Environment International 66 (2014) 174–181

Contents lists available at ScienceDirect

Environment International

journal homepage: www.elsevier.com/locate/envint

Health risks caused by short term exposure to ultrafine particles generated by residential wood combustion: A case study of Temuco, Chile

Luis A. Díaz-Robles [a,*], Joshua S. Fu [b], Alberto Vergara-Fernández [c], Pablo Etcharren [d], Luis N. Schiappacasse [a], Gregory D. Reed [b], María P. Silva [e]

[a] *Núcleo de Energías Renovables, Universidad Católica de Temuco, Chile*
[b] *Department of Civil and Environmental Engineering, University of Tennessee, Knoxville, TN, USA*
[c] *Facultad de Ingeniería y Ciencias Aplicadas, Universidad de los Andes, Chile*
[d] *Ministerio del Medio Ambiente, Araucanía, Chile*
[e] *Hospital Clínico de la Universidad Mayor, Chile*

ARTICLE INFO

Article history:
Received 9 September 2013
Accepted 23 January 2014
Available online xxxx

Keywords:
Ultrafine particles
Residential wood combustion
Relative risk
APHEA2
Temuco
Chile

ABSTRACT

Temuco is one of the most highly wood smoke polluted cities in Chile; however, there is scarce evidence of respiratory morbidity due to fine particulate matter. We aimed to estimate the relationship between daily concentration of ultrafine particles (UFP), with an aerodynamic diameter $\leq$0.1 μm, and outpatient visits for respiratory illness at medical care centers of Temuco, Chile, from August the 20th, 2009 to June the 30th, 2011. The Air Pollution Health Effects European Approach (APHEA2) protocol was followed, and a multivariate semi-parametric Poisson regression model was fitted with GAM techniques using R-Project statistical package; controlling for trend, seasonality, and confounders. The daily UFP were measured by a MOUDI NR-110 sampler. We found that results of the statistical analyses show significant associations between UFP and respiratory outpatient visits, with the elderly (population $\geq$65 years), being the group that presented the greatest risk. An interquartile increase of 4.73 μg/m^3 in UFP (lag 5 days) was associated with respiratory outpatient visits with a relative risk (RR) of 1.1458 [95% CI (1.0497–1.2507)] for the elderly. These results show novel findings regarding the relevance of daily UFP concentrations and health risk, especially for susceptible population in a wood smoke polluted city.

1. Introduction

The air quality of Temuco and other southern cities of Chile is becoming polluted due mainly to residential wood combustion (RWC). However, the species and quality of the biomass, wood burning devices, and their operation modes have tremendous differences in terms of emissions over those other cities where there are huge particulate matter (PM) episodes in winter (Diaz-Robles et al., 2008). Indeed, some of these studies indicate that the contribution of RWC to the total emissions of PM$_{10}$ is 93%. RWC and other sources, such as mobile sources, point sources, etc., emit a large portion of UFP (McDonald et al., 2000), considered the most harmful to human health. Finally, the emissions of ultrafine particles represent almost 90% of the total particles from RWC sources (Chow et al., 1998), and therefore in a city polluted mainly with wood smoke, most of the PM$_{10}$ and PM$_{2.5}$ ambient concentrations are ultrafine particles.

Temuco is among the five largest cities in Chile with high air pollution from RWC, having daily PM$_{10}$ monitoring database since 2001, and daily PM$_{2.5}$ since 2008, (see Figs. S1 and S2, Supplementary information) (SINCA, 2013). In fact, during 2012 there were 125 exceedances of daily PM$_{2.5}$ concentration of 50 μg/m^3 (Chilean Standard). That year there were seven days in which hourly concentrations of PM$_{2.5}$ exceeded 880 μg/m^3, reaching values as high as 993 μg/m^3, which took place on August 26, 2012 at 1:00 (winter). In addition, there were thirteen days when the daily concentration of PM$_{2.5}$ exceeded 200 μg/m^3, getting a value of 291 μg/m^3 on July 16, 2012 (Fig. S2 in the Supplementary data). Finally, the annual average PM$_{2.5}$ concentration reached its highest level in 2012 since 2008, with a value of 59 μg/m^3, much higher than the Chilean annual standard of 20 μg/m^3 and of 10 μg/m^3 established by the World Health Organization (WHO).

Although Temuco was designed as non-attainment area for PM$_{10}$ in 2005 and its Atmospheric Decontamination Plan came into effect in 2010, advances in reducing the days of standard are not yet achieved, but instead, a gradual increase of PM$_{10}$ is still observed, Fig. S1 in the Supplementary data. It is noteworthy that in the month of April of the year 2013, this city was designated as non-attainment area for PM$_{2.5}$, Chilean standard that became effective on January 1, 2012.

---

[*] Corresponding author. Tel.: +56 45 2205483; fax: +56 45 2205430.
E-mail address: ldiaz@uct.cl (L.A. Díaz-Robles).

http://dx.doi.org/10.1016/j.envint.2014.01.017

1.1. Characteristics and effects of fine and ultrafine particles

Over that last several years, fine and UFP have received increasing attention, due to the potential adverse effects on human health (Morawska et al., 2004). In comparison with larger particles, UFP have a larger surface in relation to volume, and can transport large quantities of toxic pollutants, absorbed or condense. These pollutants include oxidant gasses, organic compounds and transition metals (Oberdörster, 2001; Sioutas et al., 2005). UFP are commonly measured by number of particles per cubic meter (Brook et al., 2010), and also mass concentration (Khlystov et al., 2004). Their high number per m^3 and high active surface area, as well as their high deposition efficiency in the pulmonary region make that these particles very dangerous for the cardiovascular system (Ralph et al., 2005).

Although UFP represent an important fraction of fine particulate matter, studies on their health effects are scarce. At the same time, the chemical composition and the size of the ultrafine particles that produce health effects have not been investigated in depth. However, studies found that fine particulate matter and coarse particles seem to have an adverse influence on the human health, besides UFPs, given that they promote and maintain respiratory stress, provoking inflammatory processes (Andersen et al., 2010; Belleudi et al., 2010; Hertel et al., 2010; Laumbach and Kipen, 2012).

So far, numerous epidemiological studies show a positive correlation between the level of particulate contamination in the air (PM_{10} and $PM_{2.5}$) and the increase of morbidity and mortality in adults and children, especially in people with pre-existent cardiovascular and respiratory illnesses (Halonen et al., 2008; Kampa and Castanas, 2008). Coarse particulate matter, with an aerodynamic diameter in the range of 2.5 to 10 μm, can be deposited principally in the upper respiratory system and can be deactivated through mucociliary action (Cormier et al., 2006). However, fine particulate matter ($PM_{2.5}$) is capable of penetrating the alveolar regions of the lungs, while the ultrafine component can penetrate the epithelial layers (Oberdörster, 2001).

Recent studies show that UFP can be efficiently deposited in the alveolar compartment of the lungs and enter the epithelial cells and into the pulmonary system, where it can perturb vascular function, activating plaquettes and promoting the formation of thrombosis (Andersen et al., 2008; Nemmar et al., 2002; Stewart et al., 2010). Recently, other studies show that UFP present a series of specific characteristics and reactivity patterns that differ from that of larger particles, including the induction of inflammatory processes, high pulmonary deposition due to the high concentration per unit of area, the oxidative capacity and the capacity to induce damage on a DNA level (Andersen et al., 2008; Diaz-Robles et al., 2013; Wichmann, 2007). In 1995, the first studies showed that the ultrafine fraction of particulate matter is the most damaging to health (Seaton et al., 1995). From then on, research has been dedicated to the human toxicology and pathological mechanisms of UFP, with important findings in studies on cardiovascular and respiratory effects (Oberdörster et al., 2005).

In Chile the health effects associated with fine particle pollution are under study principally in Santiago and Temuco, due to the existence of a better database (Sanhueza et al., 2009). However, studies are yet to show a relationship between the exposure to ultrafine particulate matter and morbidity. Instead, these authors determined the relationship between air pollution from PM_{10} 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 APHEA2 protocol was followed, and a multivariate Poisson regression model was fitted, controlling for trend, seasonality, and confounders for Temuco during 1998–2006. The results show that PM_{10} had a significant association with daily mortality and morbidity, with the elderly being the group that presented the greatest risk. The RR for respiratory causes, with an increase of 100 $\mu g/m^3$ of PM_{10}, was 1.163 [95% CI (1.057–1.279)] for mortality, 1.137 [95% CI (1.096–1.178)] for hospital admissions, and 1.162 for ARI [95% CI (1.144–1.181)]. These results were consistent with those of comparable studies in other similar cities where wood smoke is the most important air pollution problem.

Finally, and applying the same APHEA protocol in an older study in Santiago, Chile, a study found that the RR estimates in Temuco were higher than those in Santiago, 1.061 (CI 1.017–1.106) for respiratory mortality and 1.025 (CI 1.005–1.046) for cardiovascular mortality (Sanhueza et al., 2006), suggesting that the inhabitants of Temuco would be exposed to a higher risk than those of Santiago with equal increments in PM_{10} concentration. One of the reasons for this difference in the RR could be the chemical composition and size distribution of the particles in each city. With respect to the chemical composition of the ambient particles, other studies indicated that the main air pollution sources in Temuco are residential wood combustion, while the pollution in Santiago is caused by diesel from mobile sources (Tsapakis et al., 2002). Their results presented higher levels of polycyclic aromatic hydrocarbons (PAHs) and other air-borne toxins in Temuco than Santiago. In fact, the Benzo(a)pyrene concentration in Temuco was 98.5 ng/m^3 versus 10.9 ng/m^3 in Santiago. Particle size distribution also differs between Temuco and Santiago. In the winter season, $PM_{2.5}$ presents a higher proportion of the total PM_{10} in Temuco (80–90%) than in Santiago (30–60%) (Sanhueza et al., 2009). This difference can be explained from some experiments performed in Europe. In effect, using lung cell survival analysis, a study proved that the wood smoke from a badly operated wood stove exhibited 15 times higher toxicity than diesel soot, mainly from condensable matter from the residential wood stove (Klippel and Nussbaumer, 2007). The authors also measured PAHs and size distribution from different source emissions.

Considering the results of these studies and that the main source of air pollution in Temuco is the RWC that emits finer particles, we conducted the measurement of UFP for 22 months to represent a threshold and estimated the health risks when the susceptible population was expose to daily concentrations of UFP during 22 months.

2. Methodology

With the intention of obtaining the RR of daily number of medical consultations at Temuco city, when exposed to acute concentrations of ultrafine particles, two stages were planned; (a) collection of health data, meteorology, and air quality, and (b) model development and relative risk estimation by UFP, health effects, and susceptible groups.

2.1. Health, meteorological and air quality data collection

The daily health data was collected from three healthcare municipal centers; Pedro de Valdivia, Villa Alegre, and Amanecer, located in different areas of the Temuco city shown in Fig. 1. This data was separated into three age groups, those under and equal to 5 years old (Group 1; G1), older than 5 years old and younger and equal to 64 years old (Group 2; G2), and older than 64 years old (Group 3; G3). The data considered the outpatient visits for respiratory illness classified under the letter J of ICD-10 (International Classification of Diseases, ICD-10, Chapter IX, sorting J) shown in Table 1.

The daily ultrafine particles were sampled with a Micro-Orifice Uniform-Deposit Impactor (MOUDI), 100-NR model, made by MSP. This was installed at the Catholic University of Temuco monitoring station. The mass of the filters was determined on a microbalance Radwag, model MXA 21. The hourly and daily meteorological data was collected from the Las Encinas monitoring station available at the Temuco city, (official monitoring station of Health Ministry). The meteorological data included temperature, relative humidity, and wind speed. Several indexes were generated, such as the Thermohygrometric (thi) and Steadman (si) indexes (Sanhueza et al., 2009). No other pollutants were analyzed because of the lack of data or many missing data during 2010.

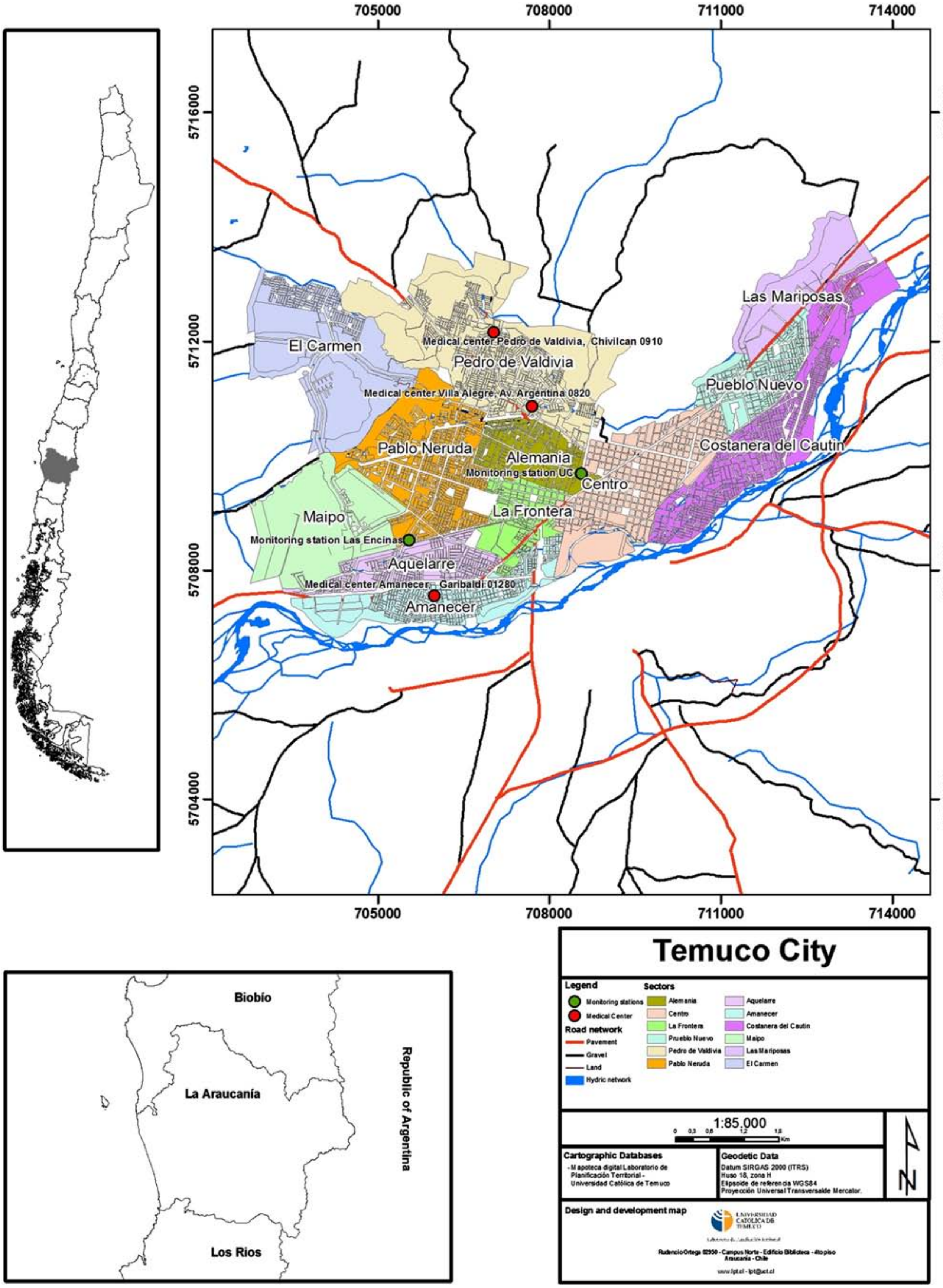

Fig. 1. Temuco urban area with the healthcare municipal centers and air monitoring station.

Table 1
Description of outpatient consultation codes.

Code	Outpatient visits
RespT	Total respiratory
RespG1	Respiratory group 1
RespG2	Respiratory group 2
RespG3	Respiratory group 3
OtherRespG1	Other causes respiratory causes group 1
OtherRespG3	Other causes respiratory causes group 3

The databases of the health and air quality time series were processed using an imputation analysis on missing data and outliers assessment. Then metric of the variables studied were built, including distribution analysis, time series, seasonality, maximum, minimum, interquartile, among others. The result of this first stage was the structuring of a global daily database of all the variables shown in Table 2.

2.2. Statistical methodology and relative risks estimation

The APHEA2 protocol was used as guide for this study. The data was fitted using a multivariate semi-parametric Poisson regression model, controlling for trends, seasonality, and confounders, using generalized additive model (GAM) parameterization on R-Project, as shown in Eq. (1) (Dominici et al., 2005; Katsouyanni et al., 2001; Peng et al., 2006; Sanhueza et al., 2009). The average of lags 1, 2, 3, 4 and 5 was used and models were developed for the total population and 3 different age groups. Finally, the RR was estimated when the ultrafine particle interquartile (IQR) increased by 4.73 μg/m^3, Eq. (2).

$$\mathrm{Log}(u) = a_o + a_1 f(T, df) + \mathrm{as.factor(DOW)} + a_2 \sum\nolimits_{i=1}^{n} f(M_i, df) + \beta X + \varepsilon \qquad (1)$$

where:

u: Health effect, morbidity
a_i: Model adjustment coefficients
T: Trend variable
DOW: Day of the week
M_i: Meteorological Variables; temperature (t), relative humidity (h), wind speed (v), Thermohygrometric index (thi), and Steadman index (si).
X: Pollutant of concern (UFP)
β: Coefficient of the pollutant of concern
f: Function obtained from semi-parametric analysis. In the case of the Trend, a cubic spline soft functions were used, with $df = 7$ degrees of freedom per year. In the case of meteorological variables, natural cubic spline functions were used, with $df = 4$ degrees of freedom for temperature, and $df = 2$ degrees of freedom for the other meteorological variables.
ε: Error term

The RR is the risk increase associated with a rise in UFP levels compared to baseline, and it is obtained as follows.

$$\mathrm{RR} = \mathrm{Exp}(\beta \times \Delta \mathrm{UFP}) \qquad (2)$$

3. Results and discussion

3.1. Descriptive statistics of ultrafine particles

Fig. 2 shows a time series of UFP concentrations in the study period (August 20, 2009 to June 30, 2011). From the figure, seasonality and trends in the time series, which coincide with the behavior of 24-h for $PM_{2.5}$ and PM_{10} could be identified. From this figure, higher concentration averages between those of 10 and 26 μg/m^3, as an average of 24 h could also be identified. These data concur with the seasonality of the daily concentrations of PM_{10} and $PM_{2.5}$ registered by the Las Encinas station during the years analyzed.

3.2. General characterization of the population in Temuco

Table 3 presents the distribution among age groups in the population of Temuco. Of these groups, it is noteworthy that the population under 5 years (G1) presents a stabilization tendency, while the 65 and over group (G3) shows an increment of 48.7% for 2011 with respect to 2001, which is consistent with the demographic transition in Chile, which is one of the most aged countries in Latin America. Considering that from the year 2009 to 2011 (during the study period), there exists an increase of 1.8% in the under 5 group, and 8.8% in the group over 65 years. This reflects that the population of Temuco projects a growing tendency in the number of aged persons (over 65) in relation to the growth of the population under 5 years.

3.3. Data analysis of morbidity in health centers

Table 4 shows the distribution of the number of respiratory primary care admissions of three public health services in the community of Temuco. Upon analyzing the number of cases for the entire 3 years, the highest number corresponded to the respiratory-type causes (RespT), which represents 30% of the total visits from these three health centers. The respiratory causes of Group 1 are the most frequent, and represent 50.1% of the total respiratory causes, while the causes of Group 2 (G2) represent 41.7% of the total respiratory causes. For Group 3 (≥65), the cases represent 9.2% of the total respiratory causes

Table 2
Descriptive statistics of the analyzed variables.

Variable	Units	Min	Mean	Max	IQR
UFP	μg/m^3	1.62	8.00	25.81	4.73
v (wind speed)	m/s	0.00	1.65	10.00	0.97
h (relative humidity)	%	14.0	81.3	100.0	14.03
t (temperature)	°C	−3.4	11.5	35.6	5.32
Respiratory outpatient					
Visits					
All ages	Outpatient visits/day	0	68	168	52
<5	Outpatient visits/day	0	29	74	23
5 ≤ persons <64	Outpatient visits/day	0	33	101	28
≥65	Outpatient visits/day	0	6	30	6
Other respiratory outpatient visits					
<5	Outpatient visits/day	0	19	56	15
≥65	Outpatient visits/day	0	5	30	5

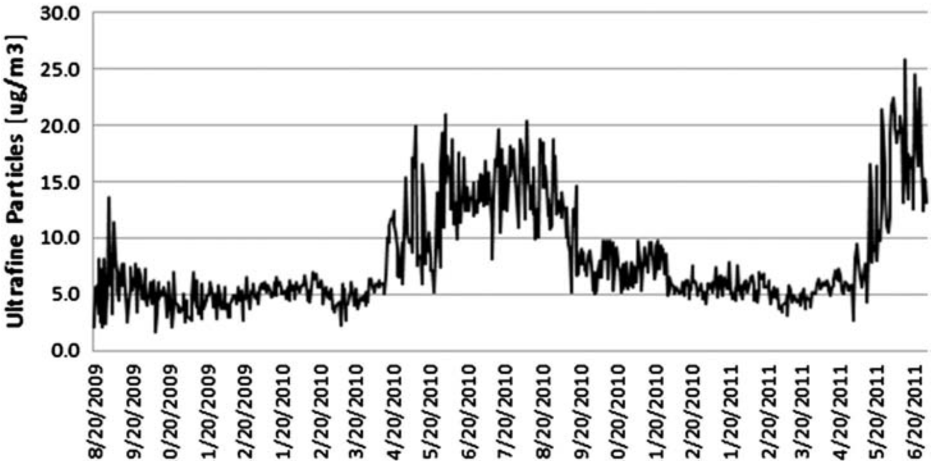

Fig. 2. Daily UFP concentration at the UCT monitoring station of Temuco from August the 20th of 2009 to June the 30th of 2011.

only. The visits for the category of "other respiratory diseases" reached 9.5% of all of the visits, taking into account just the susceptible groups G1 and G3. Upon analysis the temporal tendencies in the visits for total respiratory causes, in G1 and G3, it is concluded that there exists clear seasonality.

3.4. Times series of respiratory causes in health centers and exposure to UFP

Fig. 3 shows the times series of the total respiratory causes of the health centers and the daily UFP concentrations. The time series show clear seasonality, observing a relationship between the total number of cases and the concentrations of UFP. In particular, the period in which these relationships are seen corresponds to the months between March and September, which has the largest number of cases of respiratory origin, as well as the highest concentrations of UFP. This behavior was also seen in an older study made with this group of research (Sanhueza et al., 2009). These values coincide with the daily peaks of $PM_{2.5}$ concentrations registered in the city.

3.5. Relative risks of morbidity due to UFP exposure

Table 5 shows the results of the β parameter of the Poisson equation. The RR included confidence intervals of 95% (ICI and ICS), Standard Error (SE), lags, and the confounding variables that form part of the models for the causes analyzed. From the results obtained, it was possible to obtain what might be a significant RR, based on the statistical analysis, for the total respiratory-type causes and for the age groups G2 and G3. Additionally, important correlations were obtained for the health center visits associated with other respiratory-type causes for Groups G1 and G3. For the total respiratory causes (RespT) and for G2, (RespG2), the RR values were 1.0356 [95% CI (1.0095–1.0624)] and 1.0520 [95% CI (1.0131–1.0924)], respectively for the same day of exposure when the UFP was increased by IQR equal to 4.73 μg/m^3. These risks increase consecutively until four days after the exposure (lag 4), reaching the risk of the respiratory causes of G2 to be 58% higher than the risk of this cause in the total population with values of 1.0690 [95% CI (1.0413–1.0975)] and 1.1102 [95% CI (1.0680–1.1541)] for RespT and RespG2, respectively. However, for G3 (RespG3), the RR was 1.1458 [95% CI (1.0497–1.2507)] with a lag of 5 days (lag5), which the UFP increased by IQR equal to 4.73 μg/m^3. This risk was the most elevated in this study, indicating that there is a higher risk associated a person over the age of 65 to be exposed to UFP.

On the other hand, it might be a significant risk, based on the statistical analysis, for the classification of "Other Respiratory Causes", for which, RR of 1.0535 [95% IC (1.0025–1.1072)] (lag4) and of 1.1374 [95% IC (1.0360–1.2488)] (lag5) was obtained for those aged less than 5 years and those over 65, respectively. These results indicate that the risk is 151% higher in those aged over 65, than in those less than 5 years, when exposed to increments of 4.73 μg/m^3 of UFP IQR. Although the RR result was higher for G3 in both causes, (REspT and OtherResp), the confidence interval result was more ample in these cases, which indicates a larger uncertainty associated with the calculations obtained. This uncertainty could be related to the fact that the daily health and UFP data embarked a 22 month period, which is sufficient to estimate relative risks with semi-parametric Poisson models, and GAM techniques. Given longer period time of study, it would be possible to decrease this uncertainty.

In Fig. 4, it is observed that the associations are higher for a delay than on the same day of the exposure. For 3 days of lag, RR resulted in 1.0424 [95% IC (1.0159–1.0697)]. Lag 4 resulted in an RR of 1.0690 [95% IC (1.0413–1.0975)], while at lag 5 the RR was reduced to 1.0518 [95% IC (1.0245–1.0799)]. For the case of the respiratory causes in G2, the RR was 1.0393 [95% IC (1.007–1.0793)] with lag 2 of exposure. This RR was increased with lag 3 to 1.0582 [95% IC (1.0187–1.0993)].

Table 3
Population distribution by age group in Temuco, years 2001–2011.

Year	Total	Age group	
		<5	≥65
2001	255,594	21,655	16,950
2002	260,928	21,504	17,583
2003	266,207	21,342	18,230
2004	271,535	21,183	18,879
2005	276,883	21,037	19,529
2006	282,279	21,232	20,406
2007	287,711	21,429	21,333
2008	293,169	21,645	22,244
2009	298,575	21,846	23,132
2010	304,026	22,052	24,051
2011	309,354	22,249	25,170

Table 4
Classification of outpatient visit causes at Temuco, 2009–2011.

Clasification	2009	2010	2011	TOTAL (no cases)
	(SEP–DEC)	(JAN–DEC)	(JAN–JUN)	
RespT	5033	19,449	9030	33,512
RespG1	2522	7861	3849	14,232
RespG2	2101	9879	4260	16,240
RespG3	421	1735	919	3075
OtherRespG1	1851	5366	2309	9526
OtherRespG3	410	1572	720	2702

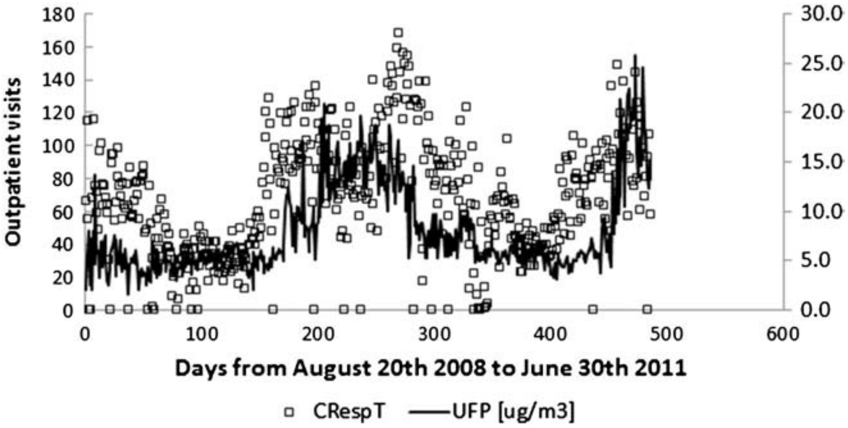

Fig. 3. Time series of total respiratory outpatient visits with UFP, in Temuco from August 20th 2009 to June 30th 2011.

With 4 days of lag, the RR increased to 1.1102 [95% IC (1.0680–1.1541)], while with a lag of 5 days, the RR was reduced to 1.0725 [95% IC (1.0315–1.1151)] with an IQR of 4.73 μg/m^3 increments in the concentrations of UFP. These results show novel findings regarding the relevance of daily UFP concentrations and the health risk, especially for the susceptible population in a wood smoke polluted urban area. There are some epidemiological studies that have linked the health effects and number of UFP instead of mass concentration; however, their relative risks are quite lower than ours. This could be explained because so far none of these studies considered the risks in a polluted city with wood smoke.

In effect, according to a case–crossover study with logistic regression developed in Rome by Belleudi et al. (2010), upon analyzing the hospitalizations for cardiac failure, it was found that there was an association with the number of UFP particles when they were increased to 9293 particles cm^{-3}, obtaining a risk increment of 1.5% [95% CI (0.4%–3.2%)]. Increases in chronic obstructive pulmonary disease (COPD) were positively related to UFP, obtaining a risk increment equal to 1.6% [95% CI (0.0%–3.2%)] (Belleudi et al., 2010). All results were obtained for the same day of exposure (lag 0) and all of effects were found, especially in patients over 65 years (Group 3, G3) (Belleudi et al., 2010).

Table 5
RR for outpatient visits at Temuco associated with IQR of UFP.

Effect	β	EE(β)	RR	ICI	ICS	Lag	Confoundents
RespT	0.00740	0.00276	1.0356	1.0095	1.0624	0[a]	t.2, h.3[b]
	−0.00318	0.00278	0.9851	0.9600	1.0108	1	t.2, h.3
	0.00160	0.00275	1.0076	0.9822	1.0336	2	t.2, h.3
	0.00879	0.00279	1.0424	1.0159	1.0697	3[a]	t.2, h.3
	0.01411	0.00284	1.0690	1.0413	1.0975	4[a]	t.2, h.3
	0.01068	0.00284	1.0518	1.0245	1.0799	5[a]	t.2, h.3
RespG1	0.00646	0.00422	1.0310	0.9915	1.0721	0	t.2, h.3
	−0.00038	0.00428	0.9982	0.9594	1.0386	1	t.2, h.3
	−0.00383	0.00424	0.9821	0.9442	1.0214	2	t.2, h.3
	0.00666	0.00431	1.0320	0.9916	1.0741	3	t.2, h.3
	0.00550	0.00437	1.0264	0.9856	1.0688	4	t.2, h.3
	0.00396	0.00436	1.0189	0.9785	1.0609	5	t.2, h.3
RespG2	0.01071	0.00407	1.0520	1.0131	1.0924	0[a]	t.2, h
	−0.00372	0.00412	0.9826	0.9458	1.0208	1	t.2, h
	0.00814	0.00408	1.0393	1.0007	1.0793	2[a]	t.2, h
	0.01196	0.00411	1.0582	1.0187	1.0993	3[a]	t.2, h
	0.02210	0.00418	1.1102	1.0680	1.1541	4[a]	t.2, h
	0.01479	0.00420	1.0725	1.0315	1.1151	5[a]	t.2, h
RespG3	0.00377	0.00896	1.0180	0.9368	1.1062	0	t.2
	−0.00927	0.00913	0.9571	0.8795	1.0416	1	t.2
	−0.00817	0.00917	0.9621	0.8836	1.0475	2	t.2
	0.00100	0.00924	1.0047	0.9223	1.0946	3	t.2
	0.01414	0.00942	1.0692	0.9798	1.1667	4	t.2
	0.02877	0.00945	1.1458	1.0497	1.2507	5[a]	t.2
OtherRespG1	0.00422	0.00521	1.0201	0.9720	1.0707	0	t.1, h.5, thi.1, si.2
	−0.00640	0.00523	0.9702	0.9242	1.0184	1	t.1, h.5, thi.1, si.2
	0.00028	0.00525	1.0013	0.9538	1.0512	2	t.1, h.5, thi.1, si.2
	0.00537	0.00524	1.0257	0.9771	1.0768	3	t.1, h.5, thi.1, si.2
	0.01103	0.00536	1.0535	1.0025	1.1072	4[a]	t.1, h.5, thi.1, si.2
	0.00033	0.00539	1.0015	0.9527	1.0528	5	t.1, h.5, thi.1, si.2
OtherRespG3	−0.00044	0.00966	0.9979	0.9124	1.0914	0	t.1
	−0.01645	0.00976	0.9252	0.8451	1.0128	1	t.1
	−0.01178	0.00986	0.9458	0.8632	1.0364	2	t.1
	−0.00178	0.00994	0.9916	0.9043	1.0873	3	t.1
	0.00787	0.01011	1.0379	0.9451	1.1400	4	t.1
	0.02722	0.01008	1.1374	1.0360	1.2488	5[a]	t.1

[a] Significant, $p < 0.001$.

[b] The number after the dot is the number of lag.

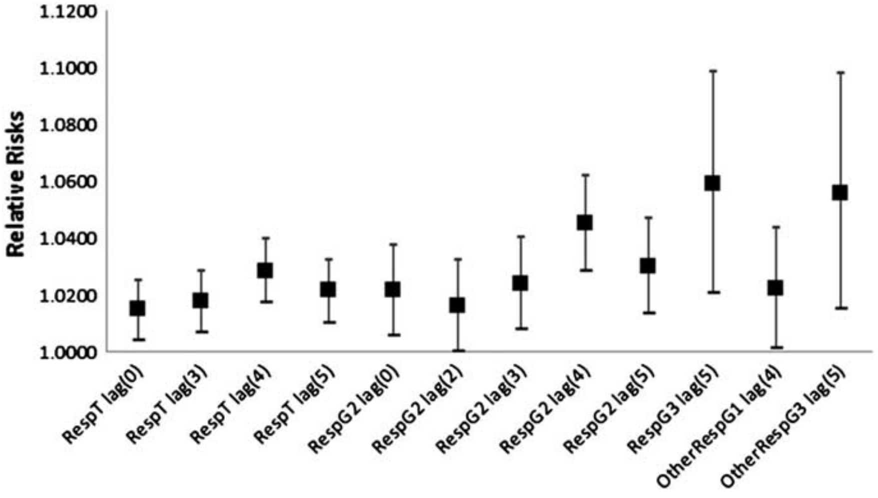

Fig. 4. Relative risks for respiratory diseases due to daily exposure to UFP at Temuco, August 20th 2009 to June 30th 2011, when the IQR of UFP increases 4.73 μg/m^3.

Studies carried out in Copenhagen, Denmark, (Andersen et al., 2010) through cross-correlation and using regression logistics, found an association between the exposure to ultrafine particulate matter and cerebral ischemic disease. In particular, it was found that 4 days after the interquartile range the increase of 3918 UFP cm^{-3} resulted in risk increase (Odds Ratio) of 1.14 [95% CI (1.04–1.25)] in the hospital admissions because of severe stroke. A short term study was conducted using a time-series Poisson regression model which found a positive relation in the exposure to the number of UFP and the increment in hospital admissions for respiratory problems in seniors over the age of 65, with a relative risk (RR) of 1.04 [95% IC (1.00–1.07)] for an interquartile range increase of 3907 UFP cm^{-3} (Andersen et al., 2008).

In a study in Beijing, China, (Leitte et al., 2011), a time series analysis was developed using a generalized additive Poisson time series regression. This study showed an increase of 5% [RR = 1.05 (95% IC, 1.01–1.08)], one day after the exposure (lag 1), in the emergency room for respiratory difficulties associated to the exposure of 3600 particles per cm^{-3} of ultrafine particulate matter between 0.05 and 0.1 μm. The results of this study are consistent with the results of other studies reporting that levels of PM$_{10}$, PM$_{2.5}$ and UFP are associated with short term increases in emergency visits for respiratory causes (Knol et al., 2009; Leitte et al., 2012; Stieb et al., 2009).

Recently, the number of hospital visits for cardiovascular and UFP exposure was observed between March of 2004 and December of 2006 in Beijing (Liu et al., 2013). Using a Poisson regression, with control of confounding variables, the risk was estimated to be associated to particles in a range of 3–100 nm. The authors found a significant risk of 7.2% [95% IC (1.1–13.7%)] for the total emergency visits for cardiovascular problems for an interquartile range increase (IQR) of 9040 cm^{-3} in 11 days average number of UFP concentration.

The exposure to UFP has also been associated to causes of death. A study by Stoelzel et al. (2007) in Erfurt, Germany, between 1995 and 2001 found significant statistics between the high level of UFP and death caused by cardio-respiratory illnesses with a lag of 4 days (Stoelzel et al., 2007). Using a Poisson regression and control of confounding variables, the authors found a relative risk of 1.029 [95% IC (1.003–1.055)] for total mortality with an increment of 9748 cm^{-3} of UFP between 0.01 and 0.1 μm. For the case of mortality by cardio-respiratory causes, a RR of 1.031 [95% IC (1.003–1.060)] was found. It was shown that UFP, represented by particles of combustion, can be an important component of urban air pollution associated with health effects.

An important study carried out by Breitner et al. (2009) found that the accumulative exposure to UFP (0.01–0.1 μm) was associated with the increase in mortality in Erfurt, Germany. An increase in 15 days of the measure of UFP of 7649 cm^{-3} was associated with an RR of 1.060 [95% IC (1.008–1.114)] (Breitner et al., 2009). In another study by Breitner et al. (2011), associations were shown between death by cardiovascular causes, ischemic cardiac disease, and UFP with a lag of 2 days in Beijing, China, between March of 2004 and August 2005 (Breitner et al., 2011). Using a Poisson semi-parametric regression model with control of confounding variables, the authors concluded that there was a significant relationship between particles in the size range of 0.03–0.1 μm, and an increase of daily mortality by cardiac ischemia of 7.1% [95% CI (2.9%–11.5%)] and an increase of daily death by cardiovascular disease of 4.0% [95% CI (1.2%–7.0%)], when the UFP increased in 6250 cm^{-3}. It is noteworthy that none of the mentioned studies considered an urban area contaminated with wood smoke.

These results reinforce those found in Copenhagen, Rome, and Beijing (Andersen et al., 2010; Belleudi et al., 2010; Leitte et al., 2011; Liu et al., 2013), where the highest RR for hospital admissions and emergencies were for the group of aged individuals when they were exposed to UFP measured as the number of particles. In this study, the concentrations were measured in μg/m^3. Noteworthy is that for G1, RR was not found for total respiratory causes. This result could be attributed to the fact that viral diseases and respiratory bacteria are more frequent among this group, complicating the results of relative risk for the daily exposure to UFP. This coincides with studies carried out in Asia and Europe (Andersen et al., 2010; Belleudi et al., 2010; Branis et al., 2010; Liu et al., 2013), where the RR are not significant for this susceptible group.

Our RR were higher for the elderly population, which is a susceptible group of concern, mainly for specific respiratory diseases, agreeing with the results obtained by Belleudi et al. (2010), in which an association was found between the UFP respiratory causes and age of those over 65 years old. These findings were obtained for obstructive chronic respiratory disease (COPD) for the same day of the exposure. These present results are also in concordance with those found in Bejing, China by Leitte et al. (2011), in which increments of 5% (RR = 1.05) were shown for the exposure to UFP, understood as between 0.1 and 0.3 μm, associated with respiratory cases in the emergency room.

We believe that our results have clear implications for the Chilean policy on air quality for those cities with residential wood combustion. Specifically, they suggest that the current 24-h daily limit value for PM$_{2.5}$ in Chile is not sufficient to protect the population from short term effects, and a stronger PM$_{2.5}$ standard must be analyzed.

4. Conclusions

Significant associations were found between daily medical consultations in Temuco for respiratory diseases and daily ultrafine particles, mainly in elder populations, based on the performed statistical analysis. The relative risk for respiratory medical consultations of persons between 5 and 65 years old increases with exposure lag when ultrafine particles increase by 4.73 $\mu g/m^3$. The results suggest that residential wood combustion may be responsible for increased morbidity in that urban area. This has an important implication in relation with the size and chemical composition of the particles, but also for monitoring and control strategies. Policy-makers should take into account these aspects along with economic considerations if protection of the public health is one of their major concerns.

Although this study was focused on UFP sampled with a MOUDI, further analysis should be done using size-segregated particle number concentration sampling when funding resources are sufficient, and this fraction has been shown to have the most serious effects on human health. The latest investigations in Europe and Asia focus on ultrafine particles ($\leq$0.1 μm) as the size particle of concern. Our results show novel findings regarding the relevance of daily ambient UFP exposure and health risk, especially for susceptible population in a wood smoke polluted city.

Acknowledgments

We acknowledge the FONIS Project No SA08I20055 for supporting this study.

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.envint.2014.01.017.

References

Andersen ZJ, Wahlin P, Raaschou-Nielsen O, Ketzel M, Scheike T, Loft S. 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 2008;65:458–66.

Andersen ZJ, Olsen TS, Andersen KK, Loft S, Ketzel M, Raaschou-Nielsen O. Association between short-term exposure to ultrafine particles and hospital admissions for stroke in Copenhagen, Denmark. Eur Heart J 2010;31:2034–40.

Belleudi V, Faustini A, Stafoggia M, Cattani G, Marconi A, Perucci CA, et al. Impact of fine and ultrafine particles on emergency hospital admissions for cardiac and respiratory diseases. Epidemiology 2010;21:414–23.

Branis M, Vyskovska J, Maly M, Hovorka J. Association of size-resolved number concentrations of particulate matter with cardiovascular and respiratory hospital admissions and mortality in Prague, Czech Republic. Inhal Toxicol 2010;22:21–8.

Breitner S, Stölzel M, Cyrys J, Pitz M. Short-term mortality rates during a decade of improved air quality in Erfurt, Germany. Environ Health Perspect 2009;117:448–54.

Breitner S, Liu LQ, Cyrys J, Bruske I, Franck U, Schlink U, et al. Sub-micrometer particulate air pollution and cardiovascular mortality in Beijing, China. Sci Total Environ 2011;409:5196–204.

Brook RD, Rajagopalan S, Pope CA, Brook JR, Bhatnagar A, Diez-Roux AV, Holguin F, Hong YL, Luepker RV, Mittleman MA, Peters A, Siscovick D, Smith SC, Whitsel L, Kaufman JD, Epidemiol AHAC, Dis CKC, Metab CNPA. Particulate Matter Air Pollution and Cardiovascular Disease An Update to the Scientific Statement From the American Heart Association. Circulation. 2010;121:2331–78.

Chow JC, Watson J, U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards. Guideline on speciated particulate monitoring; 1998.

Cormier SA, Lomnicki S, Backes W, Dellinger B. Origin and health impacts of emissions of toxic by-products and fine particles from combustion and thermal treatment of hazardous wastes and materials. Environ Health Perspect 2006;114:810–7.

Diaz-Robles LA, Ortega JC, Fu JS, Reed GD, Chow JC, Watson JG, et al. A hybrid ARIMA and artificial neural networks model to forecast particulate matter in urban areas: the case of Temuco, Chile. Atmos Environ 2008;42:8331–40.

Diaz-Robles LA, Fu JS, Reed GD. Emission scenarios and the health risks posed by priority mobile air toxics in an urban to regional area: an application in Nashville, Tennessee. Aerosol Air Qual Res 2013;13:795–803.

Dominici F, McDermott A, Daniels M, Zeger SL, Samet JM. Revised analyses of the National Morbidity, Mortality, and Air Pollution Study: mortality among residents of 90 cities. J Toxicol Environ Health A 2005;68:1071–92.

Halonen J, Lanki T, Yli-Tuomi T, Kulmala M, Tiittanen P, Pekkanen J. Urban air pollution, and asthma and COPD hospital emergency room visits. Thorax 2008;63:635–41.

Hertel S, Viehmann A, Moebus S, Mann K, Brocker-Preuss M, Mohlenkamp S, et al. Influence of short-term exposure to ultrafine and fine particles on systemic inflammation. Eur J Epidemiol 2010;25:581–92.

Kampa M, Castanas E. Human health effects of air pollution. Environ Pollut 2008;151:362–7.

Katsouyanni K, Touloumi G, Samoli E, Gryparis A, Le Tertre A, Monopolis Y, et al. 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 2001;12:521–31.

Khlystov A, Stanier C, Pandis SN. An algorithm for combining electrical mobility and aerodynamic size distributions data when measuring ambient aerosol. Aerosol Sci Tech 2004;38:229–38.

Klippel N, Nussbaumer T. Health relevance of particles from wood combustion in comparison to Diesel soot. 15th European Biomass Conference and Exhibition; 2007. [Berlin].

Knol A, Hartog J, Boogaard H, Slottje P, Van der Sluijs J, Lebret E. Expert elicitation on ultrafine particles: likelihood of health effects and causal pathways. Part Fibre Toxicol 2009;24:19.

Laumbach RJ, Kipen HM. Respiratory health effects of air pollution: update on biomass smoke and traffic pollution. J Allergy Clin Immunol 2012;129:3–13.

Leitte AM, Schlink U, Herbarth O, Wiedensohler A, Pan XC, Hu M, et al. Size-segregated particle number concentrations and respiratory emergency room visits in Beijing, China. Environ Health Perspect 2011;119:508–13.

Leitte AM, Schlink U, Herbarth O, Wiedensohler A, Pan XC, Hu M, et al. Associations between size-segregated particle number concentrations and respiratory mortality in Beijing, China. Int J Environ Health Res 2012;22:119–33.

Liu LQ, Breitner S, Schneider A, Cyrys J, Bruske I, Franck U, et al. Size-fractioned particulate air pollution and cardiovascular emergency room visits in Beijing, China. Environ Res 2013;121:52–63.

McDonald JD, Zielinska B, Fujita EM, Sagebiel JC, Chow JC, Watson JG. Fine particle and gaseous emission rates from residential wood combustion. Environ Sci Technol 2000;34:2080–91.

Morawska L, Moore M, Ristovski Z. Desktop literature review and analysis: health impacts of ultrafine particles. For the Australian Department of the Environment and Heritage; 20041–207.

Nemmar A, Hoylaerts MF, Hoet PHM, Dinsdale D, Smith T, Xu HY, et al. Ultrafine particles affect experimental thrombosis in an in vivo hamster model. Am J Respir Crit Care Med 2002;166:998–1004.

Oberdörster G. Pulmonary effects of inhaled ultrafine particles. Int Arch Occup Environ Health 2001;74:1–8.

Oberdörster G, Oberdörster E, Oberdörster J. Nanotoxicology: an emerging discipline evolving from studies of ultrafine particles. Environ Health Perspect 2005;113:823–39.

Peng RD, Dominici F, Louis TA. Model choice in time series studies of air pollution and mortality. J R Stat Soc Ser A 2006;169:179–98.

Ralph J, Delfino CS, Malik Shaista. Potential role of ultrafine particles in associations between airborne particle mass and cardiovascular health. Environ Health Perspect 2005;113:934–46.

Sanhueza P, Vargas C, Mellado P. Impact of air pollution by fine particulate matter (PM10) on daily mortality in Temuco, Chile. Rev Med Chil 2006;134:754–61.

Sanhueza PA, Torreblanca MA, Diaz-Robles LA, Schiappacasse LN, Silva MP, Astelle TD. 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 2009;59:1481–8.

Seaton A, MacNee W, Donaldson K, Godden D. Particulate air pollution and acute health effect. Lancet 1995;345.

SINCA. Sistema Nacional de Calidad del Aire. Chile: Ministerio del Medio Ambiente; 2013.

Sioutas C, Delfino RJ, Singh M. Exposure assessment for atmospheric ultrafine particles (UFPs) and implications in epidemiologic research. Environ Health Perspect 2005;113:947–55.

Stewart JC, Chalupa DC, Devlin RB, Frasier LM, Huang LS, Little EL, et al. Vascular effects of ultrafine particles in persons with type 2 diabetes. Environ Health Perspect 2010;118:1692–8.

Stieb D, Szyszkowicz M, Rowe B, Leech J. Air pollution and emergency department visits for cardiac and respiratory conditions: a multi-city time-series analysis. Environ Health Perspect 2009;8.

Stoelzel M, Breitner S, Cyrys J, Pitz M, Woelke G, Kreyling W, et al. Daily mortality and particulate matter in different size classes in Erfurt, Germany. J Expo Sci Environ Epidemiol 2007;17:458–67.

Tsapakis M, Lagoudaki E, Stephanou EG, Kavouras IG, Koutrakis P, Oyola P, et al. The composition and sources of PM2.5 organic aerosol in two urban areas of Chile. Atmos Environ 2002;36:3851–63.

Wichmann HE. Diesel exhaust particles. Inhal Toxicol 2007;19:241–4.