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Seasonal variations in airborne heavy metals in Vanderbijlpark, South Africa

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In this study, monthly settleable dust samples were collected in four seasons using the modified crude single open bucket units at five sites within the Vanderbijlpark area. The samples were captured in distilled water medium, filtered, air-dried, weighed to determine mass concentrations, digested with aqua regia acid solution and analyzed for heavy metals by inductively coupled plasma (ICP) 6000 Thermo-Electron model technique. The average ranges of ten metals identified were: zinc (Zn,1.12 – 5.87 mg/L), iron (Fe, 2.25 – 8.94 mg/L), chromium (Cr, 0.87 – 3.99 mg/L), titanium (Ti, 0.58 – 3.1 mg/L), manganese (Mn, 2.31 – 9.01 mg/L), lead (Pb, 0.5 – 2.34 mg/L), vanadium (V, 0.94 – 4.18) mg/L), copper (Cu, 0.78 – 3.01 mg/L), nickel (Ni, 0.7 – 3.64 mg/L) and cadmium (Cd, 1.0 – 2.92 mg/L) respectively. Average daily mass concentrations obtained ranged from 2.42 – 1270 mg/m^2/day in autumn, 520 – 1520 mg/m^2/day in winter, 142 – 683 mg/m^2/day in spring and 156 – 846 mg/m^2/day in summer. Both the South African and the American industrial limits were exceeded in winter and autumn seasons at a farm site. A South African standard for residential limit, 600 mg/m^2/day, was also exceeded at a school site during the same seasons.

Key words: Settleable dust,mass concentration; heavy metals, Vanderbijlpark.

INTRODUCTION

The atmosphere in most urban areas contains billions of pollutants that include particulate matter and gaseous substances. Particulates are very small liquid droplets or solid particles that are suspended in the air. They originate from natural sources (e.g. surface soil or dust, sea salt, volcanic dust and biological debris) or anthropogenic sources which include combustion activities in homes, vehicles and industries (Annegarn and Scorgie, 2002).

Dust is a generic term used to describe particles of less than 1000 µm and is found everywhere, suspended in air or deposited on surfaces (Kuhn and Loans, 2012). There is over a billion tons of dust and other particles at any given time suspended in the air (DustWatch, 2012). Dust contains pollutants such as polycyclic aromatic compounds, biological material and heavy metals. It is kept suspended in air or transported from sources by wind and can travel long distances across regions, and continents transferring contaminants to ecosystems (Adriaenssens, 2011). Evaluating the composition of dust material will give an indication of the dangers it may pose and where it originates.

Breathing poisonous heavy metals attached to airborne dust increases the potential health effects of occupants in any environment. Common health effects associated with metal poisoning include organ damage, blindness and

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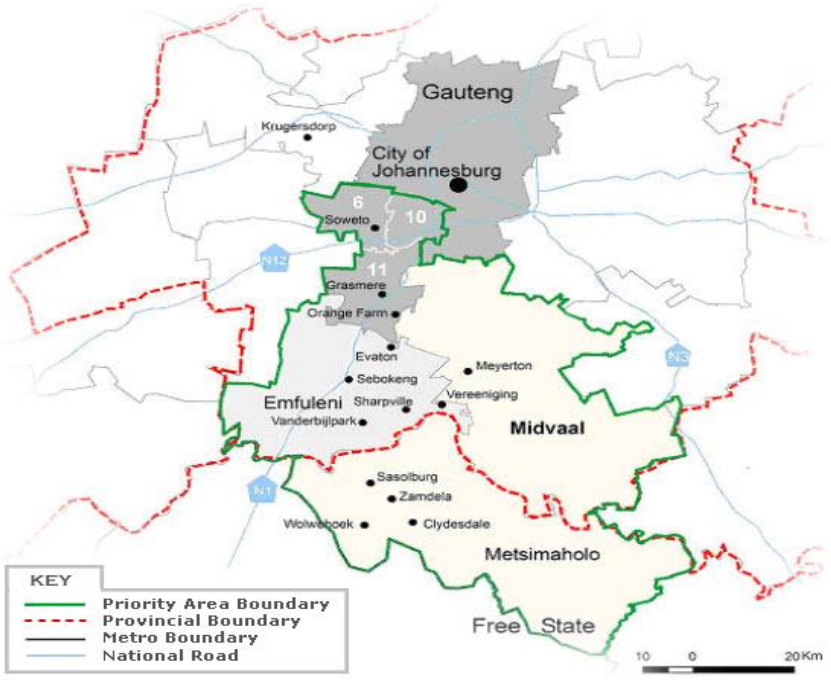

Figure 1. The location of the study area (DEA, 2011).

metal poisoning include organ damage, blindness and deafness, breathing and sexual problems or death. Lead (Pb), chromium (Cr), cadmium (Cd), mercury (Hg), and Arsenic (As) are examples of poisonous heavy metals that are non-biodegradable, indestructible and would accumulate by being adsorbed on suspended dust.

The toxicity of metals depends on the chemical form or species of element. For example, Pb is more toxic in the ion Pb^{2+} form. While, Hg is more toxic when attached onto the alkyl group due to their solubility in animal tissues and can be transmitted through biological membranes (Baird, 1995). Usually, poisonous metal levels would increase gradually within the ecological food chain. Aquatic organisms like mussels, oysters and fish, tend to bio-concentrate Hg and Cd. Major anthropogenic sources of airborne toxic heavy metals include emissions that are related to transport, residential and industrial fuel burning activities (Annegarn and Scorgie, 2002).

Different sampling techniques are available for sampling ambient dust, but most are not readily accessible due to their high costs. Electronic dust samplers are useful in collecting real time samples of specific particle sizes continuously. However, their high cost makes them inaccessible to most researchers, particularly in developing countries. Hence, there is a need to use simple cost effective sampling methods that can produce reliable and representative samples for analysis.

The single open bucket system is cost effective and easier to deploy, even in remote areas where there is no electricity and can collect samples continuously. Crude open single bucket samplers with no wind shield have been used during this study. According to Kuhn and Loans (2012), these types of samplers are able to collect all settleable dust particles on all days irrespective of the wind condition. The method has also been used successfully for evaluating settleable dust levels in different scenarios (DustWatch, 2012; Annegarn and Scorgie, 2002).

This study took place in Vanderbiljpark, a heavily industrialized and polluted region of Gauteng Province (GP) in South Africa. Vanderbijlpark is part of the Emfuleni Municipality, a home to anthropogenic pollution sources such as mining, coal power stations, steel companies, residential coal combustion and oil refineries (VECN, 2011).

Vanderbijlpark together with Sasolburg and Meyerton form a triangle and are known as the Vaal Triangle (Figure 1). Air in this region is known to be poor due to the close proximity between residential areas and a large number of industrial pollution sources (VECN, 2011). The South African government has identified this region as one of the air pollution hot spots in terms of Section 18 (1) of the National Environmental Management, Air Quality Act No. 39 (VECN, 2011; NAAQS, 2009). It is one of the mostly populated regions in GP (about one million people) and is about 60 km south of Johannesburg. Local climate is of subtropical nature with hot, wet summers and very cold winters.

This study evaluated mass concentrations and heavy

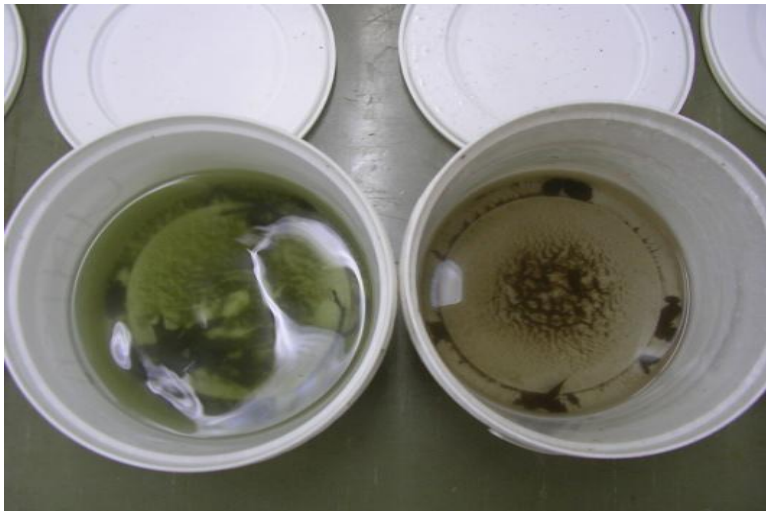

Figure 2. Bucket samplers after dust sample collection.

metals associated with seasonal settleable dust samples. Previous settleable dust studies in this region concentrated mainly on mass concentrations (Kuhn, 2011; Annegarn and Scorgie, 2002). Heavy metals were mostly addressed in studies which investigated particulate matter samples (PM10 and PM2.5) using electronic samplers and not on settleable dust samples (Moja et al., 2013; Scorgie et al., 2009). In this work, both the mass and heavy metal concentrations associated with settleable or precipitating dust samples were presented.

MATERIALS AND METHODS

Site selection, sample collection, treatment and analysis

Chosen sampling sites are located at Boipatong township school (A), a CE5 suburban house (B), a farm (C), a petrol station (D) and a traffic light (E) in Vanderbijlpark. Two sites located at the two residential areas are sites A and B. Site A is about 3.02 km on the east north-eastern directions at 26°40′07″S, 27°50′55″E; while site B is about 4.45 km south-east of site D at 26°42′01″S, 27°51′15″E. Site D is about 7 km north-west of Vaal University of Technology at 26°40′31″S, 27°49′09″E. R57 road runs parallel to site C which is about 4.44 km north western direction of site D at 26°38′16″S, 27°48′13″E. Site E is opposite to the Golden highway road at about 2.16 km on the western direction of the site D at 26°41′21″S, 27°48′14″E.

Seasonal precipitating dust samples were collected in autumn (March), winter (June), spring (September) and summer (December) from five sites and four (4) samples were collected from each site (one per 30 days). Site selection was influenced by land use, safety, easy access and sample representativeness. Modified crude single open bucket samplers were used to collect the settleable dust samples based on the American Standard Test Method (ASTM) D1739 of 1998 as described by Annegarn and Scorgie, 2002. The modified method did not have a wind shield around the bucket so that both course and fine precipitating dust samples could be collected irrespective of the wind situation. Settling dust material were collected in 5L cylindrical plastic buckets half filled with distilled water spiked with about 5 ml (3.5 %) of domestic quality bleach (hypochorite) solution which served as an algaecide. Buckets were hoisted on steel holders at heights of about 2.5 m from the surface. Exposed buckets were replaced after 30 days of sampling period and transported to the laboratory for further treatment (Figure 2). Water – dust sample mixtures were filtered through pre-weighed 0,47mm Whatmann filter papers using the Buchner flask filtering system. The samples were desiccated for 24 h or air dried in partially open petri discs in a “dust free lab” and weighed. The mass concentrations of settleable dust were determined by dividing the mass difference or collected dust mass by the cross-sectional area of buckets (0.02545 m^2) in 30 days.

High purity HNO_3 (Merck Suprapur 65%) and HCl (Merck GR 32%) were used to prepare aqua regia solution. Heavy metal content in about 0.1 g dust samples were extracted with 30.0 ml aqua regia solution into a porcelain crucible on a hotplate in the fume hood until near dryness. The material was further digested in another 30.0 ml of the acid mixture until near dryness. A warm dilute nitric acid solution (0.5 M) was used to transfer the material quantitatively from a crucible, filtered into a 50.0 ml volumetric flask and filled to the mark.

Sample solutions were stored in a refrigerator or analysed immediately by an inductively coupled plasma (ICP) 6000 Thermo-Electron model under the following operating conditions: Pump rate = 45 rpm, Nebuliser argon pressure = 0.3 $L.min^{-1}$, Centre tube = 2 mm, RF forward power = 1150 W, Coolant gas flow = 12 $L.min^{-1}$, Auxiliary gas flow = 0.5 $L.min^{-1}$ and Integration time = 15 s. Averages of three repeats (3 times) of all samples were reported.

The performance of the analytical method was validated against a European soil certified reference material (CRM-EURO876-1 from Industrial Analytical Society in South Africa). A series of certified reference material (CRM) solutions were prepared from a 1.000 g/L multi-elemental stock solution under the same experimental conditions as the samples. Acceptable linear calibration graphs were produced and detection limits of all heavy metals were < 0.01 µg/L.

Glassware was soaked for 24 h in dilute nitric acid, rinsed with deionised water (5.4 us/cm produced by Millipore Milli-Q purifier) and dried in oven before use. Blank solutions were prepared parallel with CRM and dust residue samples. Wind speed and direction, as well as precipitation during the sampling period were measured at the Arcelor Mittal South Africa (AMSA) weather station no. 350.

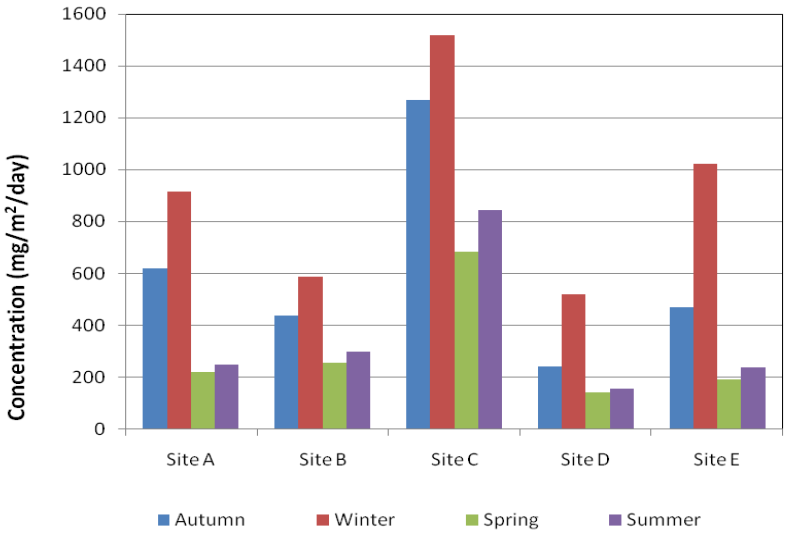

Figure 3. Seasonal variation of dust deposit per sites.

RESULTS AND DISCUSSION

Settled dust mass concentration

Concentrations of settleable dust at all sites per season decreased in the following order: winter > autumn > summer > spring (Figure 3). Average rainfall per season was 10.8 mm in autumn, 5.7 mm in winter, 275.21 mm in spring and 163.73 mm in summer. Relatively low amount of rainfall and humidity experienced in winter and autumn would explain the high amounts of mass concentrations obtained during these seasons. Secondly, the high winter mass concentrations could be due to the stable climatic condition (that is common in this region) which retains suspended dust closer to the ground based sources (VECN, 2011). Also, fine dust is known to sink in still atmospheric conditions, while the coarse particles are influenced by gravity (DustWatch, 2012). Heavy spring and summer rains could have suppressed dust generation potential from the surface, particularly when different wind thresholds of variable dust particle sizes are not exceeded.

Highest average daily mass concentrations were obtained at site C with 1270 mg/m^2/day in autumn, 1517 mg/m^2/day in winter, 683 mg/m^2/day in spring and 846 mg/m^2/day in summer. Using the American (ASTM-D1739 of 1998) classification systems, concentrations obtained in winter and autumn were very heavy (> 1200 mg/m^2/day), while those obtained in spring and summer were less, but still heavy (Annegarn and Scorgie, 2002). The South African settleable dust standard calls for remedial action when an industrial limit of 1200 mg/m^2/day is exceeded as it is the case at site C (DustWatch, 2012; SANS, 2005). Also, the department of environmental affairs (DEA) has a guideline which shows that people start complaining when exposed to dust of about 2000 mg/m^2/day (Kuhn, 2011; Annegarn and Scorgie, 2002).

Composite total dust concentrations at all sites in decreasing order were 4316 mg/m^2/day at site C, 2009 mg/m^2/day at site A, 1923 mg/m^2/day at site E, 1581 mg/m^2/day at site B and 1060 mg/m^2/day at site D. Site C is located within the farm area and near a busy industrial waste dump site. It is also close to busy streets where surface dust is lifted all the time by road transport. A second highest total mass concentration was obtained at site A, a township school site, which could be explained by large field of uncovered earth surface in and outside school yard. A residential limit of 600 mg/m^2/day is prescribed and it is worrying that both winter and autumn mass concentration at the school site have exceeded this standard (SANS, 2005). The lowest concentrations were obtained at sites D (petrol station) and B (suburban residence), which could be attributed to good housekeeping (frequent sweeping and cleaning).

The lifting of dust from the surface of the earth usually occurs when wind speed is higher than threshold value, which is minimum speed required to initiate the motion of particles. It depends on the size of the erodible particles and the effect of the wind shear stress of the surface. Annegarn and Scorgie (2002) reported that dust particle sizes ranging from 1 – 500 µm have threshold friction velocities of between 0.24 m/s and 3.5 m/s. It was also reported that 5.4 m/s is the wind speed threshold to lift surface dust in South Africa. Dust particle sizes greater than 80 µm is unlikely to be carried to a great distance unless a severe windstorm is being experienced (DustWatch, 2012). Also, dust of a 30 µm diameter will only be transported about 300 m by a wind velocity of 5, 0 m/s in about 60 s. Average monthly maximum wind

Table 1. Ranges of total metals per season (mg/L).

Heavy Metals	Autumn	Winter	Spring	Summer
Zn	2.1 - 8.02	1.42 - 10.21	0.32 - 3.42	0.53 - 1.83
Fe	2.45 - 8.56	2.7 - 13.11	3.54 - 6.84	2.3 - 7.23
Cr	0.75 - 4.2	1.04 - 5.5	0.82 - 3.21	1.01 - 3.04
Ti	0.66 - 4.1	1.2 - 4.65	0.14 - 2.61	0.32 - 1.23
Mn	5.06 - 10.23	2.51 - 15.02	1.89 - 6.47	2.5 - 4.31
Pb	0.51 - 2.45	0.6 -2.45	0.4 - 2.43	0.3 - 2.01
V	2.01 - 4.72	0.72 - 5.82	0.83 - 3.83	0.5 - 2.26
Cu	1.01 - 2.57	1.52 - 4.75	0.4 - 2.44	0.1 - 2.26
Ni	1.2 - 3.45	0.86 - 4.02	0.6 - 3.55	0.14 - 3.54
Cd	1.2 - 3.8	1.42 - 4.23	0.86 - 2.5	0.41 - 1.1

speeds during the study period were all below the threshold wind speed of 5.4 m/s. Episodes of highest daily wind speeds observed were 5.26 and 5.98 m/s in March 1 and 5; 7.36 and 7.94 m/s in June 8 and 9; 5.05 m/s in September 10; 5.09 and 5.19 m/s in December 4 and 14, respectively. The winter month of June experienced the highest wind speed, which could also explain the high amount of dust obtained in winter.

Variable wind directions were experienced and the directions with twenty (20) or more possibilities for occurance were taken as the predominant wind directions. The predominant wind directions were to the eastern (E) and south eastern (SE) directions in autumn, north western (NW), south western (SW) and western (W) directions in winter, western (W) and north western (NW) directions for both spring and summer respectively. Generally, the extent of impacts will depend on the location of dust sources and receptor sites, wind direction and wind speed. To the public, nuisance potential symbolizes possible health risk and the number of complaints may rise during the dry and windy seasons (Annegarn and Scorgie, 2002).

Heavy metal concentrations

The heavy metal concentrations obtained from the analysis of the EURO 876-1 CRM were very close to the theoretical concentrations. The percentage recoveries ranged from 85.87 to 111.54%, which confirmed the acceptability of the analytical method. Detection limits of < 0.01 µg/L and R-values of between 0.995 and 0.999 were obtained during the standard calibration process. The following ten heavy metals were identified at all sites, Zn, Fe, Cr, Ti, Mn, Pb, V, Cu, Ni, Cd and their ranges per season are listed in Table 1.

Generally, total metal ranges decreased seasonally in the order winter > autumn > spring > summer for most elements, except for a few (Figure 4a–e). Deviations from the general trends at Site A are that Fe autumn levels were higher than winter levels; Zn and Pb summer levels were higher than their spring levels. Site B had autumn > winter > spring > summer levels for most elements. Exceptions are that Ti, Pb, Cu and Cd winter levels were the highest. Zn, Ti and Mn summer levels were higher than the spring levels. Metal levels at site C followed the general trends, except that Fe summer levels were higher than their spring levels. Deviations at site D are Fe, Mn, V and Ni; autumn levels were the highest for all seasons. Also, Cr, Ti, Mn and Cu spring levels were lower than summer levels. Most elements followed the established distribution trends at site E. But V, Cu and Ni autumn levels were higher than the winter levels. The low levels of heavy metals found in summer and spring seasons overlaps with the heavy rains recorded during this period. This confirms the role of rain in removing the metal contaminated airborne dust. These results are supported by an observed decrease of heavy metals concentrations such as Cd, Cr and Pb associated with ambient air samples during wet seasons (Melaku et al., 2008). The distribution of these heavy metals within the study expressed in percentage coefficient of variance (CV) were 54% Zn, 25% Fe, 31% Cr, 26% Ti, 17% Pb, 48% V, 42% Cu, 28% Ni and 38% Cd. Lower % CV means that metal concentrations are comparable at all different sampling points within the study area, for example, Pb, Fe, Ti and Ni.

Most metallic elements evaluated exist in low levels in nature, but human activities have increased them to unacceptable high levels (Kalay and Canli, 2000). Within the study area, Mn is mainly used in steel production to improve quality, in the manufacture of alloys and as an additive in petrol to improve octane rating. About 50% of Mn emitted from vehicles are retained after being deposited on the soil surface, while the other half is reflected back into the air (Bhuie et al., 2005). But exposure to high levels of airborne Mn cause neurotoxins (Rollin et al., 2005). Despite high levels of Fe in the environment, Fe deficiency causes approximately half of all anemia cases worldwide and affects women more. Fe is used in the manufacturing of a number of metal alloys, including steel. Pure Fe is chemically reactive and

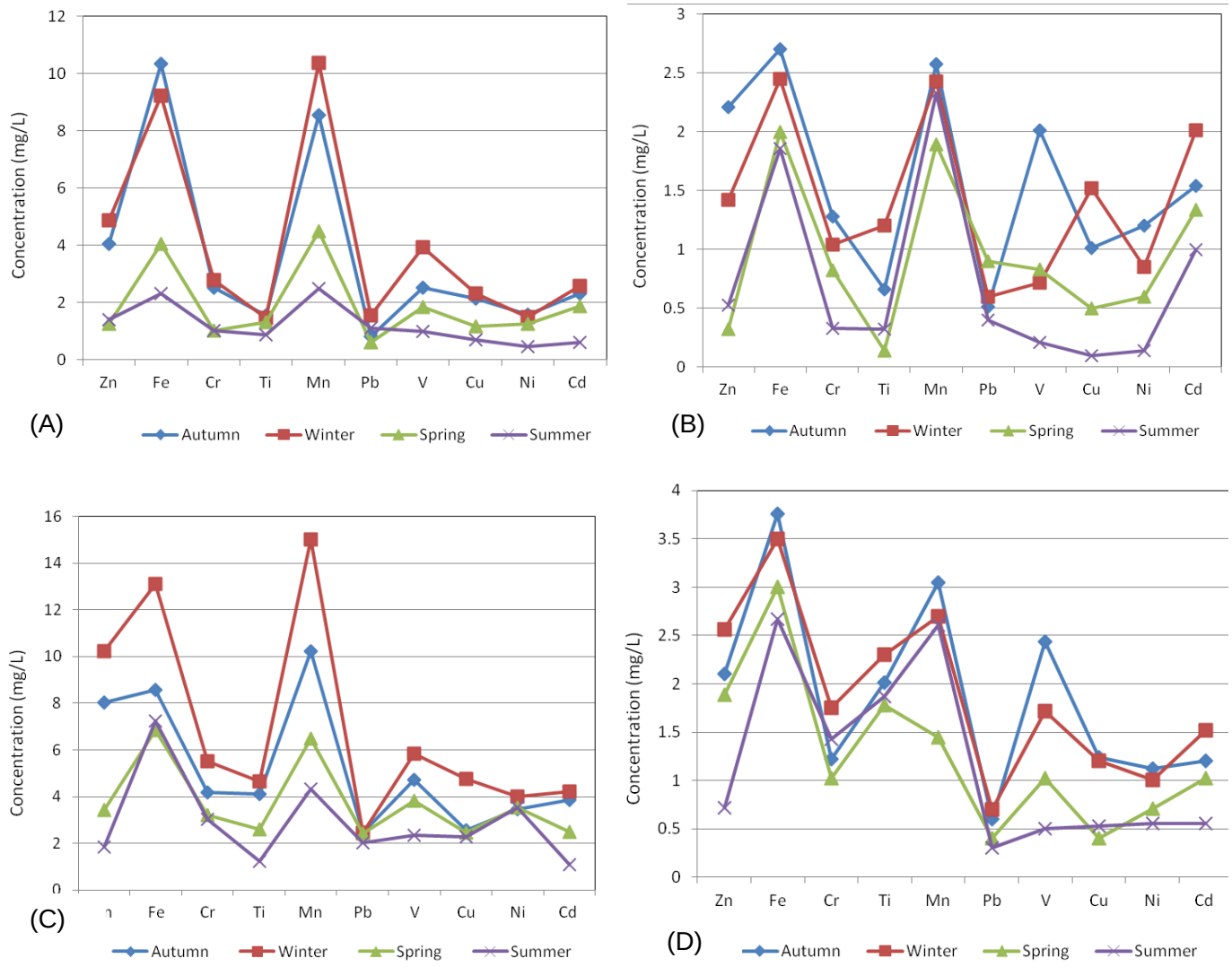

Figure 4. (a) Seasonal elemental distribution at site A. **(b)** Seasonal elemental distribution at site B. **(c)** Seasonal elemental distribution at site C. **(d)** Seasonal elemental distribution at site D.

rapidly corrodes, according to the Merck-Index. Inhalation of Cr^{6+} contaminated dust or coal ash has been shown to cause lung cancer, according to the US agency for toxic substances and disease registry toxicological profile for chromium. Breathing high levels of Cr^{6+} can also cause breathing problems, such as asthma, cough, and shortness of breath or wheezing.

The steel making process benefits from Cr's high corrosion resistance and hardness nature. Inhalable Cu is found on dust particles that contain organic matter and breathing it can cause irritation of the nose and throat, according to centers for disease control in the USA. Cu is used in metal alloys, plumbing materials, and has also been found in sewage sludge and in coal waste. Excessive absorption of Zn may suppress Cu and Fe absorption in humans. Zn has many commercial uses and is released into the environment by natural processes, but mostly comes from anthropogenic activities like mining, steel production, burning of coal and waste, according to the US agency for toxic substances and disease registry toxicological profile for zinc.

Cases of human poisoning by heavy metals are common. For example, exposure to high levels of vanadium in air can result in lung damage, as reported by the US agency for toxic substances and disease registry toxicological profile for vanadium. Vanadium pentoxide has been classified as a possible carcinogenic to humans. Anthropogenic vanadium (V) is emitted from combustion of coal and oil fuel activities. Ni and V benefits to human health are unknown, but their toxicities have been widely reported. Ni may cause dermatitis in sensitive individuals. Since about 20% of the population is sensitive to Ni, any exposure to it may lead to allergic reactions and chronic bronchitis, as reported according to

the US agency for toxic substances and disease registry, toxicological profile for nickel.

Anthropogenic sources of Ni in the atmosphere include emissions from coal and oil burning power plants, waste incinerators, manufacturing of stainless steels and other metal alloys. Cadmium (Cd) poisoning causes kidney failure, soften bones and is said to be responsible for the "itai-itai" disease due to the severe pain in the joints and the spine, as reported by the US agency for toxic substances and disease registry, toxicological profile for cadmium.

Modern day exposures to airborne Cd commonly occur in industrial settings where electroplating and manufacturing of alloys of these metals occur (Whitaker and Fowler, 2002; Hawkes, 1997). Some phosphate fertilizers contain Cd which can lead to an increase in the concentration of this metal in soil and dust (Taylor, 1997). The neuro-toxicity and nephrotoxicity of Pb has been highlighted as a major problem for humans (Whitaker and Fowler, 2002; Hawkes, 1997; Taylor, 1997). Lead is toxic at all levels, hence lead based petrol, toys and paint has been banned (DEAT and DME, 2003). The drift from leaded petrol in South Africa since 2006 is expected to contribute significantly to the decline in Pb levels in the air as reported by Okonkwo et al. 2006. This is very significant in the light of poor public transport system in the country.

Conclusion and Recommendations

Winter and autumn dust concentrations at site A, a township school site, was worrisome since the South African standard for residential limit was exceeded. Additional continuous monthly monitoring was required to ascertain if there were any sequential months of accidences that will occur so that remedial action could be taken. The levels of naturally occurring metals such as Mn, Fe, Cr, Cu and Zn from the study area could have been elevated by anthropogenic sources such as the steel making and metal alloy processing activities. Other possible contributors could be the coal burning within residential areas, industrial oil fuel combustion and vehicular fuel burning.

Fossil fuel also contributed to the high levels of V and Ni. Relatively high levels of Cd from site C (a farm area) could be linked to the Cd containing phosphate fertilizers which is commonly used near this site. Since heavy metals in the environment are persistent and not biodegradable, some metals which are no longer used currently could still be detected in the future. This would explain the significant presence of Pb in dust samples, which could be linked to the banned lead based fuel and lead based paint from old houses which are common in Vaderbijlpark area. Some of these metals could be linked to several metal works industries, as well as a nearby industrial waste dump.

This study recommends that a wider and larger scale study be undertaken to compare the performances of the cost effective passive samplers which were used here against the expensive passive samplers.

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