Human health risk assessment of airborne metals to a potentially exposed community: a screening exercise

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Abstract

Exposure to high concentrations of inhalable particulate matter (PM) is a known human health risk, depending on the chemical composition of the PM inhaled. Mogale City (Gauteng) is known for having several sources of airborne PM, however, less is known about the metals in the airborne PM. The aim of this study was to determine the metals in measured PM at Kagiso, Mogale City. An independent PM_{10} monitor was installed at the municipality's existing monitoring site. This monitor continuously monitored PM_{10} between 23 August and 9 October 2013 and simultaneously sampled particles below 20 µm in diameter onto a glass fibre filter. This filter was replaced once towards the middle of the monitoring period. These two filters were chemically analysed to determine their metal content (30 metals) by the South African Bureau of Standards accredited laboratory at the Council for Scientific and Industrial Research by means of Inductively Coupled Plasma Spectroscopy (ICPS) based on the US EPA Method IO-3.1. To provide an estimate of possible health risk, the metal concentrations were used in a screening US-EPA human health risk assessment (HHRA). Since the analysed metals were reportedly below the detection limit, three hypothetical exposure scenarios (S) based on US-EPA recommendations were created for the HHRA. In S1, concentrations were considered to be the same as the detection limit for each metal; S2 assumed concentrations to be 50% of the detection limit; and S3 put concentrations at 10% of the detection limit. Potential risks (should pollution worsen) of developing respiratory and neurological effects were identified depending on the hypothetical scenarios. Continuous long-term monitoring and chemical characterisation are necessary to confirm these preliminary findings.

Keywords

Human Health Risk Assessment, South Africa, mining, metals, PM_{10}, air pollution

Introduction

Metals are natural components of the Earth's crust. Many of these metals are needed in the human body in small amounts, such as iron (Fe) which is contained in haemoglobin, copper (Cu) and manganese (Mn) which are in enzymes, and chromium (Cr), which is a co-factor in the regulation of sugar levels (CDC, 2011). However, depending on concentrations, these trace elements may have detrimental health effects. Heavy metals such as lead (Pb), cadmium (Cd) and mercury (Hg) are detrimental to human health as they may bio-accumulate in the body, while others are carcinogenic, such as arsenic, beryllium, cadmium and nickel (CDC, 2011). Metals emitted from mining activities accumulate in soil in surrounding areas and contaminated soil then poses a hazard to human health (Kumar et al., 2014), as the metals may be absorbed by vegetables grown in the contaminated soil, leach into underground water sources, or become airborne through wind-blown dust. Humans may therefore, be exposed to these metals via inhalation, ingestion and/or dermal contact. Mogale City Local Municipality (MCLM) in Gauteng Province

(South Africa) has a long history of gold mining. As a result, it has a number of mine dumps, some of which have not been rehabilitated. Activities within the MCLM that may contribute to concentrations of particulate matter (PM) in air include mining of minerals, quarrying of stone, extraction of clay and sandpits, use of motor vehicles, various heavy and light industrial activities, as well as domestic fuel burning (AQMP, 2013).

Focus-group discussions with MCLM residents in 2013 revealed that residents' perceived dust (PM) emissions in the area was responsible for most of their illnesses, including respiratory illness and cancer, and that dust was soiling their properties and damaging their appliances (Phala et al., 2012; Wright et al., 2014). One of the study recommendations was that the metal content of the PM should be characterised and possible health impacts from PM inhalation quantified using Human Health Risk Assessment (HHRA).

HHRA links environmental exposure to potential human health

effects. The potential for detrimental health effects is assessed based on the United States Environmental Protection Agency (US-EPA) Human Health Risk Assessment Framework (US-EPA, 2014) as it relates to the physical and/or chemical properties of air pollutants and their concentrations. The framework comprises the following steps: (1) Hazard identification to determine whether exposure to a particular substance may result in detrimental human health effects; (2) Exposure assessment to determine environmental concentrations through source and emissions characterization, monitoring, and / or environmental fate, transport, and deposition modelling, and to estimate the magnitude, duration, and frequency of human exposure; (3) Dose-response assessment to estimate the relationship between dose, or level of intake of a substance, and the incidence and severity of an effect (Several agencies such as the US-EPA (IRIS, 2015), the World Health Organization (WHO) (WHO, 1999) and the Centre for Disease Control (CDC) (ATSDR, 2015) have developed databases for benchmark values, which are used to describe the dose-response relationships determined for various chemicals); and (4) Risk characterisation which combines all the information obtained in the previous three steps to describe whether a risk to public health is predicted.

Therefore, the aim of this article was to describe the chemical composition of PM measured at Kagiso, MCLM and to apply the HHRA in a screening exercise to estimate possible human health risks with the purpose of identifying metals for further investigation.
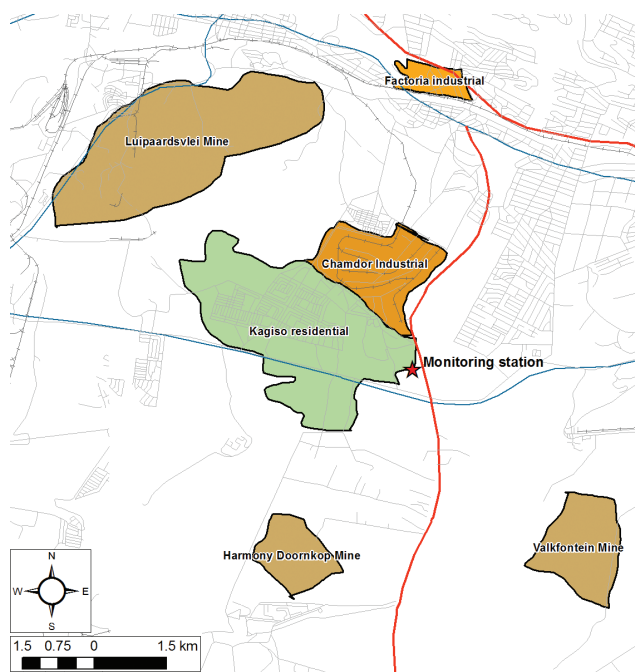
Methods

Study area
Kagiso, the community surrounding the monitoring station, formed the study area (Figure 1). The population density in Kagiso varies from 700 to 20 500 people per km^2 (AQMP, 2013). Nearly 30% of the population of 362 422 were considered vulnerable because they were either below 15 years or above 65 years of age (StatsSA, 2011). The majority (54.8%) of households had access to piped water inside the dwelling; also to electricity (85.9%) and weekly refuse removal (79.7%) (StatsSA, 2011).

Meteorological data from 2007 to 2012 indicated that the prevailing wind direction during spring (August and September), the period in which the current study was conducted, was from a north-westerly to northerly direction (AQMP, 2013). Meteorological data further indicated that the wind in the MCLM is calm for about 40% of the time and when the wind is blowing it is mostly at a speed of < 3 ms^{-1} (AQMP, 2013). There are mine dumps within four to five kilometres to the north and the north-west, as well as the south of the monitoring site, and two industrial areas (Factoria and Chamdor) resides to the north and north-west, respectively.

Monitoring and chemical analyses
The West Rand District Municipality operates an air quality monitoring station in MCLM and monitors air pollution including

PM$_{10}$, however, chemical composition of PM$_{10}$ is not routinely analysed. This site was chosen as the measurement location in this study for security reasons and to access electricity. A TOPAS Sira MC 090158/00 PM$_{10}$ monitor was installed from 23 August 2013 to 9 October 2013. This instrument continuously monitored PM$_{10}$ concentration. It also simultaneously sampled PM$_{20}$ onto a glass fibre filter, recorded the volume of air pumped through as well as the mass deposited on the filter. The filter was changed once during this period to have two filters for analyses. The filters and a blank were chemically analysed for their metal content by the South African Bureau of Standards accredited laboratory at the CSIR, based on the US EPA Method IO-3.1 and using Inductively Coupled Plasma Spectroscopy (ICPS). The instrument also heated to 60 °C to drive off moisture and volatiles. The concentrations of each metal in air were subsequently calculated and used to determine potential exposure in a HHRA.

Figure 1: Location of Kagiso, other communities, mines and industrial areas in relation to the location of the measurement instrument used in this study located at the MCLM monitoring station.

Applying the HHRA framework
Three types of risk estimates were calculated in this quantitative HHRA: The (1) Hazard Quotient (HQ), which describes the potential for developing detrimental effects (other than cancer) from exposure to a hazardous substance; (2) Incremental (over and above the background prevalence) Cancer Risk, which is the probability of individuals developing cancer from exposure to a hazardous substance; and (3) Hazard Index (HI) was calculated to determine the total incremental cancer risk for the area (community).

For Hazard Identification, reliable databases, including those of the US-EPA, were reviewed to determine whether the elements identified during chemical analyses of the PM samples may be

detrimental to human health. It was assumed in a worst-case scenario, that the Kagiso community, specifically, in the MCLM study area were continuously exposed (for example, mothers looking after small children at home) to the concentrations of PM_{10} and metals in air as determined by the monitoring and analyses of the short-term PM samples. Air was the medium and inhalation the route of exposure assessed. Where concentrations of metals were found to be below detection limits, hypothetical scenarios of exposure were created based on recommendations by the US-EPA (US-EPA, 2000), as follows: (a) Scenario 1 (S1): concentrations are the same as the detection limit for each metal; (b) Scenario 2 (S2): concentrations are 50% of the detection limit and; (c) Scenario 3 (S3): concentrations are 10% of the detection limit.

To obtain relevant benchmarks for the identified elements, the focus was on the most recently published standards or guidelines from reliable databases, including the US-EPA, WHO and CDC. A benchmark value is a value of exposure to pollutants that is believed to not be detrimental to even sensitive individuals in a population. In the case of air pollutants, the benchmark value is a "safe" concentration expressed as mass per volume.

A quantitative risk characterisation was performed, providing a numeric estimate of the potential for public health consequences from exposure to the metals concerned. This quantified potential was expressed as an HQ, which is unitless. The HQ is determined by the ratio between the expected exposure concentration of the metal and the benchmark value, which is an exposure that is assumed not to be associated with detrimental health effects. In the case of carcinogens, the incremental (over and above the background) cancer risk, which is a function of the 'Inhalation Concentration' and the 'Inhalation Unit Risk', was quantified. The Inhalation Unit Risk (risk per $\mu g/m^3$) is the unitless upper bound estimate of the probability of tumour formation per unit concentration of a chemical (Mitchell, 2004).

Results are given for the TOPAS measured PM_{10} concentrations and chemical composition followed by the results for each HHRA step and risk estimates for S1, S2 and S3.

Results and discussion

Particulate monitoring

PM_{10} concentrations measured by the TOPAS instrument exceeded the then South African 24-h ambient air quality standard of 120 $\mu g/m^3$ three times during the monitoring period of 23 August 2013 to 9 October 2013 (Figure 2). The current standard (since January 2015) of 75 $\mu g/m^3$ was exceeded 14 times during the same period. Sharp peaks were detected on 24 and 30 August 2013.

The South African Air Quality Information System (SAAQIS) website (www.saaqis.org.za) was consulted for results of monitored PM_{10} data from the municipality's monitoring station. Raw data were not available but a graph for the period

1 August to 30 September 2013 (Figure 3) showed incomplete data and although the 24-h standard of 120 $\mu g/m^3$ was not exceeded during the specified period, the current standard of 75 $\mu g/m^3$ was exceeded on at least six occasions. The highest peak of about 110 $\mu g/m^3$ was around the 30th of August, which coincided with the TOPAS peak, although lower in magnitude.
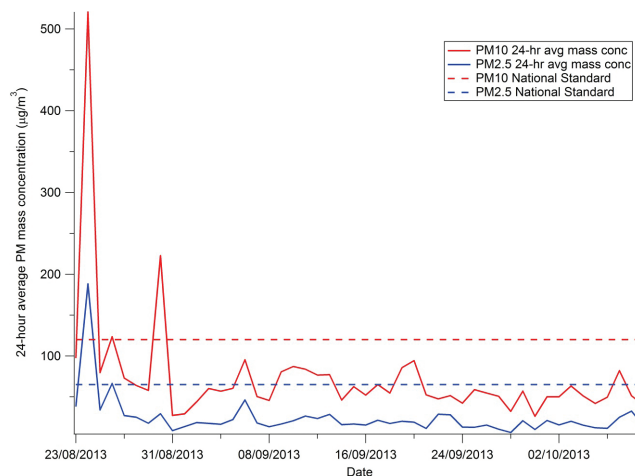

Figure 2: PM_{10} concentrations monitored during the period 23 August 2013 to 9 October 2013.
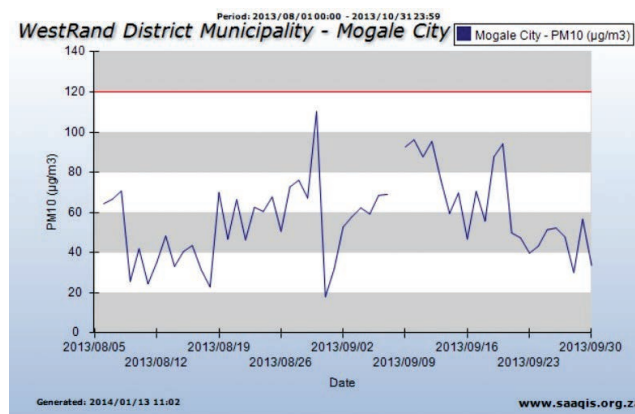

Figure 3: PM_{10} concentrations monitored by the municipality monitoring station during the period 1 August to 30 September 2013 (graph generated in SAAQIS www.saaqis.org.za).

Metal analyses

The two filters and a blank were chemically analysed for the presence of 30 metals, expressed in mg/kg. The results obtained were all below the detection limits for the individual metals (namely, calcium, magnesium, total phosphorous, potassium, nickel, silver, tin, titanium, bismuth, sodium, sulphur, aluminium, antimony, arsenic, boron, barium, beryllium, cadmium, cobalt, chromium, copper, iron, lead, lithium, manganese, selenium, silica, strontium, vanadium and zinc). The total weight of PM_{10} accumulated on the filter over each monitoring period was divided by the total volume of air sampled during that period to obtain a concentration ($\mu g/m^3$) per metal. Due to the fact that concentrations were below the detection limits, these concentrations were considered to be at the detection limit as a worst case scenario (S1). Two other scenarios were also

assessed, namely concentrations at 50% of the detection limit (S2) and at 10% of the detection limit (S3).

Health risks assessed

Hazard identification
Non-cancer health effects caused by the metals analysed are mostly respiratory-related (metals included bismuth, boron, calcium, chromium, copper, iron, phosphorus, sulphur, titanium, tin, aluminium, antimony, beryllium, cadmium, cobalt, lithium, silica and vanadium, zinc and nickel), while some may cause neurological effects (lead, manganese and selenium) (CDC, 2011). Silver may cause argyria (bluish-grey skin), while arsenic, beryllium, cadmium and nickel have been identified as being carcinogenic (CDC, 2011).

Exposure assessment
The communities of concern were considered to be those around the monitoring station in the MCLM, with specific emphasis on Kagiso since it was the closest to the monitoring site. The concentrations for each scenario used in this HHRA are given in Table 1. In this worst-case scenario exercise, it was assumed that individuals were constantly (24 hours per day) exposed to the varying concentrations calculated for the different exposure scenarios. In terms of cancer risk assessment, it was assumed that the calculated concentrations were the concentrations that individuals were exposed to over a lifetime. It is not possible to completely avoid contact with these metals naturally found in the environment. The smaller the particles, the deeper they will penetrate into the lungs. The amount absorbed into the bloodstream will again depend on how well the particles dissolve. The more easily the particles dissolve, the more easily they may enter the blood stream. If the particles do not dissolve easily, they may remain in the lungs for longer periods of time. Some of the particles may leave the lungs through the normal clearing process while some may be swallowed.

Dose-response assessment
Numeric benchmark values obtained from reliable databases were used to describe the dose-response relationships. The preferred benchmark values used were those set on the basis of health effects in human beings and not those incorporating economic or social factors. Since people are normally exposed to metals in the air, predominantly in an occupational environment, and since the main route of exposure to metals is ingestion, a number of the elements did not have benchmark values for inhalation. Where no benchmark value could be found from reliable databases surveyed, the South African Occupational Exposure Limit (SA OEL) (SA Occupational Standards, 1995) was considered to indicate the level of toxicity of the element. These OELs are indicated in bold in Table 1. These were for aluminium, arsenic, antimony, barium, beryllium, bismuth, calcium, copper, iron, lithium, magnesium, silica, silver, selenium, sulphur, tin and zinc. In cases where there was no OEL for the element but only for a species of that element, that species is given in brackets in Table 1.

It must be noted that the application of occupational standards is not technically applicable in this case, because the samples were taken in ambient air and for a period longer than eight hours. In addition, occupational standards are set with healthy workers in mind, who are only exposed for eight hours a day and forty hours a week. Therefore, occupational standards are higher than ambient standards and they will not protect sensitive individuals (such as asthmatics and children) which ambient standards are supposed to do. However, it was decided to keep the pollutants for which only occupational standards were available as part of this assessment, because if the HQs of any of these pollutants with relatively high benchmark values were >1 it would indicate possible drivers of risk that can then be further investigated. Those metals for which no benchmark values could be found, namely sodium, potassium and strontium, were excluded from the HHRA.

Risk characterisation
Non-cancer risk estimates, expressed as HQs, were calculated for the metals and are presented in Table 2. HQ values > 1 indicate that the likelihood of detrimental non-cancer effects is enhanced while HQ values <1 indicate that the potential for detrimental health effects is minimal. The HQs of most (about 80%) of the metals analysed were below one, indicating that non-cancer detrimental health effects were unlikely. For six of the analysed metals (i.e. cadmium, cobalt, manganese, nickel, lead and vanadium), the HQs indicated a risk to human health in one or more of the exposure scenarios.

(a) Risk characterisation for S1
In S1, where the concentrations were assumed to be at the detection limit, the HQs indicated a risk of developing respiratory effects from cadmium, cobalt, nickel and vanadium and neurological effects from manganese and lead.

(b) Risk characterisation for S2
In S2, HQs indicated a health risk for respiratory effects due to exposure to cadmium, cobalt and nickel as well as neurological effects due to exposure to manganese and lead. The HQ for vanadium did not indicate a risk of respiratory effects in S2.

(c) Risk characterisation for S3
In S3, assuming that the exposure concentrations were 10% of the detection limit, only cadmium and cobalt posed a risk of respiratory effects as only the HQs for these two metals were above one.

(d) Cancer risk characterisation
The incremental cancer risks (Table 3) were estimated using the determined concentrations and the inhalation unit risk for each metal (from the US-EPA) known to be a confirmed human carcinogen. The HI is also presented in Table 3. The total incremental risk to develop cancer ranges from 1.73 in 1 000 in S3, to 17.7 in 1 000 in S1. Arsenic was found to be the main driver of the risk to cancer. The calculated incremental cancer risks are thus not only above 1 in a million, but also above 1 in 10 000. Evaluation of these risks against the US-EPA air office criteria

which "strives to reduce risk for as many people as possible to 1 in a million, while assuming that the maximally-exposed individual is protected against risks greater than 1 in 10 000" indicates that these acceptable risks have been exceeded here.

Uncertainties and limitations

Several limitations and uncertainties should be considered when interpreting these findings. HHRA is a predictive process, therefore it only estimates what could occur. Due to the fact that concentrations were below the detection limits, these concentrations were considered to be at the detection limit as a worst case scenario at 50% of the detection limit and at 10% of the detection limit. While these scenarios are not representative of the status quo, should current air pollution interventions and management change, and PM levels increase, such scenarios may be experienced. Being prepared by having predicted, likely worst-case scenario health impacts supports the need for continuous improvement in air quality management and air pollution control in this area.

It is acknowledged that PM_{10} was monitored only during a short period and at one site only. Personal monitoring and time-activity data are the optimal method for exposure assessment, however, they were not possible in this small study. Although the instrument was calibrated, concentrations may not be representative for all the communities, particularly those furthest away from the monitoring site. However, this screening study aimed to understand the metals present in the dust. It did not aim to collect information on pollution sources. Although the monitoring data from the municipality at the same site may give additional data on PM_{10} exposure, the simultaneous capturing of the PM onto a filter for metal analysis was not possible from the instrumentation used by the municipality, and this was crucial for this exercise.

To address model uncertainty in this study, equations from the US-EPA were used, and applied benchmark values were based on national and international standards and guidelines which were set based on human health effects. In terms of selection of the pollutants of concern, the pollutants were pre-identified as those metals which the accredited laboratory could analyse for and which was present in the PM. Finally, in terms of the exposure pathway and route used, the risk assessment was based on inhalable PM and therefore the inhalation route was selected.

Conclusions

PM_{10} concentrations measured in this study exceeded the previous South African 24-h ambient air quality standard of 120 µg/m³ three times during the monitoring period and the current standard (since January 2015) of 75 µg/m³ 14 times. Although the concentrations of metals determined in this study were below detection limits, and were not representative of all exposure periods and all seasons, or for all individuals in

the local municipality, using US-EPA recommended scenarios, screening for possible carcinogenic and non-carcinogenic risks based on concentration and toxicity from metal exposure through inhalation was performed. Results provide an indication of which metals may drive human health risks in the area. Those pollutants identified as being potential risks should be investigated further.

Acknowledgements

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[i] *The size of the particles collected for chemical analysis was ≤ 20 µm.*

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Table 1: Concentrations of compounds of potential concern and their benchmark values.

Element	Scenario 1		Scenario 2		Scenario 3	
	Concentration (µg/m³)	Benchmark Value (µg/m³)	Concentration (µg/m³)	Benchmark Value (µg/m³)	Concentration (µg/m³)	Benchmark Value (µg/m³)
Aluminium (respirable)	2.70	**5000**	1.36	**5000**	0.27	**5000**
Arsenic	2.70	**10**	1.36	**10**	0.27	**10**
Antimony (and compounds)	6.90	**500**	3.43	**500**	0.69	**500**
Barium compounds	6.90	**500**	3.43	**500**	0.69	**500**
Beryllium (and compounds)	1.4	**2**	0.68	**2**	0.14	**2**
Bismuth (undoped)	6.90	**10000**	3.43	**10000**	0.69	**10000**
Boron	10.20	300*	5.08	300*	1.02	300*
Cadmium	1.40	0.03*	0.68	0.03*	0.14	0.03*
Calcium oxide	68.40	**2000**	34.20	**2000**	6.84	**2000**
Cobalt	1.40	0.10*	0.68	0.10*	0.14	0.10*
Chromium III	1.40	5*	0.68	5*	0.14	5*
Copper	6.90	**1000**	3.43	**1000**	0.69	**1000**
Iron oxide (dust)	1.40	**5000**	0.68	**5000**	0.14	**5000**
Lead	2.70	0.5**	1.36	0.5**	0.27	0.5**
Lithium hydride	6.90	25	3.43	25	0.69	25
Magnesium	13.70	5000	6.83	5000	1.37	5000
Manganese (and compounds)	1.40	0.03*	0.68	0.03*	0.14	0.03*
Nickel	1.40	0.20*	0.68	0.20*	0.14	0.20*
Phosphorus (white)	13.70	20*	6.83	20*	1.37	20*
Silica	13.70	**100**	6.83	**100**	1.37	**100**
Silver	6.90	**100**	3.43	**100**	0.69	**100**
Selenium (and compounds)	1.40	**100**	0.68	**100**	0.14	**100**
Sulphur	13.70	**6000 000**	6.83	**6000 000**	1.37	**6000 000**
Tin	6.90	**200**	3.43	**200**	0.69	**200**
Titanium	6.90	10*	3.43	10*	0.69	10*
Vanadium	1.40	0.8*	0.68	0.8*	0.14	0.8*
Zinc	1.40	**500**	0.68	**500**	0.14	**500**

Note. *MRL   **SA National Standard   SA-OEL figures in bold.

Table 2: Non-cancer risk estimates of the elements analysed for the different exposure scenarios.

Element	Scenario 1 Concentration (µg/m³)	Scenario 1 Hazard Quotient (HQ)	Scenario 2 Concentration (µg/m³)	Scenario 2 Hazard Quotient (HQ)	Scenario 3 Concentration (µg/m³)	Scenario 3 Hazard Quotient (HQ)
Aluminium	2.70	0.0005	1.36	0.0003	0.27	<0.0001
Antimony	6.90	0.01	3.43	0.007	0.69	0.001
Arsenic	2.70	0.27	1.36	0.14	0.27	0.03
Barium	6.90	0.01	3.43	0.007	0.69	0.001
Beryllium (and compounds)	1.40	0.68	0.68	0.34	0.14	0.07
Bismuth	6.90	0.0007	3.43	0.0003	0.69	<0.0001
Boron	10.20	0.03	5.08	0.02	1.02	0.003
Cadmium	1.40	45.48	0.68	22.70	0.14	4.55
Calcium	68.40	0.03	34.20	0.02	6.84	0.003
Cobalt	1.40	13.64	0.68	6.82	0.14	1.36
Copper	6.90	0.007	3.43	0.003	0.69	0.0007
Chromium III	1.40	0.27	0.68	0.14	0.14	0.03
Iron	1.40	0.0003	0.68	0.0001	0.14	<0.0001
Magnesium	13.70	0.003	6.83	0.001	1.37	0.0003
Manganese (and compounds)	1.40	4.55	0.68	2.27	0.14	0.45
Nickel	1.40	6.82	0.68	3.41	0.14	0.68
Phosphorus (white)	13.70	0.68	6.83	0.34	1.37	0.07
Selenium	1.40	0.01	0.68	0.007	0.14	0.001
Silica	13.70	0.14	6.83	0.07	1.37	0.01
Silver	6.90	0.07	3.43	0.03	0.69	0.007
Sulphur	13.70	0.002	6.83	0.001	1.37	0.0002
Tin	6.90	0.03	3.43	0.02	0.69	0.003
Titanium	6.90	0.69	3.43	0.34	0.69	0.07
Lead	2.70	5.46	1.36	2.73	0.27	0.55
Lithium	6.90	0.27	3.43	0.14	0.69	0.03
Vanadium	1.40	1.71	0.68	0.85	0.14	0.17
Zinc	1.40	0.003	0.68	0.001	0.14	0.0003

Table3: Incremental cancer risks estimates for the different exposure scenarios.

Element	Scenario 1 Concentration (µg/m³)	Scenario 1 Inhalation Unit Risk	Scenario 1 Incremental cancer risk	Scenario 2 Concentration (µg/m³)	Scenario 2 Inhalation Unit Risk	Scenario 2 Incremental cancer risk	Scenario 3 Concentration (µg/m³)	Scenario 3 Inhalation Unit Risk	Scenario 3 Incremental cancer risk
Arsenic	2.70	4.3×10^{-3}	11.6 in 1000	1.36	4.3×10^{-3}	5.8 in 1000	0.27	4.3×10^{-3}	1.1 in 1000
Beryllium	1.40	2.4×10^{-3}	3.3 in 1000	0.68	2.4×10^{-3}	1.6 in 1000	0.14	2.4×10^{-3}	0.3 in 1000
Cadmium	1.40	1.8×10^{-3}	2.5 in 1000	0.68	1.8×10^{-3}	1.2 in 1000	0.14	1.8×10^{-3}	0.3 in 1000
Nickel	1.40	2.4×10^{-4}	0.3 in 1000	0.68	2.4×10^{-4}	0.2 in 1000	0.14	2.4×10^{-4}	0.03 in 1000
Total HI			**17.7 in 1000**			**8.8 in 1000**			**1.73 in 1000**