Geologic and anthropogenic sources of contamination in settled dust of a historic mining port city in northern Chile: health risk implications

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Chile is the leading producer of copper worldwide and its richest mineral deposits are found in the Antofagasta Region of northern Chile. Mining activities have significantly increased income and employment in the region, however there has been little assessment of the resulting environmental impacts to residents. The port of Antofagasta, located 1430 km north of Santiago, the capital of Chile, functioned as mineral stockpile until 1998 and has served as a copper concentrate stockpile since 2014. Samples were collected in 2014 and 2016 that show elevated concentrations of As, Cu, Pb, and Zn in street dust and in residents' blood (Pb) and urine (As) samples. To interpret and analyze the spatial variability and likely sources of contamination, existent data of basement rocks and soil geochemistry in the city as well as public-domain airborne dust were studied. Additionally, a bioaccessibility assay of airborne dust was conducted and the chemical daily intake and hazard index were calculated to provide a preliminary health risk assessment in the vicinity of the port. The main conclusions indicate that the concentrations of Ba, Co, Cr, Mn, Ni and V recorded from Antofagasta dust likely originate from intrusive, volcanic, metamorphic rocks, dikes, or soil within the city. However, the elevated concentrations of As, Cd, Cu, Mo, Pb, and Zn do not originate from these geologic outcrops, and are thus considered anthropogenic contaminants. The average concentrations of As, Cu, and Zn are possibly the highest in recorded street dust worldwide at 239, 10821, and 11869 mgkg $^{-1}$, respectively. Furthermore, the contaminants As, Pb, and Cu exhibit the highest bioaccessibilities and preliminary health risk indices show that

As and Cu contribute to elevated health risks in exposed children and adults chronically exposed to dust in Antofagasta, whereas Pb is considered harmful at any concentration. Therefore, an increased environmental awareness and greater protective measures are necessary in Antofagasta and possibly in other similar mining port cities in developing countries.

1 **Title page**

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ABSTRACT

Chile is the leading producer of copper worldwide and its richest mineral deposits are found in the Antofagasta Region of northern Chile. Mining activities have significantly increased income and employment in the region, however there has been little assessment of the resulting environmental impacts to residents. The port of Antofagasta, located 1430 km north of Santiago, the capital of Chile, functioned as mineral stockpile until 1998 and has served as a copper concentrate stockpile since 2014. Samples were collected in 2014 and 2016 that show elevated concentrations of As, Cu, Pb, and Zn in street dust and in residents' blood (Pb) and urine (As) samples.

To interpret and analyze the spatial variability and likely sources of contamination, existent data of basement rocks and soil geochemistry in the city as well as public-domain airborne dust were studied. Additionally, a bioaccessibility assay of airborne dust was conducted and the chemical daily intake and hazard index were calculated to provide a preliminary health risk assessment in the vicinity of the port. The main conclusions indicate that the concentrations of Ba, Co, Cr, Mn, Ni and V recorded from Antofagasta dust likely originate from intrusive, volcanic, metamorphic rocks, dikes, or soil within the city. However, the elevated concentrations of As, Cd, Cu, Mo, Pb, and Zn do not originate from these geologic outcrops, and are thus considered anthropogenic contaminants. The average concentrations of As, Cu, and Zn are possibly the highest in recorded street dust worldwide at 239, 10821, and 11869 $mg \cdot kg^{-1}$, respectively. Furthermore, the contaminants As, Pb, and Cu exhibit the highest bioaccessibilities and preliminary health risk indices show that As and Cu contribute to elevated health risks in exposed children and adults

chronically exposed to dust in Antofagasta, whereas Pb is considered harmful at any concentration. Therefore, an increased environmental awareness and greater protective measures are necessary in Antofagasta and possibly in other similar mining port cities in developing countries.

1 INTRODUCTION

The process of economic development often results in large-scale anthropogenic impacts to the environment and inhabitants of developing countries. Previous well-reported examples of environmental pollution caused by industrial activities include: (i) the 1984 Bhopal incident in India after more than 40 tons of methyl isocyanate gas leaked from a pesticide plant (Broughton, 2005), (ii) Chernobyl, Ukraine, in 1986, after the nuclear energy plant explosion of Reactor 4 (Devell et al., 1986; Gale, 1987), (iii) the Baia Mare spill, Romania, in 2000, where nearly 100000 m^3 of cyanide and metal-rich liquid waste was released into the river system near this city (Lucas, 2001; Soldán et al., 2001), and (iv) Sukinda, India, where the most harmful form of chromium (Cr[VI]) pollutes water and inhabitants due to mining activities (Dubey, Sahoo & Nayak, 2001). As such, industrial incidents have been related to severe health issues and fatalities (Khan & Abbasi, 1999).

Despite harmful consequences that industry and uncontrolled anthropogenic activities have caused and are still contributing to (Reddy & Yarrakula, 2016), an awareness of the environmental impact of these activities is growing in many developing countries. Chile, a developing country located along the western border of southern South America, is one example where an environmental awareness has increased with time on a national scale. Among its developing characteristics is the fact that the country predominantly relies on the exploration, exploitation, and exportation of mineral resources as one of its main economic activities (De

Solminihac, Gonzales & Cerda, 2017). Although often associated with environmental degradation, these activities support an elevated gross domestic product (GDP) that has allowed Chile to be included in the Organization for Economic Co-operation and Development (OECD; Ruiz-Rudolph et al., 2016).

1.1 Antofagasta Region background

Copper (Cu) is the most significant metallic resource of Chile: the country has contributed approximately 28% of the world's Cu production since 1985 (COCHILCO, 2014). This element is naturally distributed in northern Chile, principally in the Antofagasta Region (Fig. 1a; Aroca, 2001), where large-to-giant porphyry copper deposits are found (e.g. Chuquicamata, La Escondida, and Radomiro Tomic, among others; Fig. 1b). As such, numerous large-scale mining operations and mineral deposits in the Antofagasta Region have supported the economic development of the region to the point where the per capita GDP is the highest in the country.

Following the War of the Pacific (1879 to 1884), Chile and Bolivia signed a trade agreement in 1904 which allowed Bolivian products to be exported from the Train Station and Port of Antofagasta, where resultantly, unprotected stockpiles containing metals (mainly Pb) became common at those facilities (Sepúlveda, Vega & Delgado, 2000).

It is also important to consider other natural and anthropogenic sources of contamination in Antofagasta that accompanied heightened mining and a related increase in population (by a factor of nearly 6.5 between 1895 (13530 inhabitants; Censo, 1895) and 1960 (87860 inhabitants; Censo, 1960)): (i) of the sparse fresh water sources in the region, the Toconce and Holajar rivers were naturally enriched in As, resulting in chronic contamination of drinking water until 1970 (Marshall et al., 2007), and (ii) in the late 1970s, La Negra Industrial Complex was established 20 km southeast of Antofagasta which included cement production (Industria

Nacional de Cemento SA; Inacesa), lithium (Li) processing (Sociedad Chilena del Litio and SQM), and a smelter (Refimet, currently Altonorte of Xstrata Copper; Minería Chilena, 2013).

At the end of 1980s, elevated Pb levels in blood were reported in children attending schools and living close to the Antofagasta Port and Antofagasta Train Station facilities (Sepúlveda, Vega & Delgado, 2000; Tchernitchin et al., 2006). As a result, in the early 1990s, the stockpiles of Pb-rich materials originating from Bolivia were transferred to Portezuelo, 18 km southeast of downtown Antofagasta, in close proximity to La Negra Industrial Complex. Protective measures related to the transport and deposition of concentrate from Portezuelo to Antofagasta Port were not known to be established prior to 2010 (MTT, 2015).

Combined lung and bladder cancer mortality rates in the Antofagasta Region were the highest reported for the whole country between 1992 and 1994 (153 and 50 per 100000 men and women, respectively), attributable to the chronic ingestion of As between 1958 and 1970 (Marshall et al., 2007). In 2014, despite documented adverse health impacts to the population of Antofagasta and relocation of the Bolivian stockpiles, the development of Cu, Mo, and Ag exploitation in the Sierra Gorda district, 140 km northeast of the city of Antofagasta (Fig. 1b; Minería Chilena, 2014), led to the construction of a holding facility inside the Antofagasta Port called the *galpón* (warehouse), located close to the center of the city, which currently stores Chilean Cu concentrate. Aside from Cu (nearly 32%), the concentrate contains other elements such as S, Fe, Zn, As, Pb, Mo, Sb, and Cd (Table 1 in Fuentes, Viñals & Herreros, 2009). As port operations continued, local inhabitants noticed the increase of dust on buildings etc., which led to criticism of the operations associated with the *galpón* and the port. Resultantly, social organizations were established, such as *Este polvo te mata* ("This dust kills you"), leading to ongoing environmental conflicts within the city.

Currently, the city of Antofagasta has 361873 inhabitants (INE, 2017), which is nearly 4 times greater than the 1960 population (Censo, 1960). The city has grown and developed around the port (Fig. 1c); as a result, 5.7% of the national state load and 17.3% of all regional transport passes through the port (MTT, 2015). Important locations situated near the Antofagasta Port include the downtown area of Antofagasta, hospitals, schools, preschools, the municipal square, the city shopping mall, and commercial centers (Fig. 1c).

1.2 Previous sampling campaigns

Due to increased social pressure associated with the Antofagasta dust, in 2014, the Chilean State sampled city dust and human blood and urine from locations in close proximity to, and up to 9 km distance from the Antofagasta Port (Table 1). Contaminant concentrations were quantified in the < 63 μm size fraction by the Chilean Public Health Institute (ISP-Instituto de Salud Pública; ISP, 2014). Elevated levels of Pb in blood and of As in urine were found in children attending preschools close to the Antofagasta Port, and elevated concentrations of a number of elements were also reported in street dust, including As, Cu, Pb, and Zn (Vergara, 2015). More recently, the Chilean College of Physicians (CM-Colegio Médico) undertook another sampling campaign in 2016 to compare concentrations with the values from 2014 for determining which contaminants were still present in the < 63 μm size fraction of dust (Tchernitchin & Bolados, 2016). Details regarding the methodologies used in these previous studies are found in Table 2.

1.3 Study objectives

Despite the recent 2014 and 2016 sampling events, no analyses were conducted to evaluate and understand the spatial distribution, variation, and sources associated with health risks by exposure to these metallic and metalloid (As) contaminants apart from their bioaccessibility. Also, regardless of well-reported improvements in the technology used to transport and store

material inside the Antofagasta Port (MTT, 2015), currently there is little evidence that the environmental issues and negative health impacts on Antofagasta residents have improved, and regulations have not been put into effect to control the contamination. Therefore, considering the need to better understand and establish (i) the spatial-temporal variability of the present contaminants, (ii) the likely source of contamination through the comparison of natural background concentrations, (iii) human health risk standards associated with exposure to the dust from the Antofagasta Port, and (iv) the bioaccessibility of the contaminant elements, a health risk assessment and bioaccessibility assay were conducted, and raw data collected in 2014 (ISP, 2014) and 2016 (Tchernitchin & Bolados, 2016) was further interpreted and analyzed to improve the understanding of pollution in Antofagasta and better inform regulators and interested parties in the support of new environmental policies and regulations.

2 METHODOLOGY

2.1 Data compilation

Information related to the elemental concentrations of metals was compiled from previous studies of (i) street dust and (ii) the composition of geologic outcrops, weathered products, and soil in the area of Antofagasta.

2.1.1 Antofagasta street dust

Starting 1993, leaded fuels were banned by law in Santiago, Chile (Faiz, Weaver & Walsh, 1996), as well as in other regions of the country thereafter. In medium- to low-size coastal cities of Chile, such as Tocopilla, located at 175 km north of Antofagasta, it has been shown that vehicle emissions do not significantly contribute to ambient particulate matter concentrations in the city (Jorquera, 2009). Therefore, street dust in urban areas of Antofagasta is considered to be an indicator of metal and metalloid contamination from atmospheric deposition, and public dust

157 samples from a number of key city locations were compiled from ISP (ISP, 2014) and CM
158 (Tchernitchin & Bolados, 2016) (Tables 1 and 2). These sampling data are simply reported from
159 those sources and reinterpreted in the context of other factors such as the background geology,
160 which depending on the host rock mineral composition, can weather and contribute to increased
161 concentrations of the same contaminant elements.

162 **2.1.2 Geologic outcrops, weathered products, and soil**

163 The main geologic formations of Antofagasta correspond to Jurassic volcanic basic to
164 intermediate rocks of the La Negra Formation and Neogene marine sedimentary rocks of La
165 Portada Formation, constituted by sandstones and coquinas, of which the latter is a form of
166 limestone represented by an agglomerate of shells (SERNAGEOMIN, 2003). Concentrations of
167 As, Ba, Cd, Co, Cr, Cu, Fe, Mn, Mo, Ni, Pb, V, and Zn of volcanic, intrusive, metamorphic
168 rocks, and dikes of Antofagasta were compiled from Lucassen & Franz (1994), Oliveros et al.
169 (2007), and Rogers & Hawkesworth (1989) (Fig. 2a). In addition, unpublished data (As and Mo)
170 of Oliveros et al. (2007) was utilized. Element concentrations from sedimentary rocks of the La
171 Portada Formation were not available, however the composition of soil, an exogenous matrix
172 associated with the weathering of parent rocks, was obtained from De Gregori et al. (2003; 2
173 soils around Antofagasta city) and CENMA (2014; 15 background samples and 2 contaminated
174 samples from Antofagasta). A brief summary of the methodologies used in these studies is
175 presented in Table 2.

176 **2.2 Data analysis**

177 **2.2.1 Spatial distribution of the geologic outcrops and dust samples**

178 The spatial distribution of regional volcanic, intrusive, metamorphic rocks, dikes, and soils was
179 plotted using the Geographic Information System QGIS (2.6.1 Brighton) software (Fig. 2a).

Local-scale surficial geology (1:50000), provided by the program *Geología para el Ordenamiento Territorial de Antofagasta* of the National Service of Geology and Mining (Servicio Nacional de Geología y Minería SERNAGEOMIN, 2014), was used to plot the distribution of Antofagasta dust samples and their corresponding relationship to the geologic outcrops (Fig. 2b).

2.2.2 Metal and metalloid statistics

To determine basic characteristics of the As, Ba, Cd, Cr, Cu, Mn, Mo, Ni, Pb, V, and Zn data, univariate statistics (e.g. mean, standard deviation, median, minimum, maximum, lower and upper limit of confidence interval (95%)) were calculated in SYSTAT (2009). Principal component analysis (PCA) was conducted on $\log_e(x+1)$-transformed, normalized data in PRIMER 7 (Clarke & Gorley, 2006). In addition, comparisons to the upper continental crust (UCC; from Rudnick & Gao, 2003) are also presented.

2.2.3 Geo-accumulation index and enrichment factor

To infer contaminant elements and sources of street dust, numerous studies have utilized the geoaccumulation index (I_{geo}; Equation 1; e.g., Lu et al., 2009; Li et al., 2013) and enrichment factor (EF; Equation 2; e.g., Zoller, Gladney & Duce, 1974; Lu et al., 2009), respectively.

$$I_{geo} = log_2\left(\frac{C_n}{1.5 \times B_n}\right) \quad (1)$$

In Equation 1, C_n corresponds to the measured concentration of metal n in the sediment and B_n is the local background value of the metal n. A factor of 1.5 was used for possible variations of the local background due to variable lithologies (Muller, 1979; Nowrouzi & Pourkhabbaz, 2014). The local background value (B_n) of each metal was obtained from volcanic, intrusive, metamorphic rocks, dikes, and soils in the region. Specific values of the I_{geo} index indicate the following:

$I_{geo} \leq 0$, the sample is not contaminated;

$0 < I_{geo} \leq 1$, the sample is non- to slightly contaminated;

$1 < I_{geo} \leq 2$, the sample is moderately contaminated;

$2 < I_{geo} \leq 3$, the sample is moderately to highly contaminated;

$3 < I_{geo} \leq 4$, the sample is highly contaminated;

$4 < I_{geo} \leq 5$, the sample is highly to extremely contaminated;

$I_{geo} > 5$, the sample is extremely contaminated.

Enrichment Factors (EFs; Equation 2) were calculated using the geochemical composition of basement rocks and soil of Antofagasta, with Fe as the normalizing element.

$$EF = \frac{M_1}{E_{ref1}} \Big/ \frac{M_2}{E_{ref2}} \quad (2)$$

In Equation 2, EF is equivalent to the Enrichment Factor (Zoller, Gladney & Duce, 1974). M_1 is the metal or metalloid concentration in the sample, E_{ref1} is the reference element in the sample (Fe), M_2 is the background concentration of the metal or metalloid, and E_{ref2} is the background concentration of the reference element (Fe in intrusive, volcanic, metamorphic rocks, dikes, and soil of Antofagasta). Fe, Al, or Ti have been used as reference elements in previous studies (Rule, 1986; Ergin et al., 1991; Tapia et al., 2012), however Fe was chosen as the reference element here because it was the only conservative element present in all of the utilized data sets.

2.3 Health risk assessment

To obtain a preliminary health risk assessment associated with exposure to Antofagasta dust, the chemical daily intake (*CDI*; Equation 3) of the studied elements was calculated by ingestion exposure while considering the start of the *galpón* and chronic exposure as 2 and 70 years, respectively. Dust consumption and body weight were obtained from the US EPA (1989).

$$CDI_{ingestion} = \frac{C \times IngR \times E_{freq} \times E_{dur}}{BW \times AT} \quad (3)$$

In Equation 3, $CDI_{ingestion}$ corresponds to the chemical daily intake (in $mg \cdot kg^{-1} \cdot day^{-1}$), while C is the 95% confidence upper limit of the metal or metalloid concentration (in $mg \cdot kg^{-1}$), $IngR$ is the dust ingestion rate (200 $mg \cdot day^{-1}$ for children under 6 and 100 $mg \cdot day^{-1}$ for adults), E_{freq} is the exposure frequency ($days \cdot year^{-1}$), E_{dur} is the exposure duration (years), BW is the average body weight, and AT is the average time of exposure (ED × 365 days for non-chronic exposure; 70 × 365 days for chronic exposure). As suggested by the US EPA (1989) the hazard index by oral ingestion ($HI_{ingestion}$; Equation 4) of the Antofagasta dust was preliminarily obtained using the $CDI_{ingestion}$ and reference doses (RfD) from literature (As: US EPA & IRIS (1991); Ba: Dallas & Williams (2001); Cd: US EPA & IRIS (1989); Co: Finley et al. (2012); Cr(VI): US EPA & IRIS (1998); Mo: US EPA & IRIS (1992); Ni (soluble salts of Ni): US EPA (1991); V: RAIS (1991); Zn: US EPA & IRIS (2005)). For Cu, the $HI_{ingestion}$ was obtained with the minimal risk level (MRL; ATSDR, 2004), and for Pb, the $HI_{ingestion}$ was not calculated because the RfD_{Pb} (Pb reference dose) does not exist given that Pb is considered toxic at any concentration (US EPA, 2004).

$$HI_{ingestion} = \frac{CDI_{ingestion}}{RfD} \quad (4)$$

In Equation 4, $HI_{ingestion}$ is the hazard index (by ingestion), $CDI_{ingestion}$ is the chemical daily intake (by ingestion), and RfD is the reference dose (US EPA, 1989). The greater the value of HI above unity (1), the greater the level of concern. Therefore $HI \leq 1$ suggests unlikely adverse health effects whereas $HI > 1$ suggests the probability of adverse health effects (Luo et al., 2012; Massey, Kulshrestha & Taneja, 2013). For As, in the case of $HI > 1$, health effects have been related to cellular necrosis and cancer (US EPA & IRIS, 1991).

2.4 Bioaccessibility assay

Health effects caused by exposure to the different components of dust mainly occur by ingestion

of small-sized particulate matter (PM). Fine and ultrafine particles (lower than 2.5 μm in diameter; ≤ $PM_{2.5}$) present a health risk due to their entrance into the bloodstream from lung alveoli, whereas larger particles (> $PM_{2.5}$) are momentarily retained in the bronchi and bronchiole and are then expelled from the cilia of epithelial cells in the respiratory tract. Subsequently, these particles are swallowed, and once in the stomach, gastric hydrochloric acid at pH 2.0 partially solubilize components which constitute the bioaccessible fraction of the dust used to estimate the amount available for absorption across the gastrointestinal barrier (Bradham et al., 2017). Therefore, a higher bioaccessibility of a contaminant element signifies that a larger fraction will enter the human bloodstream.

To compliment the health risk assessment, a bioaccessibility assay was performed at 3 sites close to the Antofagasta Port. Following existent methodologies (eg., Cortés et al., 2015), this assay was performed in the dust fraction of a diameter less than 63 μm (mesh #230). Dust was digested with chloridric acid at a pH of 2 and a temperature of 37ºC, for 2 hours, in order to resemble the digestive conditions of the human stomach. Three replicates were used for every sample and the standard recovery varied between 82% and 98%. Elements within the digested residue were quantified by ICP-OES.

3 RESULTS

3.1 Statistical summary and spatial variation

Compared to the UCC mean concentrations (Rudnick & Gao, 2003; Table 3), Antofagasta dust concentrations of Co, Cr, Mn, and V from 2014 and 2016 were similar, whereas Ba and Ni were lower. These elements also exhibited a lower relative standard deviation (σ ÷ mean × 100) when compared to the mean concentration of all considered data. This variability ranged from 21% to 69% for V and Ba, respectively (Table 3). Comparing concentrations from 2014 and 2016 at

locations within a 0.5 km distance from the Antofagasta Port, mean values of Ba (+96%), Co (+71%), Ni (+33%) and V (+32%) increased, and Cr (-3%) and Co (-3%) showed a slight decrease.

The elements As, Cd, Cu, Mo, Pb, and Zn showed mean concentrations that were 2 (As, Mo, and Pb) and 3 (Cd, Cu, and Zn) orders of magnitude higher than the UCC. They also displayed the highest relative standard deviations when all data were considered, ranging from 91% for Cu and Zn to 120% for Pb (Table 3). Concentrations of As (-49%), Cu (-17%), and Pb (-87%) showed a significant decrease in their mean values between 2014 and 2016, while Cd (+44%), Mo (+72%), and Zn (+56%) showed increased mean concentrations within a 0.5 km radius of the Antofagasta Port (Table 3). Despite the high observed variability in element concentrations of the Antofagasta dust, the highest concentration values of As, Cd, Cu, Mo, Pb, and Zn were evident near the facility (Fig. 3). Negative Pearson correlations between these elements and distance to the Antofagasta Port range between -0.28 and -0.43 (considering all data), indicating that the element concentrations decreased with distance from the port (Table 3). The elements Co, Ni, and V showed slightly higher values near Antofagasta Port that are different from Ba, Cr, and Mn which tended to increase as a function of distance from the port. However, all concentrations are within the same order of magnitude. For these elements, Pearson correlations are lower than 0.57 (Table 3).

Finally, the principal component analysis of the data identified two main associations. These were related to samples located less than 0.5 km and greater than 0.5 km away from the Antofagasta Port (Fig. 4).

3.2 Contaminants and sources

Results for the I_{geo} index (Muller, 1979) are shown in Table 4. Two groups are clearly observed:

295 the non-contaminants, including Ba, Co, Cr, Mn, Ni, and V, and the contaminants, which include
296 As, Cd, Cu, Mo, Pb, and Zn. With the exception of As indices calculated with a soil background,
297 all elements classified as contaminants are considered extreme contaminants within a 0.5 km
298 distance from the Antofagasta Port (Table 4).
299 The enrichment factor (EF) results are summarized in Table 5. The same elements classified as
300 non- contaminants exhibit low EFs (generally lower than 3), indicating that they likely originate
301 from volcanic, intrusive, metamorphic rocks, dikes, or soils within the immediate areas of
302 Antofagasta. Conversely, contaminant elements exhibit higher EFs near the port, which suggests
303 that the source is not related to the geology of Antofagasta and is more concentrated close to this
304 facility (Table 5).
305 **3.3 Health risk assessment and bioaccessibility of dust**
306 The *CDIs* and *HIs* related to Antofagasta dust ingestion are summarized in Table 6. As
307 previously defined, if a child between 0 to 6 years of age ingests 200 $mg \cdot day^{-1}$ of Antofagasta
308 dust, on average, during 2 years of exposure (or from the start of operation of the *galpón*), the
309 *HIs* related to As and Cu are elevated at all sampling points, even 3 km away from the
310 Antofagasta Port. Conversely, Cd and Zn represent a low hazard 1 km and 0.5 km from the port,
311 respectively (Table 6b). For adults living in Antofagasta with a mean body mass of 70 kg that
312 consume 100 $mg \cdot day^{-1}$ of dust, on average, over 70 years (signifying chronic exposure), As and
313 Cu are moderately hazardous elements within 0.5 km of the port and slightly hazardous within 1
314 km of the facility (Table 6b). With the exception of Pb, which does not have a *RfD* value, given
315 that it is highly hazardous at any concentration (US EPA, 2004), the elements Ba, Co, Cr, Mn,
316 Mo, Ni, and V are not considered harmful to infants and adults.
317 Results of the mean bioaccessibility of contaminant elements at 3 sites close to the port are

318 presented in Table 7. It is apparent that As has the highest bioaccessibility (50%), signifying that
319 it can more readily enter the human bloodstream in comparison to the other contaminant
320 elements. The highest bioaccessibility of As is followed by Pb (26%), Cu (20%), Zn (16%), and
321 Cd (10%), whereas Mo does not present a heightened potential of bioaccessibility (Table 7).

322 **4 DISCUSSIONS**

323 **4.1 Comparison to worldwide city dust**

324 The results of this analysis of existing street dust data from Antofagasta indicate that As, Cd, Cu,
325 Mo, Pb, and Zn are contaminants and their notably high concentrations are not related to the
326 geochemistry of outcrops or soil of the city. To compare these results in a national and
327 international context, the mean and standard deviation of these contaminants and the non-
328 contaminant elements (Ba, Co, Cr, Mn, Ni, and V) were compared to: (i) dust accumulated in
329 *Platanus orientalis* leaves of the main east-west highway of Santiago (Alameda), the capital of
330 Chile (13 sites, Tapia et al., 2009), (ii) resuspended dust from Fushun, China (17 dust samples
331 collected from unfrequently cleaned windowsills or plat floors), a coal-based city (Kong et al.,
332 2011), (iii) street dust from Baoji, an important industrial city in Northwest China which has
333 experienced a rapid urbanization and industrialization during the last decades (Lu et al., 2009),
334 (iv) street dust from Zhuzhou, a heavily industrialized city in central China (Li et al., 2013), (v)
335 dust from the industrial area of Qingshan district (QS) in Wuhan, China, one of the largest
336 metropolises in that country (Zhu et al., 2013), and (vi) road dust from the Islamabad
337 Expressway (Faiz et al., 2009), one of the busiest roads in the capital of Pakistan (Table 8).
338 The concentrations of the contaminants As, Cu, and Zn in Antofagasta dust are strikingly high in
339 comparison to street or urban dusts from heavily industrialized cities of Chile, China, or
340 Pakistan. Mn and Mo concentrations are similar to Santiago city dust, while Pb and Cd

concentration values are lower and comparable, respectively, to Zhouzhou dust. On the contrary, resuspended dust from the coal-based city of Fushun contains the highest values of Co, Cr, and Ni, and dust from the industrial QS district presents the highest concentrations of Ba (Table 8).

4.2 Anthropogenic sources of contamination

Based on the results of this study, there exists a strong correlation in Antofagasta between elevated concentrations of several key contaminants and their proximity to the Antofagasta Port. This indicates that materials stored in the port represent a source of As, Cd, Cu, Mo, Pb, and Zn that is measurable in Antofagasta city dust. In Table 4, these contaminants show higher I_{geo} values near the port; therefore, contamination increases close to the facility. In addition, their EFs were higher near the port (Table 5). This relationship is not shown by the non-contaminant elements (Tables 4 and 5), as some of their EFs are equal to 1, indicating that Ba might originate from volcanics, Co from volcanics or intrusives, Cr from intrusives or soil, Mn from metamorphic, volcanic, or intrusive rocks, Ni from soil, and V from all rocks of the city (Table 5). Conversely, EFs obtained for As, Cd, Cu, Mo, Pb, and Zn were generally greater than 20, indicating that their sources cannot be from rocks and soil present in the city (Table 5). To support this conclusion, other studies conducted in marine environments of the Bay of Antofagasta have showed that enrichment of metals such as Cu, Pb and Zn in coastal waters, bottom sediments, and benthic organisms are related to industrial activities developed along the coastal border of the bay (Salamanca et al., 2000; Lépez, Furet & Aracena, 2001; Salamanca, Jara & Rodríguez, 2004; Valdés et al., 2010, 2011, 2014, 2015; Calderón & Valdés, 2012).

As potential sources of anthropogenic contamination, Chilean Cu concentrates and Bolivian stockpiles are addressed individually in the two following subsections.

4.2.1 Cu concentrate

Chilean Cu concentrates are notably rich in As, containing on average from 1% or 10000 $mg \cdot kg^{-1}$ (Cantallopts, 2015) to 2.5% or 25000 $mg \cdot kg^{-1}$ (Herreros et al., 2003) of this metalloid, indicating that the Cu concentrate stored in the Antofagasta Port represents an important source of As. Indeed, the average As in Antofagasta dust (239 $mg \cdot kg^{-1}$) represents only 2.4% of the average As concentration of Chilean Cu concentrate (10000 $mg \cdot kg^{-1}$).

The Sierra Gorda Mine, which is situated within a Cu-Mo porphyry deposit (Brunetti, 2011), produces 120000 tons of Cu, 50 million pounds of Mo, and 10 million pounds of Au each year (Sierra Gorda website), meaning that the Cu concentrates of this mine (and the materials stored in the *galpón*) are rich in Mo. This is supported by the fact that many of the most important porphyry copper deposits of Chile exploit Mo as a byproduct; for instance, there has been an average of (i) 9212 tons every year, since 1997, of Mo in the concentrate of Chuquicamata and Radomiro Tomic and (ii) 8572 tons, since 2000, in the concentrate of Los Pelambres (COCHILCO, 2016). Chilean porphyries can also be related to Zn anomalies (e.g., La Escondida; Garza, Titley & Pimentel, 2001). Therefore, Cu concentrate stored in the Antofagasta Port likely also acts as a source of the high concentrations of As, Mo, and Zn recorded in street dust.

4.2.2 Bolivian stockpiles

The elementary concentrations of stockpiles from Bolivia that were stored in the port until the 1990s are unknown, yet they contained Pb and Zn (El Mercurio, 2010). This reflects the fact that Bolivian mineral deposits are typically polymetallic. For instance, Cerro Rico from Potosí was the largest silver (Ag) deposit known and is associated with base minerals such as cassiterite (SnO_2), sphalerite ((Zn,Fe)S), and galena (PbS); the San Cristóbal District contains 2.0 $oz \cdot t^{-1}$ Ag, 1.67% Zn, and 0.58% Pb with minerals such as galena, sphalerite, pyrite (FeS_2), and chalcopyrite

387 ($CuFeS_2$); and Pulacayo hosts minerals such as sphalerite, tetrahedrite ($(Cu,Fe)_{12}Sb_4S_{13}$),
388 freibergite ($(Ag,Cu,Fe)_{12}(Sb,As)_4S_{13}$), argentiferous galena, and chalcopyrite (Kamenov,
389 Macfarlane & Riciputi, 2002). These polymetallic mineral ores are related to Pb, Zn, and Cu,
390 which could explain the presence of these elements in dust sampled in close proximity to the
391 Antofagasta Port. In addition, minerals containing Zn, Pb, and Cu are natural sources of Cd
392 (ATSDR, 2012), which could explain its enrichment near the Antofagasta Port.

393 **4.2.3 Health risk**

394 The bioaccessibility of dust components was investigated in solid materials (i.e. mud and
395 sediment) of Chañaral, Atacama Region (300 km south of Antofagasta), an environment similar
396 to Antofagasta, revealing that the bioaccessibility percentages of some contaminant elements
397 range from: 26% to 49% for As, 24% to 84% for Cu, 67% to 96% for Pb, 19% to 79% for Zn
398 (Cortés et al., 2015). In the case of the 3 sites studied in relation to dust of the Antofagasta Port
399 (Table 7), As is highly bioaccessible (50%), followed by Pb (26%) and Cu (20%). These
400 significant bioaccessibilities are supported by the fact that As and Cu show the highest *HIs* in the
401 street dust (Table 6b).

402 The health effects of chronic exposure to toxic metals and metalloids are well-known. For
403 example, following chronic exposure of 30 or more years to As, the probability of mortality
404 increases dramatically due to lung and bladder cancers. In addition, prenatal or infant exposure to
405 low levels of these elements, through the mechanism of epigenetic imprinting, can cause
406 irreversible biochemical changes that promote the development of various organic diseases or
407 neurobehavioral alterations in later years (Tchernitchin et al., 2013; Tchernitchin & Gaete,
408 2015). Also, the prenatal exposure to As increases the probability of mortality due to
409 bronchiectasis between the ages of 30 to 49 years (Smith & Steinmaus, 2009); prenatal or infant

410 exposure to Pb affects reproductive functions, decreases intelligence, and causes serious
411 neurobehavioral changes in later stages of life (Tchernitchin et al., 2013).

412 **4.3 Recommendations and considerations**

413 Street dust from Antofagasta likely represents the highest recorded concentrations of As, Cu, and
414 Zn in city dust worldwide. This result is not surprising, given that Chile is the most important
415 producer of Cu, Chilean ores, and concentrates, acting as a plentiful source of As, Mo, and to a
416 lesser extent Zn. Mining development in Chile continues to be predominant; for example, La
417 Negra Industrial Complex has grown to 200 hectares and 120 companies, including a Cu smelter.
418 Previous studies, such as Ruiz-Rudolph et al. (2016), show that Chilean communes with this type
419 of facility suffer higher mortality rates.

420 Although the Antofagasta Port is an important employer in the city, storing Cu concentrate and
421 polymetallic stockpiles there for long-term periods is not appropriate due to health concerns.
422 These materials have caused disease in children in the past (Sepúlveda, Vega & Delgado, 2000)
423 and continue to cause elevated concentrations of pollutants in children's blood and urine
424 (Vergara, 2015). The results of sampling show that street dust contains concentrations of
425 dangerous contaminants that are high enough to threaten the health of people (especially
426 children) living and working in close proximity to Antofagasta Port. A precautionary measure
427 previously undertaken as of 2010 for some concentrates stored in Portezuelo is that they have
428 been and are now transported to the Antofagasta Port by hermetically sealed and cleaned trucks
429 (MTT, 2015). Nonetheless, once stationary in Antofagasta, concentrates could be displaced by
430 wind that moves preferentially in the SW direction; hence, special attention must be paid to the
431 dispersion of contaminants from concentrates and stockpiles to the NE, especially during windy
432 days.

As seen in resuspended dust from Fushun, China, some elements can be highly accumulated in dust (e.g., Cr 5334 ± 10667 $mg \cdot kg^{-1}$; Kong et al., 2011), especially when it is from long-term uncleaned surfaces. Antofagasta is located within the world's driest desert, and as such, there is a very low probability for rain to remove dusts. Cleaning surfaces with wet mops twice a month and frequently washing children's hands in locations close to the port are precautionary measures that can be taken on an individual level to prevent the ingestion of city dust, lowering Pb values in blood (Charney et al., 1983).

As a long-term goal, existing and prospective developments located in close proximity to the port should consider the high risk of contamination and develop relevant strategies to minimize exposure to contaminated dust. Operations within the port itself should be modified (through regulations, safety guidelines, and practical measures). Additionally, it will be important for the regional and national governments to invest in suitable health care (e.g. the training of specialists and development of suitable medical facilities).

The future management and prevention of contamination in Antofagasta could also be strengthened by the implementation of a local high-quality geochemistry laboratory that allows for fast, efficient, and accurate measurements of contaminants through the utilization of specific environmental matrices where pollutant speciation can be efficiently determined. Considering that the mining industry will continue to be a profitable venture in Chile that provides jobs and income as other commodities become extensively exploited in this region (e.g., Li), the implementation and management of a laboratory of this type is necessary in this growing and highly contaminated city and region.

Finally, it is important to note that the contamination and problems described are likely not restricted to Antofagasta, or to Chile. Wherever metals are extracted, stored, and transported,

456 there is a risk that workers and the public can be exposed to contamination and it is important
457 that this is recognized by all parties. Chile is rapidly moving towards the status of a developed
458 country, however environmental protection measures have not yet developed at the same rate as
459 its economy. Industrial regulations must ensure safe levels of pollutants as is defined in the
460 Constitution of Chile which states that Chilean citizens "have the right to live in a
461 contamination-free environment" (Gobierno de Chile, 1980).

462 **5 CONCLUSIONS**

463 Main conclusions related to Antofagasta dust contamination are summarized below while
464 considering the local geology, health risks, and bioaccessibility of the contaminant elements.
465 These items are followed by potential short- and long-term recommendations and considerations
466 that could reduce associated adverse health impacts to Antofagasta residents.

467 1. Street dust from the city of Antofagasta in northern Chile contains Ba, Co, Cr, Mn, Ni,
468 and V that likely originate from intrusive, volcanic, metamorphic rocks, dikes, or soil of
469 the city.
470 2. Antofagasta city dust is contaminated with As, Cd, Cu, Mo, Pb, and Zn, and this
471 contamination likely originates from the Antofagasta Port (an anthropogenic source).
472 3. The mean concentrations of As (239 $mg \cdot kg^{-1}$), Cu (10821 $mg \cdot kg^{-1}$), and Zn (11869
473 $mg \cdot kg^{-1}$) are likely the highest in street dust worldwide.
474 4. Heightened bioaccessibility in Antofagasta dust was found for As, Cu and Pb, indicating
475 that comparatively high fractions could be absorbed into the human bloodstream.
476 5. The bioaccessibility and hazard indices indicate that As, Cu, and Pb in dust represent a
477 health risk to children or adults chronically exposed to Antofagasta dust.

478 Potential recommendations and considerations include:

1. Short term:
    - Discussions with regulators and relevant parties in industry regarding the significance of the results in relation to the city's occupants. Prospective developments and businesses in close proximity to the Antofagasta Port should be fully aware of the risk of contamination.
    - Houses and children living in close proximity to the Antofagasta Port should be constantly cleaned to avoid dust ingestion.
2. Long term:
    - Stricter regulations on a national or regional level should be adopted to reduce dust-borne contamination. Also, businesses, schools, and prospective developments in close proximity to the port should develop relevant long-term strategies to minimize exposure to contaminated dust.
    - Polymetallic stockpiles and Cu concentrates should not be kept in the Antofagasta Port for long-term periods.
    - Investments should be made in medical training and infrastructure to properly remedy potential health impacts.
    - The implementation of a real-time monitoring program and high-quality geochemistry laboratory should be considered to improve the quantification of contaminant elements and their speciation in Antofagasta dust.

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507 REFERENCES

508 Aroca P. 2001. Impacts and development in local economies based on mining: the case of the
509 Chilean II region. Resources Policy 27:119–134. DOI: 10.1016/S0301-4207(01)00013-7.
510 ATSDR 2004. ATSDR - Toxicological Profile: Copper. Available at
511 https://www.atsdr.cdc.gov/ToxProfiles/tp.asp?id=206&tid=37 (accessed March 22, 2018).
512 ATSDR 2012. ATSDR - Public Health Statement: Cadmium. Available at
513 https://www.atsdr.cdc.gov/phs/phs.asp?id=46&tid=15 (accessed January 29, 2018).
514 Bradham KD., Nelson CM., Kelly J., Pomales A., Scruton K., Dignam T., Misenheimer JC., Li
515 K., Obenour DR., Thomas DJ. 2017. Relationship Between Total and Bioaccessible Lead
516 on Children's Blood Lead Levels in Urban Residential Philadelphia Soils. Environmental
517 science & technology 51:10005–10011. DOI: 10.1021/acs.est.7b02058.
518 Broughton E. 2005. The Bhopal disaster and its aftermath: a review. Environmental Health 4:6.
519 DOI: 10.1186/1476-069X-4-6.
520 Brunetti P. 2011. Geología, Alteración y Mineralización del Yacimiento Tipo Pórfido Cuprífero
521 Pampa Lina, Distrito Sierra Gorda, Región Antofagasta. Memoria para optar al título de
522 geóloga Thesis. Santiago: Universidad de Chile. Available at:
523 http://repositorio.uchile.cl/handle/2250/104305 (accessed September 10, 2016).
524 Calderón C., Valdés J. 2012. Contenido de metales en sedimentos y organismos bentónicos de la

525 bahía San Jorge, Antofagasta, Chile. Revista de biología marina y oceanografía 47:121–
526 133. DOI: 10.4067/S0718-19572012000100011.
527 Cantallopts J. 2015. Chile en el mercado de los concentrados de cobre: Elementos para un
528 análisis estratégico. Available at: http://www.revistaquimica.cl/wp-
529 content/uploads/2015/12/Chile-en-el-mercado-de-concentrados-de-cobre.-J-
530 Cantallopts1.pdf (accessed June 25, 2016).
531 CENMA 2014. Diagnóstico regional de suelos abandonados con potencial presencia de
532 contaminantes. Available at: http://www.cenma.cl/Pagina%20web-LQA/4-
533 Gesti%C3%B3n%20de%20SPPC/2014-FNDR%20Antofagasta-
534 INFORME%20FINAL.pdf (accessed March 15, 2016).
535 Censo. 1895. Sesto censo jeneral de la población de Chile. Available at:
536 http://www.memoriachilena.cl/602/w3-article-82450.html (accessed April 12, 2016).
537 Censo. 1960. Población del país, Características básicas de la población. Available at:
538 http://www.memoriachilena.cl/602/w3-article-86206.html (accessed April 12, 2016).
539 Charney E., Kessler B., Farfel M., Jackson D. 1983. Childhood Lead Poisoning. New England
540 Journal of Medicine 309:1089–1093. DOI: 10.1056/NEJM198311033091804.
541 Clarke KR., Gorley RN. 2006. PRIMER v6: User Manual/Tutorial. Plymouth, UK: Plymouth
542 Marine Laboratory.
543 COCHILCO. 2014. Estadísticas - Producción Minera. Available at
544 http://www.cochilco.cl/estadisticas/produccion.asp (accessed July 8, 2016).
545 COCHILCO 2016. Anuario de estadísticas del cobre y otros minerales 1997-2016. Available at:
546 https://www.cochilco.cl/Paginas/Estadisticas/Publicaciones/Anuario.aspx (accessed
547 September 10, 2017).

548 Cortés I., Ríos J., Gaete L., Tchernitchin A. 2015. Metales pesados en muestras ambientales de la
549 Región de Atacama. Situación postaluviones. In: Geología para el siglo XXI. La Serena,
550 Chile, 302–305.
551 Dallas CE., Williams PL. 2001. Barium: rationale for a new oral reference dose. Journal of
552 Toxicology and Environmental Health. Part B, Critical Reviews 4:395–429. DOI:
553 10.1080/109374001753146216.
554 De Gregori I., Fuentes E., Rojas M., Pinochet H., Potin-Gautier M. 2003. Monitoring of copper,
555 arsenic and antimony levels in agricultural soils impacted and non-impacted by mining
556 activities, from three regions in Chile. Journal of environmental monitoring: JEM 5:287–
557 295.
558 De Solminihac H., Gonzales LE., Cerda R. 2017. Copper mining productivity: Lessons from
559 Chile. Journal of Policy Modeling. DOI: 10.1016/j.jpolmod.2017.09.001.
560 Devell L., Tovedal H., Bergström U., Appelgren A., Chyssler J., Andersson L. 1986. Initial
561 observations of fallout from the reactor accident at Chernobyl. Nature 321:192–193. DOI:
562 10.1038/321192a0.
563 Dubey CS., Sahoo BK., Nayak NR. 2001. Chromium (VI) in Waters in Parts of Sukinda
564 Chromite Valley and Health Hazards, Orissa, India. Bulletin of Environmental
565 Contamination and Toxicology 67:541–548. DOI: 10.1007/s001280157.
566 El Mercurio. 2010. Urgentes medidas por contaminación en “Ruta del Plomo” en Antofagasta. El
567 Mercurio de Antofagasta. Available at:
568 http://www.mercurioantofagasta.cl/prontus4_noticias/site/artic/20100729/pags/20100729000523.html (accessed July 8, 2016).
570 Ergin M., Saydam C., Baştürk Ö., Erdem E., Yörük R. 1991. Heavy metal concentrations in

surface sediments from the two coastal inlets (Golden Horn Estuary and İzmit Bay) of the northeastern Sea of Marmara. Chemical Geology 91:269–285. DOI: 10.1016/0009-2541(91)90004-B.

Faiz A., Weaver C., Walsh M. 1996. Air Pollution from Motor Vehicles Standards and Technologies for Controlling Emissions. Washington: The World Bank. Available at: http://documents.worldbank.org/curated/en/606461468739185298/Air-pollution-from-motor-vehicles-standards-and-technologies-for-controlling-emissions (accessed March 15, 2018).

Finley BL., Monnot AD., Paustenbach DJ., Gaffney SH. 2012. Derivation of a chronic oral reference dose for cobalt. Regulatory toxicology and pharmacology: RTP 64:491–503. DOI: 10.1016/j.yrtph.2012.08.022.

Fuentes G., Viñals J., Herreros O. 2009. Hydrothermal purification and enrichment of Chilean copper concentrates. Part 2: The behavior of the bulk concentrates. Hydrometallurgy 95:113–120. DOI: 10.1016/j.hydromet.2008.05.004.

Gale R. 1987. Immediate medical consequences of nuclear accidents: Lessons from chernobyl. JAMA 258:625–628. DOI: 10.1001/jama.1987.03400050067029.

Garza RAP., Titley SR., Pimentel F. 2001. Geology of the Escondida Porphyry Copper Deposit, Antofagasta Region, Chile. Economic Geology 96:307–324. DOI: 10.2113/gsecongeo.96.2.307.

Gobierno de Chile. 1980. Constitución de la República de Chile.

Herreros O., Fuentes G., Quiroz R., Vinals J. 2003. Lixiviación de concentrados de cobre con alto contenido de arsénico en medio cloro-cloruro. Revista de metalurgia 39:90–98.

INE 2017. Cantidad de personas por sexo y edad: Total Comunal Antofagasta. Available at:

http://www.censo2017.cl/mapa-resultados-comunas/ (accessed January 15, 2018).

ISP. 2014. Material particulado sedimentable de Antofagasta.

Jorquera H. 2009. Source apportionment of PM10 and PM2.5 at Tocopilla, Chile (22°05'S, 70°12'W). Environmental Monitoring and Assessment 153:235. DOI: 10.1007/s10661-008-0352-0.

Kamenov G., Macfarlane AW., Riciputi L. 2002. Sources of Lead in the San Cristobal, Pulacayo, and Potosí Mining Districts, Bolivia, and a Reevaluation of Regional Ore Lead Isotope Provinces. Economic Geology 97:573–592. DOI: 10.2113/gsecongeo.97.3.573.

Khan FI., Abbasi SA. 1999. Major accidents in process industries and an analysis of causes and consequences. Journal of Loss Prevention in the Process Industries 12:361–378. DOI: 10.1016/S0950-4230(98)00062-X.

Kong S., Lu B., Ji Y., Zhao X., Chen L., Li Z., Han B., Bai Z. 2011. Levels, risk assessment and sources of PM10 fraction heavy metals in four types dust from a coal-based city. Microchemical Journal 98:280–290. DOI: 10.1016/j.microc.2011.02.012.

Lépez I., Furet L., Aracena O. 2001. Población de Emerita Analoga (Stimpson 1857) en playas Amarilla y Rinconada, Antofagasta: Aspectos abióticos, bióticos y concentración de cobre. Gayana (Concepción) 65:55–76. DOI: 10.4067/S0717-65382001000100008.

Li Z., Feng X., Li G., Bi X., Zhu J., Qin H., Dai Z., Liu J., Li Q., Sun G. 2013. Distributions, sources and pollution status of 17 trace metal/metalloids in the street dust of a heavily industrialized city of central China. Environmental Pollution 182:408–416. DOI: 10.1016/j.envpol.2013.07.041.

Lu X., Wang L., Lei K., Huang J., Zhai Y. 2009. Contamination assessment of copper, lead, zinc, manganese and nickel in street dust of Baoji, NW China. Journal of Hazardous

617 Materials 161:1058–1062. DOI: 10.1016/j.jhazmat.2008.04.052.
618 Lucas C. 2001. Baia Mare and Baia Borsa Accidents: Cases of Severe Transboundary Water
619 Pollution. Environmental Policy and Law 31:106.
620 Lucassen F., Franz G. 1994. Arc related Jurassic igneous and meta-igneous rocks in the Coastal
621 Cordillera of northern Chile/Region Antofagasta. Lithos 32:273–298. DOI: 10.1016/0024-
622 4937(94)90044-2.
623 Luo X-S., Ding J., Xu B., Wang Y-J., Li H-B., Yu S. 2012. Incorporating bioaccessibility into
624 human health risk assessments of heavy metals in urban park soils. Science of The Total
625 Environment 424:88–96. DOI: 10.1016/j.scitotenv.2012.02.053.
626 Marshall G., Ferreccio C., Yuan Y., Bates MN., Steinmaus C., Selvin S., Liaw J., Smith AH.
627 2007. Fifty-Year Study of Lung and Bladder Cancer Mortality in Chile Related to Arsenic
628 in Drinking Water. Journal of the National Cancer Institute 99:920–928. DOI:
629 10.1093/jnci/djm004.
630 Massey DD., Kulshrestha A., Taneja A. 2013. Particulate matter concentrations and their related
631 metal toxicity in rural residential environment of semi-arid region of India. Atmospheric
632 Environment 67:278–286. DOI: 10.1016/j.atmosenv.2012.11.002.
633 Minería Chilena. 2013. Barrio Industrial La Negra se consolida como eje estratégico de
634 servicios. Available at http://www.mch.cl/2013/07/11/barrio-industrial-la-negra-se-
635 consolida-como-eje-estrategico-de-servicios/ (accessed July 28, 2016).
636 Minería Chilena. 2014.Empezó producción de cobre en la mina de Sierra Gorda. Available at
637 http://www.mch.cl/2014/07/31/empezo-produccion-de-cobre-en-la-mina-de-sierra-gorda/
638 (accessed August 2, 2016).
639 MTT. 2015. Comisión especial investigadora de la actuación de los organismos públicos

encargados de la salud y medio ambiente en la Comuna de Antofagasta. Available at: https://www.camara.cl/pdf.aspx?prmID=32851&prmTIPO=DOCUMENTOCOMISION (accessed December 15, 2017).

Muller G. 1979. Schwermetalle in den Sedimenten des Rheins - Veränderungen seit. Umschau 79:778–783.

Nowrouzi M., Pourkhabbaz A. 2014. Application of geoaccumulation index and enrichment factor for assessing metal contamination in the sediments of Hara Biosphere Reserve, Iran. Chemical Speciation & Bioavailability 26:99–105. DOI: 10.3184/095422914X13951584546986.

Oliveros V., Morata D., Aguirre L., Féraud G., Fornari M. 2007. Magmatismo asociado a subducción del Jurásico a Cretácico Inferior en la Cordillera de la Costa del norte de Chile (18°30'-24°S): geoquímica y petrogenesis. Revista geológica de Chile 34:209–232. DOI: 10.4067/S0716-02082007000200003.

RAIS. 1991. Formal Toxicity Summary for VANADIUM. Available at https://rais.ornl.gov/tox/profiles/vanadium_f_V1.html (accessed August 10, 2016).

Reddy K., Yarrakula K. 2016. Analysis of accidents in chemical process industries in the period 1998-2015. International Journal of ChemTech Research 9:177–191.

Rogers G., Hawkesworth CJ. 1989. A geochemical traverse across the North Chilean Andes: evidence for crust generation from the mantle wedge. Earth and Planetary Science Letters 91:271–285. DOI: 10.1016/0012-821X(89)90003-4.

Rudnick RL., Gao S. 2003. 3.01 - Composition of the Continental Crust A2 - Holland, Heinrich D. In: Turekian KK ed. Treatise on Geochemistry. Oxford: Pergamon, 1–64.

Ruiz-Rudolph P., Arias N., Pardo S., Meyer M., Mesías S., Galleguillos C., Schiattino I.,

663 Gutiérrez L. 2016. Impact of large industrial emission sources on mortality and morbidity
664 in Chile: A small-areas study. Environment International 92–93:130–138. DOI:
665 10.1016/j.envint.2016.03.036.
666 Rule JH. 1986. Assessment of trace element geochemistry of Hampton Roads harbor and lower
667 Chesapeake Bay area sediments. Environmental Geology and Water Sciences 8:209–219.
668 DOI: 10.1007/BF02524948.
669 Salamanca MA., Camaño A., Jara B., Rodríguez T. 2000. Distribución de Cu, Pb y Zn en aguas
670 costeras de Bahía San Jorge en el norte de Chile. Gayana (Concepción) 64:195–204. DOI:
671 10.4067/S0717-65382000000200009.
672 Salamanca MA., Jara B., Rodríguez T. 2004. Niveles de Cu, Pb y Zn en agua y Perumytilus
673 Purpuratus en Bahía San Jorge, Norte de Chile. Gayana (Concepción) 68:53–62. DOI:
674 10.4067/S0717-65382004000100005.
675 Sepúlveda V., Vega J., Delgado I. 2000. Exposición severa a plomo ambiental en una población
676 infantil de Antofagasta, Chile. Revista médica de Chile 128:221–232. DOI:
677 10.4067/S0034-98872000000200014.
678 SERNAGEOMIN. 2003. Mapa geológico de Chile: versión digital. Available at:
679 http://www.ipgp.fr/~dechabal/Geol-millon.pdf (accessed July 15, 2017).
680 SERNAGEOMIN. 2014. Geología para el ordenamiento territorial: área Antofagasta. Available
681 at: http://biblioteca.sernageomin.cl/opac/DataFiles/CGCH_SGA_19_text.pdf (accessed
682 November 10, 2017).
683 Smith AH., Steinmaus CM. 2009. Health Effects of Arsenic and Chromium in Drinking Water:
684 Recent Human Findings. Annual Review of Public Health 30:107–122. DOI:
685 10.1146/annurev.publhealth.031308.100143.

686 Soldán P., Pavonič M., Bouček J., Kokeš J. 2001. Baia Mare Accident—Brief Ecotoxicological
687 Report of Czech Experts. Ecotoxicology and Environmental Safety 49:255–261. DOI:
688 10.1006/eesa.2001.2070.
689 SYSTAT 2009. SYSTAT Software Inc. Richmond, CA, USA.
690 Tapia J., Arriagada C., Townley B., Luca R., Nacif D. 2009. Estudio piloto utilizando
691 propiedades magnéticas de partículas de polvo depositado en hojas de *Platanus orientalis*
692 *L*. para determinar contaminación atmosférica.
693 Tapia, J., Audry, S., Townley, B., Duprey, JL. 2012. Geochemical background, baseline and
694 origin of contaminants from sediments in the mining-impacted Altiplano and Eastern
695 Cordillera of Oruro, Bolivia. GEEA, 12(1), 3-20.
696 Tchernitchin A., Bolados A. 2016. Informe sobre material particulado sedimentable (MPS) en
697 colegios cercanos al puerto de Antofagasta, muestras 2016. Available at:
698 http://www.colegiomedico.cl/wp-content/uploads/2016/05/MaCM16a5doc-AFB-MPS-
699 colegios-Antof-Informe-27-10-2015.pdf (accessed July 10, 2016).
700 Tchernitchin AN., Gaete L. 2015. Prenatal Exposures to Environmental Agents or Drugs
701 Promote the Development of Diseases Later in Life. Biology and Medicine 7. DOI:
702 10.4172/0974-8369.1000236.
703 Tchernitchin A., Gaete L., Bustamante R., Sorokin Y. 2013. Adulthood Prenatally Programmed
704 Diseases: Health Relevance and Methods of Study. In: Protein purification and analysis I.
705 Methods and applications. Hong Kong, 217–258.
706 Tchernitchin AN., Lapin N., Molina L., Molina G., Tchernitchin NA., Acevedo C., Alonso P.
707 2006. Human Exposure to Lead in Chile. In: Reviews of Environmental Contamination
708 and Toxicology. Reviews of Environmental Contamination and Toxicology. Springer, New

York, NY, 93–139. DOI: 10.1007/0-387-30638-2_4.

US EPA 1989. Risk assessment guidance for superfund Volume I human health evaluation manual (Part A). Available at: https://www.epa.gov/risk/risk-assessment-guidance-superfund-rags-part (accessed November 2, 2016).

US EPA 1991. Nickel, soluble salts. Available at https://cfpub.epa.gov/ncea/iris2/chemicalLanding.cfm?substance_nmbr=271 (accessed August 10, 2016).

US EPA O. 2004.Lead and compounds (inorganic) CASRN 7439-92-1 | IRIS | US EPA, ORD. Available at https://cfpub.epa.gov/ncea/iris2/chemicalLanding.cfm?substance_nmbr=0277 (accessed August 9, 2016).

US EPA, IRIS 1989.Cadmium CASRN 7440-43-9 | IRIS | US EPA, ORD. Available at https://cfpub.epa.gov/ncea/iris2/chemicalLanding.cfm?substance_nmbr=141 (accessed August 10, 2016).

US EPA., IRIS. 1991.Arsenic, inorganic CASRN 7440-38-2 | IRIS | US EPA, ORD. Available at https://cfpub.epa.gov/ncea/iris2/chemicalLanding.cfm?substance_nmbr=278 (accessed August 9, 2016).

US EPA, IRIS 1992.Molybdenum CASRN 7439-98-7 | IRIS | US EPA, ORD. Available at https://cfpub.epa.gov/ncea/iris2/chemicalLanding.cfm?substance_nmbr=425 (accessed August 10, 2016).

US EPA, IRIS. 1998.Chromium(VI) CASRN 18540-29-9 | IRIS | US EPA, ORD. Available at https://cfpub.epa.gov/ncea/iris2/chemicalLanding.cfm?substance_nmbr=144 (accessed August 10, 2016).

US EPA, IRIS 2005.Zinc and Compounds CASRN 7440-66-6 | IRIS | US EPA, ORD. Available

732 at https://cfpub.epa.gov/ncea/iris2/chemicalLanding.cfm?substance_nmbr=426 (accessed
733 August 10, 2016).
734 Valdés J., Guiñez M., Castillo A., Vega SE. 2014. Cu, Pb, and Zn content in sediments and
735 benthic organisms from San Jorge Bay (northern Chile): Accumulati on and
736 biotransference in subtidal coastal systems. Ciencias Marinas 40:45–58. DOI:
737 http://dx.doi.org /10.7773/cm.v40i1.2318.
738 Valdés J., Román D., Guiñez M., Rivera L., Ávila J., Cortés P., Castillo A. 2015. Trace metal
739 variability in coastal waters of San Jorge Bay, Antofagasta, Chile: An environmental
740 evaluation and statistical approach to propose local background levels. Marine Pollution
741 Bulletin 100:544–554. DOI: 10.1016/j.marpolbul.2015.08.035.
742 Valdés J., Román D., Guiñez M., Rivera L., Morales T., Ávila J., Cortés P. 2010. Distribution
743 and temporal variation of trace metal enrichment in surface sediments of San Jorge Bay,
744 Chile. Environmental Monitoring and Assessment 167:185–197. DOI: 10.1007/s10661-
745 009-1041-3.
746 Valdés J., Román D., Rivera L., Ávila J., Cortés P. 2011. Metal contents in coastal waters of San
747 Jorge Bay, Antofagasta, northern Chile: a base line for establishing seawater quality
748 guidelines. Environmental Monitoring and Assessment 183:231. DOI: 10.1007/s10661-
749 011-1917-x.
750 Vergara P. 2015.Contaminación de arsénico y plomo: Un conflicto que crece en Antofagasta.
751 Available at http://www.cooperativa.cl/noticias/pais/region-de-antofagasta/contaminacion-
752 plomo/contaminacion-de-arsenico-y-plomo-un-conflicto-que-crece-en-antofagasta/2015-
753 02-04/140741.html (accessed July 27, 2016).
754 Zhu Z., Sun G., Bi X., Li Z., Yu G. 2013. Identification of trace metal pollution in urban dust

755 from kindergartens using magnetic, geochemical and lead isotopic analyses. Atmospheric
756 Environment 77:9–15. DOI: 10.1016/j.atmosenv.2013.04.053.
757 Zoller WH., Gladney ES., Duce RA. 1974. Atmospheric Concentrations and Sources of Trace
758 Metals at the South Pole. Science 183:198–200. DOI: 10.1126/science.183.4121.198.

Figure 1

Antofagasta location

Location maps with features, at varying scales. (A) Location of the Antofagasta Region in northern Chile; (B) Main mineral deposits and mining activities within Antofagasta Region (1. Mantos de la Luna Mine; 2. Michilla Mine; 3. Juanita Mine; 4. Mantos Blancos Mine; 5. La Negra Industrial Complex; 6. Pedro de Valdivia (ex-nitrate mine); 7. El Abra Mine; 8. Radomiro Tomic, Chuquicamata, and Ministro Hales Mines; 9. Faride, Spence, Sierra Gorda Mines, and Aconcagua treatment plant; 10. Esperanza Mine; 11. Zaldívar and La Escondida Mines; 12. Francke Mine), and Antofagasta city (black square); (C) Downtown Antofagasta. A. Port gate; B. Clínica Antofagasta (health institution); C. Parque Brasil (Children's playground); D. Commercial center; E and F. Schools (Liceo de Hombres and Liceo de Niñas, respectively); G. Housing complex; H. City Mall; I. Municipal square; J. Hospital (health institution); K. Fruit and vegetable market; L. Supermarket. The red circle represents an area with a 1 km distance (radius) from the port gate.

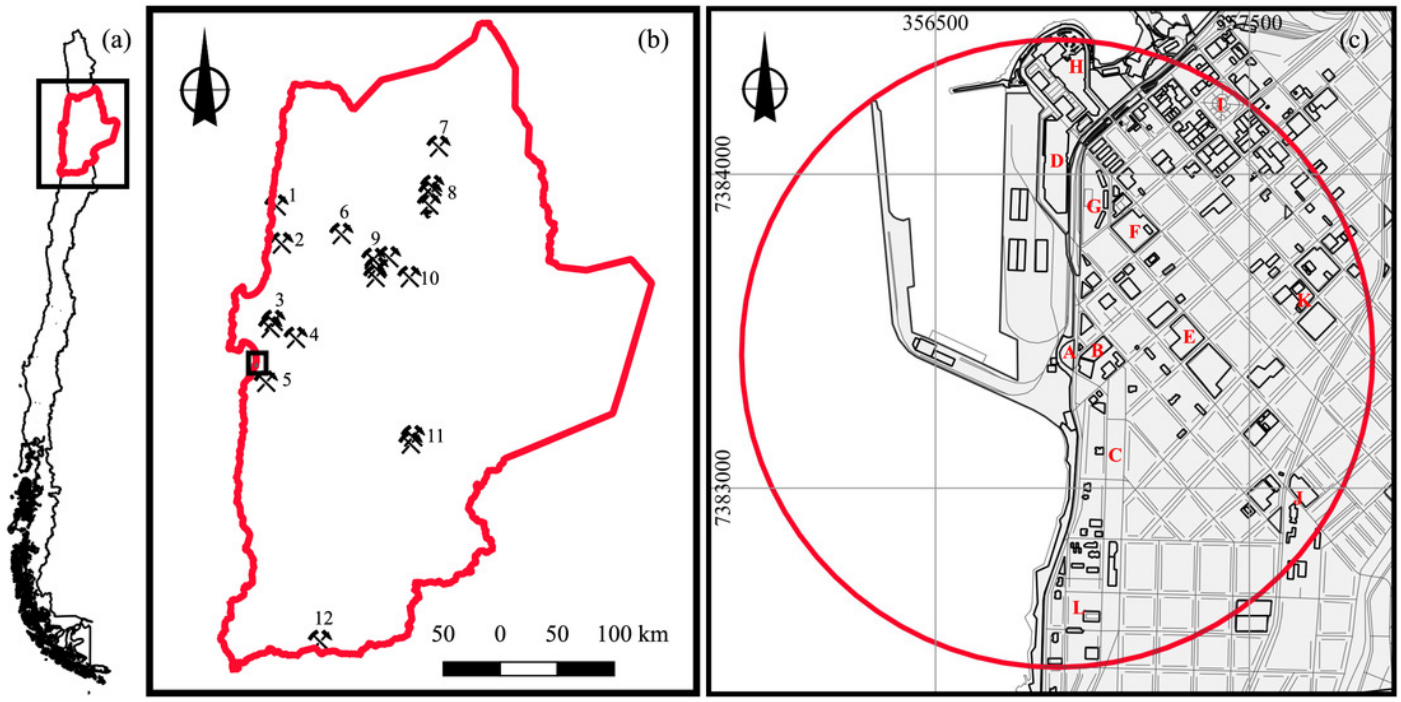

Figure 2

Antofagasta geology

Locations with geochemical data. (A) Regional volcanic, intrusive, metamorphic rocks, dikes, and soils found in the vicinity of the city of Antofagasta; (B) Local geology and dust samples from the city of Antofagasta. The red circle represents an area with a 1 km distance (radius) from the port.

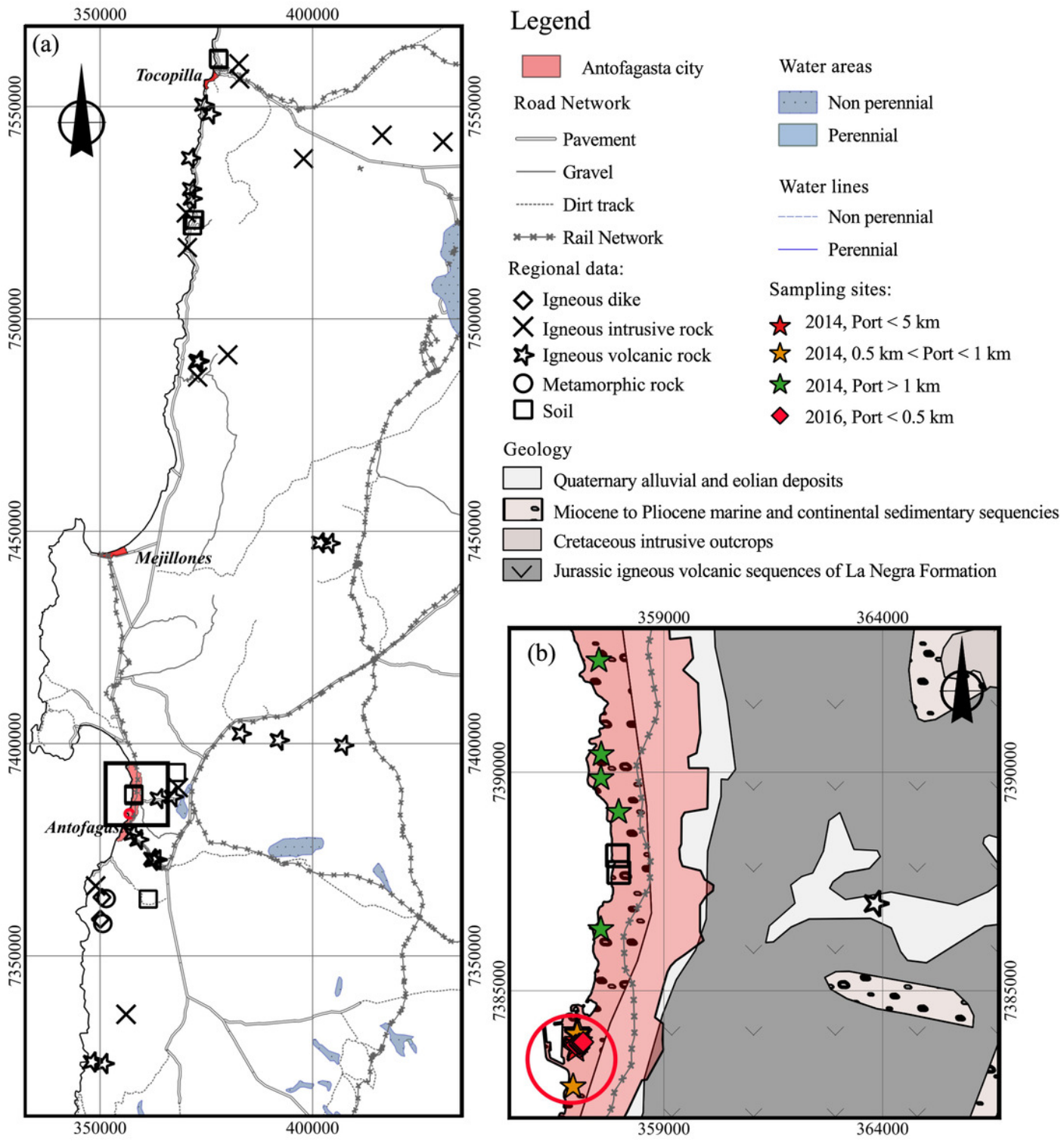
(a)
Tocopilla
Mejillones
Antofagasta
350000
400000
7550000
7500000
7450000
7400000
7350000
Legend
Antofagasta city
Road Network
Pavement
Gravel
Dirt track
Rail Network
Regional data:
Igneous dike
Igneous intrusive rock
Igneous volcanic rock
Metamorphic rock
Soil
Water areas
Non perennial
Perennial
Water lines
Non perennial
Perennial
Sampling sites:
2014, Port < 5 km
2014, 0.5 km < Port < 1 km
2014, Port > 1 km
2016, Port < 0.5 km
Geology
Quaternary alluvial and eolian deposits
Miocene to Pliocene marine and continental sedimentary sequencies
Cretaceous intrusive outcrops
Jurassic igneous volcanic sequences of La Negra Formation
(b)
359000
364000
7390000
7385000

Figure 3

Box plots of Antofagasta dust

Element concentrations versus distance from the Antofagasta Port (AP). The Box-Whisker plots show the variation in metal concentrations in street dust samples collected at various locations in Antofagasta based on their relative distance from the Port of Antofagasta (nb: In the box-whisker plots, the center vertical line shows the median value, while the length of each box shows the range within which the central 50% of the values fall, with the box edges showing the first and third quartiles (the interquartile range). Whiskers show values that the range of observed values that fall within 1.5 x the interquartile range. The y-axis shows a $\log_{10}$-scale which differs between individual figures). Test statistics reflect results of the Kruskal-Wallis non-parametric ANOVA.

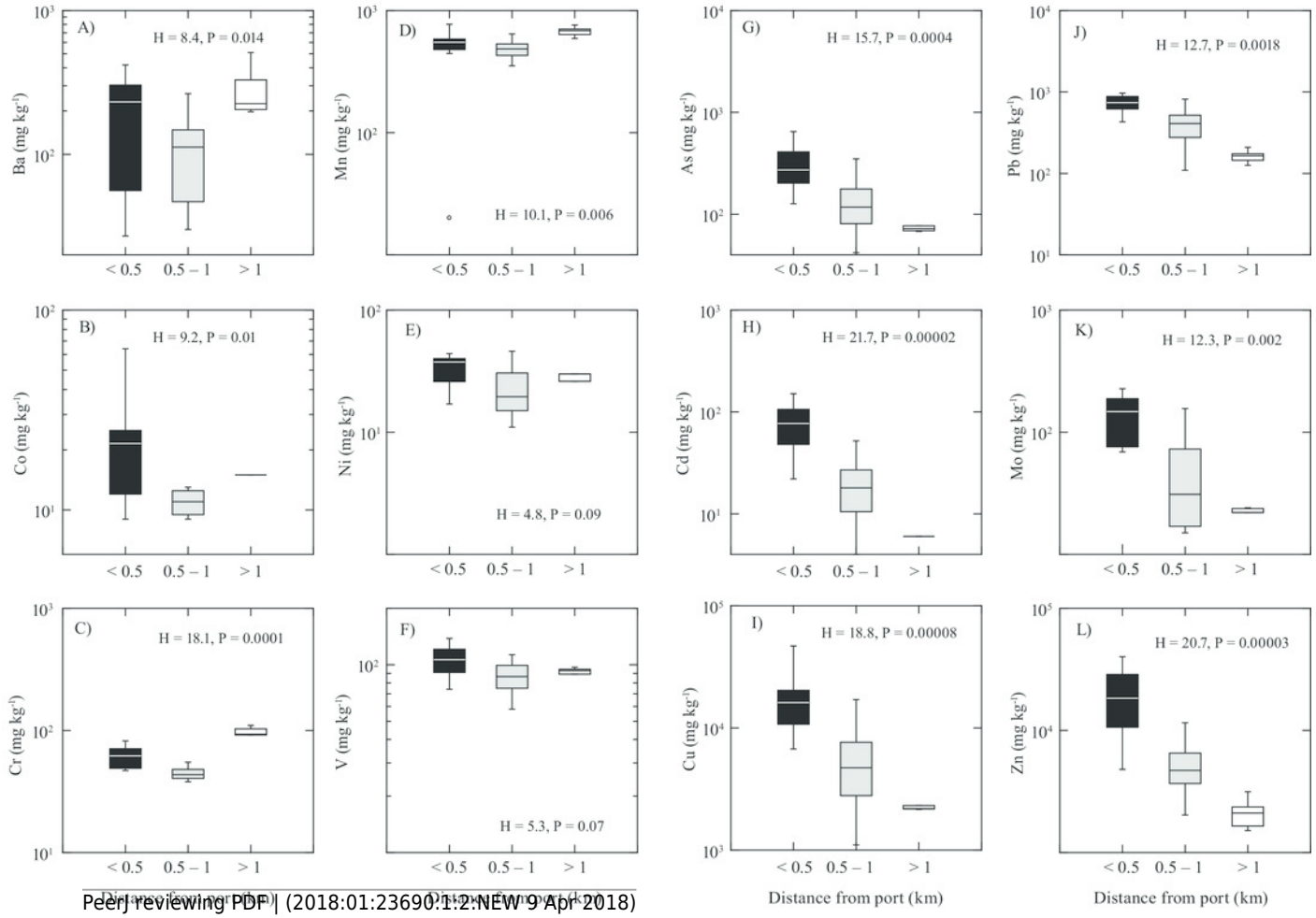

Figure 4

Principal component analysis of Antofagasta dust

Principal component analysis (PCA) ordination including vectors showing relative correlation strength between principal components (PCs) and concentrations of different metals in street dust collected in Antofagasta. The circles are filled with distinct colors to show their relative proximity to the main gate of the Port of Antofagasta.

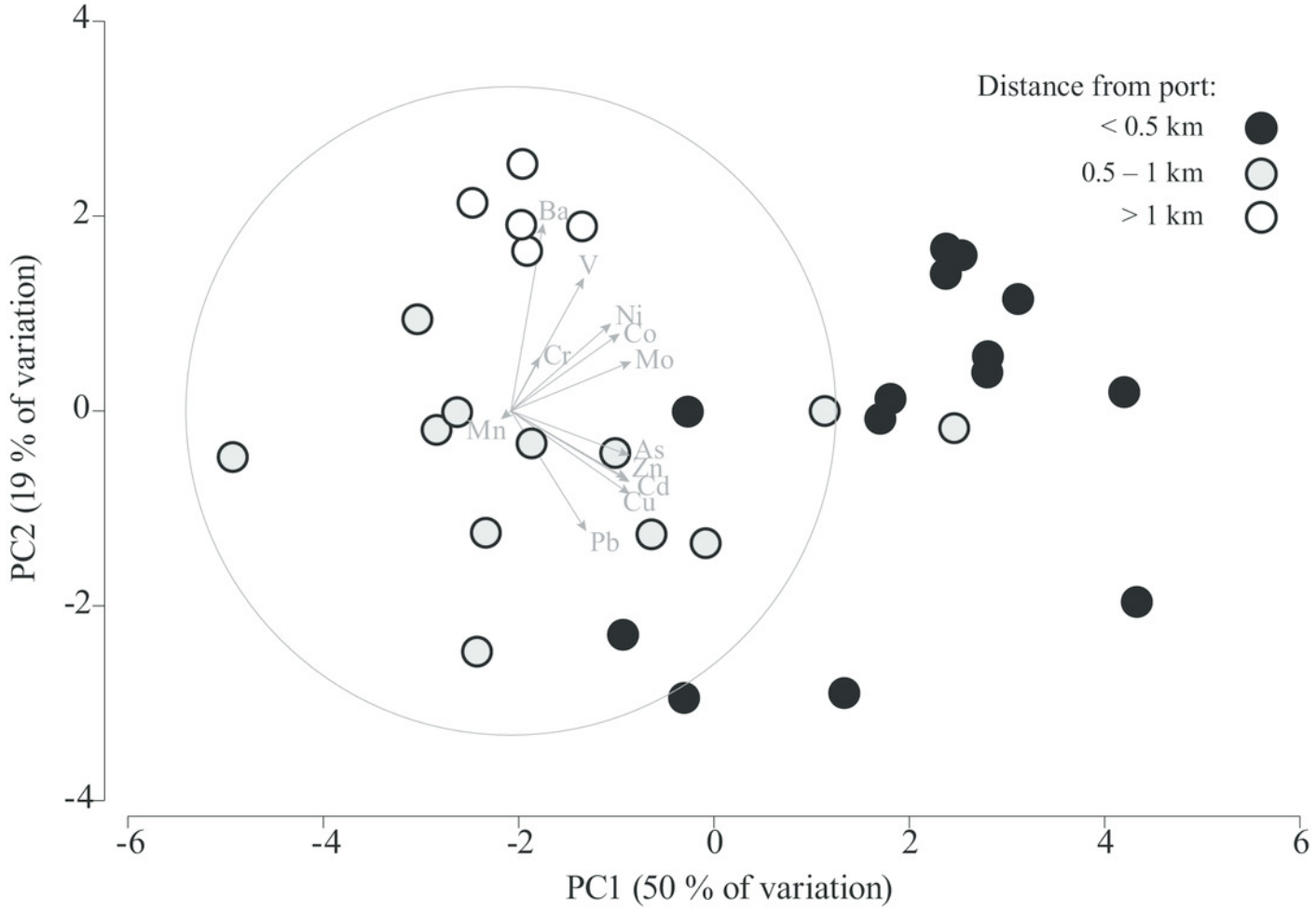

Table 1(on next page)

Sampling sites

Summary of street dust sampling locations: name of streets, sampling years, sampling institutions, UTM coordinates (WGS-84), and elevations (m. a.s.l.). AP: distance to the Antofagasta Port in kilometers (km).

Table 1. Summary of street dust sampling locations: name of streets, sampling years, sampling institutions, UTM coordinates (WGS-84), and elevations (m.a.s.l.). AP: distance to the Antofagasta Port in kilometers (km).

Sample	Location	Year	Institution	UTM E	UTM N	Elevation (m.a.s.l)	AP (km)
1	Grecia/Salvador Reyes	2014	ISP	356925	7382802	10	0.62
2	Grecia/Salvador Reyes	2014	ISP	356925	7382802	10	0.62
3	Grecia 1816/21 Mayo	2014	ISP	356973	7383725	8	0.31
4	Grecia/21 Mayo	2014	ISP	357075	7383834	11	0.45
5	Uribe/Balmaceda Pabellón 1	2014	ISP	357008	7384065	9	0.65
6	Uribe/Balmaceda Pabellón 1	2014	ISP	357008	7384065	9	0.65
7	Uribe/Pabellón 2	2014	ISP	357025	7384049	10	0.64
8	Uribe/Pabellón 2	2014	ISP	357025	7384049	10	0.64
9	Uribe/Pabellón 3	2014	ISP	357049	7384024	10	0.62
10	Uribe/Pabellón 3	2014	ISP	357049	7384024	10	0.62
11	Uribe/Washington Pabellón 4	2014	ISP	357062	7384012	10	0.61
12	Uribe/Washington Pabellón 4	2014	ISP	357062	7384012	10	0.61
13	MOP building in front of the Port	2014	ISP	356969	7383667	7	0.26
14	MOP building inside in front of the Port	2014	ISP	356988	7383637	8	0.24
15	Liceo Técnico in front of the Port	2014	ISP	357027	7383797	11	0.40
16	Colegio Técnico, frontis	2014	ISP	356996	7383771	10	0.37
17	Edificio Colectivo Argentina	2014	ISP	357023	7383850	10	0.45
21	Edificio Colectivo Perú 1er piso	2014	ISP	357027	7383967	10	0.56
22	Edificio Colectivo Perú 2o piso	2014	ISP	357017	7384003	10	0.59
23	14 de Febrero/Edmundo Pérez Zujovic	2014	ISP	357560	7386404	12	3.10
24	Av. Edmundo Pérez Zujovic 7344	2014	ISP	357968	7389089	14	5.77
25	Av. Edmundo Pérez Zujovic 8126-9114	2014	ISP	357562	7389860	13	6.47
26	Colegio San Agustín	2014	ISP	357568	7390407	8	7.02
27	Condominio Jardines del Norte VI	2014	ISP	357514	7392555	18	9.15
AF-1	Jardín Infantil Semillita	2016	CM	357072	7383698	9	0.33
AF-2	Jardín Infantil Semillita	2016	CM	357072	7383698	9	0.33
AF-3	Ex Liceo Técnico A14	2016	CM	356986	7383759	9	0.35
AF-4	Lice Técnico A14	2016	CM	357071	7383760	10	0.38
AF-5	Lice Técnico A14	2016	CM	357071	7383790	11	0.41
AF-6	Liceo Marta Narea A1	2016	CM	357128	7383791	12	0.44
AF-7	Liceo Marta Narea A1	2016	CM	357156	7383822	12	0.48

Table 2(on next page)

Summary of analytical methodologies utilized in previous studies

Summary of analytical methodologies utilized in previous studies. Antofagasta dust: ISP (2014) and Tchernitchin & Bolados (2016); rocks: Lucassen & Franz (1994), Oliveros et al. (1997), and Rogers & Hawkesworth (1989); soil and sediments: CENMA (2014) and De Gregori et al. (2003). N/A: information not provided.

Table 2. Summary of analytical methodologies utilized in previous studies. Antofagasta dust: ISP (2014) and Tchernitchin & Bolados (2016); rocks: Lucassen & Franz (1994), Oliveros et al. (1997), and Rogers & Hawkesworth (1989); soil and sediments: CENMA (2014) and De Gregori et al. (2003). N/A: information not provided.

	Detection limit ($mg \cdot kg^{-1}$)												Instrument	Laboratory
	As	Ba	Cd	Co	Cr	Cu	Mn	Mo	Ni	Pb	V	Zn		
Antofagasta Dust														
ISP 2014	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	ICP	ISP, Chile
Tchernitchin and Bolados (2016)	1.7	2.1	1.0	0.8	1.8	2.1	2.5	0.9	2.2	2.1	0.7	3.2	ICP-OES	CENMA, Chile
Rocks of Antofagasta														
Lucassen and Franz (1994)	N/A												Major elements XRF and trace element ICP	TU-Berlin
Oliveros et al. (2007)	1.0	3.0	0.4	0.2	4.0	0.2	3.9	0.5	4.0	1.2	1.5	8.0	ICP-AES and ICP-MS	CRPG, France
Rogers and Hawkesworth (1989)	N/A												Major elements ED XRF and trace elements WD XRF	Open University and Nottingham University
Soil/sediments														
CENMA 2014	14.6	8.9	1.8	2.4	4.6	1.0	8.5	8.3	1.4	3.7	1.5	2.7	ICP-OES	CENMA, Chile
De Gregori et al. 2003	N/A												FAAS (Cu), HG-AFS (As)	N/A

Table 3(on next page)

Basic statistical summary of Antofagasta dust

Basic statistical summary. Number of data (n), mean, standard deviation (σ), relative σ (σ ÷ mean × 100), median, range, lower and upper 95% confidence interval. All values are in $mg \cdot kg^{-1}$. AP: Antofagasta port; Antofagasta dust data from CM (Tchernitchin and Bolados, 2016) and ISP (ISP, 2014); UCC: upper continental crust values (Rudnick and Gao, 2003).

Table 3. Basic statistical summary. Number of data (n), mean, standard deviation (σ), variability (σ/average·100), median, range, lower and upper 95% confidence interval. All values are in mg·kg^{-1}. AP: Antofagasta port; Antofagasta dust data from CM (2016) and ISP (2014); UCC: upper continental crust values (Rudnick and Gao, 2003).

	Distance to AP km	n	Mean (mg·kg^{-1})	σ (mg·kg^{-1})	Relative σ %	Median (mg·kg^{-1})	Range (mg·kg^{-1})		95% confidence limits (mg·kg^{-1})		Correlation to distance to AP	2016-2014 (mg·kg^{-1})	UCC (mg·kg^{-1})	Element /UCC
As	< 0.5	14	376	347	92	272	127	1492	175	577	-0.46	-186	4.8	78
	0.5 - 1	12	148	99	67	117	42	350	81	194	-0.63	-		31
	> 1	5	77	14	18	72	68	101	40	94	0.18	-		16
	< 0.5 to > 1	31	239	269	112	175	42	1492	141	338	-0.28	-		50
Ba	< 0.5	14	216	136	63	234	27	418	137	294	0.33	207	624	0.3
	0.5 - 1	12	112	71	63	113	30	264	67	157	-0.71	-		0.2
	> 1	5	293	132	45	225	198	510	129	457	0.50	-		0.5
	< 0.5 to > 1	31	188	129	69	181	27	510	141	236	0.39	-		0.3
Cd	< 0.5	14	81	37	46	77	22	151	59	102	-0.25	35	0.09	895
	0.5 - 1	12	20	13	68	18	4	52	11	28	-0.48	-		221
	> 1	5	6	1	17	6	4	7	4	7	-0.56	-		63
	< 0.5 to > 1	31	45	42	93	28	4	151	30	60	-0.43	-		500
Co	< 0.5	14	22	14	61	21	9	64	15	30	0.00	16	17.3	1.3
	0.5 - 1	12	12	4	34	11	6	20	9	14	0.01	-		0.7
	> 1	5	15	1	8	15	13	15	13	16	-0.76	-		0.8
	< 0.5 to > 1	31	17	11	62	15	6	64	13	21	-0.13	-		1.0
Cr	< 0.5	14	62	12	19	62	47	82	55	69	-0.68	-2	92	0.7
	0.5 - 1	12	46	12	25	43	28	76	38	53	-0.04	-		0.5
	> 1	5	94	15	16	93	71	110	76	112	-0.95	-		1.0
	< 0.5 to > 1	31	61	20	33	55	28	110	53	68	0.57			0.7
Cu	< 0.5	14	17914	9897	55	16114	6725	46898	12199	23628	-0.62	-3044	28	640
	0.5 - 1	12	6103	5059	83	4735	1103	17047	2889	9317	-0.61	-		218
	> 1	5	2287	187	8	2188	2153	2601	2054	2519	-0.06	-		82

	< 0.5 to > 1	31	10821	9816	91	7874	1103	46898	7221	14422	-0.40	-		386
Mn	< 0.5	14	540	183	34	550	20	771	435	646	-0.20	-18	774	0.7
	0.5 - 1	12	475	98	21	485	280	643	412	537	0.28	-		0.6
	> 1	5	675	66	10	683	590	761	594	756	0.70	-		0.9
	< 0.5 to > 1	31	537	153	28	545	20	771	481	593	0.40	-		0.7
Mo	< 0.5	14	128	70	55	147	12	227	88	169	-0.11	93	1.1	117
	0.5 - 1	12	35	48	135	15	5	156	5	66	-0.74	-		32
	> 1	5	22	2	9	22	19	24	19	25	0.87	-		20
	< 0.5 to > 1	31	75	74	98	31	5	227	48	102	-0.33	-		68
Ni	< 0.5	14	33	9	29	37	44	17	27	38	0.01	11	47	0.7
	0.5 - 1	12	23	11	49	20	11	46	16	30	-0.50	-		0.5
	> 1	5	32	5	16	30	26	38	26	39	-0.61	-		0.7
	< 0.5 to > 1	31	29	11	36	30	11	46	25	33	0.06	-		0.6
Pb	< 0.5	14	1071	1091	102	739	28	3968	441	1700	-0.71	-936	17	63
	0.5 - 1	12	518	481	93	412	109	1924	212	824	-0.49	-		30
	> 1	5	164	31	19	165	125	209	125	203	-0.87	-		10
	< 0.5 to > 1	31	710	852	120	486	28	3968	398	1023	-0.30	-		42
V	< 0.5	14	104	23	22	106	57	138	91	117	0.21	33	97	1.1
	0.5 - 1	12	86	17	20	87	58	113	75	97	0.01	-		0.9
	> 1	5	90	8	9	93	77	97	80	100	0.86	-		0.9
	< 0.5 to > 1	31	95	20	21	94	57	138	87	102	-0.07	-		1.0
Zn	< 0.5	14	20351	10378	51	18320	4792	40062	14359	26343	-0.15	11392	67	304
	0.5 - 1	12	6022	3943	65	4721	2029	15868	3517	8527	0.04	-		90
	> 1	5	2155	653	30	2102	1513	3149	1344	2965	-0.46	-		32
	< 0.5 to > 1	31	11869	10743	91	6543	1513	40062	7929	15810	-0.41	-		177

Table 4(on next page)

Geo-accumulation index of Antofagasta dust

Geo-accumulation index (Muller, 1979). $I_{geo} \leq 0$ non-contaminated; $0 < I_{geo} \leq 1$ non- to slightly contaminated; $1 < I_{geo} \leq 2$ moderately contaminated; $2 < I_{geo} \leq 3$ moderately to highly contaminated; $3 < I_{geo} \leq 4$ highly contaminated; $4 < I_{geo} \leq 5$ highly to extremely contaminated; $I_{geo} > 5$ extremely contaminated. Note: below the sample type, distances are given from the Antofagasta Port.

Table 4. Geo-accumulation index (Muller, 1979). $I_{geo} \leq 0$ non-contaminated; $0 < I_{geo} \leq 1$ non- to slightly contaminated; $1 < I_{geo} \leq 2$ moderately contaminated; $2 < I_{geo} \leq 3$ moderately to highly contaminated; $3 < I_{geo} \leq 4$ highly contaminated; $4 < I_{geo} \leq 5$ highly to extremely contaminated; $I_{geo} > 5$ extremely contaminated. Note: below the sample type, distances are given from the Antofagasta Port.

	Soil			Metamorphic rocks			Volcanic rocks			Intrusive rocks			Igneous dikes		
	< 0.5 km	0.5-1 km	> 1 km	< 0.5 km	0.5-1 km	> 1 km	< 0.5 km	0.5-1 km	> 1 km	< 0.5 km	0.5-1 km	> 1 km	< 0.5 km	0.5-1 km	> 1 km
As	4	3	2	-	-	-	5	4	3	7	6	5	-	-	-
Ba	-	-	-	0	-1	0	-1	-2	-1	-1	-2	0	-1	-2	0
Cd	5	3	1	-	-	-	-	-	-	-	-	-	-	-	-
Co							-1	-2	-1	-1	-2	-1	-	-	-
Cr	1	0	1	0	-1	0	0	-1	0	-1	-2	-1	-1	-2	-1
Cu	6	5	4	7	6	4	7	6	4	7	6	4	6	5	4
Mn	-	-	-	-2	-2	-1	-2	-2	-2	-1	-2	-1	-1	-1	-1
Mo	-	-	-	-	-	-	6	4	3	5	4	3	-	-	-
Ni	0	0	0	0	-1	0	0	-1	0	0	-1	0	0	-1	0
Pb	6	5	3	-	-	-	6	5	4	8	7	5	-	-	-
V	-	-	-	-2	-2	-2	-2	-2	-2	-2	-2	-2	-2	-2	-2
Zn	7	5	4	7	6	4	6	4	3	8	6	5	8	6	5

Table 5(on next page)

Enrichment Factors obtained in Antofagasta dust

Enrichment Factors obtained in Antofagasta from volcanic, intrusive, and metamorphic rocks as well as from dikes and soils. Values close to 1 indicate that the material originates from the same parent material (Zoller et al., 1974). High values signify that they do not originate from that parent material. Note: below the sample type, distances are given from the Antofagasta Port.

Table 5. Enrichment Factors obtained in Antofagasta from volcanic, intrusive, and metamorphic rocks as well as from dikes and soils. Values close to 1 indicate that the material originates from the same parent material (Zoller et al., 1974). High values signify that they do not originate from that parent material. Note: below the sample type, distances are given from the Antofagasta Port.

	Soil			Metamorphic rocks			Volcanic rocks			Intrusive rocks			Igneous dikes		
	< 0.5 km	0.5-1 km	> 1 km	< 0.5 km	0.5-1 km	> 1 km	< 0.5 km	0.5-1 km	> 1 km	< 0.5 km	0.5-1 km	> 1 km	< 0.5 km	0.5-1 km	> 1 km
As	7	4	2	-	-	-	91	50	22	235	130	55	-	-	-
Ba	-	-	-	2	1	3	1	1	2	1	1	2	3	2	4
Cd	20	7	2	-	-	-	-	-	-	-	-	-	-	-	-
Co	-	-	-	-	-	-	1	1	1	1	1	1	-	-	-
Cr	1	1	1	2	2	3	2	2	3	1	1	1	2	2	3
Cu	45	21	7	313	150	46	340	163	50	236	113	35	364	174	53
Mn	-	-	-	1	1	1	1	1	1	1	1	1	2	2	2
Mo	-	-	-	-	-	-	135	52	26	70	27	14			
Ni	1	1	1	2	2	2	2	2	2	2	2	2	4	4	4
Pb	35	24	6	-	-	-	196	133	34	365	248	64	-	-	-
V	-	-	-	1	1	1	1	1	1	1	1	1	1	2	1
Zn	74	31	9	365	151	44	172	71	21	476	198	58	934	388	113

Table 6(on next page)

Chemical daily intake and Hazard Index of Antofagasta dust

Chemical daily intake and Hazard Index. (A) *CDI*: chemical daily intake (ingestion); (B) *HI*: hazard index (ingestion); values above 1 indicate health issue risks (Luo et al., 2012). *RfD*: reference dose (references of *RfD* values are in the text).

Table 6. Chemical daily intake and Hazard Index. (a) *CDI*: chemical daily intake (ingestion); (b) *HI*: hazard index; values above 1 indicate health issue risks (Luo et al., 2012). *RfD*: reference dose (references of *RfD* values are in the text).

(a)

	95% confidence upper limit ($mg \cdot kg^{-1}$)			2 year exposure *CDI* ($mg \cdot kg^{-1}day^{-1}$)			Lifetime exposure *CDI* ($mg \cdot kg^{-1}day^{-1}$)		
	< 0.5 km	0.5 to 1 km	> 1 km	< 0.5 km	0.5 to 1 km	> 1 km	< 0.5 km	0.5 to 1 km	> 1 km
As	577	194	94	0.007	0.002	0.001	0.001	0.000	0.000
Ba	294	157	457	0.004	0.002	0.006	0.000	0.000	0.001
Cd	102	28	7	0.001	0.000	0.000	0.000	0.000	0.000
Co	30	14	16	0.000	0.000	0.000	0.000	0.000	0.000
Cr	69	53	112	0.001	0.001	0.001	0.000	0.000	0.000
Cu	23628	9317	2519	0.295	0.116	0.031	0.034	0.013	0.004
Mn	646	537	756	0.008	0.007	0.009	0.001	0.001	0.001
Mo	169	66	25	0.002	0.001	0.000	0.000	0.000	0.000
Ni	38	30	39	0.000	0.000	0.000	0.000	0.000	0.000
Pb	1700	824	203	0.021	0.010	0.003	0.002	0.001	0.000
V	117	97	100	0.001	0.001	0.001	0.000	0.000	0.000
Zn	26343	8527	2965	0.329	0.107	0.037	0.038	0.012	0.004

(b)

		RfD ($mg \cdot kg^{-1}day^{-1}$)	2 year exposure *HI*			Lifetime exposure *HI*		
			< 0.5 km	0.5 to 1 km	> 1 km	< 0.5 km	0.5 to 1 km	> 1 km
As	*RfD*	0.0003	24	9	4	3	1	0
Ba	*RfD*	0.0700	0	0	0	0	0	0
Cd	*RfD*	0.0010	1	1	0	0	0	0
Co	*RfD*	0.0300	0	0	0	0	0	0
Cr	*RfD* [Cr(VI)]	0.0030	0	0	0	0	0	0
Cu	*MRL*	0.0100	30	12	3	3	1	0
Mn	*RfD*	0.1400	0	0	0	0	0	0
Mo	*RfD*	0.0050	0	0	0	0	0	0
Ni	*RfD*	0.0200	0	0	0	0	0	0
Pb	-	-	-	-	-	-	-	-
V	*RfD*	0.0070	0	0	0	0	0	0
Zn	*RfD*	0.3000	1	0	0	0	0	0

Table 7 (on next page)

Bioaccessibility of Antofagasta dust

Bioaccessibility assay of three dust samples located in close proximity to the Antofagasta Port.

1 Table 7. Bioaccessibility assay of three dust samples located in close proximity to the Antofagasta Port.

		AFA-237 Port Gate	AFA-238 Window in front of the *galpón*	AFA-239 Building	Mean bioaccessibility (%)	Detection limit ($mg \cdot kg^{-1}$)	Quantification limit ($mg \cdot kg^{-1}$)	Reference material recovery (%)
	UTM E	356907	356899	357208				
Coordinates	UTM N	7383395	7383305	7384279				
	total ($mg \cdot kg^{-1}$)	243	92	85				
	bioaccessible ($mg \cdot kg^{-1}$)	27	58	65				
As	% of bioaccessibility	11	63	76	50	1.7	5.5	81.6
	total ($mg \cdot kg^{-1}$)	62	34	17				
	bioaccessible ($mg \cdot kg^{-1}$)	2	2	3				
Cd	% of bioaccessibility	3	6	21	10	1.0	3.4	83.2
	total ($mg \cdot kg^{-1}$)	15246	4157	3737				
	bioaccessible ($mg \cdot kg^{-1}$)	78	1024	1342				
Cu	% of bioaccessibility	1	25	36	20	2.1	6.9	95.4
	total ($mg \cdot kg^{-1}$)	197	31	14				
	bioaccessible ($mg \cdot kg^{-1}$)	0	0	0				
Mo	% of bioaccessibility	0	0	0	0	0.9	3.0	87.8
	total ($mg \cdot kg^{-1}$)	666	371	335				
	bioaccessible ($mg \cdot kg^{-1}$)	4	127	146				
Pb	% of bioaccessibility	1	34	44	26	2.1	7.0	89.4
	total ($mg \cdot kg^{-1}$)	19692	8821	3021				
	bioaccessible ($mg \cdot kg^{-1}$)	49	941	1135				
Zn	% of bioaccessibility	0	11	38	16	3.2	10.6	98.1

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Table 8(on next page)

City dust from Antofagasta

City dust from Antofagasta (ISP, 2014; Tchernitchin and Bolados, 2016) and Santiago (Tapia et al., 2009), Chile; Fushun (Kong et al., 2011), Baoji (Lu et al., 2009), Zhuzhou (Li et al., 2013), and the Qingshan district (QS) in Wuhan (Zhu et al., 2013), China; and the Islamabad Expressway in Pakistan (Faiz et al., 2009).

Table 8. City dust from Antofagasta (ISP, 2014; Tchernitchin & Bolados, 2016) and Santiago (Tapia et al., 2009), Chile; Fushun (Kong et al., 2011), Baoji (Lu et al., 2009), Zhuzhou (Li et al., 2013), and the Qingshan district (QS) in Wuhan (Zhu et al., 2013), China; and the Islamabad Expressway in Pakistan (Faiz et al., 2009).

	Chile		China				Pakistan
	Antofagasta ($mg \cdot kg^{-1}$)	Santiago ($mg \cdot kg^{-1}$)	Fushun ($mg \cdot kg^{-1}$)	Baoji ($mg \cdot kg^{-1}$)	Zhuzhou ($mg \cdot kg^{-1}$)	QS ($mg \cdot kg^{-1}$)	Islamabad ($mg \cdot kg^{-1}$)
As	239 ± 269	12 ± 1.9	-	-	89 ± 183	32 ± 20	-
Ba	188 ± 129	411 ± 124	-	-	-	1,610 ± 984	-
Cd	45 ± 42	0.8 ± 0.19	-	-	41 ± 117	2.8 ± 1.8	5.0 ± 1.0
Co	17 ± 11	11 ± 2.3	139 ± 179	-	13 ± 11	20 ± 12	-
Cr	61 ± 20	38 ± 11	5,334 ± 10,667	-	125 ± 54	172 ± 96	-
Cu	10,821 ± 9,816	669 ± 567	149 ± 177	123 ± 43	139 ± 148	213 ± 180	52 ± 18
Mn	573 ± 153	619 ± 109	-	-	-	-	-
Mo	75 ± 74	73 ± 27	-	-	6.4 ± 12.4	7.2 ± 3.9	-
Ni	29 ± 11	24 ± 6.4	302 ± 555	49 ± 30	40 ± 16	38 ± 14	23 ± 6
Pb	710 ± 852	127 ± 50	-	408 ± 296	956 ± 2,815	336 ± 191	104 ± 29
V	95 ± 20	54 ± 11	14.6 ± 4.1	-	-	-	-
Zn	11,869 ± 10,743	943 ± 411	-	715 ± 320	2,379 ± 5,145	1,250 ± 889	116 ± 35