmental problems that face other large metropolitan areas.

## INTRODUCTION

Aerosol coarse particulate matter ($PM_{10}$) is produced by natural and human activities and can be inhaled by organisms. Particles $<10$ $\mu m$ in aerodynamic diameter are called respiratory particulates or $PM_{10}$. Typical natural sources include saline particles from the sea and windblown dust. On the other hand, particulate matter (PM) is also generated as a result of industrial activities, traffic, and combustion processes.[1]

Recent air pollution studies have demonstrated statistical associations between health effects and ambient concentrations of PM.[2–6] Most of these studies have been performed in cities of industrialized countries, generally presenting similar results.[7–10] Results coming from cities located in developing countries present a large variability, probably because of soft control rules and different industrialization stages.[11] It has become clear in the industrial world that air pollution affects not only large metropolitan areas but also medium-sized urban areas.[1–7] Air pollution studies in Chile concerning PM have been principally focused in the capital city of Santiago, although little research has been carried out in urban areas with population ranges between 50,000 and 250,000 inhabitants.[12,13] Two preliminary studies in surroundings areas of Chillán indicated a significant presence of aerosol particles with an aerodynamic diameter $<10$ $\mu m$.[14,15] The lands surrounding Chillán are the result of fluvial and volcanic deposits transported by rivers from the Andes Mountain and are

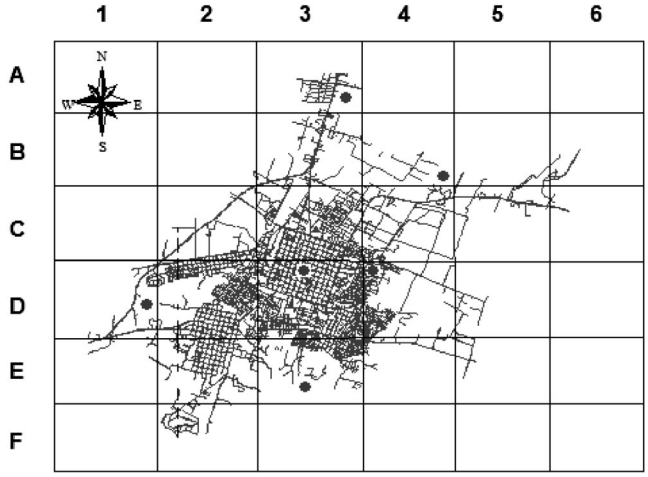


**Figure 1.** Location of the $PM_{10}$ samplers in the city of Chillán (monitoring station located at downtown, D3; hospital location, D4).

characterized by a flat topography with smooth slopes. The climate is Mediterranean with a prolonged dry season (6 months) followed by a wet season with an annual average precipitation of 1100 mm.

These Chilean cities are among the fastest growing cities in Latin America and present the greatest possibilities of sustainable development.[16] Consequently, it is imperative that efforts to improve air quality are enforced, so these middle-sized cities, such as Chillán, do not necessarily develop the same environmental problems of contaminated metropolitan areas. This study represents a first endeavor to evaluate the aerosol concentration and composition in the city of Chillán. Additional work is required to develop a complete understanding of the composition, size, particulate distribution, and meteorological effects, among other factors. The objective of this study was to analyze the concentration and composition of the $PM_{10}$ in the city of Chillán, considering its temporal and spatial variability.

## EXPERIMENTAL WORK

This study was performed between September 1, 2001, and September 30, 2003, in the city of Chillán, Chile (36° 34′S latitude, 72° W longitude, 144 m altitude above sea level). $PM_{10}$ concentrations were collected in six fixed locations within the city (Figure 1) using IMPROVE monitors equipped with a Sierra Andersen 246-b cyclone on 25-mm and 47-mm Teflon filters (Gelman Scientific) operating at a flow rate of 16.7 L/min. Sampler inlets were located 3 m above the ground. The sampling sites were selected considering the movable and stationary sources with significant emission contribution and the potential effects of prevailing north–south wind direction. Daily collection was programmed from 12:00 a.m. to 11:59 p.m. for the period between September 1, 2001, and December 31, 2002. From January 1, 2003, to August 31, 2003, samplers operated for 12-hr diurnal periods from 6:00 a.m. to 6:00 p.m. and nocturnal periods from 6:00 p.m. to 6:00 a.m.

The integrity of each filter was inspected before use. The $PM_{10}$ concentration was determined using a Cahn 31 microbalance. Both blank and field filter samples were conditioned for 24 hr before being weighed at constant temperature (22 ± 3 °C) and relative humidity (RH; 40% ± 5%). Filters were weighed before and after each continuous monitoring sampling. To characterize the chemical compounds in the filters, 25 elements (S, Cl, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, As, Pb, Se, Br, Rb, Sr, Zr, Mo, Na, Mg, Al, Si, and P) were analyzed using energy-dispersive X-ray fluorescence spectrometry at the Natural Resources Laboratory of the Universidad de Concepción and at the Crocker Nuclear Laboratory of the University of California, Davis. Filters were used to determine organic carbon using a Desert Research Institute thermal optical analyzer (model 4000X). The organic matter concentration was obtained by multiplying the organic carbon content by a factor 1.4.[17,18] Elemental carbon (EC) was determined by a thermographic method following the procedure detailed in the reference.[8] Ammonium ($NH_4^+$) detection analysis was performed using a spectrometer Lambda 2 Perkin-Elmer. As suggested,[19] the ions nitrate ($NO_3^-$), sulfate ($SO_4^{2-}$), and chlorine ($Cl^-$) were determined by ion chromatography (Dionex DX 100).

Each monitor was equipped to handle four cassettes plus an internal programmable clock, which permitted filter changing and equipment maintenance every 5 days. Approximately 1800 filters were collected throughout the study and were used to determine the $PM_{10}$ mass. Only one sample was taken at each site per day. At each monitoring site, a mixed sample from this set of filters was prepared once a month for chemical analyses: 10 filters for ion analysis, 3 filters for carbon determination, and 10 filters for elemental concentration determination. For the mixed samples, the selected filters were shredded into small pieces of ~1 $cm^2$, macerated with nitric acid (65%), and centrifuged. The concentration values resulting from the mixed samples were monthly average values for each of the components measured.

As suggested,[20] the proportion of soil (wind-blown dust) in $PM_{10}$ was determined by eq 1, where Si, Al, Fe, Ca, and Ti are the concentrations of those elements in micrograms per cubic meter.

$$\text{Soil} = 2.20\,\text{Al} + 2.49\,\text{Si} + 1.6\,\text{Ca} + 1.94\,\text{Ti} + 2.38\,\text{Fe} \quad (1)$$

In Chillán, wood is a common fuel for residential heating, producing visually dense smoke clouds during cold months. To calculate the amount of nongeologic potassium, eq 2 was used as suggested,[16] where K and Fe are the concentrations of those elements ($\mu g/m^3$).

$$K_{nos} = K - 0.6\,\text{Fe} \quad (2)$$

Finally, meteorological data, such as wind speed, wind direction, temperature, rain fall, and RH were obtained during particle sampling from the weather station of the University of Concepción, located northeast of the Chillán city limit (point B4 in Figure 1).

## RESULTS AND DISCUSSION

Figure 2 presents the temporal and spatial variability of $PM_{10}$ mass concentration. Results show that downtown station D3 registered the highest concentrations during

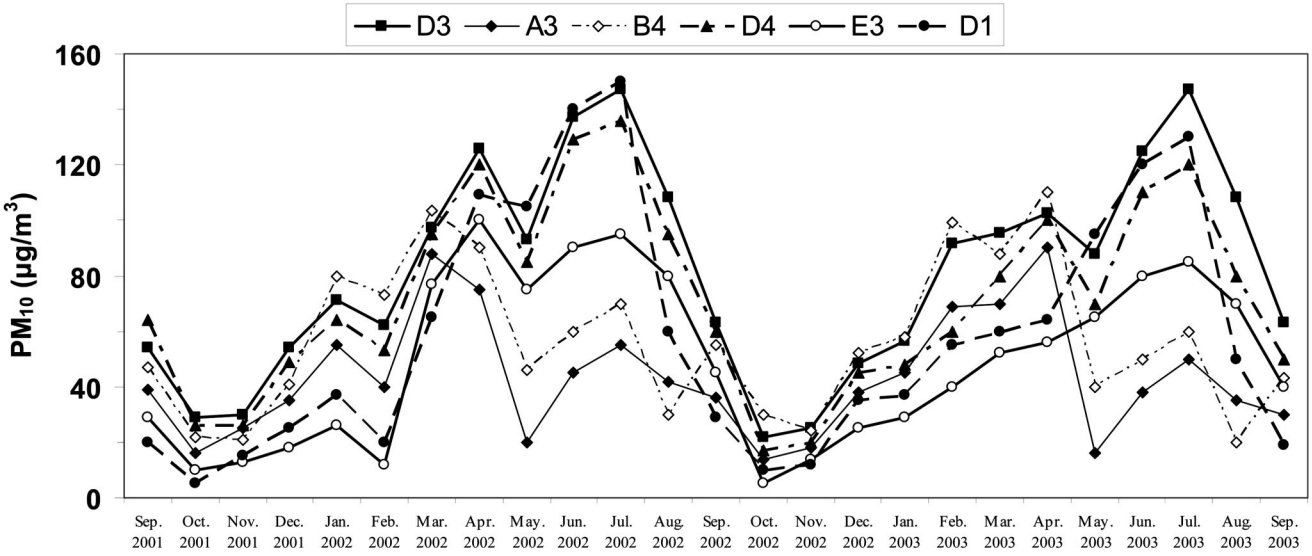


**Figure 2.** Average monthly $PM_{10}$ mass concentration measured at six different locations within the city of Chillán from September 1, 2001, to September 30, 2003.

the warm and cold periods. Station D1, located southwest, presented high values similar to D3 in winter, depending on the prevailing wind direction in those months (northeast). Wind effects were also observed in the concentration values during the warm months: the stations located in the north and northeast of the city (A3 and B4, respectively) registered higher aerosol concentrations than the stations located to the south of Chillán (E3 and D1). During that period, wind-blown dust comes from the southwest, dragging the accumulated fugitive dust out of the city to the northwest area increasing the total mass concentration. It is important to note that, in general, the $PM_{10}$ mass concentration declined in February and May 2002 because of increased rainfall during those months. The annual average $PM_{10}$ mass concentration measured downtown (monitoring station D3) from January 1 to December 31, 2002, was 83.4 ± 8.7 $\mu g/m^3$, which exceeded both the U.S. air quality standard of 50 $\mu g/m^3$ and the European limit of 30 $\mu g/m^3$.[21,22] Additionally, it is higher than the level reported in other Chilean cities, such as Temuco (67.7 ± 3.2 $\mu g/m^3$), Rancagua (73.8 ± 4.2 $\mu g/m^3$), Valparaíso (77.5 ± 2.7 $\mu g/m^3$), Iquique (62,1 ± 3,5 $\mu g/m^3$), and Viña del Mar (55.5 ± 3.5 $\mu g/m^3$),[12] as well as being lower than the level measured in México City (115 $\mu g/m^3$). It was higher than the $PM_{10}$ mass concentration registered in Montreal (27.8 $\mu g/m^3$), Toronto (28.1 $\mu g/m^3$), Vancouver (26.9 $\mu g/m^3$), and Ottawa (22.6 $\mu g/m^3$).[23] Moreover, it was higher than the values measured in Boston (19 $\mu g/m^3$), Washington, DC (28 $\mu g/m^3$), Memphis (30.9 $\mu g/m^3$), and Philadelphia (26 $\mu g/m^3$).[24] The annual average $PM_{10}$ mass concentration measured at the edge of the city (monitoring station A3) was 43.8 ± 9.3 $\mu g/m^3$. Considering all six of the monitoring sites, the $PM_{10}$ mean annual mass concentration (63.5 ± 14.6 $\mu g/m^3$) was 27% higher than the $PM_{10}$ U.S. air quality standard of 50 $\mu g/m^3$, which is also the Chilean standard.

Figure 3 shows the temporal daily correlation between the mass $PM_{10}$ concentrations downtown (point D3 in Figure 1) versus atmospheric temperature in the city of Chillán from September 1, 2001, to April 30, 2003. A clear seasonal variation in $PM_{10}$ concentration was observed, with higher values occurring in winter in comparison with summer. An inverse correlation between $PM_{10}$ mass concentration and air temperature was particularly notable, following the same pattern as has been previously found by other researchers.[8,10] The observed variability is quite plausible, because the main factors controlling $PM_{10}$ concentration are temperature dependent. The greater emissions could be because of residential heating methods (fire stoves) and lower mixing heights in the cold season.

Average $PM_{10}$ concentrations were 40% lower during weekends (Figure 4), which suggests that air pollution is significantly affected by road traffic, construction work, and industrial processes that generally take place during working days. These results are similar to those cited in other research, where it was shown that street traffic contributes strongly to this variation through the resuspension of street dust and fine particle emission by combustion motors.[8] The large difference between the workday and weekend-day concentrations is probably because of the fact that the majority of Chillán local stores and industries are closed on Sunday.

The difference in ambient $PM_{10}$ concentration between warm and cold months appears to be principally the result of nighttime influences as opposed to daytime influences. Figure 5 shows that whereas both daytime and nighttime concentrations increase as the months become colder, the increase in ambient concentrations is greater for the nighttime than the daytime data. Therefore, during warm months, the higher $PM_{10}$ concentrations occur during the day, whereas higher $PM_{10}$ concentrations during cold months take place during the night. This result can be explained by the fact that most residential heating systems are based on wood burning, which incorporates soot and other products into the atmosphere. Heating begins operating in the evening and runs until dawn. Additionally, there are strong winds in the afternoon during the winter, from 12:00 p.m. to ~6:00 p.m.,

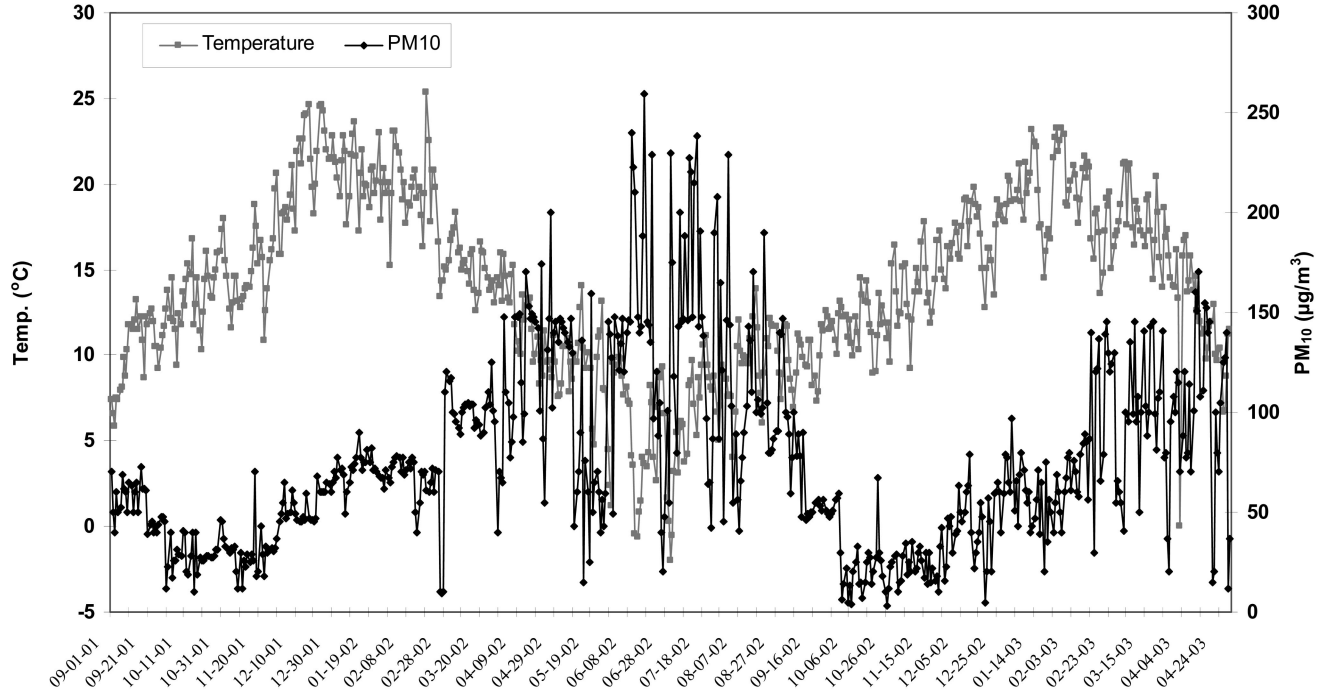


**Figure 3.** Temporal daily time series between the mass $PM_{10}$ concentration measured at downtown vs. atmospheric temperature in the city of Chillán from September 1, 2001, to April 30, 2003.

whereas stronger winds occur later in the evening in summer, from ~6:00 p.m. to 9:00 p.m.

Table 1 presents the seasonal $PM_{10}$ averages for chemical compounds in Chillán during 2001, 2002, and 2003. Each seasonal value is the average of six stations with continuous monitoring. OM, $NO_3$, EC, $NH_4$, and $SO_4$ concentrations were higher in the cold season than during warm months. As shown in Table 2, the most prominent elements were Si, Fe, Al, Ca, Mg, Cu, K, Ti, Cl, and Zn. K, Ti, Cl, Zn, Mn, V, and Mo concentrations were higher in the cold season than during warm months. On the other hand, Si, Fe, Al, Ca, Mg, and Sr concentrations were higher during summertime (December to March) than during cold months. The high Si, Fe, Al, and Ca concentrations found in summertime can be explained by resuspension of dust from roads and surrounding agricultural lands. This finding is similar to results registered in recent studies abroad, suggesting that air pollution during the cold season originates from anthropogenic sources, whereas that in the summertime comes from natural sources.[7,8,25,26] The components OM, EC, and $NO_3$ result from combustion gases of vehicles and firewood burning,[24,25,27] whereas $NH4^+$ originates in agricultural activities as well as from vehicles with catalyst and industrial sources.[6] Particularly, OM is of primary as well as secondary origin.[8] The $NO_3^-$ concentration is directly associated with city traffic because it is formed from the nitric oxide ($NO_2$).

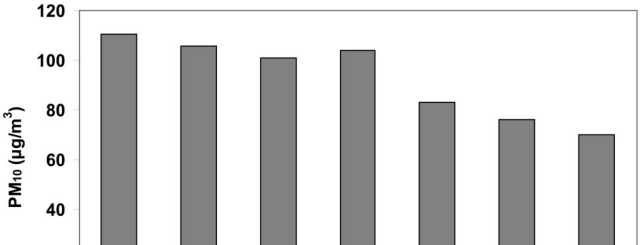


**Figure 4.** Mean $PM_{10}$ concentration during workdays and weekends measured in downtown Chillán during February 2003.

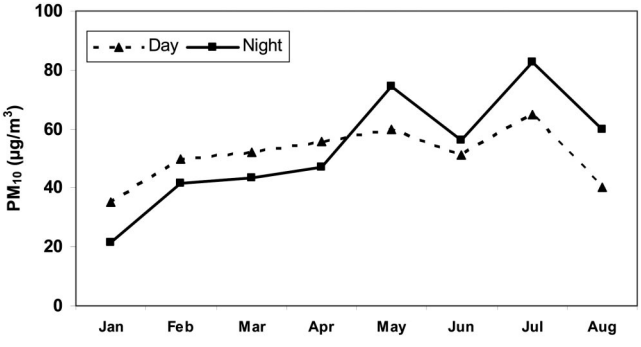


**Figure 5.** Monthly average mass $PM_{10}$ concentration during daily and nocturnal monitorings in downtown Chillán during 2003.

Figure 6 presents the temporal variability in the chemical compounds conforming the $PM_{10}$. A clear temporary variability for OM, $NO_3$, $NH_4$, $SO_4$, and soil was observed. For the anthropogenic compounds, the concentrations were higher during the cold months. On the other hand, the highest soil concentrations were measured in warm months. A seasonal pattern for EC was not clearly observed, similar to results reported in a similar study.[8]

Figure 7 indicates for each station the spatial variability of the chemical compounds that form part of the $PM_{10}$. It can be observed that at downtown and hospital locations (monitoring stations D3 and D4, respectively) the OM, EC, $NO_3$, $SO_4$, and $NH_4$ concentrations were higher than the geologic compound levels (denoted by

**Table 1.** Average chemical compounds $PM_{10}$ ($\mu g/m^3$) in the atmosphere of the city of Chillán between September 2001 and September 2003.

| Chemical Compound | Spring September 22–December 21 | Summer December 22–March 21 | Autumn March 22–June 21 | Winter June 22–September 21 | Mean Value |
|---|---|---|---|---|---|
| OM | 12.56 ± 9.02 | 16.37 ± 10.72 | 38.35 ± 16.46 | 35.29 ± 20.11 | 25.64 ± 14.08 |
| $NO_3$ | 4.55 ± 2.13 | 6.13 ± 1.99 | 18.01 ± 6.03 | 11.98 ± 7.43 | 10.17 ± 4.39 |
| EC | 5.21 ± 2.73 | 5.28 ± 1.87 | 6.54 ± 2.05 | 6.02 ± 2.07 | 5.76 ± 2.18 |
| $NH_4$ | 3.64 ± 1.17 | 3.41 ± 1.12 | 6.23 ± 1.58 | 7.85 ± 2.23 | 5.28 ± 1.53 |
| $SO_4$ | 2.12 ± 0.44 | 1.53 ± 1.03 | 5.97 ± 1.86 | 6.52 ± 2.03 | 4.04 ± 1.34 |

*Notes:* Seasonal values were calculated averaging the monthly values obtained at each site; for instance, Spring represents the average of the values corresponding with September, October, November, and December.

soil). The observed $SO_4^{2-}$ particles were probably originated in the $SO_2$ gas-to-particle conversion process. Here, the highest $SO_4^{2-}$ concentrations were detected at the monitoring station D4 located at the local hospital, which has an energy plant with a coal-fired furnace and a local trash incinerator operating throughout the year. Higher $K_{nos}$ concentrations were observed in downtown Chillán (D3) and at the more populated sites within the city (D4

**Table 2.** Average chemical composition of $PM_{10}$ ($\mu g/m^3$) in the urban atmosphere of Chillán between September 2001 and September 2003.

| Chemical | Spring September 22–December 21 | Summer December 22–March 21 | Autumn March 22–June 21 | Winter June 22–September 21 | Mean Value |
|---|---|---|---|---|---|
| Si | 3.11 ± 1.05 | 9.17 ± 2.33 | 1.36 ± 3.44 | 1.03 ± 2.01 | 3.67 ± 7.12 |
| Fe | 2.37 ± 0.23 | 4.71 ± 0.52 | 0.97 ± 0.11 | 0.31 ± 0.09 | 2.09 ± 0.35 |
| Al | 1.94 ± 0.04 | 3.23 ± 0.50 | 0.39 ± 0.05 | 0.21 ± 0.14 | 1.44 ± 0.15 |
| Ca | 2.16 ± 0.22 | 3.62 ± 0.23 | 0.44 ± 0.12 | 0.24 ± 0.03 | 1.62 ± 0.20 |
| Mg | 0.39 ± 0.04 | 0.40 ± 0.05 | 0.26 ± 0.05 | 0.13 ± 0.04 | 0.30 ± 0.04 |
| Cu | 0.25 ± 0.02 | 0.26 ± 0.06 | 0.17 ± 0.11 | 0.12 ± 0.13 | 0.20 ± 0.03 |
| Sr | 0.05 ± 0.27 | 0.09 ± 0.16 | 0.03 ± 0.11 | 0.06 ± 0.23 | 0.06 ± 0.15 |
| As | 0.00 ± 0.04 | 0.00 ± 0.03 | 0.00 ± 0.04 | 0.08 ± 0.16 | 0.02 ± 0.03 |
| K | 1.01 ± 0.22 | 0.54 ± 0.31 | 4.24 ± 0.83 | 2.00 ± 0.04 | 1.95 ± 0.61 |
| Ti | 0.12 ± 0.03 | 0.18 ± 0.12 | 0.03 ± 0.51 | 0.76 ± 0.50 | 0.27 ± 0.23 |
| Cl | 0.35 ± 0.04 | 0.31 ± 0.04 | 0.44 ± 0.21 | 0.25 ± 0.21 | 0.34 ± 0.11 |
| Zn | 0.09 ± 0.03 | 0.25 ± 0.13 | 0.41 ± 0.15 | 0.54 ± 0.21 | 0.32 ± 0.10 |
| Pb | 0.01 ± 0.03 | 0.01 ± 0.14 | 0.03 ± 0.14 | 0.03 ± 0.20 | 0.02 ± 0.12 |
| Mn | 0.02 ± 0.16 | 0.03 ± 0.11 | 0.05 ± 0.13 | 0.09 ± 0.03 | 0.05 ± 0.13 |
| Se | 0.00 ± 0.02 | 0.02 ± 0.23 | 0.01 ± 0.32 | 0.01 ± 0.15 | 0.01 ± 0.21 |
| Ni | 0.01 ± 0.04 | 0.02 ± 0.04 | 0.01 ± 0.05 | 0.00 ± 0.21 | 0.01 ± 0.05 |
| V | 0.01 ± 0.02 | 0.01 ± 0.03 | 0.03 ± 0.03 | 0.01 ± 0.09 | 0.02 ± 0.04 |
| Ba | 0.13 ± 0.02 | 0.12 ± 0.02 | 0.14 ± 0.04 | 0.16 ± 0.03 | 0.14 ± 0.01 |
| S | 0.02 ± 0.03 | 0.01 ± 0.03 | 0.05 ± 0.03 | 0.11 ± 0.02 | 0.05 ± 0.02 |
| Mo | 0.01 ± 0.02 | 0.02 ± 0.01 | 0.07 ± 0.04 | 0.09 ± 0.02 | 0.05 ± 0.01 |
| Br | ND | ND | 0.024 ± 0.011 | ND | 0.02 ± 0.01 |

*Notes:* ND indicates not determined.

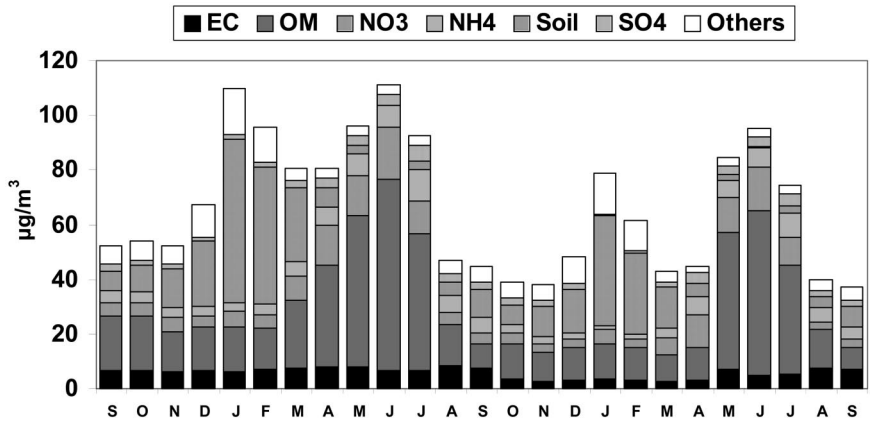


**Figure 6.** Average monthly chemical compound concentrations ($\mu g/m^3$) within the $PM_{10}$ in the city of Chillán from September 1, 2001, to September 30, 2003.

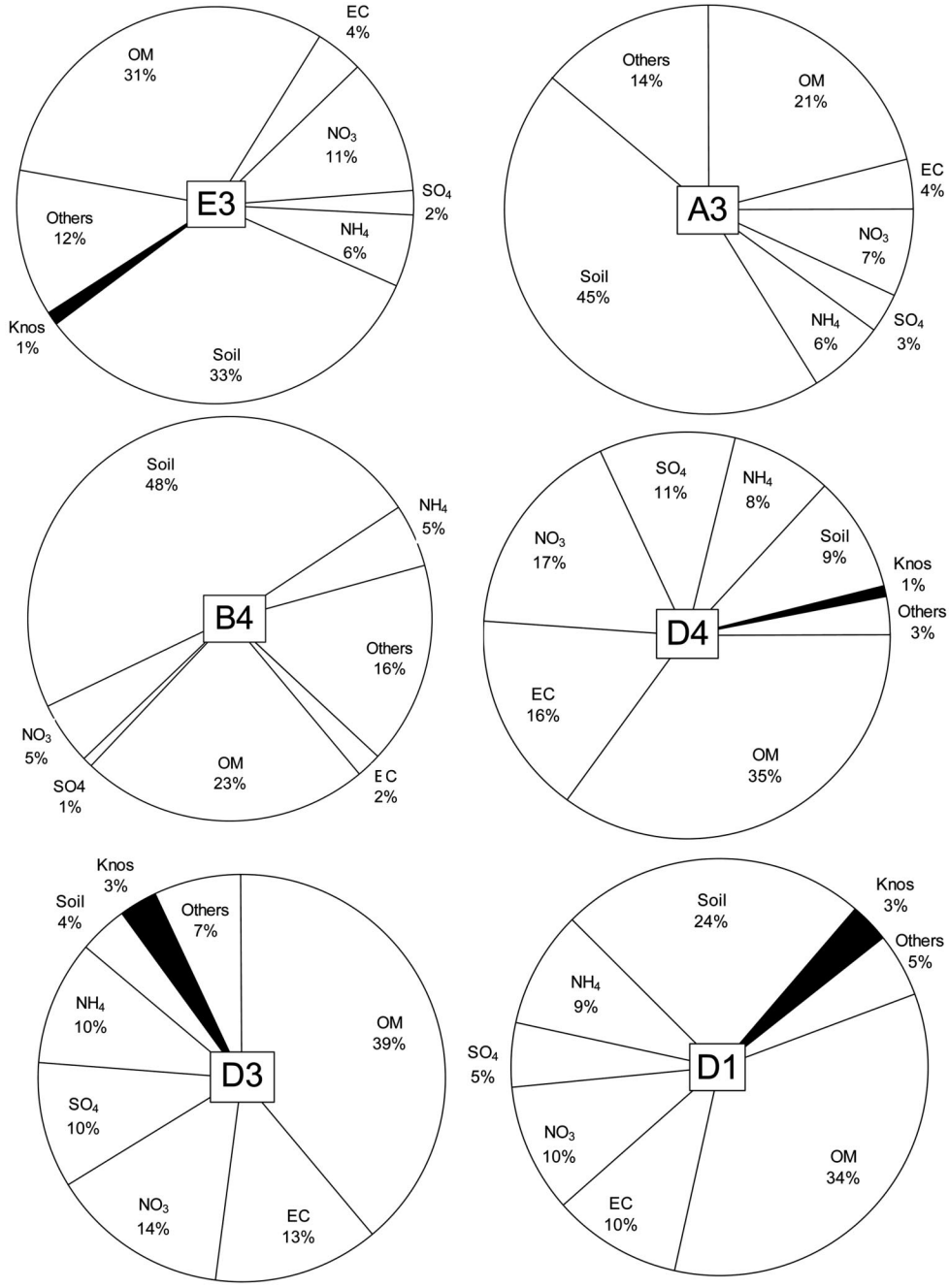


**Figure 7.** Spatial variation of the $PM_{10}$ chemical composition in the city of Chillán from September 1, 2001, to September 30, 2003.

and D1). As potassium (K) originates principally from vegetative biomass,[8] it may be concluded that K originates mainly from wood combustion during winter. This situation changed in lower population density sites at the city edge (A3 and B4), where the most dominant compound was soil. In downtown Chillán, all of the streets are completely paved, which undoubtedly explains the low soil dust concentration measured.

The results found in this study indicated that in Chillán, during the cold season (from May to August), average ambient temperature in Chillán was $<10$ °C, which implies reduced thermal convection. For that reason, the consequently shallow atmospheric boundary layer will significantly reduce vertical mixing by turbulence, resulting in stagnation of ground-based emissions. Moreover, firewood burning also increases during this season, and, consequently, the observed contamination is because of increased human impact amplified by local meteorological conditions.

## CONCLUSIONS

During the field study, $PM_{10}$ mass concentration measured in Chillán showed higher levels in the cold months (March to August) than the warm months (September to February). The yearly average was higher than the U.S. Environmental Protection Agency and European Union limit value. An intricate system of $PM_{10}$ sources and meteorological conditions regulates particulate pollution levels. The principal

reasons that seem to explain the observed situation, although there were not hourly measurements, are the influence of traffic during rush hours and the influence of elevated stack emissions in the city during the early afternoon and at dawn because of convective conditions, as well as the formation of a stable boundary layer at nighttime.

Carbonaceous substances (EC and OM) and inorganic substances of secondary origin ($NO_3$, $NH_4$, and $SO_4$) are the predominant components of $PM_{10}$ in the city of Chillán during the cold season, whereas soil dust prevails in the Chillán atmosphere during the summer. $PM_{10}$ mass concentrations and chemical compound levels were found to be higher in downtown Chillán in comparison with the areas of the city edge, presenting a clear spatial variability.

These results indicate that the respirable atmosphere in the city of Chillán is a problem of anthropogenic origin during autumn and winter. It is principally because of the massive amount of wood used as fuel for residential heating, producing a dense smoke cloud within urban areas on days of atmospheric stability.

In Chillán, local authorities need to enforce a law to control wood combustion on those days with low temperature, low wind speed, and high RH. This control should be combined with public transportation and industry regulations. For instance, defining new vehicle routes to avoid passing through downtown and restraining the use of vehicles that do not satisfy strict regulations like those enforced in Santiago, the capital city.

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