

Particulate matter in urban areas of south-central Chile exceeds air quality standards

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Abstract This study analyzed air quality in terms of the concentrations of sub-10 µm (PM₁₀) and sub-2.5 µm particulate matter (PM_{2.5}) recorded at 23 automated public monitoring stations located in 16 cities in south-central Chile (Rancagua, Rengo, San Fernando, Curicó, Talca, Maule, Chillán and Chillán Viejo, Gran Concepción, Coronel, Los Ángeles, Temuco and Padre Las Casas, Valdivia, Osorno, Puerto Montt, Coyhaique, and Punta Arenas). In each city, the spatial and temporal distributions of the PM₁₀ and PM_{2.5} concentrations were recorded at daily, monthly, and yearly intervals. Air quality was evaluated by comparing the annual average concentrations and the maximum daily concentrations of PM₁₀ and PM_{2.5} with the World Health Organization (WHO) and national standards. The results showed that the limits established in the WHO guidelines and the national standards were systematically exceeded at all the study sites. The highest concentrations of both PM₁₀ and PM_{2.5} were observed during the fall and winter months (April to September), i.e., the cold period of the year, whereas the lowest concentrations were recorded in the spring and summer months (October to March), i.e., the warm period of the year. Analysis of variance (ANOVA) of the data collected in the warm and cold periods showed that all stations in this study exhibited statistically significant differences between these two periods. During cold periods, burning firewood for heating produces emissions that are a main source of PM. Furthermore, firewood is primarily burned at night when the lowest temperatures occur and when the atmospheric

Keywords Particulate matter \cdot Air pollution assessment \cdot Chile \cdot Urban areas

Introduction

"Chile, your sky is a pure blue; pure breezes blow across you; and your field, embroidered with flowers; is a happy copy of Eden." This excerpt from the national anthem of Chile highlights its natural beauties (Lillo Robles and de Vera y Pintado 1847). However, the main cities in Chile are currently far from lying beneath an azure sky. Chilean cities such as Santiago, Rancagua, Gran Concepción, Temuco, Chillan, Los Ángeles, Osorno, and Coyhaique have deteriorating air quality (Díaz-Robles et al. 2011; MMA 2012, 2013, 2014; WHO 2016). This deterioration has mainly been due to an increase in emissions from rapid urban expansion (86.6% Chile's population lives in urban areas) (INE 2016a), biomass burning (high



conditions are generally unfavorable for dispersion; thus, pollution accumulates the above cities. The levels of PM_{2.5}, the most important type of pollution, exceeded the limit established by the WHO on at least one third of the days of the year (>120 days) in the cities of Rancagua, Rengo, Curicó, Talca, Chillan, Los Angeles, Temuco, Valdivia, Osorno, Puerto Montt, and Coyhaigue. Therefore in the cities in southern Chile, the population is exposed to particulate matter concentrations that can have negative health impacts. To improve the air quality conditions in the studied cities, research on heaters and combustion techniques should be promoted, home energy efficiency should be increased to reduce firewood consumption, the firewood certification process should be improved at the national level with a better auditing processes, and the introduction of alternative fuels should be considered for greater energy efficiency at competitive costs.

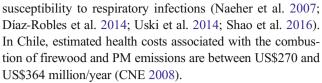
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firewood use, mainly in southern cities), an increase in the size and number of vehicles, and industrial activity (Pino et al. 2015; Schueftan et al. 2016). In addition, geographic and meteorological conditions that are unique to cities located in the middle basin of the southern region of Chile (between the Andes Mountains Range and the Costal Mountain Range), such as low temperatures and recurring inversion layers during the cold season, are important factors that make these cities vulnerable to air pollution (Garreaud 2009; Mena-Carrasco et al. 2014; Reizer and Juda-Rezler 2016). Thus, atmospheric particulate matter (PM) is one of the most relevant pollutants in the urban areas of Chile as well as in many cities around the world (WHO 2014a, 2016).

Atmospheric PM pollution is defined as solid and liquid particles that are part of an aerosol mixture in which these particles are suspended in the air (Colbeck and Lazaridis 2010; EPA 2016). Based on its negative impacts on human health (EPA 2012; Heal et al. 2012; IARC/WHO 2013; Vanos et al. 2015), PM is usually divided into particles with an aerodynamic diameter of less than 2.5 µm (PM_{2.5}) and particles with an aerodynamic diameter of less than 10 μ m (PM₁₀). On a global scale, the World Health Organization (WHO) estimated that at least 3.7 million premature deaths were related to atmospheric pollution in 2012 (WHO 2014b). In Chile, premature deaths have been estimated to be approximately 4000 each year (MMA 2012). Studies conducted in the city of Santiago showed positive associations between an increase in PM_{2.5} and an increase in mortality from respiratory diseases (1.75% for each 10 μ g m⁻³ increase in PM_{2.5}) (Valdes et al. 2012); another study showed a positive relationship between an increase in cerebrovascular damage and an increase in exposure to PM_{2.5} (1.29% for each 10 μ g m⁻³ increase in PM_{2.5}) (Leiva et al. 2013). In the city of Temuco, located south of Santiago in southern Chile, positive associations were found between the PM₁₀ concentration and three types of mortality: respiratory, total cardiovascular, and cardiorespiratory (Sanhueza et al. 2006).

Today, 10 million people in Chile (60% of the total population) are exposed to PM_{2.5} concentrations that are higher than the annual limit of 20 µg m⁻³ set by the National Ambient Air Quality Standards (NAAQS-Cl) (MMA 2013). These high concentration levels are mainly the result of 90% of the population in south-central Chile using wood-burning devices for heating or cooking; this wood burning emits fine and coarse particles that contain toxic atmospheric pollutants (benzene, aldehydes, polycyclic aromatic hydrocarbons, etc.) (Adler et al. 2011) as well as other pollutants (Williams et al. 2012; Reyes et al. 2015). Studies have associated long-term exposure to smoke from the burning of firewood with reduced pulmonary function, the development of asthma and chronic bronchitis, heart problems, and premature mortality. Shortterm exposure has been associated with acute bronchitis, asthma attacks, worsening of pulmonary diseases, and greater



In this study, we analyzed PM₁₀ and PM_{2.5} concentrations recorded at 23 automated public-monitoring stations operated by the Ministry of Environment of the Chilean government. The stations are located in 16 southern cities in Santiago de Chile (Rancagua, Rengo, San Fernando, Curicó, Talca, Maule, Chillán and Chillán Viejo, Gran Concepción, Coronel, Los Ángeles, Temuco and Padre Las Casas, Valdivia, Osorno, Puerto Montt, Coyhaique, and Punta Arenas) (SINCA 2012). Based on the data available in the National Air Quality Information System (Sistema Información Nacional de Calidad del Aire (SINCA)), the spatial and temporal distributions of the PM₁₀ and PM_{2.5} concentrations of each city were determined on daily, monthly, and annual scales. The temporal variability was summarized by the hour of the day, the day of the week, and the month of the year to characterize each location and to establish differences between the different cities. The results were evaluated by comparing the annual average concentrations and the maximum daily concentrations of PM₁₀ and PM_{2.5} with the levels outlined in the NAAQS-Cl (DS-N°12/MMA 2011; DS-N°59/ SEGPRES 1998) and World Health Organization Guidelines (WHOG) (WHO 2006). The temporal behavior of atmospheric PM pollution was analyzed to evaluate measures for improving the air quality in certain cities that already have antipollution plans in effect and to determine and provide information on the exposure levels of the population in different cities in southern Chile.

Experimental method

Study area

The study area included the main small- and medium-sized cities in south-central Chile (Fig. 1). The studied cities comprise a population of 2 million inhabitants, which is approximately 10% of the total population of the country, and cover a total area of 511 km² (see Table 1) (INE 2016a, b). The largest urban centers studied were the cities of Rancagua (RG), Concepción (CP) and Temuco and Padre Las Casas (TM), which have populations of between 200,000 and 300,000 inhabitants. The cities of Talca (TL), Chillán and Chillán Viejo (CC), Los Ángeles (LA), Valdivia (VL), Osomo (OS), Puerto Montt (PM), Puerto Aisén (PA), Coronel (CR), and Curicó (CU) have populations of between 90,000 and 200,000 inhabitants. The towns of Rengo (RN), San Fernando (SF), and Coyhaique (CH) have populations of between 30,000 and 50,000 inhabitants (INE 2016b).



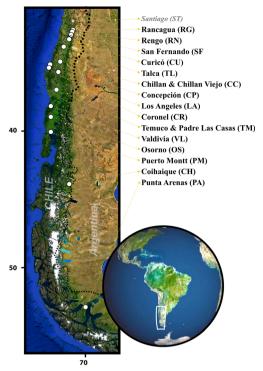


Fig. 1 Locations IDs and station numbers (ID/no. station) of the cities under study. More information is provided in Table 1

PM concentrations and meteorological data

Hourly air quality (PM₁₀ and PM_{2.5}) and meteorological (temperature, relative humidity, wind speed and wind direction) data from 23 automated monitoring stations located in 15 cities throughout central and southern Chile were analyzed over a period of 8 years (2007–2014) (see Table 1). The data were obtained from SINCA (2012), which is currently administered by the Ministry of Environment of the Chilean government, via an online web portal. This portal utilizes Airier software that was developed by the Swedish Meteorological and Hydrological Institute in Norrköping, Sweden (SMHI 2016). The data were downloaded from the webpage in April 2015.

The monitoring stations were installed to evaluate the impacts of pollution on the population, so their measurements are representative of the population. Two cities have three monitoring stations (Talca and Temuco-Padre las Casas), four cities have two monitoring stations (Rancagua, Chillan and Chillan Viejo, Los Ángeles, and Coyhaique), and the nine remaining cities have one station in each urban area (Table 1).

In general, the monitoring stations have quality assurance and control systems that control the flow velocity, as well as automatically detect leaks, zero the instruments, and identify instrumental noise; verifications are conducted on a weekly basis, and the data are validated

to correct for null entries, duplicates, and/or anomalies in the data. Disregarding the above issues, aspects related to the quality of the measurements available from SINCA are discussed.

More information about the PM and meteorological instrumentation and the locations and operation of the stations can be found in the SINCA web portal.

Data analysis

The available data were tabulated in a spreadsheet (MS-Excel® 2016 for Mac, Microsoft Corporation, Redmond, WA, USA). Descriptive statistical analysis was conducted using the same software (MS-Excel® 2011) with the pivot table tool. Some charts were generated using the time series graphing software Origin Pro v2015® for Windows (OriginLab Corporation, Northampton, MA, USA). In addition, the open source software R was used (RCoreTeam 2013) along with the RStudio-integrated development environment for R (RStudioTeam 2015) and the tool package for air quality analysis in R called OpenAir (Carslaw and Ropkins 2012; Carslaw 2013).

Assessment of air quality standards and guidelines

In this study, the basis for judging the possible health impacts of atmospheric pollutants was compliant with the NAAQS-Cl (DS-N°59/SEGPRES 1998; DS-N°12/MMA 2011). The concentration limits stipulated in the NAAQS-Cl are an annual mean of 50 $\mu g \ m^{-3}$ for PM $_{10}$ and 20 $\mu g \ m^{-3}$ for PM $_{2.5}$. The 24-h concentration limits are fixed at 150 and 50 $\mu g \ m^{-3}$ for PM $_{10}$ and PM $_{2.5}$, respectively. The WHOG (WHO 2006) establishes an annual mean threshold of 20 $\mu g \ m^{-3}$ and a 24-h mean of 50 $\mu g \ m^{-3}$ for PM $_{10}$; for PM $_{2.5}$, the thresholds for the annual and 24-h means are 10 and 25 $\mu g \ m^{-3}$, respectively.

Results and discussion

Annual PM₁₀ and PM_{2.5} concentrations

Table 2 shows the annual average concentrations at the monitoring stations in this study. In addition, the availability of the data is enclosed in parenthesis. In general, more data were available for PM₁₀ than for PM_{2.5}. This is because in Chile, the NAAQS for PM₁₀ was established in Chile in 1998 (DS-N°59/SEGPRES 1998) but the corresponding standards for PM_{2.5} were established in 2012 (DS-N°12/MMA 2011). Notably, in this study, the annual average PM₁₀ or PM_{2.5} concentration was considered valid if at least 70% of the hourly measurements were available for each year. As shown in Table 2, several stations and years do not meet this



 Table 1
 Studied monitoring stations in different cities in central and southern Chile (see Fig. 1)

City	Region	City	Location (lat; lon)	Population (no.)	Area (km²)	Station name	Station ID	Station location (lat; lon)	Altitude (m)	Measurement variables
Rancagua	VI O'Higgins	RG	34° 10′ 00″ S; 70° 45′ 00″ W	206,971	50.36	Rancagua 1	RG1	34° 09′ 44.24″ S; 70° 42′ 50.15″ W	514	PM ₁₀ , PM _{2.5} , T, RH, ws, wd
						Rancagua 2	RG2	34° 08′ 37.90″ S; 70° 44′ 13.50″ W	486	PM ₁₀ , PM _{2.5} , T, RH, ws, wd
Rengo	VI O'Higgins	RN	34° 25′ 00″ S; 70° 52′ 00″	30,891	9.28	Rengo	RN1	34° 23′ 40.30″ S; 70° 51′ 10.70″ W	330	PM ₁₀ , T, RH, ws, wd
San	VI O'Higgins	SF	34° 35′ 00″ S; 70° 59′ 00″ W	49,519	12.37	San Fernando	SF1	4′ 47.10″ S; 70° 59′ 22.90″	337	PM ₁₀ , T, RH, ws, wd
Curicó	VII Maule	CO	34° 59′ 00″ S; 71° 14′ 00″ W	93,447	20.5	Curicó	CU1	34° 58′ 29.80″ S; 71° 14′ 02.20″ W	216	PM ₁₀ , PM _{2.5} , T, RH, ws, wd
Talca	VII Maule	TL	35° 26′ 00″ S; 71° 40′ 00″ W	189,505	46.04	U.C. Maule	TL1	35° 26′ 08.60″ S; 71° 37′ 10.40″ W	131	PM ₁₀ , PM _{2.5} , T, RH, ws, wd
						La Florida	TL2	35° 26′ 06.81″ S; 71° 40′ 41.23″ W	66	PM ₁₀ , PM _{2.5} , T, RH, ws, wd
						U. Talca	TL3	35° 24′ 23.67″ S; 71° 37′ 59.71″ W	112	PM ₁₀ , PM _{2.5} , T, RH, ws, wd
Chillán	VIII Bio Bio	CC	36° 36′ 00″ S; 72° 07′ 00″ W	165,528	41.03	INIA Chillán	CC1	36° 35′ 41.42″ S; 72° 05′ 21.52″ W	135	PM ₁₀ , PM _{2.5} , T, RH, ws, wd
						Puren	CC2	36° 36′ 58.38″ S; 72° 05′ 35.12″ W	129	PM ₁₀ , PM _{2.5} , T, RH, ws, wd
Concepcion	VIII Bio Bio	CP	36° 49′ 41″ S; 73° 03′ 05″ W	212,003	55.95	Punteras	CP1	36° 55′ 23.94″ S; 73° 02′ 10.04″ W	30	PM_{10}
Coronel	VIII Bio Bio	CR	37° 01′ 00″ S; 73° 13′ 00″ W	91,469	24.52	Cerro Merquín	CR1	37° 01′ 16.29″ S; 73° 08′ 58.42″ W	57	PM_{10}
Los Ángeles	Los Ángeles VIII Bio Bio	LA	3′ 00″ S; 72° 21′ 00″	117,972	27.35	Cesfam Los Ángeles	LA1	37° 27′ 49.03″ S; 72° 21′ 42.29″ W	139	$PM_{10}, PM_{2.5}$
						21 de Mayo	LA2	8′ 16.29″ S; 72° 21′ 41.29″	138	PM ₁₀ , PM _{2.5} , T, RH, ws, wd
Temuco	IX Araucania TM	MI	38° 45′ 00″ S; 72° 40′ 00″ W	260,783	53.23	Museo Ferroviario	TM1	38° 43′ 30.91″ S; 72° 34′ 16.11″ W	199	PM ₁₀ , PM _{2.5} , T, RH, ws, wd
						Las Encinas	TM2	38° 44′ 55.38″ S; 72° 37′ 14.54″ W	107	PM ₁₀ , PM _{2.5} , T, RH, ws, wd
						Padre Las Casas	TM3	38° 45′ 53.03″ S; 72° 35′ 55.65″ W	119	PM ₁₀ , PM _{2.5} , T, RH, ws, wd
Valdivia	XIV Los Rios	VL	39° 48′ 50″ S; 73° 14′ 45″ W	127,750	42.39	Valdivia	VL1	39° 49′ 52.69″ S; 73° 13′ 42.67″ W	∞	PM ₁₀ , PM _{2.5} , T, RH, ws, wd
Osomo	X Los Lagos	SO	40° 34′ 21″ S; 73° 08′ 07″ W	132,245	31.82	Osorno	OS1	40° 35′ 4.19″ S; 73° 07′ 07.20″ W	52	PM ₁₀ , PM _{2.5} , T, RH, ws, wd
Puerto Montt	X Los Lagos	PM	41° 28′ 00″ S; 72° 56′ 00″ W	153,118	39.58	Mirasol	PM1	41° 28′ 46.30″ S; 72° 58′ 07.78″ W	104	PM ₁₀ , PM _{2.5} , T, RH, ws, wd



PM₁₀, PM_{2.5}, T, RH, ws, Measurement variables PM₁₀, PM_{2.5}, T, RH, wd PM_{10} 356 58 341 45° 34′ 44.57″ S; 72° 02′ 59.88″ 45° 34′ 47.77″ S; 72° 03′ 39.91″ 53° 09' 30.05" S; 70° 55' 17.42" Station location (lat; lon) Station ID CHI PA1 CHI Station name Coyhaique 2 Punta Arenas Coyhaique 18.27 39.03 Population 44,850 116,005 53° 10' 00" S; 70° 56' 00" 00 ,40 S; 72° Location (lat; lon) 00, 45° 34′ (≽ CH PA XI Aisen Region Fable 1 (continued) Coyhaique City

requirement. Significant gaps are present in the data that affect the comparability and representativeness of the information. The Ministry of Environment needs to revise the mechanisms of quality assurance and control for the measurements taken at the air quality monitoring stations that it oversees, which we have clearly demonstrated in previous studies (Toro et al. 2015).

The stations can be listed from highest to lowest average annual PM_{10} concentration as follows: \geq 80 µg m⁻³: RG-2 and CH-1; between 80 and 70 µg m⁻³: OS and RG-1; between 70 and 60 µg m⁻³: TM-3, CC-2, LA-2, and TM-2; between 60 and 50 µg m⁻³: TM-1, TL-2, TL-3, CU, VL, and RN; between 50 and 30 µg m⁻³: SF, TL-1, LA-1, PM, CP, and CR; and less than 5 µg m⁻³: PA. Comparing the annual PM_{10} concentrations recorded in the cities of Rancagua (RG-1 and RG-2) and Coyhaique (CH-1 and CH-2) with those observed in the metropolitan area of Santiago de Chile (an annual average of 72 µg m⁻³ for the station in Podhale) reveals that the PM pollution is higher in these southern cities than in the metropolitan area (Toro et al. 2014).

The annual PM_{2.5} concentrations show that the highest annual concentration of 57 μg m⁻³ occurred in Coyhaique (CH-1), followed by Chillán (CH-2), OS, and LA-2, with concentrations between 45 and 40 μg m⁻³. Concentrations between 40 and 30 μg m⁻³ were detected in Temuco (TM-1, TM-2, and TM-3), PM, VL, and CU. A previous study found that the annual PM₁₀ concentration recorded in the Santiago metropolitan area was 33 μg m⁻³ during the period from 2000 to 2012 (Toro et al. 2014). Compared with these previous results, most of the cities to the south of Santiago exhibit annual PM_{2.5} concentrations that are higher than those in the metropolitan region.

Monthly and daily PM₁₀ and PM_{2.5} levels

Figure 2 compares the time series of the PM_{10} and $PM_{2.5}$ concentrations and the $PM_{2.5}/PM_{10}$ ratios for the stations in the study area during 2014. This figure can be used to analyze both the seasonal fluctuations and the daily variability in the PM_{10} and $PM_{2.5}$ concentrations and the $PM_{2.5}/PM_{10}$ ratios. The highest concentrations of both PM_{10} and $PM_{2.5}$ were observed in the fall and winter months (April to September), i.e., the cold period of the year, whereas the lowest concentrations were observed during the spring and summer months (October to March), i.e., during the warm period of the year. The analysis of variance (ANOVA) results revealed statistically significant differences between the warm and cold periods at the stations in this study: the p value was smaller than the significance level of 0.05.

On average, in all the cities in this study, the ratio of $PM_{2.5}$ to PM_{10} was $50 \pm 10\%$; in the warm and cold months, values of $40 \pm 10\%$ and $60 \pm 10\%$ were obtained,



 Table 2
 Annual concentrations of PM₁₀ and PM_{2.5} in micrograms per cubic meter at the stations under study

Year	RG-1	RG-2	RN RN	SF	CU	TL-1	TL-2	TL-3	CC-1	CC-2	CP-1	CR-1
PM ₁₀ (µg m ⁻³ (%)) >2007 2008 2008 2009 2010 2011 2013 2014 Average PM _{2.5} (µg m ⁻³ (%)) 2007 2008 2009 2010 2011 2013 2013 2013	73 (93) ^a 66 (84) 85 (78) 101 (21)* 93 (57) * 78 (90) 78 (86) 72 (98) 75 (90) 32 (41)* 39 (79) 36 (80) 36 (80) 38 (86) 38 (86) 35 (88)		63 (94) 49 (98) 49 (99) 45 (96) 50 (98) 50 (99) 1	53 (78) 52 (98) 53 (99) 24 (94) 46 (99) 46 (90) 46 (90) 6 (94)					- 45 (20)* 58 (63)* 40 (26)* - 6 (20)* 52 (10)* 43 (50)* - 7 (10)* 15 (20)* 33 (63)* 17 (50)* 20 (82) 20 (82)			25 (77) 33 (61) 31 (61) 32 (95)
Year	LA-1	LA-2	TM-1	TM-2	TM-3	AL	SO	P	PM	CH-1	CH-2	PA-1
PM ₁₀ (µg m ⁻³ (%)) >2007 2008 2009 2010 2011 2013 2013 2014 Average 2009 2009 2009 2011 2011 2012 2013 2014 Average			56 (80) 31 (57) 80 (61) 557 (91) 68 (71) 55 (92) 55 (92) 58 (75) 58 (75) 44 (64)* 44 (55)* 38 (93) 37 (88)	46 (93) ^b (64 (83)) (64 (83)) (64 (83)) (73 (75)) (75 (71)) (75 (26 (88) 56 (88) 56 (50) 60 (69) 61 (87) 62 (67) 63 (86) 64 (38) 65 (38) 67 (67) 67 (67) 68 (88) 68 (88) 69 (88) 60 (89) 60 (89) 60 (89) 61 (87) 62 (87) 63 (86) 64 (88) 65 (88) 66 (88) 67 (88) 68 (88) 69 (88) 60 (89) 60	010 125 125 125 126 127 128 128 129 129 129 129 129 129 129 129		1	100 (73) 83 (86) 72 (94) 83 (96) 86 (85) 84 (88) 69 (69) 84 (68) 84 (88) 84 (88) 77 (96) 57 (96)		1

The percentage of data available is enclosed in parenthesis * data avaibility less than 70%

^a Average 2004–2007

^b Average 2001–2007



respectively. In general, the annual average PM_{2.5}/PM₁₀ ratio varied between 0.46 and 0.63 for the cities in this study; the city of Puerto Montt was an exception with an observed ratio of 0.9, suggesting that this station is poorly situated and that the measuring equipment at this station should be inspected, as this ratio is unusually high. The ANOVA results also showed that the differences in the PM_{2.5}/PM₁₀ ratios between the cold and warm months were statistically significant. The proportions of PM_{2.5} contained within PM₁₀ were very high, demonstrating that fine particles are the greatest contributor to PM air pollution in the cities of south-central Chile. Moreover, the higher ratios of PM_{2.5}/PM₁₀ during the cold periods indicate an increase in combustion sources, which is logical.

Examining the daily variability in the PM₁₀ and PM_{2.5} concentrations revealed that higher PM concentrations were measured after sunrise (6:00–10:00) and in the evening (18:00–21:00) during the warm months (October to March) (Fig. 2). During the cold period (April to September), a more prominent daily variability was observed compared with the warm period. In general, the highest concentrations were observed in the morning (6:00–9:00) and during the evening and night (18:00–03:00); these results are similar to those obtained in New York and Beijing (Degaetano and Doherty 2004; Liu et al. 2015).

These differences in the PM levels between warm and cold periods can be explained by the fact that in warm periods, emissions mainly originate from vehicular sources during the warm periods. Thus, emissions are highest during peak traffic hours: in the morning (06:00–9:00) and evening (17:00–21:00). Otherwise, daytime atmospheric conditions (higher wind speed, higher temperature, and more development of the mixing layer) are favorable for diluting pollution. In contrast, during cold periods, in addition to the emissions from vehicle exhaust, the burning of firewood for heating produces emissions that are a significant source of PM, especially at night, which is when the lowest temperatures occur and unfavorable atmospheric conditions for dispersion are commonly present; thus, pollution accumulates the above cities.

Effects of meteorological variables

Table 3 shows the meteorological parameters (temperature, relative humidity, and wind speed) recorded in the cities under study in 2014 for both the whole year and during the warm and cold periods. The average temperature in all the cities under study was 13 \pm 2 °C, with a range from 15 to 9 °C. The highest temperature was observed in the warm period (October to March), whereas the minimum temperature was observed in the cold period (April to September). Low temperatures, particularly during the cold period, can form thermal inversions due to surface cooling, which prevent vertical air mixing and promote the occurrence of air pollution

episodes (Hussein et al. 2006; Liu et al. 2015). The relative humidity in all the cities under study was $72 \pm 7\%$ in 2014, with a range from 74 to 60%. The relative humidity was highest during the cold periods. High relative humidity is associated with low temperature and hence stable atmospheric conditions. The wind speed reached values greater than 1.0 m s^{-1} in all the cities under study. In general, the wind speed was higher during the warm periods than during the cold periods. Low wind speed implies that stagnation of air masses would be favored in the cold period (Jelić and Klaić 2010). Temperature, relative humidity, and wind speed are thought to influence air pollutant concentrations (Sánchez-Ccoyllo and de Fátima 2002; Tai et al. 2010).

Table 4 shows the correlation coefficient between the PM_{2.5}/PM₁₀ ratio and the meteorological variables (temperature, relative humidity, and wind speed and velocity). A negative correlation was found between the PM_{2.5}/PM₁₀ ratio and the annual average temperature: the correlation coefficient ranged from -0.72 to -0.38 in all the cities under study. In contrast, a positive correlation was observed with relative humidity: the correlation coefficient ranged from 0.73 to 0.22 in all the cities under study. In general, for both temperature and relative humidity, the correlation was stronger for the cool period than for the warm period. The correlation coefficient between the PM_{2.5}/PM₁₀ ratio and wind speed was generally weak and negative: the average correlation coefficient was -0.23 for all the cities under study. This correlation was stronger for the cool period than for the warm period. These results are similar to other results found in the literature (Dunea et al. 2015; DeGaetano and Doherty 2004)

The correlation coefficient between the PM_{2.5}/PM₁₀ ratio and the meteorological variables can be explained by the effects of temperature on the formation of new particles through the processes of gas-particle conversion as well as the effects of relative humidity on the coalescence and settling of suspended particles, in which atmospheric moisture helps fine suspended particles to stick together to form heavier particles that then fall down (Tiwari et al. 2014). Moreover, at lower temperatures, increased emissions are associated with the combustion of firewood for heating (Ancelet et al. 2013; Celis et al. 2007; Meza et al. 2010; Sun et al. 2016). The correlation coefficient obtained for the warm period may have been higher than for the cold period because the increased emissions from firewood burning during cold periods increase the emissions of larger primary particles, given that most combustion processes occur in open heaters, relative to the formation or emission of fine particles. In contrast, during warm periods, primary emissions decrease, and the factors associated with the processes of secondary particle formation are more significant (Yue et al. 2009). Furthermore, stagnant wind conditions allow air pollutants to accumulate, resulting in elevated and localized concentrations of air pollutants (DeGaetano and Doherty 2004). Therefore, low wind speeds are generally associated with high levels of pollution



due to poor dilution and dispersion of pollutants. Conversely, fast winds near the surface are linked to high PM levels due to the resuspension of ground particles and the long-range transport of particulates (Hosiokangas et al. 2004; Jelić and Bencetić 2010; Liu et al. 2015).

A bivariate polar plot of the wind speed and direction and the PM_{2.5} concentrations at stations with available data for 2014 is shown in Fig. 3. These plots account for possible transport phenomena. Point sources of emission near the station are indicated by higher pollutant concentrations occurring with a higher wind speed from the direction of the source. In contrast, if the maximum concentrations are observed when wind speeds are low, i.e., in conditions of atmospheric stability, then the emissions are local in origin. Except under calm winds, there was not a statistically significant tendency for high PM concentrations to be associated with wind speed. The results presented in Fig. 3 indicate that the highest concentrations were observed during periods of low wind speed at most of the stations. This indicates that the PM concentration maxima were local in origin and associated with emissions from firewood combustion close to the monitoring stations; thus, the maximum concentrations were not primarily associated with transport phenomena. However, some stations may be affected by another source that may correspond to firewood combustion from distant neighborhoods; stations LA-2, TM-1, CH-1, and CH-2 presented a different pattern. More extensive research is needed in each city to evaluate the wind direction and PM concentration to determine the possible pollution sources and the portion that is due to transport (Klaić et al. 2012). Currently, we are analyzing the available information to characterize the sources that can impact each station; however, this investigation is beyond the scope of the present study.

Air pollution assessment

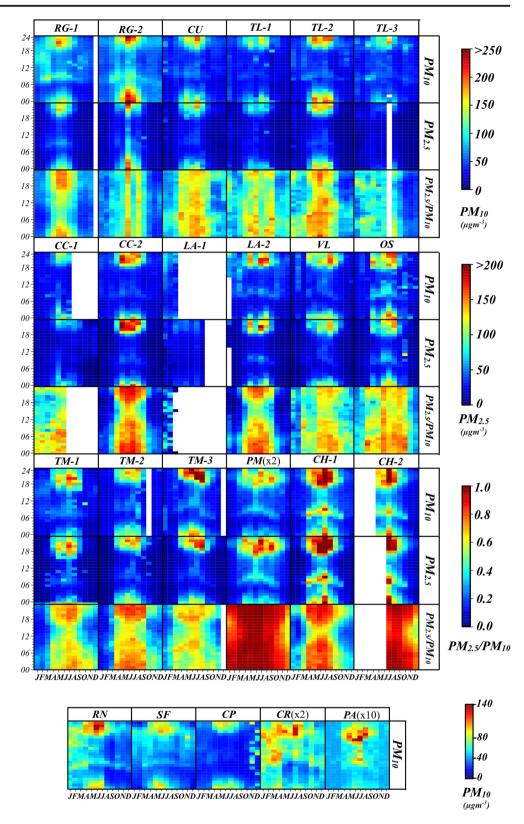
Figure 4 shows the ranking of the PM₁₀ and PM_{2.5} concentrations detected in different countries and cities around the world based on the exterior air pollution database from the WHO (2016); in addition, the average PM concentrations in 2014 are shown for the Chilean cities in this study and a statistical summary of the PM concentrations (maximum, minimum, average, and standard deviation) is provided. For PM₁₀, Chile is ranked 22nd out of 89 countries, with an average concentration in the WHO database of 64 µg m⁻³ (see Fig. 4a). This concentration is similar to that observed in countries such as Mexico (MEX, 78 µg m⁻³), Mauritius (MRT, 72 μ g m⁻³), Myanmar (MMR, 69 μ g m⁻³), Vietnam (VNM, 65 μg m⁻³), Sri Lanka (LKA, 64 μg m⁻³), Israel (ISR, 64 μg m⁻³), Lebanon (LBN, 63 μg m⁻³), Peru (PER, 63 μ g m⁻³), Turkey (TUR, 58 μ g m⁻³), and Honduras (HND, 58 μ g m⁻³). Moreover, the ranking of Chilean cities compared with the 1553 cities listed in the WHO database (Fig. 4b) is as follows: Rancagua, 194 (84 μ g m⁻³); Coyhaique, 198 (83 µg m⁻³); Osorno, 232 (75 µg m⁻³); Temuco, 356 (57 μ g m⁻³); Chillan, 363 (56 μ g m⁻³); Curicó, 370 (55 µg m⁻³); Los Ángeles, 371 (55 µg m⁻³); Valdivia, 373 (55 μg m⁻³); Talca, 408 (51 μg m⁻³); Rengo, 416 (50 μg m⁻³); San Fernando, 452 (46 μg m⁻³); Puerto Montt, 560 (37 μg m⁻³); Coronel, 607 (34 μg m⁻³); and Concepción, 637 (32 µg m⁻³). As a reference, the city of Santiago de Chile, the capital of Chile, is ranked 273rd for PM_{10} (68 µg m⁻³), which is a lower position than that observed for the cities of Rancagua, Coyhaique, and Osorno. Considering the directives from the WHO (2006) and Chilean regulations (DS-N°12/MMA 2011; DS-N°59/ SEGPRES 1998), the average annual concentrations of PM₁₀ in all cities in this study exceeded the WHO limit of 20 μ g m⁻³ by a factor of at least 1.6 and a maximum of 4.2. The annual standard from the Ministry of Environment was exceeded in nine of the studied cities by a factor ranging from 1.02 to 1.7.

In the case of PM_{2.5}, Chile, with a concentration of $28 \mu g \, m^{-3}$ (see Fig. 3a), is ranked 29th among the 91 countries reported in the WHO (2016) database, with a concentration of 28 µg m⁻³ (see Fig. 3a). This concentration is similar to that reported for Oman (OMN, 31 μg m⁻³), MMR (30 μg m⁻³), VNM (30 μg m⁻³), Saudi Arabia (SAU, 28 μg m⁻³), LKA (28 μg m⁻³), Bolivia (BOL, 27 μg m⁻³), South Africa (ZAF, $27 \mu g m^{-3}$), Poland (POL, $27 \mu g m^{-3}$), MEX ($27 \mu g m^{-3}$), and Hungary (HUN, 27 μg m⁻³). Moreover, ranking the Chilean cities with respect to the 1619 cities listed in the WHO database (Fig. 4b) for PM_{2.5} resulted in the following ranking: Coyhaique, 72 (57 $\mu g \text{ m}^{-3}$); Rancagua, 234 (39 $\mu g \text{ m}^{-3}$); Osomo, 255 (37 μ g m⁻³); Temuco, 294 (34 μ g m⁻³); Puerto Montt, 295 (34 μ g m⁻³); Chillan, 307 (33 μ g m⁻³); Los Ángeles, 328 (32 μ g m⁻³); Valdivia, 344 (31 μ g m⁻³); Curicó, 364 (30 $\mu g \text{ m}^{-3}$); and Talca, 437 (26 $\mu g \text{ m}^{-3}$). Santiago de Chile is ranked 363rd (30 µg m⁻³), which is lower than most of the cities in the study with data for PM_{2.5}. Considering the directives from the WHO and Chilean regulations, the average annual concentrations of PM2.5 in all the studied cities exceeded the limits of the WHO (10 µg m⁻³) and the Ministry of Environment (20 µg m⁻³) by factors ranging from 5.7 to 2.6 times and from 2.7 to 1.3 times, respectively.

Table 5 shows the number of days when the 24-h moving average of the PM₁₀ and PM_{2.5} concentrations exceeded the threshold set in the WHOG and the NAAQS-C1. The established limits were systematically exceeded at all the study sites. For example, in some cities, the WHOG limit for PM₁₀ was exceeded on more than half of the days of the year: Rancagua, Rengo, San Fernando, Los Ángeles, Chillán, and Coyhaique. The cities of Coyhaique, Rancagua, Los Angeles, and Temuco exceeded the Chilean standard for PM₁₀, which is more permissive than the WHOG, on more than 30 days of the year. On at least one third of the days of the year (>120 days), the limit established by the WHO for PM_{2.5} was exceeded in



Fig. 2 Time series of PM_{10} and $PM_{2.5}$ levels and the $PM_{2.5}/PM_{10}$ ratio for the stations under study. The plots show the average concentrations beginning at the hour



the cities of Rancagua, Rengo, Curicó, Talca, Chillan, Los Angeles, Temuco, Valdivia, Osorno, Puerto Montt, and Coyhaique. The highest number of days in which the WHOG and the NAAQS-Cl were exceeded in both PM categories was recorded during the coldest part of the year, the period from April to September. These values are related to the increased emissions due to firewood combustion for heating and more stable atmospheric conditions (weaker



Table 3 Average temperature (T, °C), relative humidity (RH, %), and wind speed (ws, ms⁻¹) recorded at the stations under study in 2014

Station	All			Warm			Cool	Cool		
	T(°C)	RH (%)	ws (ms ⁻¹)	T(°C)	RH (%)	ws (ms ⁻¹)	T(°C)	RH (%)	ws (ms ⁻¹)	
RG-1	17	59	_a	21	51	_a	13	68	_a	
RG-2	15	63	2.4	20	53	2.8	11	73	1.9	
RN	14	62	_a	18	52	_a	9.9	72	_a	
SF	14	67	1.8	18	54	2.0	10	80	1.6	
CU	15	67	1.8	19	55	2.1	11	78	1.5	
TL-1	14	72	1.5	18	60	1.7	11	85	1.3	
TL-2	15	69	1.6	19	56	1.8	11	82	1.4	
TL-3	14	73	1.2	18	61	1.4	9.9	85	1.0	
CC-1	13	_a	1.8	17	_a	1.8	9.6	_a	1.8	
CC-2	14	71	2.5	18	60	2.5	11	82	2.2	
LA-2	14	63	3.1	17	54	3.3	11	72	2.8	
TM-1	12	_a	5.0	15	_a	1.4	11	_a	1.7	
TM-2	11	_a	1.7	12	_a	2.0	9.9	_a	1.6	
TM-3	11	81	1.8	13	74	1.9	11	85	1.7	
VL	12	_a	1.8	14	_a	1.9	9.3	_a	1.8	
OS	11	73	1.6	13	66	1.8	8.7	80	1.4	
PM	11	83	3.0	12	69	3.2	9.5	86	2.8	
CH-1	8.9	69	2.5	12	62	3.3	6.4	76	1.8	
CH-2	8.9	65	3.2	12	55	4.0	7.0	71	2.4	

^a Annual data available was less than 70%

mixing and lower boundary layer), which decrease pollutant dilution. All the data presented above indicate that the population in southern Chilean cities is exposed to PM concentrations that can produce negative health impacts.

Table 4 Pearson's correlation analysis between the PM_{2.5}/PM₁₀ ratio, temperature (*T*), and relative humidity (RH) during cool and warm periods in 2014

Wood burning and PM

Firewood is the main fuel used for heating and cooking in the southern region of Chile and has a strong cultural and social

Station	All			Warm	Warm			Cool		
	T(°C)	RH (%)	ws (ms ⁻¹)	T(°C)	RH (%)	ws (ms ⁻¹)	T(°C)	RH (%)	ws (ms ⁻¹)	
RG-1	-0.700	0.7	_a	-0.48	0.58	_a	-0.63	0.67	_a	
RG-2	-0.38	0.36	-0.22	-0.45	0.61	-0.19	-0.38	0.47	-0.11	
CU	-0.55	0.49	-0.22	-0.29	0.27	-0.18	-0.27	0.27	-0.07	
TL-1	-0.38	0.42	-0.18	-0.2	0.24	-0.05	-0.22	0.29	-0.13	
TL-2	-0.53	0.54	-0.28	-0.2	0.22	-0.16	-0.45	0.47	-0.23	
TL-3	_a	0.22	-0.15	_a	0.07	-0.16	_a	_a	_a	
CC-2	-0.68	0.69	-0.21	-0.49	0.53	-0.31	-0.61	0.67	-0.23	
LA-2	-0.62	0.67	-0.29	-0.37	0.44	-0.23	-0.56	0.67	-0.26	
TM-1	-0.61	_a	_a	_a	_a	_a	-0.48	_a	-0.02	
TM-2	-0.54	_a	-0.34	_a	_a	_a	-0.45	_a	-0.36	
TM-3	-0.42	0.44	-0.16	_a	_a	_a	-0.28	0.33	-0.1	
VL	-0.46	_a	-0.13	-0.34	_a	-0.17	-0.39	_a	-0.06	
OS	-0.47	0.43	-0.11	-0.19	0.3	-0.14	-0.41	0.28	0.09	
PM	-0.52	0.56	-0.07	-0.18	0.56	0	-0.49	0.42	0.09	
CH-1	-0.72	0.73	-0.62	-0.64	0.62	-0.47	-0.53	0.71	-0.57	

^a The data available for one or both variables were less than 70%



Table 5 Annual exceedances (in number of days per year) of the 24-h Chilean air quality standards (NAAQS-CI) and World Health Organization Guidelines (WHOG) at the monitoring sites under study

CR-1		5/844 0/46a 1/98 0/81	PA-1 0/0
CP-1	0/0 _a	47764 0/40 0/96 3/62 	CH-2 50/149 ^a 6/7
RN SF CU TL-1 TL-2 TL-3 CC-1 CC-2 CP-1 CR-1	1 1 1 1	46/180 28/177 26/152 - - 120/176 103/167	CH-1 57/158 74/199 59/197 73/212 68/176 68/206 19/169 75/223 - - - - - - - 148/219
CC-1		3/11 ^a 1/21 ^a 0/41 ^a - 0/11 ^a 57/109 ^a 14/24 ^a - 5/9 ^a 5/17 ^a 5/17 ^a	PM
TL-3	1 1 1 1		
TL-2	1 1 1 1		OS 38/154 33/99 13/52 ^a 54/124 ^a 114/263 52/175 48/161 44/74 0/7 ^a 26/44 ^a 147/234 86/144
TL-1	1 1 1 1	2/129 8/143 	VL - 32/127 1/41 ^a 2/15 ^a 20/104 ^a 47/181 14/117 18/168 13/44 ^a 7/15 ^a 28/48 ^a 109/210 63/127
CU	1 1 1 1	22/52 ^a 6/163 11/180 - - - 46/57 ^a 79/144 87/168	TM-3
SF	1 _	16/285 6/282 8/278 0/320 0- - - - -	G) 34/138 34/138 34/175 34/175 34/146 43/150 43/164 53/170 32/113 38/155 OG) 75/135 36/65 ^a 36/65 ^a 105/149 ^a 105/149 ^a
RN	26/228 1/315 7/324 0/309	0/307 0/325 2/312 0/320 over the WHOG) - - - -	s over the WHOG) - 34/ 39/129 34/ 2/38 43/ 32/148 43/ 34/168 53/ 34/169 38/ 35/144 32/ 30/169 38/ 35/144 32/ 30/169 38/ 35/144 32/ 30/169 38/ 119/173 105/ 119/173 105/ 112/172 104
	days ove		Edays over the 39/129 2/38 32/148 34/168 35/144 30/169 of days over the 83/129 ^a 83/129 ^a 59/113 ^a 59/113 ^a 119/173
	indard/no. oi	andard/no. c	LA-2 undard/no. oi 38/168 21/216 30/196 andard/no. c
RG-1 RG-2	PM ₁₀ (no. of days over the NAAQS-CI standard/no. of days over the WHOG) 2007 38/308 – 26/228 2008 13/280 – 1/315 2009 22/309 – 7/324 2010 18/88 ⁸ – 0/309	2011 28/250	Year LA-1 LA-2 TM-1 TN-1 TN-1 TN-1 TN-1 TN-1 TN-1 TN-1 TN
	days over the	days over th	days over the
Year	PM ₁₀ (no. of 2007 2008 2009 2010	2011 2012 2013 2014 PM _{2.5} (no. of 2007 2008 2009 2010 2011 2012 2013	Year PM ₁₀ (no. of 2007 2008 2009 2011 2012 2013 2014 PM _{2.5} (no. of 2009 2010 2011 2013 2013 2013 2013

^a Available annual data were less than 70%



attachment that is sustained by the low cost of this energy source. In Chile, firewood and biomass are currently the second most-consumed sources of primary energy, following crude oil and reaching 18% of the primary energy used in Chile in 2012 (estimated in calories generated by each type of fuel) (DEE 2015). The percentage of firewood used by the population in urban areas has reached 57.8% in the sixth region (where the cities of Rancagua, Rengo, and San Fernando are located), 64.1% in the seventh region (where the cities of Curicó and Talca are located), 73.7% in the eighth region (where the cities of Chillán, Los Ángeles, and Concepción are located), 91.2% in the ninth region (where the city of Temuco is located), 94.6% in the 14th region (where the city of Valdivia is located), 96.3% in the 10th region (where the cities of Osorno and Puerto Montt are located), 99.3% in the 11th region (where the city of Coyhaigue is located), and only 13.0% in the 12th region (where Punta Arenas is located) due to the availability of natural gas (CNE 2008, 2015). In general, the household consumption of firewood increases with increasing latitude, as the average daily temperatures decrease and the number of cold hours increases; thus, the hours of operation of heating units increase.

To minimize the impact of firewood on air quality, the Chilean Ministry of Environment has designed a strategy with four facets to control pollution from firewood: (i) reducing emissions and improving the efficiency of devices for burning solid forest biomass, (ii) improving the ability to dry firewood and other fuels derived from firewood, (iii) improving thermal insulation in dwellings, and (iv) raising awareness and educating communities about the health impacts associated with firewood (Naeher et al. 2007; Sarigiannis et al. 2015). To implement these measures, programs have been developed to exchange heating units for higher-efficiency devices; a system of firewood certification to control firewood origin and the percentage of humidity has been developed; a restriction has been placed on the use of firewood during pollution episodes; plans for the improvement of thermal insulation in new and existing homes have been developed; and the competitiveness of alternative fuels, such as pellets and briquettes, has increased.

The measures to improve air quality have not all been effective (Reyes et al. 2015; Schueftan and González 2015). Efforts to date have concentrated on exchanging heating units, implementing a firewood certification system and prohibiting the use of firewood during air pollution episodes. The use of firewood has important social and economic impacts (SEN 2008; CNE 2015; DEE 2015); firewood is widely used by low-income sectors of the population for whom it is the only means of cooking and heating. In addition, the sale and production of firewood is an important source of jobs; therefore, its replacement with higher-cost fuels or its prohibition has a high social impact. As a result, the population does not follow prohibitions on the use of firewood or on the use of uncertified

firewood, given that the informal firewood market decreases costs even further (CNE 2008).

o improve air quality conditions in the cities in southern Chile, the following measures are suggested: (i) encourage research on heating and combustion techniques (some effort has been made in this respect, but further work is needed); (ii) prioritize the heating unit exchange program and encourage improvements in home energy efficiency to reduce firewood consumption; (iii) improve the firewood certification process at the national level with better auditing processes; (iv) introduce higher-efficiency alternative fuels at competitive costs; (v) conduct extension and education programs that can be implemented by public, private, and nonprofit organizations and that can benefit from the participation of a wide range of interested parties, such as volunteer groups, professional associations, and decision makers; and (vi) use tools to establish statutes and/or rules regarding firewood burning from current regulations.

Conclusions and summary

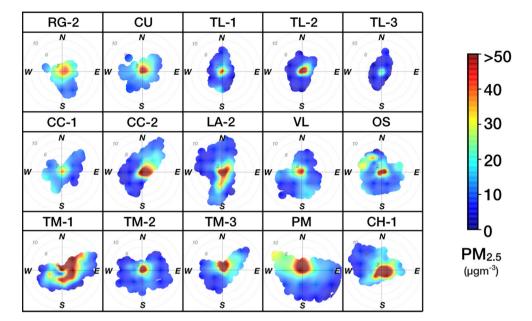
The main cause of environmental pollution in the cities of south-central Chile is the burning of firewood as the primary source of home and industrial heating. The extensive use of firewood is due to cultural factors (in the south, heating with firewood is historically associated with the warmth of the home), economic factors (firewood has been used in cities because it is an inexpensive fuel), and geographic factors (increased firewood use is associated with locations farther to the south and a colder and wetter climate).

Analysis of the temporal variability demonstrated that the highest PM concentrations were observed during the morning hours (6:00–9:00) and during the evening and night (18:00–03:00) in the cold periods. During these periods, in addition to the emissions from fuel exhaust, emissions from firewood burning result in the accumulation of PM pollution over cities at night when atmospheric conditions are unfavorable for dispersion.

The annual and daily PM_{10} and $PM_{2.5}$ limits established by the WHO and the Ministry of Environment were systematically exceeded at all the study sites. The average annual $PM_{2.5}$ concentrations in all the studied cities exceeded the WHO ($10~\mu g~m^{-3}$) and Ministry of Environment ($20~\mu g~m^{-3}$) limits. In the cities of Rancagua, Rengo, Curicó, Talca, Chillán, Los Ángeles, Temuco, Valdivia, Osorno, Puerto Montt, and Coyhaique, the daily limit established by the WHO was exceeded on at least 120~days out of the year. Therefore, in cities in the southern region of Chile, the population is exposed to PM concentrations with potential negative health impacts. The challenge is finding ways to control the use of firewood at the household level to minimize the environmental impacts and to reduce the influence of firewood



Fig. 3 Bivariate plot for the PM_{2.5} concentration in micrograms per cubic meter for 2014. Wind speed is given in milliseconds



combustion on the health of individuals without affecting the economy of the households that currently use firewood.

This work also demonstrated the lack of studies that have determined the current PM source apportionment in the studied cities. Therefore, more data collection is required. Future work could be directed towards the integration of emission inventory information, dispersion modeling, time series data, and chemical monitoring data in source distribution studies of PM in a systematic and integrated way. In addition, it is necessary and urgent to implement a quality assurance and control system for the measurement stations in the SINCA network so that information and resources are not lost and to

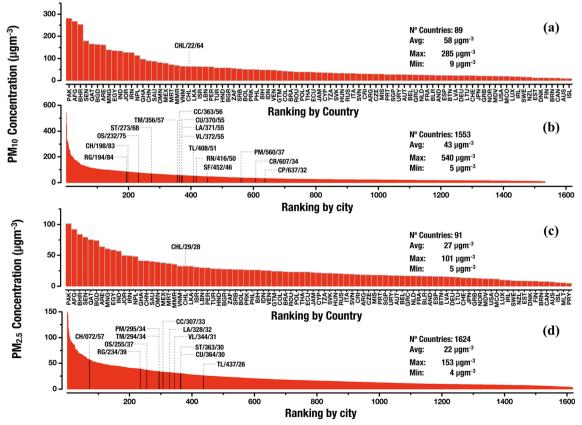


Fig. 4 Ranking of the PM₁₀ and PM_{2.5} concentrations for different countries and cities around the world

ensure comparability and traceability of air quality measurements in Chile.

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