

# Short Term Health Effects of Particulate Matter: A Comparison between Wood Smoke and Multi-Source Polluted Urban Areas in Chile

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# ABSTRACT

Temuco and Pudahuel are two urban areas in Chile that are among the highest in particulate matter (PM<sub>10</sub>) air pollution in Chile. In fact, Temuco is also classified as one of the most polluted cities in Latin America by the World Health Organization. Both cities show important differences in the sources of this  $PM_{10}$  pollution. For Temuco, a southern city, the main source is the residential wood combustion (RWC), and for Pudahuel, located in the central zone, the main sources are mobile and point sources. The relationship between PM<sub>10</sub> air pollution and health effects measured as the daily number of deaths and hospital admissions for cardiovascular and respiratory causes was determined. The Air Pollution Health Effects European Approach (APHEA-2) protocol was followed, and a multivariate Poisson regression model with gam.exact algorithm was fitted for these cities during 2001-2006. The results show that PM<sub>10</sub> had a significant association with daily mortality, where the relative risks (RR) for cardio respiratory mortality of the elderly age group was 1.0126 [95% (CI: 1.0004–1.0250)] at Temuco and 1.0086 [95% (CI: 1.0007–1.0165)] for Pudahuel when PM<sub>10</sub> increased by 10  $\mu$ g/m<sup>3</sup>. For the hospital admissions due to chronic obstructive pulmonary disease (COPD), the RR were 1.0198 [95% (CI: 1.0030-1.0369)] at Temuco and 1.0097 [95% (CI: 1.0000-1.0204)] at Pudahuel. There is evidence in these cities of positive relationships between ambient particulate levels and the rates of mortality and morbidity for cardiovascular and respiratory causes; being the excess risk 47% and 104.1% higher in Temuco than Pudahuel for cardiorespiratory mortality of the elderly population and COPD hospital admissions, respectively. These results demonstrate that there is greater risk when people are exposed to air polluted with wood smoke.

Keywords: Soot; Residential wood combustion; Mortality; Hospital admissions; Time series.

# INTRODUCTION

Chile occupies a long, narrow coastal strip between the Andes Mountains to the east and the Pacific Ocean to the west of South America. Small coastal mountains are found in the central part of the country, called the Coast Mountains (CM). The climate varies geographically in Chile, ranging from the world's driest desert in the north, to a Mediterranean climate in the central zone, and a rainy climate in the south. The relatively small central area dominates in terms of population and agricultural resources, where the main cities lie between the Andes and the CM.

Weather patterns of the majority of cities in Chile located in the central depression are detrimental to the removal of pollutants from the airshed, especially during fall and winter.

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The presence of the Pacific subtropical anticyclone causes for much of the year the emergence of the phenomenon of temperature inversion and a heavy coastal fog. This favors the generation of a very stable layer of air near the surface, which inhibits turbulence and vertical air movement in these basins. During the summer, surface heating allows the erosion of the inversion layer on the airshed, resulting in a significant improvement in the ventilation of these cities.

Pudahuel is an urban area in the western sector of Santiago of Chile with about 300,000 inhabitants, being part of the Metropolitan Area. It has a Mediterranean climate with a long dry season. The maximum and minimum temperatures in summer and winter are usually 30 and 13°C, respectively, while in winter the maximum and minimum are 13 and 2°C, respectively; having a total annual average rainfall of 317 mm (Grass and Cane, 2008). Based on data collected between 1993 and 1995, in August 1996 the Central Government designated the Metropolitan Region, where Pudahuel is allocated, as a non-attainment area for PM<sub>10</sub>, ozone (O<sub>3</sub>), carbon monoxide (CO), and nitrogen dioxide (NO<sub>2</sub>). The principal sources of pollution in this city are

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automobiles and industry, and in a very small comparative quantity, residential heating.

In contrast, Temuco city, the capital of the Cautín Province and of the Araucanía Region, is located 670 Km south of Santiago (38.5°S) equidistant between the Pacific Ocean and the Andes. Its climate is more humid and temperate than Chile's Central Valley Mediterranean region. Through the year, cyclonic and anticyclonic influences alternate, with a shorter dry summer period than Santiago or other Central Valley cities. Its mean annual temperature is 12°C, with highest median during the warmest month of 23.5°C and lowest median during the coldest month of 3.9°C. Annual mean rain is about 1,157 mm (Arias and Mader, 2010). Annually, Temuco suffers from severe woodsmoke particulate pollution events, especially during the period from April to September. In fact, almost 80% of the population uses wood for cooking or heating in winter, and it is estimated that 93% of PM<sub>10</sub> annual emissions originate from RWC. In spite that Temuco was declared a nonattainment area due to PM10 in 2005, and whose Atmospheric Decontamination Plan was enacted in 2010 by the President of Chile, still there are no visual advances towards the reduction of days that surpass regulation (Díaz-Robles *et al.*, 2011). On the contrary, a steady increase of PM<sub>10</sub> is still seen, Fig. 1. It is necessary to add that recently this city was declared a non-attainment area for PM<sub>2.5</sub>, in daily as well as annual Chilean standards in March 2013.

#### Health and Particulate Atmospheric Pollution

Particulate matter concentrations produce significant damage to human health. The negative effects of particulate concentrations in the air depend on the physico-chemical composition of the pollutant, its concentration (time and level of exposure) (Pope and Dockery, 2006), but also due to its atmospheric aerosol size and surface area (Andersen *et al.*, 2010; Belleudi *et al.*, 2010; Branis *et al.*, 2010; Díaz-Robles *et al.*, 2014).

Numerous time series or ecological studies estimate the



(a)



**Fig 1.** Daily  $PM_{10}$  concentration  $[\mu g/m^3]$  from 2001 to 2006 at (a) Temuco and (b) Pudahuel.

effects provoked by the daily exposure variations (Sanhueza *et al.*, 2009). The results of these studies about mortality and morbidity are consistent in identifying particulates as principally responsible for premature deaths and hospital admissions, with a secondary role for the ozone and other pollutants.

Epidemiological models have been based on short and long-term studies, using time series analysis (TSA) and cohort studies (CS), respectively. The magnitude of association between the concentration of atmospheric pollutants with mortality and morbidity of TSA and CS are different. For example, several SDT have estimated relative risks of mortality of less than 1% for an increment of PM<sub>10</sub> of 10  $\mu$ g/m<sup>3</sup> in the previous day (Pope and Dockery, 2006). At the same time, the principal CS (Dockery *et al.*, 1993; Pope *et al.*, 1995, 2002, 2004; Pope and Dockery, 2006) have estimated relative risks from 4 to 10%, due to the increase of PM<sub>10</sub> to 10  $\mu$ g/m<sup>3</sup> over several years.

#### Statistical Methods

There are various functional forms that are used for functions of concentration-response (C-R) for TSA models, such as linear models, the Poisson model (log-linear), Cox model (log-linear), and logistic regression, among others. For the regression of Poisson and Cox, the natural logarithm of the response in health is a linear function of the PM concentration or other pollutant, while the logarithmic regression is used to estimate the probability of an occurrence of an adverse health effect, where the natural logarithm of the probability ratio (odds ratio) is a linear function of the concentration of PM or other pollutant. However, for over-dispersed counts with daily number of deaths or hospital admissions as outcomes, the Poisson model is recommended for its simplicity to estimate the relative risks (RR) (Dominici and Burnett, 2003; Peng *et al.*, 2006).

In fact, the semiparametric Poisson regression models with Generalized Additive Model (GAM) parametrization are used in TSA to estimate the increase of the risk for an adverse health effect, such as mortality or morbidity, associated to an increase in atmospheric pollution over a short-term period (Dominici and Burnett, 2003; Dominici *et al.*, 2004; Peng *et al.*, 2006). The statistical methods most frequently used for TSA with confounding variable adjustment, include regression models with smooth time and temperature functions to adjust for seasonal variations, long-term tendencies and temporary changes in the temperature that can bias the estimation of the health risk (Peng *et al.*, 2006).

## Atmospheric Pollution and Its Short-term Association with Mortality and Morbidity

One of the first epidemiological TSA studies of acute exposure using  $PM_{10}$  was carried out in 1985–86 (Dockery and Schwartz, 1992). A consistent and robust relation between exposure to particulate matter and lung cancer and other cardiovascular illnesses is published in over 90 epidemiological studies in humans on an international level (Lipsett and Campleman, 1999; Pope *et al.*, 2002; Pope *et al.*, 2006; Pope

and Dockery, 2006; Jimenez *et al.*, 2009; Mate *et al.*, 2010; Nitta *et al.*, 2010; Jimenez *et al.*, 2011; Cao *et al.*, 2012; Lopez-Villarrubia *et al.*, 2012). Adverse effects due to prolonged exposure to high concentrations of particulate matter are also observed in susceptible groups of the population, such as asthma sufferers, children, and the elderly (Fischer *et al.*, 2003; Tainio *et al.*, 2005; Cakmak *et al.*, 2007; Jimenez *et al.*, 2009; Jimenez *et al.*, 2011).

A compilation of the principal international epidemiological studies about atmospheric pollution by particulate matter can be found in four important works (Dab *et al.*, 2001; Samet, 2002; Pope and Dockery, 2006; Nitta *et al.*, 2010). Recent, publications concentrate on studies that incorporate various cities, by which uniform statistical analyses are generated for daily databases (Schwartz and Zanobetti, 2000; Katsouyanni, 2003; Dominici *et al.*, 2005; Peng *et al.*, 2005; Analitis *et al.*, 2006; Ostro *et al.*, 2006; Wong *et al.*, 2008; Chen *et al.*, 2012). The advantage of these analyses is the uniform statistical approximation and the exclusion of biases, because all of the data is presented for specific cities.

It has been found that ultra-fine particles (UFP), particles with an aerodynamic diameter equal or less than 100 nm, can deeply penetrate the lungs and enter into the bloodstream, taking with it toxins to the rest of the body (Donaldson et al., 2001; Nemmar et al., 2002; Biswas and Wu, 2005). Within the body, these toxins can affect the respiratory, cardiovascular, and other systems (Pope et al., 2004; Delfino et al., 2005; Krewski et al., 2005; Schulz et al., 2005; Sioutas et al., 2005; Zanobetti and Schwartz, 2005). UFP dominates the number of particulates and surface area, principally in the emissions from combustion systems, and is capable of carrying high concentrations of adsorbed or condensed toxic pollutants (Ibald-Mulli et al., 2002; Delfino et al., 2005; Sioutas et al., 2005). The UFP is a significant fraction that can be characterized by the number of the particles, however, UFP exposure data and health effects are still scarce (Andersen et al., 2008; Lefranc and Larrieu, 2008; Andersen et al., 2010; Belleudi et al., 2010; Leitte et al., 2011; Liu et al., 2013; Díaz-Robles et al., 2014). In addition, in cities highly contaminated with wood smoke from firewood, an important size fraction, between 70-85%; of total particulate matter is UFP (Lightya et al., 2000; Chang et al., 2004; Hosseini et al., 2010).

In Chile, the current regulation establishes maximum limits of  $PM_{10}$ , which in 24 hours corresponds to 150 µg/m<sup>3</sup> and as an annual medium 50 µg/m<sup>3</sup>, while for  $PM_{2.5}$ , regulations in Chile limit the 24 hour maximum to 50 µg/m<sup>3</sup> and annually of 20 µg/m<sup>3</sup>. These are quite reasonable limits from the populational health protection point of view.

The objective of the present study was to determine the relationship between  $PM_{10}$  air pollution and health effects measured as the daily number of deaths and hospital admissions for cardiovascular and respiratory causes in a wood-smoke polluted city and a multisource air-polluted urban area. The APHEA-2 protocol was followed, and a multivariate Poisson regression model with *gam.exact* algorithm was fitted, controlling for trend, seasonality, and confounders for Temuco and Pudahuel during 2001–2006.

# METHODOLOGY

With the intention of obtaining the mortality and hospital admission relative risks (RR) of the population in Pudahuel and Temuco when exposed to acute concentrations of  $PM_{10}$ , three stages were planned; (a) collection of health data and air quality, (b) statistical processing of health and air quality information, and finally, (c) model development and relative risk estimation by type health effects and susceptible groups.

#### Health and Air Quality Data Collection

Death and hospital admission data were collected from the Statistics and Health Information Department (DEIS in Spanish) of the Health Ministry of Chile for each county from 2001 to 2006. This data was separated into three age groups, those under and equal to 5 years old (G1), older than 5 years old and younger and equal to 15 years old (G2), older than 15 years old and younger than and equal to 65 years old (G3), and older than 65 years old (G4), (Table 1). For these urban areas, a 7.8% and 8.5% of the population was from G1, a 19.7% and 20.9% from G2, 65.5% and 65.7% from G3, and 6.9% and 4.9% from G4, to Temuco and Pudahuel, respectively.

For these data, deaths and hospital admissions were considered that were caused by cardiovascular, respiratory and external diseases. In particular, digestive causes were used as a control in the statistical model. For cardiovascular causes, the age groups were considered that suffered from circulatory system illnesses (International Classification of Diseases (ICD), Chapter 10, I classification, ICD-10). For respiratory causes, the effects were considered for the same age groups, taking into account the diagnostics that corresponded to the J classification of the ICD-10. In addition, and for a specific analysis of mortality and hospital admissions, the following groups of specific cardiovascular causes were included from the ICD:

- Arrhythmias : I44-I49.9
- Cerebrovascular Disease : I60-I69+ G45-G46
- Cardiac Ischemia : I20-I25
- Hypertensive Disease : I10-I15

For the rest of the mortalities and hospital admissions, the following categories were considered, according to pathology:

Other causes: (code "O") includes all of the diagnostics of ICD-10 coded in A00-B99, C00-D48, D50-D89, E00-E90, F00-F99, G00-G99 (except those coded under the codes G45 and G46 which are included in the analysis of cardiac affections) H00-H59, H60-H95, L00-L99, M00-M99, N00-N99, O00-O99, P00-P96, Q00-Q99, R00-R99, Z00-Z99,

 Table 1. Age group classification.

Group	Code
< 5 years	G1
$\geq$ 5 and $\leq$ 15 years	G2
$\geq$ 16 and $\leq$ 65 years	G3
> 65 years	G4
Total	Т

U00-U99. External causes (code "E") includes all of the diagnostics coded in S00-T98 and V01-Y98 del ICD-10

Finally, numerical information was collected on the number of deaths and hospital admissions for gastrointestinal causes (K classification of the ICD-10); for the same age groups mentioned, in such a way that they were incorporated into the daily base. The pathology type K was utilized as a control in the statistical model. In Tables 2 and 3 the codes are summarized, as well as the description of each mortality and hospital admission by age group.

The  $PM_{10}$  and meteorological data summarized from Tables 4 to 7 were collected from monitoring stations available at Pudahuel and Temuco counties. The meteorological data included temperature, relative humidity, and wind speed.

## Analysis and Statistical Processing of Health Information and Air Quality

The databases of the health and air quality time series were processed using an imputation data analysis and outliers assessment. Then metrics of the variables studied was built, including distribution analysis, time series, seasonality, maximum, minimum, percentiles, among others. The result of this first stage was the structuring of a global daily database of all the variables. The software used was JMP version 7.1.

Table 2. Mortality description codes.

Code	Description
MCardT	Total Cardiovascular mortality
MCardG1	Cardiovascular mortality group 1
MCardG2	Cardiovascular mortality group 2
MCardG3	Cardiovascular mortality group 3
MCardG4	Cardiovascular mortality group 4
MRespT	Total Respiratory mortality
MRespG1	Respiratory mortality group 1
MRespG2	Respiratory mortality group 2
MRespG3	Respiratory mortality group 3
MRespG4	Respiratory mortality group 4
MCRT	Total Cardiorespiratory mortality
MCRG1	Cardiorespiratory mortality group 1
MCRG2	Cardiorespiratory mortality group 2
MCRG3	Cardiorespiratory mortality group 3
MCRG4	Cardiorespiratory mortality group 4
MCVE	Total Specific cardiovascular mortality
MECV	Total Stroke Mortality
Marrhytmia	Total Arrhythmias Mortality
MEIC	Total Ischemic Mortality
MER	Total Specific Respiratory Mortality
Mpneumonia	Total Pneumonia Mortality
MCOPD	Total COPD Mortality
MDigestT	Total Digestive Mortality
MDigestG1	Digestive Mortality group 1
MDigestG2	Digestive Mortality group 2
MDigestG3	Digestive Mortality group 3
MDigestG4	Digestive Mortality group 4
Mexternal	External Total Mortality
Mothers	Other causes of Mortality
Total	Total Mortality

Table 3. Description of Hospital Admission Codes.

Code	Description
HACardT	Cardiovascular total
HACardG1	Cardiovascular group 1
HACardG2	Cardiovascular group 2
HACardG3	Cardiovascular group 3
HACardG4	Cardiovascular group 4
HARespT	Respiratory total
HARespG1	Respiratory group 1
HARespG2	Respiratory group 2
HARespG3	Respiratory group 3
HARespG4	Respiratory group 4
HACRT	Cardiorespiratory total
HACRG1	Cardiorespiratory group 1
HACRG2	Cardiorespiratory group 2
HACRG3	Cardiorespiratory group 3
HACRG4	Cardiorespiratory group 4
HAEIC	Cardiac Ischemic Disease
HAarrhytmia	Arrhythmia
HAECV	Cerebrovascular disease
HACVE	Specific Cardiovascular
HApneumonia	Pneumonia
HACOPD	COPD
HAER	Specific Respiratory
HADigestT	Digestive
HADigestG1	Digestive group 1
HADigestG2	Digestive group 2
HADigestG3	Digestive group 3
HADigestG4	Digestive group 4
HAexternal	External (accidents)
HAothers	Other diseases

The APHEA-2 and the Spain Multicenter Study for Air Pollution and Morbidity-Mortality (EMECAM) protocols were used as guidelines for this study. These procedures involve a standardized approach to specify a multivariate Poisson regression model to assess air pollution and weather effects while controlling for trends and season using GAM parameterization (Katsouyanni *et al.*, 2001; Dominici *et al.*, 2005; Peng *et al.*, 2006; Sanhueza *et al.*, 2009) to estimate the increased risk for adverse health effects associated with increased air pollution on a short term basis.

Multiple regressions can be linear or nonlinear, depending on the characteristics of the relationships between variables, which can be checked step by step through the process with non-parametric regression using GAM techniques that have the following general form:

$$Log(u) = a_o + a_1 f(T) + a_2 f(S) + a_3 f(M) + gf(P) + \beta f(Cont) + \varepsilon$$
(1)

where:

и:	Health	effect,	mortality	or mo	rbidity
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- *a*<sub>i</sub>: Model adjustment coefficients
- *T*: Trend variables
- S: Seasonal Variables
- *M*: Meteorological Variables
- *P*: Other pollutants  $(NO_2, O_3, SO_2, CO)$

*Cont*: Pollutant of concern ( $PM_{10} \circ PM_{2.5}$ )

- $\beta$ : Coefficient of the pollutant of concern
- *f*: Function obtained from non-parametric analysis. In the case of meteorological variables, cubic spline smooth functions were used, with 4 degrees of freedom for temperature, and 2 degrees of freedom for the other meteorological variables
   *ε*: Error term

The relative risk (RR) is the risk increase associated with a rise of 10  $\mu$ g/m<sup>3</sup> in PM<sub>10</sub> or PM<sub>2.5</sub> levels compared to baseline, and it is obtained as follows.

$$RR = \operatorname{Exp}(\beta \times \Delta PM_{10}) \tag{2}$$

Finally, excess risk (ER) corresponds to the percentage increase of events (mortality or morbidity) associated with an increase in the concentrations of  $PM_{10}$  or  $PM_{2.5}$  respect to a baseline. The ER is obtained as follows.

$$ER = (RR - 1) \times 100 \tag{3}$$

The data was analyzed using multivariate Poisson regression fitted with S-Plus 8 software (Insightful Corporation) with the *gam.exact* algorithm. Trend and seasonality were introduced first and then the meteorological variables and

Valid data Concentration  $[\mu g/m^{3}]$ Available Missing City Year data data % Maximum P98 Minimum 2001 344 94.25 239.68 21 263.65 0.69 2002 330 35 90.41 263.56 231.29 8.68 9 2003 356 97.53 321.31 228.64 5.34 Temuco 2004 12 96.72 241.57 232.12 8.16 354 2005 346 19 262.30 7.51 94.80 260.77 2006 0 100.00 316.16 246.02 7.24 365 0 241.33 2001 365 100.00 241.50 10.13 2002 1 99.89 260.13 245.72 8.08 364 2003 365 0 100.00 231.46 223.71 8.04 Pudahuel 2004 0 210.08 366 100.00 185.14 13.21 2005 365 0 100.00 199.08 185.35 4.88 2006 365 0 100.00 216.00 214.03 16.79

Table 4. Daily PM<sub>10</sub> statistics from Temuco and Pudahuel 2001–2006.

City	Voor	Available	Missing	Valid data		[m/s]	
City	i eai	data	data	%	Maximum	Mean	Minimum
	2001	361	4	98.92	6.47	1.98	0.49
	2002	354	11	96.98	6.51	2.09	0.81
Tomuso	2003	365	0	100.00	6.85	1.95	0.69
Tennuco	2004	361	5	99.18	6.64	1.76	0.59
	2005	307	58	84.11	5.88	1.68	0.33
	2006	365	0	100.00	5.81	172	0.29
	2001	364	1	99.72	2.45	1.37	0.48
	2002	365	0	100.00	2.43	1.36	0.35
Dudahual	2003	365	0	100.00	2.36	1.31	0.43
Pudanuei	2004	366	0	100.00	2.24	1.26	0.45
	2005	365	0	100.00	2.28	1.31	0.51
	2006	365	0	100.00	2.25	1.25	0.39

Table 5. Wind Speed statistics in Temuco and Pudahuel 2001–2006.

Table 6. Relative Humidity statistics from Temuco and Pudahuel 2001–2006.

City	Voor	Available	Missing	Valid data		[%]	
	i eai	data	data	%	Maximum	Mean	Minimum
	2001	361	4	98.93	97.49	77.87	39.19
	2002	365	0	100.00	94.95	78.42	52.19
Tomuso	2003	365	0	100.00	97.11	79.84	52.65
Tennuco	2004	361	5	99.18	98.75	78.81	47.59
	2005	347	18	95.07	99.86	77.12	48.17
	2006	365	0	100.00	99.50	77.83	54.00
	2001	365	0	100.00	94.44	61.92	22.73
	2002	365	0	100.00	92.48	59.37	21.37
Dudahual	2003	365	0	100.00	92.58	56.12	23.45
Pudanuei	2004	365	1	99.70	91.87	61.54	27.37
	2005	365	0	100.00	94.27	66.30	26.01
	2006	365	0	100.00	96.17	61.31	24.01

Table 7. Temperature statistics from Temuco and Pudahuel from 2001 to 2006.

City	Voor	Available	Missing	Valid data	[°C]			
City	real	data	data	%	Maximum	Mean	Minimum	
-	2001	361	4	98.92	19.91	12.21	2.88	
	2002	365	0	100.00	23.96	11.95	1.38	
Tomuso	2003	365	0	100.00	20.23	12.29	3.27	
Tennuco	2004	361	5	98.63	24.21	12.47	1.04	
	2005	349	16	95.62	25.76	11.26	-1.70	
	2006	365	0	100.00	21.49	11.56	2.09	
	2001	365	0	100.00	26.20	15.77	6.03	
	2002	365	0	100.00	24.30	15.22	4.41	
Dudahual	2003	365	0	100.00	25.44	15.52	4.30	
Pudanuel	2004	364	2	99.73	23.95	15.83	4.74	
	2005	365	0	100.00	25.48	15.66	5.84	
	2006	365	0	100.00	26.15	16.30	4.79	

their transformations. Finally, the average of lags 1, 2, 3, and 4 were used for meteorological variables and  $PM_{10}$ .

#### **RESULTS AND DISCUSSION**

In Chile there are few epidemiological studies to establish the link between air quality and health, most of them made in Santiago and Temuco, but studies with gam.exact algorithm are scarce. This algorithm was developed to implement the expensive computation of the exact asymptotic standard error (Dominici and Burnett, 2003). Its use allows a more robust assessment of uncertainty of air pollution health effects.

Since the year 1995, more than 35 investigations have been published about mortality and morbidity and their association with atmospheric pollution by  $PM_{10}$ ,  $PM_{2.5}$ , and other atmospheric pollutants in Chile. Sanhueza *et al.* (2009) carried out a complete and comprehensive revision of the state of the art from the principal studies done in Chile between 1995 and 2007. Since 2008, studies have been published with new evidence associated with atmospheric pollution and mortality in Chile.

In effect, a study realized in 3 Latin American cities, and using a Poisson semi-parametric regression with confounding adjustment, found that for increments of 10  $\mu$ g/m<sup>3</sup> of PM<sub>10</sub>, the increase in risk for all non-external-cause adult mortality was 0.61% [95% IC (0.40%-0.83%)] for the city of Santiago, Chile (O'Neill et al., 2008). Sanhueza et al. (2009) determined the relationship between air pollution from PM<sub>10</sub> and health effects measured as the daily number of deaths, hospital admissions, and emergency room visits for cardiovascular, respiratory, and acute respiratory infection (ARI) diseases. The APHEA-2 protocol was followed, and a multivariate Poisson regression model was fitted, controlling for trend, seasonality, and confounders for Temuco during 1998-2004 for mortality, 2001-2004 for hospital admissions, and 2001-2006 for acute respiratory infection (ARI) diseases. The results show that PM<sub>10</sub> had a significant association with daily mortality and morbidity, with the elderly being the group that presented the greatest risk.

Finally, a study found associations between elemental concentrations of ambient particles and cause-specific mortality in Santiago between 1998 and 2007 (Valdes *et al.*, 2012). They used a time series analysis controlling for time trends, day of the week, temperature, and relative humidity. The authors found significant effects of PM<sub>2.5</sub> on all the causes analyzed, with a 1.33% increase [95% CI (0.87–1.78)] in cardiovascular mortality per 10  $\mu$ g/m<sup>3</sup> increase in the two days average of PM<sub>2.5</sub>. The authors suggested that PM<sub>2.5</sub> with high zinc, chromium, copper, sodium, and sulfur content have stronger associations with mortality than PM<sub>2.5</sub> mass alone in Santiago, Chile.

Our results of Table 8 show that  $PM_{10}$  had a significant association with daily mortality, with the elderly (G4) and younger population (G1) being the groups that presented the greatest risk, mainly for Temuco, although this city had better ventilation than Pudahuel (Table 5). The RR for cardio respiratory causes in G4 (MCRG4), increasing 10 µg/m<sup>3</sup> of PM<sub>10</sub>, was 1.0086, with a 95% confidence interval (CI) (1.0007–1.0165) for mortality in Pudahuel and 1.0126 [95% CI (1.0004–1.0250)] in Temuco. It is important to note that the ER for MCRG4 was 1.26 for Temuco, 46.5% higher than the value of Pudahuel of 0.86. Our ER also was

higher than the value of 1.11 estimated by (Sanhueza *et al.*, 2009), using gam.exact and increasing 10  $\mu$ g/m<sup>3</sup> of PM<sub>10</sub>. The daily mortality data for cardiovascular and respiratory diseases in Temuco from 1998 to 2004 was used by these authors, which had several missing data for its first 3 years. That missing data was imputed by a multivariable linear regression before estimating the RR. Therefore, the quality of that data may have influenced the estimated risk. Furthermore, our data from 2001 to 2006 had better quality and had almost none missing values. Finally, the air quality worsened from 2004 to 2006 in Temuco, and therefore those levels were reflected in our estimated RR for MCRG4.

For specific causes, the Total Specific Respiratory Mortality (MER) was more significant in Pudahuel, and the Total Ischemic Mortality (MEIC) was significant for Temuco, with RR of 1.0157 [95% CI (1.0013–1.0302)] and 1.0068 [95% CI (1.0006–1.0130)], respectively. Significant risks in the group of children under 5 years of age were not found for the Pudahuel, however, for Temuco these mortality risks were significant for MRespG1 and MCRG1, with RR of 1.0298 [95% CI (1.0076–1.0524)] and 1.0212 [95% CI (1.0016–1.0412)], respectively. For total respiratory mortality (MRespT), the RR was 1.0129 [95% CI (1.0000–1.0266)] in Pudahuel and Temuco did not show any risk. There is evidence in these cities of positive relationships between ambient particulate levels and mortality for cardiovascular and respiratory diseases.

For Temuco hospital admissions shown in Table 9, the main RR resulted significant for the HACOPD five days after the increase of 10  $\mu$ g/m<sup>3</sup> of PM<sub>10</sub> (lag 5); for the ischemic cardiac disease (HAEIC) (lag 5); for cardiorespiratory disease of group 4 (HACRG4) (lag 4); and for the respiratory disease of group 4 (HARespG4) (lag 4) with RR of 1.0198 [95% IC (1.0030–1.0369)], 1.0034 [95% IC (1.0004–1.0064)], 1.0079 [95% IC (1.0010–1.0148)], and 1.0030 [95% IC (1.0000–1.0060)], respectively. Our ER results for HARespT, HARespG4, and HACRT at Temuco were lower than those estimated by (Sanhueza *et al.*, 2009), using gam.exact and increasing 10  $\mu$ g/m<sup>3</sup> of PM<sub>10</sub>. This difference may be due to they used just 5 years of data instead of the 7 we used to estimate the RR, increasing their level of uncertainty due to the size of their data (Dominici *et al.*, 2003).

On the other hand, for Pudahuel the main RR resulted significant for the HACOPD (lag 2); for the respiratory disease of group 3 (HARespG3) (lag 1); for specific cardiovascular disease (HACVD) (lag 1); and

<b>Table 8.</b> Relative Risk of Mortality at Temuco and Pudahuel due to $PM_{10}$ from 20	)01	to 2	20	06	ć
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City	Mortality	β	$Error(\beta)$	RR	ICI	ICS	ER	Lag
;	MEIC	0.00067	0.00031	1.0068	1.0006	1.0130	0.68	0
	MRespG1	0.00293	0.00111	1.0298	1.0076	1.0524	2.98	1
Tenhuco	MCRG1	0.00210	0.00099	1.0212	1.0016	1.0412	2.12	1
	MCRG4	0.00126	0.00062	1.0126	1.0004	1.0250	1.26	6
	MCardG4	0.00087	0.00047	1.0087	1.0000	1.0180	0.87	7
	MRespT	0.00128	0.00068	1.0129	1.0000	1.0266	1.29	0
Pudahuel	MER	0.00155	0.00073	1.0157	1.0013	1.0302	1.57	0
	MCRT	0.00064	0.00033	1.0064	1.0000	1.0129	0.64	7
	MCRG4	0.00085	0.00040	1.0086	1.0007	1.0165	0.86	7

City	Morbidity	β	Error(β)	RR	ICI	ICS	ER	Lag
	HAEIC	0.00034	0.00015	1.0034	1.0004	1.0064	0.34	5
	HARespT	0.00017	0.00007	1.0017	1.0004	1.0031	0.17	5
Temuco	HARespG4	0.00030	0.00015	1.0030	1.0000	1.0060	0.30	4
	HACOPD	0.00196	0.00085	1.0198	1.0030	1.0369	1.98	5
	HAER	0.00019	0.00009	1.0019	1.0002	1.0037	0.19	4
	HACRT	0.00012	0.00006	1.0013	1.0002	1.0023	0.13	5
	HACRG4	0.00079	0.00035	1.0079	1.0010	1.0148	0.79	4
	HACVE	0.00092	0.00043	1.0092	1.0008	1.0176	0.92	1
Pudahuel	HARespG3	0.00097	0.00049	1.0097	1.0000	1.0196	0.97	1
	HACOPD	0.00096	0.00054	1.0097	1.0000	1.0204	0.97	2
	HACRG3	0.00066	0.00029	1.0066	1.0008	1.0124	0.66	1
	HACRG4	0.00069	0.00030	1.0069	1.0010	1.0128	0.69	3

**Table 9.** Relative risks of hospital admissions in Temuco and Pudahuel due to  $PM_{10} 2001-2006$ .

cardiorespiratory disease of group 4 (HACRG4) (lag 3) with RR of 1.0097 [95% IC (1.0000–1.0204)], 1.0097 [95% IC (1.0000–1.0196)], 1.0092 [95% IC (1.0008–1.0176)], and 1.0069 [95% IC (1.0010–1.0128)], respectively. It is noteworthy that the ER for COPD hospital admissions was 1.98 for Temuco, doubling the value of Pudahuel of 0.97. Significant risks of hospital admissions in the group of children younger than 5 years of age (G1) were not found for both Temuco and Pudahuel. This behavior may be because the daily cases are very scarce and usually children recover at home, especially for respiratory diseases.

Finally, the estimated relative risks for digestive diseases were not statistically significant when they were correlated with daily  $PM_{10}$  exposure, strengthening our results for respiratory and cardiovascular causes.

The possible difference in the levels of relative risk between both cities can be due to the origin of the pollution by particles. The pollution in Temuco is primarily from residential wood smoke, and in Pudahuel, the pollution is from mobile and industrial sources. In fact, the 2010 emission inventory of Temuco indicates that 96% of the total PM<sub>2.5</sub> came from the residential wood combustion, being the contribution of mobile sources and point sources, less than 2 and 3%, respectively (CENMA-CONAMA, 2010). On the other hand, the main 2005 contribution of  $PM_{25}$  in the Metropolitan Area of Santiago, where Pudahuel is allocated, came from motor vehicles (37.3%, most of them diesel buses and trucks), industrial sources (27.0%), agricultural and residential wood burning (20.1%), and area sources (15.6%). Part of this PM<sub>2.5</sub> comes from marine aerosols and soil dust (CONAMA, 2005; Jorquera and Barraza, 2012). It is important to mention that over a span of 20 years, allowable diesel sulfur content has been steadily reduced from 5,000 to 50 ppm, and lead was entirely removed from gasoline in 2001 (Jhun et al., 2013).

The burning of firewood generates much finer particles, whose chemical composition contains high levels of toxic organic compounds and other irritants, such as acrolein (Gaeggeler *et al.*, 2008), which is an aldehyde, highly irritant to the respiratory pathway (Gilbert *et al.*, 2005; Seaman *et al.*, 2009). Another toxic compound found in wood smoke is benzo[*a*]pyrene from the family of polycyclic aromatic hydrocarbons (PAH) (Nussbaumer and Hasler, 1999; Tsapakis *et al.*, 2002; Nussbaumer, 2003; Lewtas, 2007; Bari *et al.*, 2011; Cereceda-Balic *et al.*, 2012). This chemical has been recently ratified by the US Environmental Protection Agency (EPA) as cancerogenic for humans, as well as by the World Health Organization (WHO), which has established an air quality regulation with an annual maximum average of 1 ng/m<sup>3</sup> for Europe.

During 2012, two important studies were published in Chile, showing the PAH contents in PM10 for the Valdivia and Temuco urban area for the autumn-winter season. The results showed a range of 2.93–78.01 ng/m<sup>3</sup> for downtown Valdivia (Bravo-Linares *et al.*, 2012) and 48.12–248.74 for the residential area of Temuco (Las Encinas Monitoring Station) (Cereceda-Balic *et al.*, 2012). Another study found concentrations of PAH in PM<sub>10</sub> for Santiago in the spring and winter of 2000, whose values ranged from 1.39 to 59.98 ng/m<sup>3</sup> (Sienra *et al.*, 2005). These results establish that in cities where residential wood combustion is massively used (more than 80% of the population), the levels of PAHs in particulate matter were higher than those found for the city of Santiago, where the Pudahuel is located.

A recent study suggests that particles from mediumtemperature combustion of the wood stoves have a higher toxic and inflammatory potential than particles from more complete combustion conditions (Bolling et al., 2012). This study concluded that a differential deposition of PM from varying combustion conditions might influence the deposited dose. In order to properly evaluate the relative toxicity of RWC soot particles from varying combustion conditions it is consequently of major concern to perform more inhalation studies, mainly because the inhabitants of Temuco and from other Chilean wood smoke polluted cities use wet wood for RWC and have bad habits to operate their wood stoves. Finally, an important review indicated that a number of the PAH and substituted PAH components are significant sources of human exposure to mutagenic and carcinogenic chemicals that may also cause oxidative and DNA damage that can lead to reproductive and cardiovascular effects (Lewtas, 2007).

Apart from the differences in chemical composition of the particles, the cities polluted from RWC present a larger proportion of fine, UFP, and soot than in other contaminated cities (Krecl *et al.*, 2008a, b; Bari *et al.*, 2010, 2011). For

this reason, cities that use firewood as a principal source of heat and fuel for cooking, have a larger health risk for the population, given that UFP penetrate deeper into the respiratory system. This larger proportion of UFP in Temuco could explain its higher toxicity in relation to Pudahuel. In fact, the annual average ratios of  $PM_{2.5}/PM_{10}$ in those two cities are 0.6907 and 0.4458, for Temuco and Pudahuel, respectively (SINCA, 2014), indicating that a higher ratio would imply a greater inhaled dose of fine particles in Temuco over Pudahuel. Both cities are urban environment where workers spend more time indoor than outdoor; however, the air of Pudahuel is impacted with its own and surrounding industries and mobile sources and Temuco is definitively impacted by its residential wood combustion sources, where the indoor air pollution could be higher than outdoor. On the other hand, and in terms of health care, the quality and access to centers and health services in Chile have a high standard, having minimal differences between Pudahuel and Temuco centers, especially for public health.

In addition, the percentage of population older than 65 years old was slightly higher in Temuco (6.9%) than Pudahuel (4.9%) during the study period, having more cases of mortality and morbidity in this group in Temuco than Pudahuel. The age distribution of the other groups was more similar between these two cities. The population of Temuco lives in a city with better ventilation but colder and more polluted with  $PM_{10}$  than Pudahuel. In fact, the particulate matter air pollution has improved in Pudahuel during 2001 to 2006 (Fig.1), having less days over the Chilean standard; but for Temuco the air pollution due to  $PM_{10}$  has worsened, having several environmental emergency and pre-emergency days during that period.

The results of this current study are consistent with those of comparable studies in other similar cities where wood smoke and mobile sources are the most important air pollution problem.

In fact, a study in Christchurch, New Zealand, where the PM<sub>10</sub> air pollution predominantly arises from the residential wood combustion, found an ER of 3.37% (95% CI: 2.34%, 4.40%) in total respiratory admissions and 2.86% (95%: 1.23%, 4.49%) for elderly respiratory admission per interquartile increment in PM<sub>10</sub> of 14.8  $\mu$ g/m<sup>3</sup> at a lag of 2 days (McGowan et al., 2002). That study also found an association with total and elderly cardiac hospital admissions of 1.26% (95%: 0.31, 2.21%) and 1.22% (95%: 0.11, 2.33%), respectively. but they did not found other cardiac diagnoses, while a later study in 7 cities of Australia and New Zealand found association with cardiac and cardiac failure admissions per interquartile increment in PM<sub>10</sub> of 7.5  $\mu$ g/m<sup>3</sup> at a lag of 1 day in the population older than 65 years only (Barnett et al., 2006), whose ER were 1.1%(95%: 0.2%, 2.0%) and 3.4% (95%: 2.1%, 4.7%), respectively. They also found association with ischemic heart disease, cardiac, cardiac failure, myocardial infarction, and total cardiovascular disease and PM<sub>2.5</sub> for elderly (Barnett et al., 2006). A similar study from Sydney, Australia, compared associations between cardiorespiratory hospitalizations and PM<sub>10</sub> resulting from wood smoke with associations between these results and

 $PM_{10}$  derived from other sources (Morgan *et al.*, 2010). They apportioned ambient  $PM_{10}$  on biomass fire days into particulates resulting from biomass combustions and particulate matter from other sources. The authors found an ER of 1.24% (95% CI: 0.22%, 2.27%) for all respiratory hospital admissions per 10 µg/m<sup>3</sup> PM<sub>10</sub> increment due to biomass burning at lag 0. They did not find an association between cardiovascular outcomes and biomass smoke. Finally, a study from Darwin, Australia, compared associations between cardiorespiratory hospitalizations and PM<sub>10</sub> resulting from vegetation fire smoke from 1996 to 2005 (Hanigan *et al.*, 2008). They found positive associations between respiratory disease and PM<sub>10</sub> but not with cardiovascular disease; however, their estimates had wide confidence intervals.

The magnitude of the point estimates for all hospital admissions from our study, the studies discussed above, and other studies in association with biomass smoke (Lipsett et al., 1997; McGowan et al., 2002; Barnett et al., 2005; Barnett et al., 2006; Naeher et al., 2007; Hanigan et al., 2008; Ostro et al., 2008; Sanhueza et al., 2009; Morgan et al., 2010; Clark et al., 2013), are quite comparable with the multi-city studies (Samet et al., 2000; Le Tertre et al., 2002; Pope and Dockery, 2006) of associations between hospital admissions for cardio respiratory diseases with positive associations for a 10  $\mu$ g/m<sup>3</sup> change in ambient PM<sub>10</sub> from 0.34% (HAEIC) to 1.98% (HACOPD), with lags between 4 and 5 for Temuco. These values for elderly are lower than those from the McGowan et al. (2002); this difference may be due to the elderly percentage living in Temuco are almost half of those living in Christchurch. Finally, the magnitude of the point estimates for all cardio respiratory hospital admissions from our study are comparable with those from the multi-city studies, where the ER values were from 0.66% to 0.97%, with lags between 1 and 3.

On the other hand, the aim and design of this study for mortality were similar to those summarized in a Critical Review for different cities (Pope and Dockery, 2006). The results shown here for particulate matter pollution in terms of elderly cardio respiratory mortality (ER = 1.26%) were quite similar to those published in (Pope and Dockery, 2006), which ranged from 0.6% to 1.5%: However, the values found in Ostro *et al.* (2008) for 6 Californian counties were quite different (3%–5%) for cardio respiratory mortality, since these authors used just 3 years statistical modeling. It could be corroborated on their wide estimated confidence intervals.

The greater magnitude of adverse cardio respiratory effects reported in studies specifically examining wood smoke might reflect a true difference in the adverse outcomes associated with this source of particulate matter. Nevertheless, these studies of wood smoke are usually conducted in cities with around short episodes of extreme exposures and/or small population, and their results certainly have more uncertainties than those from multi-city studies, making a challenge their direct comparison and interpretation.

Finally, we cannot discard the possibility that this difference in risks between Temuco and Pudahuel is due to the artifacts of the statistical method that can originate from

a difference in the representatively of the outdoor exposure given by the measurements of  $PM_{10}$  in monitors in both cities, and the susceptibility of the population. An intercomparison between different statistical methods could help to discard the possibility of artifacts among them, mainly considering the source apportionment of each studied city. In addition, future analyses should consider indoor exposure, especially in cities with high particulate matter pollution from the residential burning of firewood.

# CONCLUSIONS

Significant statistical associations were found between daily PM<sub>10</sub> concentrations and mortality and morbidity at Temuco and Pudahuel for respiratory and cardiovascular diseases, mainly in elderly populations.

The results suggest that RWC may be responsible for increased risk in an urban area. This has an important implication in relation with the size and chemical composition of the particles, but also for monitoring, control strategies, and soot regulations. Policy-makers should take into account these aspects along with social and economic considerations if protection of the public health is one of their major concerns, especially for susceptible groups, such as children, the elderly, and those with chronic diseases.

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# REFERENCES

- Analitis, A., Katsouyanni, K., Dimakopoulou, K., Samoli, E., Nikoloulopoulos, A.K., Petasakis, Y., Touloumi, G., Schwartz, J., Anderson, H.R., Cambra, K., Forastiere, F., Zmirou, D., Vonk, J.M., Clancy, L., Kriz, B., Bobvos, J. and Pekkanen, J. (2006). Short-term Effects of Ambient Particles on Cardiovascular and Respiratory Mortality. *Epidemiology* 17: 230–233.
- Andersen, Z.J., Wahlin, P., Raaschou-Nielsen, O., Ketzel, M., Scheike, T. and Loft, S. (2008). Size Distribution and Total Number Concentration of Ultrafine and Accumulation Mode Particles and Hospital Admissions in Children and the Elderly in Copenhagen, Denmark. *Occup. Environ. Med.* 65: 458–466, doi: 10.1136/oem. 007.033290.
- Andersen, Z.J., Olsen, T.S., Andersen, K.K., Loft, S., Ketzel, M. and Raaschou-Nielsen, O. (2010). Association between Short-term Exposure to Ultrafine Particles and Hospital Admissions for Stroke in Copenhagen, Denmark. *Eur. Heart J.* 31: 2034–2040, doi: 10.1093/eurheartj/ehq 188.
- Arias, R.A. and Mader, T.L. (2010). Determination of Potential Risk of Heat Stress of Cattle in Four Locations of Central and Southern Chile. *Arch. Med. Vet.* 42: 33–39.
- Bari, M.A., Baumbach, G., Kuch, B. and Scheffknecht, G.

(2010). Temporal Variation and Impact of Wood Smoke Pollution on a Residential Area in Southern Germany. *Atmos. Environ.* 44: 3823–3832, doi: 10.1016/j.atmosenv. 2010.06.031.

- Bari, M.A., Baumbach, G., Brodbeck, J., Struschka, M., Kuch, B., Dreher, W. and Scheffknecht, G. (2011.) Characterisation of Particulates and Carcinogenic Polycyclic Aromatic Hydrocarbons in Wintertime Wood-fired Heating in Residential Areas. *Atmos. Environ.* 45: 7627–7634, doi: 10.1016/j.atmosenv.2010.11.053.
- Barnett, A.G., Williams, G.M., Schwartz, J., Best, T.L., Neller, A.H., Petroeschevsky, A.L. and Simpson, R.W. (2006). The Effects of Air Pollution on Hospitalizations for Cardiovascular Disease in Elderly People in Australian and New Zealand Cities. *Environ. Health Perspect.* 114: 1018–1023, doi: 0.1289/Ehp.8674.
- Barnett, A.G., Williams, G.M., Schwartz, J., Neller, A.H., Best, T.L., Petroeschevsky, A.L., Simpson, R.W. (2005). Air Pollution and Child Respiratory Health - A Casecrossover Study in Australia and New Zealand. *Am. J. Respir. Crit. Care Med.* 171: 1272–1278, doi: 10.1164/ rccm.200411-1586OC.
- Belleudi, V., Faustini, A., Stafoggia, M., Cattani, G., Marconi, A., Perucci, C.A. and Forastiere, F. (2010). Impact of Fine and Ultrafine Particles on Emergency Hospital Admissions for Cardiac and Respiratory Diseases. *Epidemiology* 21: 414–423, doi: 10.1097/Ede.0b013e3181d5c021.
- Biswas, P. and Wu, C.Y. (2005). 2005 Critical Review: Nanoparticles and the Environment. *J. Air Waste Manage. Assoc.* 55: 708–746.
- Bolling, A.K., Totlandsdal, A.I., Sallsten, G., Braun, A., Westerholm, R., Bergvall, C., Boman, J., Dahlman, H. J., Sehlstedt, M., Cassee, F., Sandstrom, T., Schwarze, P.E. and Herseth, J.I. (2012). Wood Smoke Particles from Different Combustion Phases Induce Similar Proinflammatory Effects in a Co-culture of Monocyte and Pneumocyte Cell Lines. *Part. Fibre Toxicol.* 9: 45, doi: 10.1186/1743-8977-9-45.
- Branis, M., Vyskovska, J., Maly, M. and Hovorka, J. (2010). Association of Size-resolved Number Concentrations of Particulate Matter with Cardiovascular and Respiratory Hospital Admissions and Mortality in Prague, Czech Republic. *Inhalation Toxicol.* 22: 21–28, doi: 10.3109/ 08958378.2010.504758.
- Bravo-Linares, C., Ovando-Fuentealba, L., Mudge, S.M., Cerpa, J. and Loyola-Sepulveda, R. (2012). Source Allocation of Aliphatic and Polycyclic Aromatic Hydrocarbons in Particulate-Phase (PM<sub>10</sub>) in the City of Valdivia, Chile. *Polycyclic Aromat. Compd.* 32: 390– 407, doi: 10.1080/10406638.2012.661829.
- Cakmak, S., Dales, R.E. and Vidal, C.B. (2007). Air Pollution and Mortality in Chile: Susceptibility among the Elderly. *Environ. Health Perspect.* 115: 524–527, doi: 10.1289/Ehp.9567.
- Cao, J.J., Xu, H.M., Xu, Q., Chen, B.H. and Kan, H.D. (2012). Fine Particulate Matter Constituents and Cardiopulmonary Mortality in a Heavily Polluted Chinese City. *Environ. Health Perspect.* 120:373–378, doi: 10.1289/Ehp.1103671.

- CENMA-CONAMA (2010). Emission Inventory Update of Temuco and Padre Las Casas.
- Cereceda-Balic, F., Fadic, X., Llanos, A.L., Dominguez, A.M., Guevara, J.L., Vidal, V., Diaz-Robles, L.A., Schiappacasse, L.N. and Etcharren, P. (2012). Obtaining Polycyclic Aromatic Hydrocarbon Concentration Ratios and Molecular Markers for Residential Wood Combustion: Temuco, A Case Study. *J. Air Waste Manage. Assoc.* 62: 44–51, doi: 10.1080/10473289.2011.617599.
- Chang, M.C.O., Chow, J.C., Watson, J.G., Hopke, P.K., Yi, S.M. and England, G.C. (2004). Measurement of Ultrafine Particle Size Distributions from Coal-, Oil-, and Gas-Fired Stationary Combustion Sources. J. Air Waste Manage. Assoc. 54: 1494–1505, doi: 10.1080/10473289. 2004.10471010.
- Chen, R.J., Samoli, E., Wong, C.M., Huang, W., Wang, Z. S., Chen, B.H., Kan, H.D. and Grp, C.C. (2012). Associations between Short-term Exposure to Nitrogen Dioxide and Mortality in 17 Chinese Cities: The China Air Pollution and Health Effects Study (CAPES). *Environ. Int.* 45: 32–38, doi: 10.1016/j.envint.2012.04.008.
- Clark, M.L., Peel, J.L., Balakrishnan, K., Breysse, P.N., Chillrud, S.N., Naeher, L.P., Rodes, C.E., Vette, A.F. and Balbus, J.M. (2013). Health and Household Air Pollution from Solid Fuel Use: The Need for Improved Exposure Assessment. *Environ. Health Perspect.* 121: 1120–1128, doi: 10.1289/Ehp.1206429.
- CONAMA (2005). Ministerio Secretaría General de la Presidencia de la República de Chile. Revisa, Reformula y Actualiza Plan de Prevención y Descontaminación Atmosférica para la Región Metropolitana, PPDA.
- Dab, W., Segala, C., Dor, F., Festy, B., Lameloise, P., Le Moullec, Y., Le Tertre, A., Medina, S., Quenel, P., Wallaert, B. and Zmirou, D. (2001). Air Pollution and Health: Correlation or Causality? The Case of the Relationship between Exposure to Particles and Cardiopulmonary Mortality. J. Air Waste Manage. Assoc. 51: 220–235.
- Delfino, R.J., Sioutas, C. and Malik, S. (2005). Potential Role of Ultrafine Particles in Associations between Airborne Particle Mass and Cardiovascular Health. *Environ. Health Perspect.* 113: 934–946.
- Díaz-Robles, L.A., Saavedra, H., Schiappacasse, L. and Cereceda-Balic, F. (2011). The Air Quality in Chile: Twenty Years of Challenge, in *Environmental Management Air & Waste Management Association*.
- Díaz-Robles, L.A., Fu, J.S., Vergara-Fernández, A., Etcharren, P., Schiappacasse, L.N., Reed, G.D. and Silva, M.P. (2014). Health Risks Caused by Short Term Exposure to Ultrafine Particles Generated by Residential Wood Combustion: A Case Study of Temuco, Chile. *Environ. Int.* 66: 174–181, doi: 10.1016/j.envint.2014.01.017.
- Dockery, D.W. and Schwartz, J. (1992). Particulate Air-Pollution and Daily Mortality in Steubenville, Ohio -Response. *Am. J. Epidemiol.* 135: 23–25.
- Dockery, D.W., Pope, C.A., Xu, X.P., Spengler, J.D., Ware, J.H., Fay, M.E., Ferris, B.G. and Speizer, F.E. (1993). An Association between Air-Pollution and Mortality in 6 United-States Cities. *New Engl. J. Med.* 329: 1753–1759.

- Dominici, F. and Burnett, R.T. (2003). Risk Models for Particulate Air Pollution. *J. Toxicol. Environ. Health Part A* 66:1883–1889.
- Dominici, F., Zanobetti, A., Zeger, S.L., Schwartz, J. and Samet, J.M. (2004). Hierarchical Bivariate Time Series Models: A Combined Analysis of the Effects of Particulate Matter on Morbidity and Mortality. *Biostatistics* 5: 341– 360.
- Dominici, F., McDermott, A., Daniels, M., Zeger, S.L. and Samet, J.M. (2005). Revised Analyses of the National Morbidity, Mortality, and Air Pollution Study: Mortality among Residents of 90 Cities. J. Toxicol. Environ. Health Part A 68: 1071–1092.
- Dominici, F., Peng, R.D., Bell, M.L., Pham, L., McDermott, A., Zeger, S.L. and Samet, J.M. (2006). Fine Particulate Air Pollution and Hospital Admission for Cardiovascular and Respiratory Diseases. J. Am. Med. Assoc. 295: 1127– 1134.
- Donaldson, K., Stone, V., Seaton, A. and MacNee, W. (2001). Ambient Particle Inhalation and the Cardiovascular System: Potential Mechanisms. *Environ. Health Perspect.* 109: 523–527.
- Fischer, P., Hoek, G., Brunekreef, B., Verhoeff, A. and van Wijnen, J. (2003). Air Pollution and Mortality in the Netherlands: Are the Elderly More at Risk? *Eur. Respir. J.* 21: 34s–38s.
- Gaeggeler, K., Prevot, A.S.H., Dommen, J., Legreid, G., Reimann, S. and Baltensperger, U. (2008). Residential Wood Burning in an Alpine Valley as a Source for Oxygenated Volatile Organic Compounds, Hydrocarbons and Organic Acids. *Atmos. Environ.* 42: 8278–8287, doi: 10.1016/j.atmosenv.2008.07.038.
- Gilbert, N.L., Guay, M., Miller, J.D., Judek, S., Chan, C. C. and Dales, R.E. (2005). Levels and Determinants of Formaldehyde, Acetaldehyde, and Acrolein in Residential Indoor Air in Prince Edward Island, Canada. *Environ. Res.* 99: 11–17, doi: 10.1016/j.envres.2004.09.009.
- Grass, D. and Cane, M. (2008). The Effects of Weather and Air Pollution on Cardiovascular and Respiratory Mortality in Santiago, Chile, during the Winters of 1988-1996. *Int. J. Climatol.* 28: 1113–1126, doi: 10.1002/Joc. 1592.
- Hanigan, I.C., Johnston, F.H. and Morgan, G.G. (2008). Vegetation Fire Smoke, Indigenous Status and Cardio-Respiratory Hospital Admissions in Darwin, Australia, 1996-2005: A Time-series Study. *Environ. Health* doi: 7: 42, doi: 10.1186/1476-069x-7-42.
- Hosseini, S., Li, Q., Cocker, D., Weise, D., Miller, A., Shrivastava, M., Miller, J.W., Mahalingam, S., Princevac, M. and Jung, H. (2010). Particle Size Distributions from Laboratory-scale Biomass Fires Using Fast Response Instruments. *Atmos. Chem. Phys.* 10: 8065–8076, doi: 10.5194/acp-10-8065-2010.
- Ibald-Mulli, A., Wichmann, H.E., Kreyling, W. and Peters, A. (2002). Epidemiological Evidence on Health Effects of Ultrafine Particles. J. Aerosol Med. 15: 189–201.
- Jhun, I., Oyola, P., Moreno, F., Castillo, M.A. and Koutrakis, P. (2013). PM<sub>2.5</sub> Mass and Species Trends in Santiago, Chile, 1998 to 2010: The Impact of Fuel-related

Interventions and Fuel Sales. *J. Air Waste Manage. Assoc.* 63: 161–169, doi: 10.1080/10962247.2012.742027.

- Jimenez, E., Linares, C., Rodriguez, L.F., Bleda, M.J. and Diaz, J. (2009). Short-term Impact of Particulate Matter (PM<sub>2.5</sub>) on Daily Mortality among the over-75 Age Group in Madrid (Spain). *Sci. Total Environ.* 407: 5486– 5492, doi: 10.1016/j.scitotenv.2009.06.038.
- Jimenez, E., Linares, C., Martinez, D. and Diaz, J. (2011). Particulate Air Pollution and Short-term Mortality Due to Specific Causes among the Elderly in Madrid (Spain): Seasonal Differences. *Int. J. Environ. Health Res.* 21: 372–390, doi: 10.1080/09603123.2011.560251.
- Jorquera, H. and Barraza, F. (2012). Source Apportionment of Ambient PM<sub>2.5</sub> in Santiago, Chile: 1999 and 2004 Results. *Sci. Total Environ.* 435: 418–429, doi: 10.1016/ j.scitotenv.2012.07.049.
- Katsouyanni, K., Touloumi, G., Samoli, E., Gryparis, A., Le Tertre, A., Monopolis, Y., Rossi, G., Zmirou, D., Ballester, F., Boumghar, A., Anderson, H.R., Wojtyniak, B., Paldy, A., Braunstein, R., Pekkanen, J., Schindler, C. and Schwartz, J. (2001). Confounding and Effect Modification in the Short-term Effects of Ambient Particles on Total Mortality: Results from 29 European Cities within the APHEA2 Project. *Epidemiology* 12: 521–531.
- Katsouyanni, K. (2003). Ambient Air Pollution and Health. Br. Med. Bull. 68: 143–156.
- Krecl, P., Larsson, E.H., Strom, J. and Johansson, C. (2008a). Contribution of Residential Wood Combustion and Other Sources to Hourly Winter Aerosol in Northern Sweden Determined by Positive Matrix Factorization. *Atmos. Chem. Phys.* 8: 3639–3653.
- Krecl, P., Strom, J. and Johansson, C. (2008b). Diurnal Variation of Atmospheric Aerosol during the Wood Combustion Season in Northern Sweden. *Atmos. Environ.* 42: 4113–4125, doi: 10.1016/j.atmosenv.2008.01.026.
- Krewski, D., Burnett, R., Jerrett, M., Pope, C. A., Rainham, D., Calle, E., Thurston, G. and Thun, M. (2005). Mortality and Long-term Exposure to Ambient Air Pollution: Ongoing Analyses Based on the American Cancer Society Cohort. J. Toxicol. Environ. Health Part A 68: 1093–1109.
- Le Tertre, A., Medina, S., Samoli, E., Forsberg, B., Michelozzi, P., Boumghar, A., Vonk, J.M., Bellini, A., Atkinson, R., Ayres, J.G., Sunyer, J., Schwartz, J. and Katsouyanni, K. (2002). Short-term Effects of Particulate Air Pollution on Cardiovascular Diseases in Eight European Cities. J. Epidemiol. Community Health 56: 773–779, doi: 10.1136/jech.56.10.773.
- Lefranc, A. and Larrieu, S. (2008). Ultrafine Particles and Health: Insights from Epidemiological Studies. *Environ. Risque Sante* 7: 349–355, doi: 10.1684/ers.2008.0165.
- Leitte, A.M., Schlink, U., Herbarth, O., Wiedensohler, A., Pan, X.C., Hu, M., Richter, M., Wehner, B., Tuch, T., Wu, Z.J., Yang, M.J., Liu, L.Q., Breitner, S., Cyrys, J., Peters, A., Wichmann, H.E. and Franck, U. (2011). Size-Segregated Particle Number Concentrations and Respiratory Emergency Room Visits in Beijing, China. *Environ. Health Perspect.* 119: 508–513, doi: 10.1289/ Ehp.1002203.

- Lewtas, J. (2007). Air Pollution Combustion Emissions: Characterization of Causative Agents and Mechanisms Associated with Cancer, Reproductive, and Cardiovascular Effects. *Mutat. Res.-Rev. Mutat. Res.* 636: 95–133, doi: 10.1016/j.mrrev.2007.08.003.
- Lightya, J.S., Verantha, J.M. and Sarofima, A.F. (2000). Combustion Aerosols: Factors Governing Their Size and Composition and Implications to Human Health. *J. Air Waste Manage. Assoc.* 50: 1565–1618, doi: 10.1080/104 73289.2000.10464197.
- Lipsett, M., Hurley, S. and Ostro, B. (1997). Air Pollution and Emergency Room Visits for Asthma in Santa Clara County, California. *Environ. Health Perspect.* 105: 216– 222, doi: 10.1289/Ehp.97105216.
- Lipsett, M. and Campleman, S. (1999). Occupational Exposure to Diesel Exhaust and Lung Cancer: A Metaanalysis. *Am. J. Public Health* 89: 1009–1017.
- Liu, L.Q., Breitner, S., Schneider, A., Cyrys, J., Bruske, I., Franck, U., Schlink, U., Leitte, A.M., Herbarth, O., Wiedensohler, A., Wehner, B., Pan, X.C., Wichmann, H.E. and Peters, A. (2013). Size-fractioned Particulate Air Pollution and Cardiovascular Emergency Room Visits in Beijing, China. *Environ. Res.* 121: 52–63, doi: 10.1016/j.envres.2012.10.009.
- Lopez-Villarrubia, E., Iniguez, C., Peral, N., Garcia, M.D. and Ballester, F. (2012). Characterizing Mortality Effects of Particulate Matter Size Fractions in the Two Capital Cities of the Canary Islands. *Environ. Res.* 112: 129– 138, doi: 10.1016/j.envres.2011.10.005.
- Mate, T., Guaita, R., Pichiule, M., Linares, C. and Diaz, J. (2010). Short-term Effect of Fine Particulate Matter (PM<sub>2.5</sub>) on Daily Mortality Due to Diseases of the Circulatory System in Madrid (Spain). *Sci. Total Environ.* 408: 5750–5757, doi: 10.1016/j.scitotenv.2010.07.083.
- McGowan, J.A., Hider, P.N., Chacko, E. and Town, G.I. (2002). Particulate Air Pollution and Hospital Admissions in Christchurch, New Zealand. *Aust. N. Z. J. Public Health* 26: 23–29, doi: 10.1111/j.1467-842X.2002.tb00266.x.
- Morgan, G., Sheppeard, V., Khalaj, B., Ayyar, A., Lincoln, D., Jalaludin, B., Beard, J., Corbett, S. and Lumley, T. (2010). Effects of Bushfire Smoke on Daily Mortality and Hospital Admissions in Sydney, Australia. *Epidemiology* 21: 47–55, doi: 10.1097/Ede.0b013e3181c15d5a.
- Naeher, L.P., Brauer, M., Lipsett, M., Zelikoff, J.T., Simpson, C.D., Koenig, J.Q. and Smith, K.R. (2007). Woodsmoke Health Effects: A Review. *Inhalation Toxicol.* 19: 67– 106, doi: 10.1080/08958370600985875.
- Nemmar, A., Hoet, P.H.M., Thomeer, M., Nemery, B., Vanquickenborne, B., Vanbilloen, H., Mortelmans, L., Hoylaerts, M.F., Verbruggen, A. and Dinsdale, A. (2002). Passage of Inhaled Particles into the Blood Circulation in Humans - Response. *Circulation* 106: E141–E142.
- Nitta, H., Yamazaki, S., Omori, T. and Sato, T. (2010). An Introduction to Epidemiologic and Statistical Methods Useful in Environmental Epidemiology. *J. Epidemiol.* 20: 177–184, doi: 10.2188/jea.JE20100010.
- Nussbaumer, T. and Hasler, P. (1999). Formation and Behavior of Aerosols from Wood Combustion. *Holz Roh Werkst.* 57: 13–22, doi: 10.1007/Pl00002614.

- Nussbaumer, T. (2003). Combustion and Co-combustion of Biomass: Fundamentals, Technologies, and Primary Measures for Emission Reduction. *Energy Fuels* 17: 1510–1521, doi: 10.1021/Ef030031q.
- O'Neill, M.S., Bell, M.L., Ranjit, N., Cifuentes, L.A., Loomis, D., Gouveia, N. and Borja-Aburto, V.H. (2008). Air Pollution and Mortality in Latin America The Role of Education. *Epidemiology* 19: 810–819, doi: 10.1097/ Ede.0b013e3181816528.
- Ostro, B.D., Feng, W.Y., Broadwin, R., Malig, B.J., Green, R.S. and Lipsett, M.J. (2008). The Impact of Components of Fine Particulate Matter on Cardiovascular Mortality in Susceptible Subpopulations. *Occup. Environ. Med.* 65: 750–756, doi: 10.1136/oem.2007.036673.
- Ostro, B., Broadwin, R., Green, S., Feng, W.Y. and Lipsett, M. (2006). Fine Particulate Air Pollution and Mortality in Nine California Counties: Results from CALFINE. *Environ. Health Perspect.* 114: 29–33.
- Peng, R.D., Dominici, F., Pastor-Barriuso, R., Zeger, S.L. and Samet, J.M. (2005). Seasonal Analyses of Air Pollution and Mortality in 100 US Cities. *Am. J. Epidemiol.* 161: 585–594.
- Peng, R.D., Dominici, F. and Louis, T.A. (2006). Model Choice in Time Series Studies of Air Pollution and Mortality. J. R. Stat. Soc. Ser. A 169: 179–198.
- Pope, C.A. and Dockery, D.W. (2006). Health Effects of Fine Particulate Air Pollution: Lines that Connect. J. Air Waste Manage. Assoc. 56: 709–742.
- Pope, C.A., Thun, M.J., Namboodiri, M.M., Dockery, D.W., Evans, J.S., Speizer, F.E. and Heath, C.W. (1995). Particulate Air-Pollution as a Predictor of Mortality in a Prospective-Study of Us Adults. *Am. J. Respir. Crit. Care Med.* 151:669–674.
- Pope, C.A., Burnett, R.T., Thun, M.J., Calle, E.E., Krewski, D., Ito, K. and Thurston, G.D. (2002). Lung Cancer, Cardiopulmonary Mortality, and Long-term Exposure to Fine Particulate Air Pollution. J. Am. Med. Assoc. 287: 1132–1141.
- Pope, C.A., Burnett, R.T., Thurston, G.D., Thun, M.J., Calle, E.E., Krewski, D. and Godleski, J.J. (2004). Cardiovascular Mortality and Long-term Exposure to Particulate Air Pollution - Epidemiological Evidence of General Pathophysiological Pathways of Disease. *Circulation* 109: 71–77.
- Samet, J.M., Zeger, S.L., Dominici, F., Curriero, F., Coursac, I., Dockery, D.W., Schwartz, J. and Zanobetti, A. (2000). The National Morbidity, Mortality, and Air Pollution Study. Part II: Morbidity and Mortality from Air Pollution in the United States. *Res. Rep. Health Eff. Inst.* 94: 5–70; Discussion 71–79.
- Samet, J.M. (2002). Air Pollution and Epidemiology: "Deja Vu All over Again?". *Epidemiology* 13:118–119.
- Sanhueza, P.A., Torreblanca, M.A., Diaz-Robles, L.A., Schiappacasse, L.N., Silva, M.P. and Astelle, T.D. (2009). Particulate Air Pollution and Health Effects for Cardiovascular and Respiratory Causes in Temuco, Chile: A Wood-smoke-polluted Urban Area. J. Air Waste

Manage. Assoc. 59: 1481–1488, doi: 10.3155/1047-3289.59.12.1481.

- Schulz, H., Harder, V., Ibald-Mulli, A., Khandoga, A., Koenig, W., Krombach, F., Radykewicz, R., Stampfl, A., Thorand, B. and Peters, A. (2005). Cardiovascular Effects of Fine and Ultrafine Particles. *J. Aerosol Med.* 18: 1–22.
- Schwartz, J. and Zanobetti, A. (2000). Using Meta-smoothing to Estimate Dose-response Trends Across Multiple Studies, with Application to Air Pollution and Daily Death. *Epidemiology* 11: 666–672.
- Seaman, V.Y., Bennett, D.H. and Cahill, T.M. (2009). Indoor Acrolein Emission and Decay Rates Resulting from Domestic Cooking Events. *Atmos. Environ.* 43: 6199–6204, doi: 10.1016/j.atmosenv.2009.08.043.
- Sienra, M.D., Rosazza, N.G. and Prendez, M. (2005). Polycyclic Aromatic Hydrocarbons and Their Molecular Diagnostic Ratios in Urban Atmospheric Respirable Particulate Matter. *Atmos. Res.* 75: 267–281, doi: 10.1016/j.atmosres.2005.01.003.
- SINCA (2014). Chilean Environmental Ministery. National Air Quality System (Sistema Nacional de Calidad del Aire, SINCA in Spanish), http://sinca.mma.gob.cl/.
- Sioutas, C., Delfino, R.J. and Singh, M. (2005). Exposure Assessment for Atmospheric Ultrafine Particles (UFPs) and Implications in Epidemiologic Research. *Environ. Health Perspect.* 113: 947–955, doi: 10.1289/Ehp.7939.
- Tainio, M., Tuomisto, J.T., Hanninen, O., Aarnio, P., Koistinen, K.J., Jantunen, M.J. and Pekkanen, J. (2005). Health Effects Caused by Primary Fine Particulate Matter ( $PM_{2.5}$ ) Emitted from Buses in the Helsinki Metropolitan Area, Finland. *Risk Anal.* 25:151–160.
- Tsapakis, M., Lagoudaki, E., Stephanou, E.G., Kavouras, I. G., Koutrakis, P., Oyola, P. and von Baer, D. (2002). The Composition and Sources of PM<sub>2.5</sub> Organic Aerosol in Two Urban Areas of Chile. *Atmos. Environ.* 36: 3851– 3863.
- Valdes, A., Zanobetti, A., Halonen, J.I., Cifuentes, L., Morata, D. and Schwartz, J. (2012). Elemental Concentrations of Ambient Particles and Cause Specific Mortality in Santiago, Chile: A Time Series Study. *Environ. Health* 11: 82, doi: 10.1186/1476-069x-11-82.
- Wong, C.M., Vichit-Vadakan, N., Kan, H.D., Qian, Z.M. and Teams, P.P. (2008). Public Health and Air Pollution in Asia (PAPA): A Multicity Study of Short-term Effects of Air Pollution on Mortality. *Environ. Health Perspect.* 116: 1195–1202, doi: 10.1289/Ehp.11257.
- Zanobetti, A. and Schwartz, J. (2005). The Effect of Particulate Air Pollution on Emergency Admissions for Myocardial Infarction: A Multicity Case-crossover Analysis. *Environ. Health Perspect.* 113: 978–982.

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