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# Cancer Burden Disease Attributable to $PM_{2.5}$ and Health Risk by $PM_{2.5}$-bound Toxic Species in Two Urban Chilean Municipalities

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## ABSTRACT

This study aimed to estimate the environmental cancer disease burden in adults attributable to fine particulate matter ($PM_{2.5}$) exposure using Ostro's function methodology, and health risk indexes for particle-bound toxic chemicals through hazard quotients (*HQ, HI*) and carcinogenic risk (*CR, CRI*) indexes from EPA guidelines, of two urban Chilean Municipalities: Coyhaique and Independencia. Quantification of chemical species (OC, EC, metals, and PAHs) was done at the Lamont-Doherty Earth Observatory of Columbia University, USA. Modern carbon in OC and EC analysis showed that the principal source of $PM_{2.5}$ emission in Coyhaique was firewood burning compared with Independencia. The total PAHs and B[a]P concentrations were 6.3 and 8.9 times higher in Coyhaique than in Independencia. In contrast, As and Pb levels were significantly greater in Independencia. The *HI* was 14.5 and 2.37 times the limit considered acceptable ($HI > 1$) in Coyhaique and Independencia, explained 92.45% by B[a]P and 66.99% by As, respectively. *CRI* exceeded the threshold ($1 \times 10^{-6}$) in Coyhaique and Independencia, explained by As (75.38%) plus B[a]P (20.30%) and As (97.01%). The attributable fraction (*AF*) of deaths due to lung cancer from long-term exposure to $PM_{2.5}$ reached 54% (95% CI: 25–72) in Coyhaique vs. 43% (95% CI: 19–46) in Independencia. The *AF* for cardiopulmonary cancer were 40% (95% CI: 17–57) and 32% (95% CI: 12–46), respectively. A relevant fraction of the cancer cases and potential expected adverse effects would be attributable to long-term exposure to $PM_{2.5}$ and the presence of chemical compounds bound to the particles. These results deserve further study to help guide policy in different environments, mainly carcinogenic $PM_{2.5}$-bound toxic species from other emission sources, particularly firewood burning.

**Keywords:** $PM_{2.5}$, Health risk assessment, Burden disease, Attributable fraction, Toxic metals, PAHs

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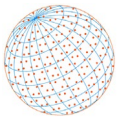
## 1 INTRODUCTION

Air pollutants, including particulate matter (PM), have become a global environmental issue over several decades (Li *et al.*, 2019a; Molina, 2008). Global burden of disease analysis lists particulate matter as the 5$^{th}$ highest cause of mortality among all health risks, leading to 4.2 million premature deaths in 2015 (Cohen *et al.*, 2017). Many studies have demonstrated that exposure to fine particulate matter $PM_{2.5}$ (particulate matter with aerodynamic diameter $\leq$ 2.5 μm) increases the risk of respiratory and cardiovascular diseases (Anderson *et al.*, 2012; Dockery, 2009; Liu *et al.*, 2019). Recently, extensive studies have reported a clear and robust association with oral, rectal, liver, skin, breast, gastric and kidney cancers due to exposure to $PM_{2.5}$ and its metallic components, especially the sulfur fraction (Weinmayr *et al.*, 2018). Air pollution by fine particles is a problem widely observed in cities which experienced rapid economic development (de Jesus *et al.*, 2019). Examples include the London haze in the 1950s, Los Angeles smog in the 1960s and Beijing haze in recent decades, where industrial combustion, vehicle exhausts and secondary aerosol are considered the primary sources of $PM_{2.5}$ (Li *et al.*, 2019b; Wang *et al.*, 2016). Chile, located in South America, has suffered severe air pollution since 1990. Since then, Chile's government has implemented a series of policies to control air pollution. As a result, $PM_{10,}$ sulfur, and nitrogen pollution have decreased significantly (Mazzeo *et al.*, 2018). However, $PM_{2.5}$ remains a problem in central and southern Chile, especially for using wood as a source of energy for house heating and cooking (Díaz-Robles *et al.*, 2014). Currently, 12 of the 15 administrative regions of Chile are decreed as areas with elevated PM levels, and 14 decontamination plans are under development (MMA, 2014).

Previous studies in Chilean cities have reported regional and seasonal variation in estimated short-term health effects of different size distributions of PM, including $PM_{2.5}$ (Boneberger *et al.*, 2011; Díaz-Robles *et al.*, 2014; Rivas *et al.*, 2008; Sanhueza *et al.*, 2009). The cities of Santiago and Coyhaique are experiencing the most severe air pollution problem. Santiago, the capital of Chile, located in the central zone, is highly populated, with more than 50% of the country's population living in the metropolitan area. The geography and climate of the Santiago basin are generally unfavorable for the diffusion of air pollutants. Consequently, extreme high pollution events occur frequently during the winter season (Bell *et al.*, 2011, 2009; Toro-Araya *et al.*, 2014). It is estimated that the transport sector in Santiago explains about 25% of $CO_2$ emissions and 40% of air pollution by fine particulate matter, also showing high and increasing levels of nitrogen dioxide ($NO_2$) above 28% in the decade 2004–2014 (Barraza *et al.*, 2017; Duncan *et al.*, 2016; Gallardo *et al.*, 2018). Villalobos and collaborators determined that approximately 70% of the mass of $PM_{2.5}$ is organic matter in the winter season. The primary source of contribution is wood smoke (almost 60%), followed in decreasing order by diesel, gasoline and natural gas emissions (Villalobos *et al.*, 2015). In contrast, in Coyhaique, the southern city most polluted by $PM_{2.5}$ in the American region, the primary source of emission (99%) of $PM_{2.5}$ is residential firewood combustion for cooking and heating (Perez *et al.*, 2020). Various studies in developed countries indicate that wood smoke is the most significant source of exposure to particles during the winter months, originating from residential stoves (Maykut *et al.*, 2003; Naeher *et al.*, 2007). It is known that, depending on the quality of combustion, the type and characteristics of the firewood used, a series of compounds that are very harmful to health can be emitted that vary spatially and seasonally, which will determine the potential adverse health effects (Bell *et al.*, 2011, 2009; Toro-Araya *et al.*, 2014). Among the compounds adsorbed to the particles are the Polycyclic Aromatic Hydrocarbons (PAHs) and some heavy metals that are particularly dangerous due to toxicity and their known carcinogenic adverse effects (Scipioni *et al.*, 2012). These are involved in cellular damage and the subsequent inflammatory response to the lung and cardiovascular system (Chang *et al.*, 2019; Diociaiuti *et al.*, 2001; Li *et al.*, 2003; Maykut *et al.*, 2003).

The characterization of public health risk of populations due to exposure to certain environmental agents is essential when risk control and management strategies are to be carried out. Estimation of the health risk due to exposure to air pollutants has generally focused on the use of the mass of particles as a metric ($PM_{2.5}$ or $PM_{10}$) to evaluate the possible impacts on health. This has allowed estimating the disease burden and through this generating different public policies, aimed at reducing concentrations to protect the population. Regardless of the achievements that



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these policies adopted and adapted to the reality of the countries have produced in various parts of the world, one of the most relevant limitations of this metric is that it does not consider the complexity of the chemical composition of the particles, which will vary substantially locally, since the sources that contribute to PM concentrations can be very diverse (Salimbene *et al.*, 2021). Several authors consider that in addition to using mass as a risk assessment metric, the use of indicators such as black carbon (BC), polycyclic aromatic hydrocarbons (PAHs) and metals should be explored as possible metrics (Chowdhury *et al.*, 2022; Krzyzanowski *et al.*, 2014; Grahame *et al.*, 2014; Peng *et al.*, 2013; Sosa *et al.*, 2017), especially in urban areas where quality of the air is dominated by combustion processes with different geographic, topographic, meteorological and social factors, as occurs in the cities of south-central Chile, where the use of firewood for cooking and heating is very intensive (Torres *et al.*, 2021; Salimbene *et al.*, 2021). There are still few studies carried out in Chile that have reported the association between the components of $PM_{2.5}$ and health effects. Elements such as K, S, Se, V, Cr, Cu, Zn, EC (Elemental Carbon) have been associated with various effects such as respiratory, cardiovascular, and cerebrovascular diseases, both in children and adults (Prieto-Parra *et al.*, 2017; Valdés *et al.*, 2012; Cakmak *et al.*, 2009; Cáceres *et al.*, 2021).

Considering this background, we carried out an exploratory study whose objective is to characterize the health risk due to exposure to fine particulate matter and its components in two urban municipalities, using two strategies, estimation of potentially carcinogenic and non-carcinogenic effects through risk assessment, and estimating the burden of cancer disease attributable to outdoor $PM_{2.5}$ exposure. The characterization and comparison of these risk assessments will allow us to understand the sources of $PM_{2.5}$ emission for local actions of environmental and public health.

## 2 METHODS

### 2.1 Urban Municipalities Studied

Coyhaique Municipality (hereafter Coyhaique) is (Fig. 1) located in Chilean Patagonia (45°34'S, 72°04'W), extreme southern Chile with about 58,000 inhabitants, spanning an area of 7290,2 $km^2$, where the urban area is only 7 $km^2$ (Azócar-García *et al.*, 2010). It is surrounded by rivers (Simpson and Coyhaique) and mountains (Mackay, Sombrero, Negro, San Martin), snow-covered throughout the year. Independence Municipality (hereafter Independencia) is located in the north of the metropolitan area of Santiago, with about 100,000 inhabitants in with an area of 7 $km^2$ (BCN, 2021a, 2021b). Santiago is located in a basin between the Andes Range and the Coast Range (33°27'S, 70°40'W). In both localities, the topographic configuration of the basin contributes to the low vertical dispersion of air pollutants during winter, resulting in rapid increases of particulate matter, a phenomenon known as atmospheric blocking (Perez *et al.*, 2020; Yun and Yoo, 2019).

### 2.2 $PM_{2.5}$ Monitoring and Data Analysis

The hourly data concentrations and the time series of $PM_{2.5}$ for this study were obtained from the Air Quality Monitoring Stations (AQMS); the AQMS/COY1 located in the center of Coyhaique, and the AQMS/IND located in Independencia close to the Hospital Psiquiatrico of the Universidad de Chile (Fig. 1) (SINCA, 2022). These AQMS continuously measure $PM_{2.5}$ in the ambient air using specific sensors, reporting the average concentration ($\mu g\ m^{-3}$) for each hour of the day. Each measure of the AQMS has a representativeness area of around 3 $km^2$. For each AQMS, the historical record of hourly concentrations of $PM_{2.5}$ spanned five years between 01/01/2013 and 31/12/2017. The descriptive and graphic exploratory analysis of temporal data of $PM_{2.5}$ was done using the R package *openair* (Carslaw and Ropkins, 2012).

### 2.3 Sampling of Outdoor $PM_{2.5}$

The outdoor samples of $PM_{2.5}$ for chemical analysis were collected in the area of representativeness of the AQMS. Twenty samples of $PM_{2.5}$ were collected, fifteen on Teflon (37 mm and 47 mm) and five on quartz (47 mm) filters. These were pre-weighed and environmentally conditioned for their use at Lamont-Doherty Earth Observatory of Columbia University. These samples were collected during a week in wintertime using two monitor samplers (44XR Universal Sample Pump, SKC Inc., and MiniVol TAS, Air Metrics Corp) between August 24 and 29, 2016, in Coyhaique, and September 13 to 16, 2016, in Independencia. Both monitor samplers were calibrated daily before use and

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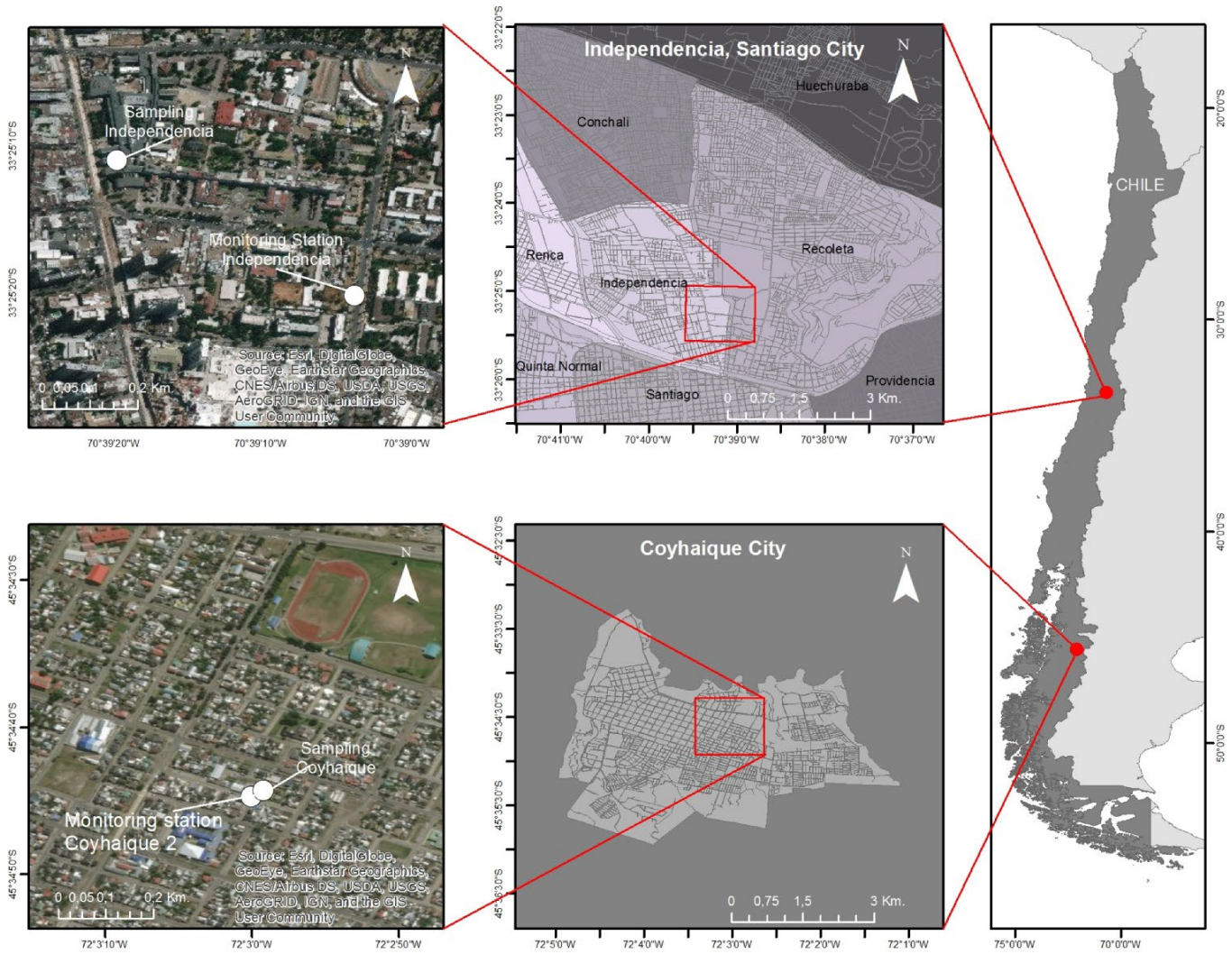


**Fig. 1.** Location of the AQMS and $PM_{2.5}$ sampling sites in Coyhaique and North of Santiago (Independencia neighborhood).

operated with a stable flow rate of 4 L $min^{-1}$. The sample collection for both kinds of monitors was programmed for 24 h, from 08:00 AM to 08:00 AM the next day. After sampling, all the filters were post-weighed to calculate the $PM_{2.5}$ concentration for each sample.

## 2.4 Chemical Analysis of $PM_{2.5}$

### 2.4.1 PAH analysis on Teflon filters

The Teflon parts of filters were cut and mixed with diatomite and copper powder. The samples were spiked with surrogate standard to monitor the recovery of the procedure. The samples were subsequently extracted by DIONEX accelerated solvent extraction (ASE) with a mixture of 60 mL acetone/dichloromethane (1:1, v/v), then the extracts were purified by silica gel column. Finally, the extracts were converted to hexane and concentrated by gentle $N_2$ to about 1 mL. Before gas chromatography-mass spectrometry (GC-MS) analysis, the internal standards were added to the final extract to calculate the concentrations of PAHs. The parameters of GC-MS were as follows: the oven temperature started at 55°C for 2 min; then it was heated to 280°C at a rate of 20°C $min^{-1}$; finally, it was heated to 300°C at a rate of 3°C $min^{-1}$ and held for 4 min. The injection volume was 2 µL with a split-less mode, and the carrier was helium. Recovery of surrogate and internal standards ranged from 62.3% to 95.6% and from 71.7% to 98.2%, respectively. All PAH data reported in this study were corrected by recoveries. To differentiate PAHs from un-burning fossil fuel and combustion sources, compound-specific carbon stable isotope analysis of PAHs was conducted following a method described in the literature (Yan *et al.*, 2016).

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### 2.4.2 Organic carbon (OC), Elemental carbon (EC) and metals

Quartz filters were used for analysis of radioactive carbon ($^{14}C$) for calculating the percentage of modern carbon in organic carbon (OC) and elemental carbon (EC) was measured in the National Ocean Sciences Accelerator Mass Spectrometry (NOSAMS), Woods Hole Oceanographic Institute, USA (Zencak *et al.*, 2007). For samples less than 30 μgC, accurate amount of $CO_2$ from OX-I was added. The diluted samples were reduced to graphite and measured by AMS. The original $^{14}C$ of samples was calculated from measured value from the mixture using the dilution factor. Metals cadmium (Cd), nickel (Ni), lead (Pb), and arsenic (As) were measured by energy-dispersive X-ray fluorescence spectrometry on Teflon filters at Lamont-Doherty Earth Observatory of Columbia University.

## 2.5 Human Health Risk Assessment

### 2.5.1 Exposure concentration ($E_{xp}C$)

The inhalation dosimetry methodology was used to assess the potential human exposure (Asante-Duah, 2017; U.S. EPA, 2009, 1989). This estimates the exposure concentration ($E_{xp}C$) for each receptor exposed to contaminants via inhalation using Eq. (1):

$$E_{xp}C_{PM2.5}\left(\mu g\ m^{-3}\ day^{-1}\right) = \frac{CA \times ET \times ED \times EF}{AT} \quad (1)$$

*CA* is the contaminant concentration in air (μg $m^{-3}$), *ET* is the exposure time (h $day^{-1}$), *EF* is the exposure frequency (days $year^{-1}$), *ED* is the exposure duration (years) of the exposed subject, and *AT* is the time (days). According to the EPA, the estimation of the level of exposure is fundamental in calculating the health risks related to air pollutant exposure (Ostro, 2004); concentration and time of exposure to $PM_{2.5}$ are relevant parameters for this. Short-term exposures are transiently higher air pollution levels that might cause adverse health events, mainly respiratory and cardiovascular illness. Chilean air quality standards consider levels dangerous when the concentration of $PM_{2.5}$ exceeds the daily norm of 50 μg $m^{-3}$ (MMA, 2014). To estimate the *ET* parameter in Eq. (1), we use the average number of hours per day (Short Time Exposure: STE) that the $PM_{2.5}$ concentration exceeds the value of the daily norm in each city studied (Table 1).

### 2.5.2 Non-carcinogenic risk

The Hazard Quotient (*HQ*) was estimated to evaluate the potential non-carcinogenic risk. *HQ* is the ratio of the potential exposure to a substance to the level at which no adverse effects are expected. The *HQ* was estimated as the ratio between $E_{xp}C$ coefficient and the reference concentration *RfC* (mg $m^{-3}$ $day^{-1}$) for inhalation of a substance using Eq. (2). *RfC* is an estimate of a continuous inhalation exposure to the human population (including sensitive subgroups) that is likely to be without an appreciable risk of deleterious effects during a lifetime. *RfC* is derived from the NOAEL (no-observed-adverse-effect level) (U.S. EPA, 1989).

When the pollutants are diverse, the total risk hazard index (*HI*) can be estimated using Eq. (3), which is the sum of the individual *HQs.* If the calculated value of *HQ* or *HI* is $> 1$, it indicates a high likelihood of potential non-carcinogenic adverse health effects impacts; otherwise, the probability is minimal.

$$HQ = \frac{E_{xp}C}{(Rfc) \times 1000\ \mu g\ mg^{-1}} \quad (2)$$

$$HI = \sum_{i=1}^{n} HQ_i \quad (3)$$

### 2.5.3 Carcinogenic risk

The carcinogenic risk (*CR*) was calculated as the product of $E_{xp}C$ and *IUR* (Eq. (4)), which is the Inhalation Unit Risk (*IUR*), i.e., as the upper-bound excess lifetime cancer risk estimated to result

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**Table 1.** Parameters used to calculate the daily exposure concentration ($E_{xp}C$) dose through the inhalation route, and the non-carcinogenic and carcinogenic risks from $PM_{2.5}$-bound toxic species.

| Parameters | | | Units | Adults | Reference |
|---|---|---|---|---|---|
| C | Chemical concentration | | (μg $m^{-3}$) | | |
| ED | Exposure Duration (years) | | Years | 24 | U.S. EPA (2009) |
| EF | Exposure Frequency | | (days $years^{-1}$) | 365 | U.S. EPA (2009) |
| ET | Exposure Time | | (h $days^{-1}$) | 16 | U.S. EPA (2009) |
| STE | Short Time Exposure* | | (h $days^{-1}$) | | |
| | STE Coyhaique on 50 μg $m^{-3}$ | | | 7 | |
| | STE Independencia on 50 μg $m^{-3}$ | | | 2.4 | |
| AT | Averaging Time | | (days) | | |
| | AT carcinogenic | | | 70 × 365 | U.S. EPA (2009) |
| | AT non-carcinogenic | | | ED × 365 | U.S. EPA (2009) |
| Metals/metalloids and PAHs (B[a]P) | | | | | |
| Symbol | Element | $RfC_{inh}$ (mg $m^{-3}$) | *IUR* (μg $m^{-3}$) | Cancer Target | |
| As | Arsenic | 1.50E-05 | 4.30E-03 | Respiratory (Group A)** | Asante-Duah (2017) |
| Cd | Cadmium | 1.00E-05 | 1.80E-03 | Respiratory (Group B1) | Asante-Duah (2017) |
| Ni | Nickel | 9.00E-05 | 2.60E-04 | Respiratory (Group B2) | Asante-Duah (2017) |
| Pb | Lead | 1.20E-02 | 1.20E-05 | Not Defined (Group B2) | Greene and Morris (2006) |
| B[a]P | Benzo[a]Pyrene | 2.00E-06 | 6.00E-04 | Gastrointestinal, Respiratory (Group A) | Asante-Duah (2017) |

*RfC* = Reference Concentration dose (non-cancer)/IRIS/EPA by inhalation; *IUR:* Inhalation Unit Risk; **IARC Classification:A = Human carcinogen - based on sufficient evidence of carcinogenicity in animals; B1 = Probable human carcinogen - based on sufficient evidence of carcinogenicity in human; B2 = Probable human carcinogen - based on sufficient evidence of carcinogenicity in animals; IUR = Inhalation Unit Risk (μg $m^{-3}$)/IRIS/EPA; EPA = Agency Protection America; IRIS = Integrated Risk Information System. *STE = estimated of air pollution data considering the average number of hours per day over the Chilean standard of $PM_{2.5}$ (50 μg $m^{-3}$) as the exposure time parameter.

from continuous exposure to an agent at a concentration of 1 μg $m^{-3}$ in the air (U.S. EPA, 1989). When the pollutants are diverse, the carcinogenic risk index (*CRI*) can be calculated using Eq. (5), which is the sum of the *CR* of each element or compound evaluated. There is no safe threshold level of exposure for carcinogenic agents; the EPA suggests that risk values between $1 \times 10^{-4}$ and $1 \times 10^{-6}$ are adequate for protecting human health (Asante-Duah, 2017). Above these values, an excess lifetime cancer risk in exposed populations is expected.

$$CR = IUR \times E_{xp}C \tag{4}$$

$$CRI = \sum_{i=1}^{n} CRi \tag{5}$$

## 2.6 Environmental Cancer Burden Disease by $PM_{2.5}$ Exposure

We used Ostro's functions (Ostro, 2004) to estimate the environmental burden of cardiopulmonary mortality and lung cancer disease attributable to long-term exposure to $PM_{2.5}$. First, the relative risk (*RR*) for each type of cancer is calculated by comparing the annual concentration of $PM_{2.5}$ with a background level without anthropogenic air pollution (Eq. (6)).

$$RR = \left[\frac{(X+1)}{X_0+1}\right]^{\beta} \tag{6}$$

X is the annual average concentration of $PM_{2.5}$ (μg $m^{-3}$), $X_0$ is the background concentration of $PM_{2.5}$ (3 μg $m^{-3}$), and $\beta$ is the coefficient of the risk function obtained from the population-based study of Ostro (2004). For *RR* estimates we used the recommended $\beta$ coefficient for cardiopulmonary mortality of 0.15515 (95% CI: 0.0562–0.2541), while for lung cancer $\beta$ mortality is 0.23218 (95% CI: 0.08563–0.37873) (Pope *et al.*, 2002). Once the relative risk (RR) has been determined, the fraction

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attributable (*AF*) to $PM_{2.5}$ exposure is estimated as shown in Eq. (7):

$$AF = \frac{(RR-1)}{RR} \tag{7}$$

The *AF* estimates the proportion of the deaths that could be avoided if $PM_{2.5}$ concentrations were reduced to 3 μg $m^{-3}$. The number of annual deaths (*I*) was calculated multiplying the annual mortality rate (*IR*) in both localities by the *AF* as shown in Eq. (8):

$$I = IR \times AF \tag{8}$$

*I* is the number of deaths attributable to long-term exposure to $PM_{2.5}$, and *IR* is the total number of annual deaths in the target population. For both localities, the data of age-adjusted (> 30 years) cardiopulmonary and lung-cancer mortality were obtained from the Department of Statistics and Information in Health (DEIS) of the Ministry of Health of Chile. The Tenth International Classification of Diseases (ICD10) code CM I26–I27 was used for cardiopulmonary mortality and code C34 for lung cancer mortality.

## 3 RESULTS

### 3.1 Concentrations of $PM_{2.5}$ (μg $m^{-3}$) and Characteristics of the $PM_{2.5}$-bound Carcinogenic Species Detected in Independencia and Coyhaique

Fig. 2 shows the time variation for an average of 24 h for $PM_{2.5}$ (μg $m^{-3}$) concentrations, and two reference lines representing the annual and daily national standard of the period studied. The

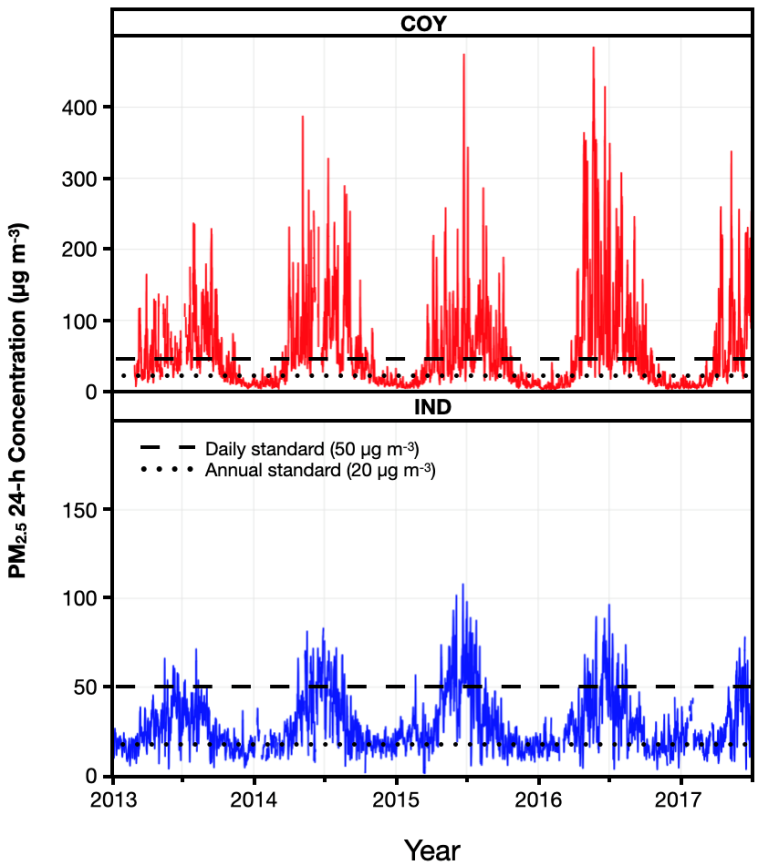


**Fig. 2.** Time series of $PM_{2.5}$ concentrations measured in the AQMS/COY (Center of Coyhaique) and AQMS/IND (downtown Independencia) between 01/01/2013–31/12/2017.

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**Table 2.** Number of days and hours per day that the average 24-hour standard (50 μg $m^{-3}$) of $PM_{2.5}$ is exceeded and reached in the Municipalities of Coyhaique and Independencia.

| AQMS | IND | COY | IND | COY | IND | COY | IND | COY | IND | COY |
|---|---|---|---|---|---|---|---|---|---|---|
| | Summer | | Autumn | | Winter | | Spring | | | |
| Days | | | | | | | | | Total (days $year^{-1}$) | |
| 2013 | **NA** | **NA** | 1 | 44 | 7 | 63 | 0 | 27 | 8 | 134 |
| 2014 | 0 | 3 | 9 | 41 | 34 | 74 | 0 | 18 | 43 | 136 |
| 2015 | 0 | 1 | 9 | 41 | 46 | 66 | 0 | 18 | 55 | 126 |
| 2016 | 0 | 0 | 8 | 43 | 30 | 68 | 0 | 24 | 38 | 135 |
| 2017 | 0 | 1 | 9 | 42 | NA | NA | NA | NA | 9 | 43 |
| Average | **0** | **1.2** | **8.7** | **41.7** | **36.6** | **69.3** | **0** | **20** | **45.3** | **132.3** |
| Hours | | | | | | | | | Total (hours $days^{-1}$) | |
| 2013 | NA | NA | 81 | 725 | 370 | 1214 | 25 | 509 | 476 | 2448 |
| 2014 | 26 | 31 | 265 | 811 | 779 | 1435 | 19 | 364 | 1089 | 2641 |
| 2015 | 39 | 40 | 489 | 747 | 866 | 1192 | 35 | 413 | 1429 | 2392 |
| 2016 | 18 | 29 | 328 | 871 | 750 | 1194 | 52 | 422 | 1148 | 2516 |
| 2017 | 15 | 28 | 245 | 753 | NA | NA | NA | NA | 260 | 781 |
| Average | **24.5** | **32.0** | **281.6** | **781.4** | **691.2** | **1258.7** | **32.7** | **427.0** | **880.4** | **2155.6** |

AQMS: Air Quality Monitoring Stations; NA = Not Available. IND: Independencia; COY: Coyhaique.

average daily concentrations of $PM_{2.5}$ for Coyhaique and Independencia were 110 μg $m^{-3}$, and 45 μg $m^{-3}$, respectively; 2.4 times higher in Coyhaique than in Independencia. The daily (50 μg $m^{-3}$) and annual (25 μg $m^{-3}$) AQS were significantly exceeded in Coyhaique compared to Independencia. The highest concentrations are concentrated in the cold months of the year. Table 2 shows the summarized data for season and year as the number of days that the daily average AQS of $PM_{2.5}$ is exceeded. The average value for the period in Independencia was 45.3 days $year^{-1}$ and 132.3 days $year^{-1}$ for Coyhaique, which represents 12.40% and 36.22% of the days the AQS is exceeded in the year, respectively. In winter these values reached 40.60% and 77.00% of days in Independencia and Coyhaique, respectively. It should be noted that in even summer Coyhaique showed values exceeding the daily AQS.

Fig. S1 shows the distribution of the hourly time series decomposition of $PM_{2.5}$ for each month, per week, for each day of the week, and daily for Independencia and Coyhaique, respectively. As can be seen, in both cases the highest concentrations occur in the cold months, from mid-March to mid-September in Coyhaique and during June in Independencia. The concentrations of $PM_{2.5}$ were lower on weekends than on weekdays in both cases. The hourly average during the week Coyhaique was relatively similar, with higher concentration on Tuesdays and Fridays. In Independencia this variation was less well defined. Finally, the hourly distribution of $PM_{2.5}$ during 24 h shows a different pattern for the cities (monitoring sites). There is a clear and marked daily bimodal profile in Coyhaique, whose concentrations began to increase on average from 4:00 to 8:30 and then decreased around midnight. It began to rise around 15:00, reaching a peak around 19:00. This behavior is less evident in Independencia, where the increase in $PM_{2.5}$ concentration started on average at 5:50, reaching a plateau at 10:00 which progressively decreased until 16:00, finally increasing after 18:00.

Table 3 and Fig. 3 presents the $PM_{2.5}$ mass-concentrations and $PM_{2.5}$-bound chemical species collected in both cities during the winter of 2016. The 24 h average concentrations of $PM_{2.5}$ in the Coyhaique and Independencia sampling sites were 120.58 (78.95–162.82) μg $m^{-3}$ and 46.08 (40.53–51.98) μg $m^{-3}$, respectively. The outdoor concentration of $PM_{2.5}$ was 2.97 times higher in Coyhaique compared to Independencia. The fraction of modern carbon analyzed in EC and OC was almost 4 and 2.5 times higher in Coyhaique than in Independencia. The levels of total PAHs measured in the $PM_{2.5}$ of the Coyhaique (15.95 ng $m^{-3}$) were more than 6.35 times the concentration measured in Independencia (2.51 ng $m^{-3}$). However, 21.8% of PAHs identified in the $PM_{2.5}$ of the Coyhaique were B[*a*]P, compared to the 15.5% B[*a*]P in Independencia. The concentration of B[a]P was 8.92 times higher in Coyhaique than in Independencia. The concentrations of the elements detected in the $PM_{2.5}$ collected in Coyhaique were, in decreasing order, arsenic (As) [1.80 ng $m^{-3}$] > lead (Pb)

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**Table 3.** $PM_{2.5}$ mass-concentration and $PM_{2.5}$-bound chemical species collected in the Municipality of Coyhaique and Independencia in winter of 2016.

| | COY | IND | COY/IND |
|---|---|---|---|
| $PM_{2.5}$ (μg $m^{-3}$) (min–max) | 120.58 (78.95–162.82) | 40.54 (40.53–51.98) | 2.97 |
| % modern carbon in EC | 80.00 | 20.10 | 3.98 |
| % modern carbon in OC | 100.00 | 40.50 | 2.46 |
| Total PAHs (ng $m^{-3}$) | 15.95 | 2.51 | 6.35 |
| B[a]P (ng $m^{-3}$) | 3.48 | 0.40 | 8.92 |
| Ni (ng $m^{-3}$) | 0.57 | 0.77 | 0.75 |
| Cd (ng $m^{-3}$) | 0.15 | 0.21 | 0.73 |
| Pb (ng $m^{-3}$) | 1.61 | 9.02 | 0.18 |
| As (ng $m^{-3}$) | 1.80 | 7.01 | 0.26 |

EC: Elemental Carbon; OC: Organic Carbon; B[a]P: Benzo-a-Pyrene; IND: Independencia; COY: Coyhaique.

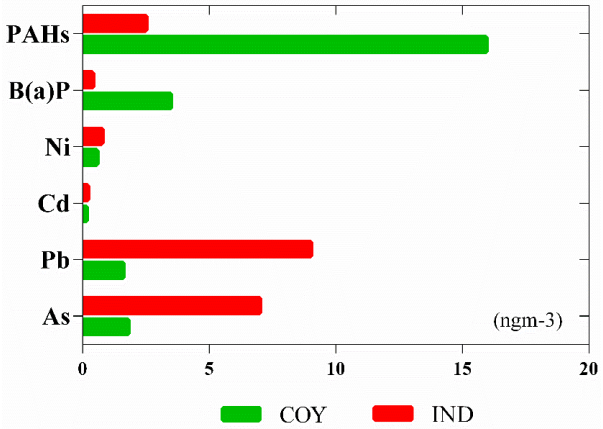


**Fig. 3.** $PM_{2.5}$-bound chemical species collected in the Municipality of Coyhaique and Independencia in winter of 2016.

[1.605 ng $m^{-3}$] > nickel (Ni) [0.573 ng $m^{-3}$] > cadmium (Cd) [0.153 ng $m^{-3}$]. In Independencia these were lead (Pb) [9.07 ng $m^{-3}$] > arsenic (As) [7.00 ng $m^{-3}$] > nickel (Ni) [0.766 ng $m^{-3}$] > cadmium (Cd) [0.211 ng $m^{-3}$]. The concentration of the metallic elements in Independencia was higher than in Coyhaique, with values ranging from 1.3 (Pb) to 5.6 (Ni) times that determined in Coyhaique.

## 3.2 Health Risk Assessment of $PM_{2.5}$-bound Chemical Species

Table 4 and Fig. 4 show both cities' non-carcinogenic (*HQ; HI*) and carcinogenic (*CR; CRI*) human health risk indexes. The estimated *HQ* values for Coyhaique in decreasing order were B[*a*]P > As > Cd > Ni, and Pb. B[*a*]P was 13.4 times higher than the safety level (*HQ* < 1), representing 92.45% of the total risk. In contrast, for Independencia these values were lower than 1 for B[*a*]P > Cd > Ni, and Pb, but 1.12 for As. As and B[a]P showed the highest percentages of the non-carcinogenic risk. The total hazard index (*HI*) was almost 6.12 times higher in Coyhaique than in Independencia. For *CR*, the $PM_{2.5}$-bound species collected in the Coyhaique were, in decreasing order, As > B[a]P > Cd > Ni > Pb, with As and B[a]P higher than the lower acceptable limit ($1 \times 10^{-6}$). Cd, Ni, and Pb were below this limit. The estimated *CR* in decreasing order for Independencia was As > Cd > B[a]P > Ni and Pb. All these values were below the risk threshold, except As. Cd, B[a]P, Ni, and Pb were below the lower limit ($1 \times 10^{-6}$). For Coyhaique 20.30% and 75.38% of the *CRI* ($2.72 \times 10^{-5}$) was explained by B[a]P and As, respectively. For Independencia, these chemicals explained 0.77% and 97.01% of the *CRI* ($2.66 \times 10^{-5}$).

## 3.3 Mortality Attributable to Long-term $PM_{2.5}$ Exposure

Eq. (8) indicates that an estimate of deaths attributable to long-term exposure to air pollution

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in a local area can be made by multiplying the attributable fraction (*AF*) by the total number of deaths annually in the local area (*IR*). According to Ostro (2004), *AF* estimates the proportion of deaths that could be avoided if $PM_{2.5}$ concentrations were reduced to 3 μg $m^{-3}$.

**Table 4.** Non-carcinogenic and carcinogenic risk by inhalation via in Municipality of Coyhaique and Independencia.

| | COY | | IND | | COY/IND |
|---|---|---|---|---|---|
| | HQ | % HI | HQ | % HI | |
| B[a]P | **1.34E+01** | 92.45 | 4.80E-01 | 28.65 | 27.92 |
| Ni | 4.91E-02 | 0.34 | 2.05E-02 | 1.22 | 2.40 |
| Cd | 1.18E-01 | 0.82 | 5.08E-02 | 3.03 | 2.32 |
| Pb | 1.03E-03 | 0.01 | 1.80E-03 | 0.11 | 0.57 |
| As | 9.26E-01 | 6.39 | **1.12E+00** | 66.99 | 0.83 |
| ∑HQ:HI | **1.45E+01** | 100.00 | **2.37E+00** | 100.00 | 6.12 |
| | CR | % CRI | CR | % CRI | |
| B[a]P | **5.51E-06** | 20.30 | 2.06E-07 | 0.77 | 26.75 |
| Ni | 3.94E-07 | 1.45 | 1.71E-07 | 0.64 | 2.30 |
| Cd | 7.29E-07 | 2.68 | 3.26E-07 | 1.23 | 2.24 |
| Pb | 5.09E-08 | 0.19 | 9.28E-08 | 0.35 | 0.55 |
| As | **2.05E-05** | 75.38 | **2.58E-05** | 97.01 | 0.79 |
| ∑CR:CRI | **2.72E-05** | 100.00 | **2.66E-05** | 100.00 | 1.02 |

HQ: Hazard Quotient; HI: Hazard Index; CR: Carcinogenic Risk; CRI: Carcinogenic Risk Index; Values over risk threshold in **bold;** IND: Independencia; COY: Coyhaique.

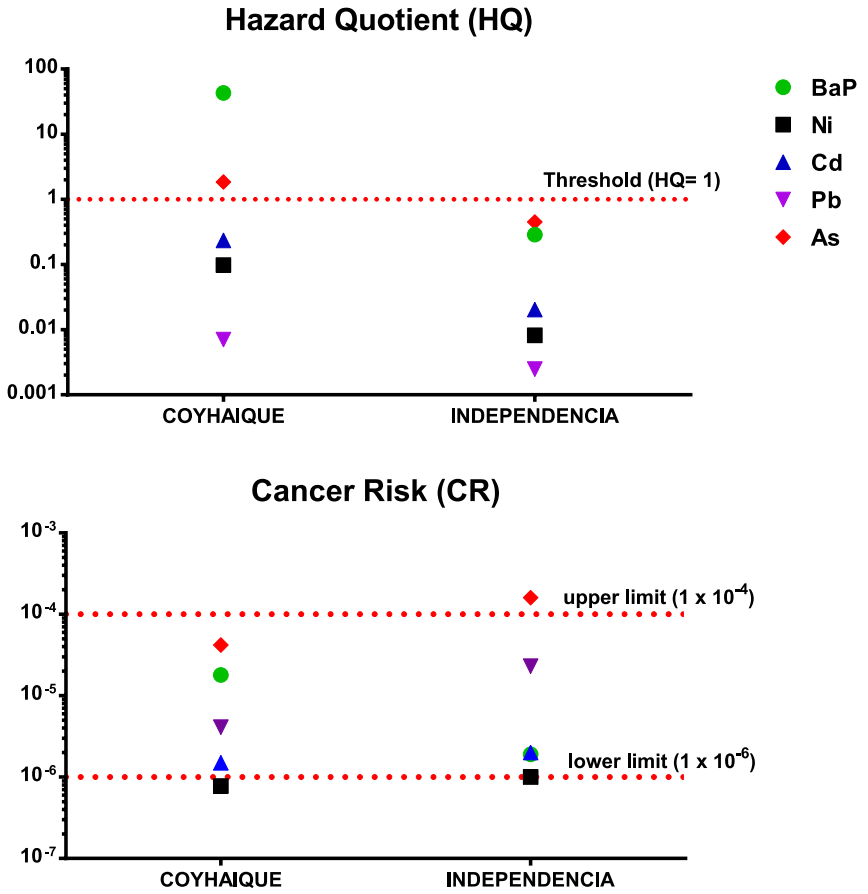


**Fig. 4.** Non-carcinogenic (*HQ*) and carcinogenic (*CR*) risk indicators in the Municipality of Coyhaique and Independencia, respectively.

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**Table 5.** Mortality rate (*IR*), attributable Fraction (*AF*) and annual number of deaths (*I*) from outdoor air pollution by $PM_{2.5}$ in adults > 30 years.

| Disease | Coyhaique | | | Independencia | | |
|---|---|---|---|---|---|---|
| | *IR*[1] | *AF*% (IC95%) | *I* (nº) (IC95%)[2] | *IR*[1] | *AF*% (IC95%) | *I* (nº) (IC95%)[2] |
| Lung cancer mortality | 1.50 | 54 (25–72) | 0.81 (0.37–1.07) | 1.37 | 43 (19–60) | 0.59 (0.26–0.83) |
| Cardiopulmonary mortality | 0.05 | 40 (17–57) | 0.02 (0.01–0.03) | 1.44 | 32 (12–46) | 0.45 (0.18–0.67) |

*IR*: Mortality Rate (× 10,000 inh); *AF*: Attributable Fraction to $PM_{2.5}$ exposure; *I*: Annual deaths; [1] Data obtained from the Department of Statistics and Health Information of the Ministry (DEIS/MS-Chile); [2] Estimated value from Ostro´s function; IND: Independencia; COY: Coyhaique.

Table 5 shows the annual mortality rate (*IR*) for cardiopulmonary and lung cancer, the attributable fraction (*AF*) and the expected number of deaths attributable (*I*) to long-term exposure to $PM_{2.5}$. Coyhaique had a slightly higher *IR* for lung cancer and a significantly lower *IR* for cardiopulmonary compared to Independencia (0.05 vs. 1.44 per 10,000 inh). The attributable fraction (*AF*) estimates show that 54% and 40% of the annual cases of lung cancer and cardiopulmonary mortality in adults over 30 years of age could be attributed to long-term exposure to concentrations of $PM_{2.5}$ in the city of Coyhaique. According to the communal reports (BCN, 2021a), the population of adults over 30 years is around 35,000 people, so almost 3 (95% CI = 1–4) deaths per year from lung cancer would be attributable to $PM_{2.5}$, and less than one case per year for cardiopulmonary disease. For Independencia, 43% and 32% of lung cancer and cardiopulmonary mortality cases were attributed to long-term exposure to $PM_{2.5}$, respectively. Considering that the population over 30 years of age in Independencia is about 56,000 (BCN, 2021b), almost 3 (95% CI = 1–5) and 2 (95% CI = 1–4) new cases per year of lung cancer and cardiopulmonary mortality, respectively, would be expected attributable to long-term exposure to $PM_{2.5}$.

# 4 DISCUSSION

## 4.1 Analysis of $PM_{2.5}$ Data

The daily profile variation of $PM_{2.5}$ observed in this, and previous studies suggests that the principal emissions during the day in Independencia are from mobile sources, while in Coyhaique they are closely related to the use of residential firewood for heating and cooking. This distribution was reported by Alvarado-Zúñiga (2006) for downtown Santiago; he analyzed the daily behavior of $PM_{10}$ during fall and winter. There is an increase in concentration from the beginning of the city's activity from 06:00 to 10:00, decreasing until 15:00–16:00, and then increasing towards the end of the afternoon until 20:00–21:00. Perez and Menares (2018), reported that between April and August in 2014 to 2016 there was a pattern of hourly distribution of $PM_{2.5}$ concentrations similar to those reported in the present study in two neighborhoods of the south of Santiago, *El Bosque* and *Cerrillos*. There was a peak around 09:00 correlated with heavy traffic (rush hour) in the morning. The 23:00 peak would be explained by residential heating and low ventilation during cold months, as in Coyhaique. Another factor that explains this behavior is the effect of the season (seasonal very low temperatures and wind speed), especially in the magnitude of $PM_{2.5}$ concentration. A study prepared by the Mario Molina center for the Ministry of the Environment in Chile, where the trend of particulate matter in Santiago and the cities of central-southern Chile were studied in an MP time series from 2014 to 2018, concluded that the factor with the greatest influence on the increase in $PM_{2.5}$ concentration is the seasonal effect, which in Coyhaique accounts for up to 220%, while in Santiago where Independencia is located this increase was 80%. Other relevant factors would be the height of the thermal inversion layer and the synoptic movements of air masses, closely related to the increase in seasonal emission sources such as residential firewood combustion (CMM, 2019). The daily standard was significantly exceeded in Coyhaique; in Independencia it was surpassed especially in the winter months, while the annual standard was exceeded throughout the period studied. Both time series show a seasonal behavior related to an atmospheric phenomenon called the thermal inversion layer that decreases the vertical dispersion of air pollutants during the winter period, increasing the concentrations of fine particles (Perez *et al.*, 2020; Perez and Menares, 2018; CMM, 2019).

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https://doi.org/10.4209/aaqr.220247

These results are in line with what was reported by Molina *et al.* (2017), who analyzed the daily, monthly, and annual concentrations of $PM_{2.5}$ at the spatial and temporal level of 16 cities in the center-south of Chile, for a period of 7 years (2007–2014), including Coyhaique, establishing that the limits established by the WHO are systematically exceeded in all cities. The highest concentrations occur in autumn and winter and are significantly higher than those determined in the warm period, estimating that at least one third of the days monitored exceed the limits established by the WHO, the primary source being the combustion of firewood. The highest annual concentrations of $PM_{2.5}$ range from 57 μg $m^{-3}$ to less than 20 μg $m^{-3}$, being the highest in Coyhaique.

## 4.2 Analysis of $PM_{2.5}$-bound Organic Compounds

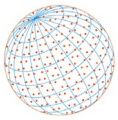
The results of the analysis of the carbon origin in the samples of $PM_{2.5}$ collected in Coyhaique indicate a high percentage of modern carbon in the EC and OC fractions compared to those determined in Independencia. This suggests that the carbonaceous sources of apportionment that prevail in the outdoor air of Coyhaique come mainly from biogenic origin, which would be explained by the use of firewood as combustion energy for residential use. In contrast, the largest source in Independencia would be from the use of fossil fuels; less than 40% would be explained by modern sources probably related to the use of residential firewood during winter, as well as other possible industrial sources that use firewood as a source of energy. These results agree with those reported in different cities of the world with emission sources similar to those described in our study (Marley *et al.*, 2009).

The concentration of total PAHs determined in Coyhaique was within the range reported by Pozo *et al.* (2015), (4–21 ng $m^{-3}$), who determined these compounds in Temuco, a city highly contaminated by wood smoke as occurs in Coyhaique. Bravo-Linares *et al.* (2016), conducted a study in five southern Chilean cities in the Los Ríos region where the primary source of air pollution was the use of wood; they found concentrations of PAHs in $PM_{2.5}$ of 27.2 ng $m^{-3}$ to 49.6 ng $m^{-3}$ during the winters of 2013 and 2014, which were significantly higher than the 1.8 ng $m^{-3}$ to 7.5 ng $m^{-3}$ determined in summer. These concentrations were 2 to 3 times higher than those determined in winter in Coyhaique and more than ten times higher than those in Independencia. Pozo *et al.* (2018), studied the total concentrations of PAHs in outdoor air in eleven different regions and seasons of the year in Chile between January 13, 2016 and March 21, 2017; they reported that average concentrations of total PAHs in Coyhaique were 8.8 times those determined in Santiago downtown in winter, with values of 149 ng $m^{-3}$ and 17 ng $m^{-3}$, respectively; these values are significantly higher than those reported in this study. In the same study, the total average total concentrations varied between 4 and 49 ng $m^{-3}$, the highest value being in the city of Coyhaique. The lowest concentration was in the northern city of Antofagasta. Regarding B[a]P, the concentrations varied between 0.02 ng $m^{-3}$ and 0.96 ng $m^{-3}$. The concentration determined in Coyhaique was 0.78 ng $m^{-3}$.

Various studies have found that the concentrations of PAHs vary depending on the emission sources, the combustion conditions and the types of firewood used (Avagyan *et al.*, 2016). It is important to note that these studies were carried out with different methodologies, monitoring times and determination of PAHs, so they are described referentially; however, they consistently show that PAHs levels are significantly higher in cities where the source of energy for heating is firewood.

## 4.3 Analysis of $PM_{2.5}$-bound Heavy Metals

The present study found relatively similar concentrations of Ni and Cd in both sites, while the concentrations of Pb and As were significantly higher in Independencia than in Coyhaique These were significantly lower than those that have been reported in different studies in the USA (Chen and Lippmann, 2009) and China (Li *et al.*, 2018a). The possible sources of emission of elements may be related to high vehicular traffic, especially in Santiago, and possible industrial emissions (Jorquera and Barraza, 2012). The United States Environmental Protection Agency (U.S. EPA) classifies Ni refinery dust and nickel subsulfide ($Ni_3S_2$) as Group A human carcinogens, and nickel carbonyl ($Ni(CO)_4$) is classified in Group B2, probable human carcinogen (EHC, 1991). The relevance of these toxic species in fine particulate matter is that they have been associated with lung



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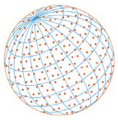
cancer, cardiovascular effects and an increase in the general mortality rate, especially in winter (Grimsrud *et al.*, 2002; Huang *et al.*, 2012; Lippmann *et al.*, 2006; Raaschou-Nielsen *et al.*, 2016). The reference value for environmental nickel in the European Union is 20 ng $m^{-3}$ (European Commission, 2000). Cadmium is classified by the U.S. EPA as a Group B1 probable human carcinogen. Ambient air Cd concentrations have generally ranged from 2 to 15 ng $m^{-3}$ in urban areas and 15 to 150 ng $m^{-3}$ in industrialized areas (EHC, 1992). Aruta *et al.* (2020) analyzed the spatial distribution of potentially toxic elements including Pb and Cd in soil samples, determining high levels of Pb and Cd in the northern area of Santiago near Independencia, which suggests natural and anthropogenic sources of industrial and urbanized (houses and streets) areas, being lower in the green areas of the city. Similar findings were reported by Rodríguez-Oroz *et al.* (2018) who determined different metals (Cr, Ni, Cu, Zn, As, Cd, and Pb) in Chile in playground soils in Concepción, a city in south-central Chile, where higher concentrations occurred in industrial sectors than in green areas. These findings may explain the concentrations of these species reported in our study to a certain extent, due to the effect of dust resuspension. Different studies carried out in Santiago, Chile have determined in the $PM_{2.5}$ analyzed that Pb would be associated with traffic emissions and As to the presence of nearby copper smelters (Ancelet *et al.*, 2014; Dirks *et al.*, 2020; Jorquera and Barraza, 2012; Moreno *et al.*, 2010). The monitoring site in Independencia is just 30 meters from a large avenue with a high traffic flow of public transport buses, which could explain the high levels of Pb (Wróbel *et al.*, 2000). The As concentrations determined in Independencia and Coyhaique were similar to those reported by Jorquera *et al.* (2018), (5 ng $m^{-3}$) in a study carried out in the city of Temuco, which is highly polluted with wood smoke. As is an element that has been associated with indoor cooking sources (Abdullahi *et al.*, 2013), and in some studies, high levels have been observed during the winter, when firewood is used for heating; this could be associated with the burning of firewood treated with chemical preservatives that contain As (Dirks *et al.*, 2020). Arsenic in fine air particulate matter has been recently identified as an important factor for lung cancer in China (Wang *et al.*, 2020).

## 4.4 Health Risk Assessment

The non-carcinogenic risk index was greater than 1 for both cities, indicating an increased risk of adverse health effects in the adult population. The *HQ* of Coyhaique (14.5) was six times more than that of Independencia (2.37), which is mostly explained by the presence of B[a]P in fine particles. The *CRI* was higher than the safety threshold ($1 \times 10^{-6}$). This would indicate possible carcinogenic effects due to inhalation of these compounds by the exposed population. These values of potential carcinogenic risk would imply an excess of 2 to 3 cases of cancer per year compared to those expected otherwise. These results align with the incidence rate of death from lung cancer and cardiopulmonary mortality recorded in both cities. As reported by the Department of Statistics and Health Information (DEIS) of the Ministry of Health of Chile, the incidence rate (IR) of death due to lung cancer for Coyhaique (1.50 × 10,000 inh) is slightly lower than Independencia (1.3 × 10,000 inh). The incidence rate of cardiopulmonary disease is much higher in Independencia (1.44 × 10,000 inh) than in Coyhaique (0.05 × 10,000 inh). However, the estimated contribution of $PM_{2.5}$ to lung cancer and cardiopulmonary mortality rates is almost twice as great in Coyhaique as in Independencia.

The high concentrations of As detected in the $PM_{2.5}$ collected in Independencia could be associated with the higher incidence of lung and cardiopulmonary cancer reported by the DEIS, compared to that Coyhaique, where the concentration of As is lower. As could be determined in our results, the carcinogenic risk index (*CRI*) in Coyhaique significantly exceeded the lower risk threshold ($1 \times 10^{-6}$), which is mainly explained by As (75.38%) and B[a]P (20.30%), respectively. In Independencia, this risk is mainly explained by As (97.01%).

Both chemical elements have been widely associated with cardiopulmonary and lung cancer (Moorthy *et al.*, 2015; Pope *et al.*, 2002; Taghvaee *et al.*, 2018). In a case-control study carried out in Mexico City, Báez-Saldaña *et al.* (2021), evaluated the association between the number of hours of daily exposure to wood smoke and lung cancer; they reported an increased risk OR of 2.6 (2.6 (95% CI: 1.05–6.44) for individuals exposed to > 100 hour-years compared to those to fewer hour-years. Similar findings were reported by Hosgood *et al.* (2010) and Kurmi *et al.* (2012), who found an association between exposure to coal and wood use and lung cancer risk, supporting the hypothesis of a carcinogenic potential of in-home wood use in a systematic review; they



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suggest that in-home burning of biomass is consistently associated with an increased risk of lung cancer (OR 1.50, 95% CI: 1.17–1.94). Another systematic review reported a stronger relationship between biomass use, cooking and/or heating and lung cancer (OR 1.17, 95% CI: 1.01–1.37) (Bruce *et al.*, 2015).

## 4.5 Cancer Burden Diseases

As observed in this study, the adjusted incidence rates of lung cancer are relatively similar between both cities. In contrast, cardiopulmonary mortality rates were significantly higher in Independencia than in Coyhaique. For both diseases, a high percentage of annual cases in the population over 30 years of age would be explained by long-term outdoor exposure to $PM_{2.5}$, considering a concentration of 3 μg $m^{-3}$ of $PM_{2.5}$ as the baseline value. Within the context of the equations used by Ostro to calculate the environmental burden of disease due to long-term exposure outdoor air pollution, these are quite stable since they use a log-linear function for exposure, which allows modulating high concentrations and allows an approximation of risk estimation, with the minimum measurements error (Ostro, 2004; Pope *et al.*, 2002).

The association between exposure to particulate matter and respiratory diseases such as lung cancer and cardiopulmonary diseases has been widely reported in epidemiological studies (Cao *et al.*, 2018; Fu *et al.*, 2015; Wu *et al.*, 2021). The IARC concluded in 2013 that air pollution is carcinogenic to humans. The mechanisms of action involved have to do with epigenetic changes such as DNA deregulation and methylation, microenvironmental alterations related to cellular processes, activation and inactivation of genes, inflammatory processes that produce oxidative stress, among other effects under study (Lee *et al.*, 2020; Li *et al.*, 2018b). In these investigated cities, high concentrations of As and B[a]P were determined, which are known carcinogenic agents for humans. In Chile, Sapunar-Zenteno *et al.* (2021), studied the association between the incidence of lung cancer and air pollution in 14 districts of Chile served by the Oncology Institute of the Fundation Arturo López Pérez (FALP), reporting a positive association between the incidence of lung cancer lung and the concentration of $PM_{2.5}$ in a study period of 4 years (2015–2019), adjusting for the human development index.

Ostro´s function was used in this study to calculate the burden of disease resulting from exposure to outdoor fine particulate matter, which is a sufficiently robust methodology based on epidemiological studies in different parts of the world that have been consistent in their findings and that allow estimating parameters that can be used as a reference to estimate disease burden in exposed populations with similar characteristics to those studied. Although the population is indeed exposed to a mixture of solid and liquid particles and chemical compounds that varies temporally and geographically, the data collected allow us to make a quantitative estimate of premature death and the risk attributable to exposure to PM in the general population and the most vulnerable age groups. Like all methodology, it has advantages and disadvantages that must be considered when interpreting these results on their merit (Ostro, 2004). The objective of applying this methodology was to compare the cities with regard to the attributable risks due to exposure to PM. However, the particularities of each place evaluated must be considered. Air quality monitoring stations have geographic and population representation, but they are not capable of discriminating the possible variations due to exposure in hot spots and the fact that the population spends a large amount of time indoors, such as in Coyhaique, so indoor exposure becomes very relevant. The $PM_{2.5}$ sampling periods for risk assessment were relatively short; the concentrations of the species and the PAH-determined compounds in most cases were similar to or lower than recent studies carried out in Chile as described in this discussion, and therefore our results may be underestimated (Wu *et al.*, 2021).

Estimating the attributable incidence of death for cancer related to long-term $PM_{2.5}$ exposure using Ostro's function for Coyhaique and northern Santiago showed different percentages; Coyhaique was worse. Estimates of the burden of disease attributed to outdoor pollution can help prioritize air $PM_{2.5}$ pollution control over other interventions that improve public health. The burden of disease in Chilean cities will vary due to the amount of fossil fuel used, weather, underlying disease rates, and population size and density. As observed in Coyhaique, the burden of disease estimates will be higher in some areas of the south of Chile, such as those heavily dependent on firewood for fuel use and those with topographic and climatic conditions that limit

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the dispersion of pollution in winter. $PM_{2.5}$ is believed to be a greater health threat since the smaller particles are more likely to be deposited deep into the lung. High concentrations of $PM_{2.5}$-bound species such as B[*a*]P might be related to the higher attributable incidence of lung cancer in Coyhaique. The function and mechanism of B[*a*]P exposure to cancer progression remain unclear (Wei *et al.*, 2016). According to the European Environment Agency (EEA), the average B[*a*]P concentration in Europe in 2012 ranged from 0.12 to 1.5 ng $m^{-3}$ (Lewandowska *et al.*, 2018). In the urban and mining area of northern Chile, the key pollution sources are copper foundries and coal-burning power plants; in the central zone, as recorded in Independencia, massive vehicular traffic is the principal source of emissions; while in southern urban zones like Coyhaique, residential wood combustion is a key source of particulate matter emission (Molina *et al.*, 2017; Mesías-Monsalve *et al.*, 2018; Torres *et al.*, 2021).

## 5 CONCLUSIONS

The analysis of the time series of the concentration of fine particulate matter suggests that the daily distribution is highly correlated with vehicular traffic activity in Independencia and the use of firewood for heating and cooking in Coyhaique. The $PM_{2.5}$ concentrations were significantly higher in Cohyaique than in Santiago for the study period, especially in the winter periods, considerably exceeding the former's daily and annual national standards. The total PAH concentrations determined in the $PM_{2.5}$ collected in winter were more than six times higher for Coyhaique, which would be explained by the extensive use of firewood as an energy source for residential use. The percent of modern carbon observed in the fine particles collected in Coyhaique showed that sources of $PM_{2.5}$ emission directly relate to firewood burning in houses for heating and cooking. The analysis of potential non-carcinogenic effects (*HI*) was only significant for Coyhaique, mainly explained by B[a]P, followed by As. Estimating potential carcinogenic chronic effects showed that arsenic, cadmium, nickel, and Benzo[a]pyrene were in the safe range. As and B[a]P explain over 95% of the integrated risk (*CRI*) of carcinogenic adverse effects in Coyhaique. In contrast, the *CRI* for Independencia is explained mainly by As (97%). Finally, Ostro's function estimated that 54% and 43% of lung cancer incidence and between 40% and 32% of the incidence of cardiopulmonary cancer could be attributed to long-term exposure $PM_{2.5}$ for Coyhaique and Santiago, respectively. $PM_{2.5}$-bound carcinogenic species might be related to the burden of death for lung and cardiopulmonary cancer in adults $>$ 30 years in both cities. Other studies could elucidate the biomedical basis of the high incidence of cancer in both highly polluted cities.

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## DISCLAIMER

The authors declare that they have no conflict of interest in this study.

## SUPPLEMENTARY MATERIAL

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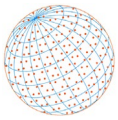



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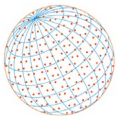



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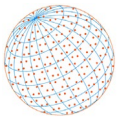



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