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RESEARCH ARTICLE

The impact of urban pollution on metal contamination of selected forest pockets in Cape Town, South Africa

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Abstract
Urban forests are exposed to metals, such as manganese, copper, and zinc in the atmosphere that originate from anthropogenic activities, that include vehicle-related traffic, industries, construction sites, fossil fuel burning for heating and cooking purposes, and resuspension processes related to urban surfaces. Not only is the rich biodiversity of plant and animal species in forests under threat, but so are the biodiversity of soil, sustaining ecosystem functions, as well as human health. The objective of this study was therefore to determine the concentrations of manganese, copper, and zinc arising from urban, industrial, and traffic-related pollution in the remote and/or untouched urban indigenous forests using soil, leaf litter, and key forest organisms (mosses, lichens, and millipedes) in three forests (Platbos, Orange Kloof, and Newlands) in the Western Cape, South Africa. Elevated concentrations of these metals were found in the forests closest to the city, as well as at sites in close proximity of vehicle traffic.

Keywords Metal: Forest · Metals · Urban pollution · Brown haze · Sentinel organisms · Soil · Leaf litter · Invertebrates

Introduction

The magnitude of ecological benefits and contribution to biodiversity conservation of urban vegetation, such as forests within urban ecosystems cannot be emphasized enough (Matthies et al. 2017). Of utmost importance is their function in the removal of air pollutants (Greene and Millward 2017) and the conservation of soil and water resources (Rey and Bullock 2012), granting these ecosystems the right to be actively protected (DAFF 2016).

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In Cape Town, remnants of indigenous afrotemperate forests occur on Table Mountain, which is a natural World Heritage Site and one of the Seven Wonders of the World (De Villiers et al. 2005). This mountain is globally known for the extraordinarily rich, diverse, and unique fauna and flora, as well as four of the remaining seven ancient forests in houses (SA-Venues.com 2017). However, it is also one of the world's most endangered ecosystems (Underwood et al. 2009). The root causes may be multiple but metals are considered one of the most critical forms of environmental contamination (Pruski and Dixon 2002) with known adverse and long-term impacts on remote ecosystems (Zhang et al. 2013), such as forests, not to mention the severe health effects on humans (Raukas 2010). Metals, such as manganese (Wei et al. 2014) and copper and zinc (Tan and Duan 2013) in the atmosphere are a consequence of anthropogenic activities. Vehicle traffic, industries, construction sites, fossil fuel burning for heating and cooking purposes, and resuspension processes related to urban surfaces (Li et al. 2014) all contribute to atmospheric particulate matter, that especially in winter become concentrated and appears as a brown colored haze, indicating severe pollution (Cheng et al. 2013). Moreover, it is a common occurrence in Cape Town's winter season, covering the whole of Cape Town and increasing in the number of episodes (CMA 2015). Xing et al. (2004) have furthermore confirmed similarities in the metal concentrations measured in haze and topsoil of remote areas.

Forests, even in remote areas, during the significant process of removal of air pollutants, intercept aerosols by means of their canopies (Gandois and Probst 2012), which indivertibly collects in the soil via precipitation or litter fall (Driscoll et al. 2007). Sizable metal basins are created in this way (Richardson et al. 2015) in soil, a habitat that is considered one of the most diverse on earth, comprising the most diverse combinations of living organisms (EU 2010). Soil biodiversity is the force behind soil-based ecosystem services, such as soil formation, nutrient cycling, carbon sequestration, and food production, but also in controlling the break-out of pests, water purification, and decontamination, as well as bioremediation of pollutants (Turbé et al. 2010). The biodiversity of soil may also be appraised economically as a natural capital asset, producing a flow of soil ecosystem services (Kareiva et al. 2011).

Soil biodiversity, however, has become severely threatened by rapid economic development and human activities, due to increased soil pollution, among others (Jacobsen and Hjelmsø 2014). A reduction in the biodiversity of soil (Breure et al. 2012) with strong possibilities of soil species becoming extinct are known consequences, necessitating it to become a priority for conservation measures (Gardi et al. 2013). This is significant, due to the strong link between the above and below ground biodiversity that interacts both direct and indirectly (Sylvain and Wall 2011). In a forest thus, the diversity of trees is instrumental in the inception of microhabitats that are able to support various species of soil biota (Schwarz et al. 2015). Organisms in forests are dependent on healthy functioning ecosystems for continuity and survival (Hurme et al. 2008) and it is, therefore, essential to sustain the forest ecosystem's function in terms of its response to air pollution (McLaughlin and Percy 1999) that includes the biodiversity of soil (Jeffery et al. 2010).

Information on the concentrations of polluting metals in Africa, either in the atmosphere or in atmospheric deposition or of its emissions are scarce (Slemr et al. 2011), as are its effect on ecosystem processes (Selonen and Setälä 2015). It is, however, widely accepted that a major part of anthropogenic metal emissions arises from vehicle traffic that infiltrates all three environmental domains, causing a pollution dilemma in the air, water, and soil that is both complex and problematic (Wåhlin et al. 2006). Therefore, representative organisms in forests, above and belowground, such as moss, lichen, and millipedes, as well as soil and leaf litter are well-known bioindicators of metal pollution, which can be used successfully to monitor metal contamination and simultaneously serve as a warning system for foreseeable problems (Greensdale 2007).

The objective of this study was therefore to determine the concentrations of manganese, copper, and zinc arising from urban, industrial, and traffic-related pollution in the remote and/or untouched urban indigenous forests using soil, leaf litter, and key forest organisms (mosses, lichens, and millipedes) in three of the seven ancient forests (Platbos, Orange Kloof, and Newlands) in the Western Cape, South Africa.

Methodology

Sampling area

Three afrotemperate forests in the Western Cape were selected as the sampling areas. Platbos forest on the slopes of the Baviaanspoort Hills served as a control area for comparison and is approximately 170 km from Cape Town. No potential sources of pollution are close to this forest. The other two areas were chosen because they are situated on the slopes of Table Mountain (Orange Kloof and Newlands forests), close to the Cape Town CBD.

Sampling sites

Three 20×20 m^2 sample sites within each of the three selected forests were chosen, as recommended by Fernandez et al. (2002) and Aboal et al. (2006), with the first sample site at the lowest point at least 300 m away from highways and 100 m from other types of roads or structures. The remaining two sample sites thereafter were at different altitudes on higher points on the mountain. Sites were planned between 150 to 200 m apart, but it was not always possible, due to rough terrain and availability of the organisms to be sampled.

Sampling procedure

The sampling for this investigation took place as a once-off event in May 2014. Five replicates of equal weight or size of each of the selected organisms at each of the sampling sites were randomly collected at least 50 cm apart.

Soil

The top 0–5 cm of soil under the leaf litter layer were collected and transported to the laboratory in clean 50-ml plastic vials (Křībek et al. 2010; Stafilov et al. 2010), where it was subjected to pH and moisture measurements. Soil samples were also collected for soil characterization analysis. The remainder of the soil samples was stored in a freezer before metal analysis.

Leaf litter

Decomposing leaf litter from various afromontane tree species were scraped from the top soil layer. Samples were transported separately in clean plastic bags to the laboratory where it was stored in a freezer before metal analysis.

Moss

Whole mosses were used in order to get an idea of metals that have accumulated over a long period of time. The species *Hypnum cupressiforme* was randomly collected from rocks at a height of 1 to 1.5 m from the ground to prevent any soil contamination (Bargagli and Nimis 2002). Samples were transported separately in clean plastic bags and cleaned from extraneous material, but not subjected to any washing in order to prevent possible changes in metal contents (Bargagli and Nimis 2002; Conti 2002). Moss samples were frozen before metal analysis.

Lichen

The lichen *Parmotrema* sp. was randomly collected and carefully removed from rocks at a height of 1 to 1.5 m from the ground to prevent any soil contamination (Bargagli and Nimis 2002). They were transported to the laboratory in clean plastic bags where they were cleaned to remove dust, leaf debris, fungus, and degraded material deposited on the surface, but not rinsed in water to prevent possible changes in metal contents (Bargagli and Nimis 2002; Conti 2002). To get an idea of metals that have accumulated over a long period of time, the whole lichen thalli were used for analysis. Lichen samples were brought back to the laboratory and frozen before metal analysis.

Millipedes

Similar sizes of adult millipedes (*Sphaerotherium compressum*) were hand-collected underneath the litter layer in the soil. They were transported separately in plastic containers in the forest soil in order to prevent unnecessary stress. In the laboratory, the millipedes were killed by putting them separately in labeled 10-ml plastic vials in the freezer before metal analysis.

Soil characterization, pH, and moisture

All three sites at Platbos have been classified as sandy as per soil characterization analysis. pH of the soil ranged from slightly acidic (6.48) to alkaline (7.28) and the moisture % of the soil ranged from a relatively dry 9.99% to a dry 4.16%.

Site 1 and site 3 at Orange Kloof forest have been classified as sandy and site 2 as loam sandy as per the soil characterization analysis. pH of the soil of Orange Kloof ranged from slightly acidic (6.48) to alkaline (7.64) and the moisture % of the soil ranged from a relatively moist 20.96% to a moist 36.77%.

Newlands, site 1 and site 3 have been classified as loam sandy and site 2 as sandy as per soil characterization analysis. pH of the soil was slightly acidic (6.19) to (6.39) and the moisture % of the soil ranged from a relatively moist 21.57% to a moist 40.33%.

Acid digestion

The soil, leaf litter, moss, lichen, and millipede samples were dried at 60 °C for 48 h and ground into powder, homogenized and weighed to approximately 0.2 and 0.3 g per sample. The weighed samples were digested with 10 ml 65% nitric acid, in accordance with the method used by Odendaal and Reinecke (1999), after which, it was filtered through Whatman no. 6 filter paper, followed by Whatman 0.45 μm cellulose nitrate membrane filter paper. The soil, leaf litter, and sentinel organisms were analyzed for manganese, copper, and zinc and the metal concentrations in the samples determined with an Inductively Coupled Plasma Mass Spectrophotometer (ICP–MS). Metal concentration is expressed as milligram per kilogram.

Statistical analysis

Kruskal–Wallis one-way analysis of variance on ranks was carried out to compare the concentrations of metals at different sites during this study in soil, leaf litter, lichen, moss, and millipede samples. The Student–Newman–Keuls method was used to do pairwise multiple comparisons and t-tests were used to compare soil and leaf litter, as well as moss and lichen concentrations. The probability level used for statistical significance was $P < 0.05$. Statistical analysis was done using the Sigmaplot 13.0 software package.

Results

Metal concentrations in soil, leaf litter, and sentinel organisms at sites within the three forests

The mean manganese, copper, and zinc concentrations in soil, leaf litter and sentinel organisms for sites 1, 2, and 3 of Platbos, Orange Kloof, and Newlands forests are presented in Table 1. No pill millipedes were found in Platbos forest, most likely due to the dry conditions and sparse layer of leaf litter observed during the time of sampling. Pill millipedes are usually found in habitats with high moisture and dense leaf litter in a loam soil, but can survive dry conditions by burrowing deeper into the soil and rolling into a ball (SANBI 2016). Metal concentrations are expressed in mg/kg.

Comparison of metal concentrations in soil, leaf litter, and sentinel organisms between the three forests

The mean metal concentrations in soil, leaf litter, moss, lichen, and millipedes for each forest for the sampling occasion in May 2014 are presented in Table 2. No millipedes were found at Platbos forest. The metal concentrations for the sites within each forest were pooled to calculate the mean concentrations

Table 1 The mean metal concentrations (mg/kg) (± SD) in soil, leaf litter, moss, lichen, and millipedes for sites 1, 2, and 3 of Platbos, Orange Kloof, and Newlands forests

Platbos		Soil			Leaf litter			Moss			Lichen			Millipedes		
Metal		Site 1	Site 2	Site 3	Site 1	Site 2	Site 3	Site 1	Site 2	Site 3	Site 1	Site 2	Site 3	Site 1	Site 2	Site 3
Mn	Mean	^{a}44.88	^{a}76.28	^{a}44.30	^{a}70.72	^{a}31.87	^{a}26.43	^{a}55.19	^{a}40.64	^{a}34.68	^{a}18.42	^{a}9.14	^{a}22.81	N/A	N/A	N/A
	SD	14.01	17.29	39.77	41.34	11.31	15.06	7.91	11.51	15.1	8.66	1.28	20.16	N/A	N/A	N/A
Cu	Mean	^{a}1.03	^{a}1.13	^{a}0.86	^{a}3.95	^{b}1.94	^{b}2.54	^{a}2.88	^{b}0.90	^{b}1.03	^{a}1.86	^{a}0.26	^{a}0.40	N/A	N/A	N/A
	SD	0.91	0.57	0.54	1.39	0.95	1.17	1.24	0.15	0.77	1.43	0.51	0.69	N/A	N/A	N/A
Zn	Mean	^{a}10.64	^{a}12.62	^{b}2.68	^{a}16.04	^{a}10.12	^{b}6.45	^{a}16.14	^{a}12.69	^{b}7.74	^{a}16.59	^{a}11.51	^{a}10.45	N/A	N/A	N/A
	SD	5.4	4.52	4.24	6.77	2.21	1.34	3.37	3.23	2.9	7.1	2.36	3.65	N/A	N/A	N/A
Orange Kloof		Soil			Leaf litter			Moss			Lichen			Millipedes		
Metal		Site 1	Site 2	Site 3	Site 1	Site 2	Site 3	Site 1	Site 2	Site 3	Site 1	Site 2	Site 3	Site 1	Site 2	Site 3
Mn	Mean	^{a}184.13	^{a}258.26	^{a}230.32	^{a}305.26	^{b}583.51	^{c}293.70	^{a}153.62	^{b}633.32	^{a}158.19	^{a}179.06	^{a}459.67	^{b}65.01	^{a}49.27	^{b}133.21	^{c}29.37
	SD	95.34	298.35	132.73	168.47	223.2	179.31	66.45	241.13	75.55	99.07	279.1	40.83	12.37	113.18	8.7
Cu	Mean	^{a}34.30	^{a}19.76	^{a}1.79	^{a}3.69	^{a}5.48	^{a}4.27	^{a}3.67	^{b}6.79	^{a}3.92	^{a}6.11	^{a}4.66	^{a}4.67	^{a}4.17	^{b}6.67	^{b}10.84
	SD	62.56	34.78	1.15	1.3	1.64	0.37	1.36	1.74	1.14	1.29	1.03	4.09	1.75	1.3	6.63
Zn	Mean	^{a}18.19	^{b}6.51	^{b}9.35	^{a}30.34	^{b}14.90	^{b}12.00	^{a}22.38	^{a}21.52	^{a}16.34	^{a}41.46	^{b}23.40	^{b}18.96	^{a}89.58	^{a}102.47	^{a}90.21
	SD	3.37	4.28	2.87	17.47	4.59	1.6	6.8	5.61	3.88	14.76	9.59	6.32	28.16	18.89	23.93
Newlands		Soil			Leaf litter			Moss			Lichen			Millipedes		
Metal		Site 1	Site 2	Site 3	Site 1	Site 2	Site 3	Site 1	Site 2	Site 3	Site 1	Site 2	Site 3	Site 1	Site 2	Site 3
Mn	Mean	^{a}454.51	^{a}672.24	^{a}997.06	^{a}564.88	^{b}855.75	^{b}1142.39	^{a}346.09	^{a}341.69	^{a}611.13	^{a}94.64	^{a}199.85	^{a}348.45	^{a}96.61	^{a}903.95	^{a}201.49
	SD	30.8	202.81	511.37	82.71	153.08	350.81	149.8	209.86	206.33	75.95	147.36	198.36	88.57	127.44	134.74
Cu	Mean	^{a}7.43	^{a}34.57	^{a}10.06	^{a}8.39	^{a}6.41	^{a}8.20	^{a}8.82	^{b}14.62	^{a}8.43	^{a}2.89	^{a}8.15	^{a}7.33	^{a}10.16	^{a}15.09	^{b}5.57
	SD	3.57	46	5.71	3.35	2.22	1.76	2.01	3.76	2.36	1.43	5.69	4.69	4.74	5.55	2.19
Zn	Mean	^{a}25.36	^{a}24.46	^{a}29.99	^{a}27.66	^{a}26.21	^{a}33.77	^{a}28.25	^{a}33.04	^{a}32.68	^{a}37.32	^{a}41.27	^{a}40.71	^{a}105.24	^{a}148.41	^{a}95.49
	SD	8.61	5.38	23.59	5.65	13.27	14.22	4.8	9.15	8.61	15.07	10.43	12.86	8.65	48.1	28

Statistical significant differences between sites within each forest are indicated with different superscripted letters. Comparisons were done separately for the soil, leaf litter, and sentinel organism

N/A = Not available; $N = 5$

Table 2 The mean manganese, copper, and zinc concentrations (mg/kg) (± SD) in soil, leaf litter, and sentinel organisms for forests

Metal			Platbos	Orange Kloof	Newlands
Mn	Soil	Mean	[a]55.15	[b]224.24	[c]707.94
		SD	28.85	184.56	374.14
	Leaf Litter	Mean	[a]43.01	[b]394.16	[c]854.34
		SD	31.72	225.3	321.51
	Moss	Mean	[a]43.5	[b]315.04	[c]432.97
		SD	14.15	271.61	219.47
	Lichen	Mean	[a]16.79	[b]234.58	[b]214.31
		SD	13.14	234.52	175.24
	Millipedes	Mean	N/A	[a]70.62	[b]159.63
		SD	N/A	76.78	119.47
Cu	Soil	Mean	[a]1.01	[b]18.62	[c]17.35
		SD	0.65	40.66	27.88
	Leaf Litter	Mean	[a]2.81	[b]4.48	[c]7.67
		SD	1.4	1.37	2.52
	Moss	Mean	[a]1.6	[b]4.79	[c]10.62
		SD	1.22	1.97	3.92
	Lichen	Mean	[a]0.9	[b]5.15	[b]6.13
		SD	1.14	2.46	4.67
	Millipedes	Mean	N/A	[a]7.23	[a]10.27
		SD	N/A	4.69	5.73
Zn	Soil	Mean	[a]8.65	[a]11.35	[b]25.92
		SD	6.25	6.11	14.05
	Leaf Litter	Mean	[a]10.87	[b]19.08	[c]29.21
		SD	5.63	12.78	11.34
	Moss	Mean	[a]12.19	[b]20.08	[c]31.32
		SD	4.62	5.84	7.54
	Lichen	Mean	[a]12.85	[b]27.94	[c]39.77
		SD	5.24	14.19	12.1
	Millipedes	Mean	N/A	[a]94.09	[a]116.38
		SD	N/A	23.02	38.38

Statistical significant differences between the three forests are indicated with different superscripted letters.

Comparisons were done separately for the soil, leaf litter, and sentinel organisms

N/A = Not available; $N = 5$

for each forest. Metal concentrations are expressed in milligram per kilogram.

Discussion

Manganese, copper, and zinc contamination found in Platbos, Orange Kloof, and Newlands forests

Orange Kloof and Newlands forests are in contrast with the remote Platbos forest located in the Cape Metropolitan Area, which is considered the most congested city in South Africa (BusinessDay 2017) with a population count of 3.74 million (StatsSA 2011) and 1.93 million registered vehicles (Wheels24 2017). As a result, the City of Cape Town produces an immense amount of particulate emissions originating from industries, agriculture, construction, and exhaust and non-exhaust emissions from vehicles and vehicle wear (Adachia and Tainoshob 2004). It is a well-known fact that the cause of higher concentrations of metals in atmospheric particulate matter is as a result of severe anthropogenic pollution (Tan and Duan 2013). High concentrations of manganese, copper, and zinc found in PM2.5 during China's severe brown haze episodes (Wang et al. 2014) have been closely linked to metal processing (Yu et al. 2013), steel mills, road dust surfaces, tail pipe emissions (Yang et al. 2013), biomass burning, waste incinerators, tires, brake linings, and brake pads (Brown et al. 2007). These are also prominent emitters in Cape Town and vehicle emissions have furthermore specifically been identified as one of the sources of brown haze in Cape Town (Piketh and Walton 2004). The brown haze is thus a reflection of the exorbitant concentrations of particulate matter produced in this city (Cheng et al. 2013; CMA 2015). The higher manganese, copper, and zinc concentrations found during the peak haze period in Newlands forest, closest to the city center, as opposed to Orange Kloof, more than 20 km away, further suggest the infiltration and larger impact of haze pollution from urban cities to urban forests. Moreover, these findings were corroborated with reports in China of PM2.5 concentrations measured in the remote Changbai Mountain forests of the adjacent city, which was slightly lower than were found within the city (Yang et al. 2011b). This is significant as particulate matter consists of inhalable particles of which the fine particles (PM2.5) are specifically harmful to health as a result of these highly toxic metals and other toxic materials it contains (Jouraeva et al. 2002). Metals in atmospheric particulate matter have adverse impacts on regional and global cycles due to deposition (Boquete et al. 2014), which amass in soil, the primary terrestrial receptacle of contaminants (Zhang et al. 2013). Soil microorganisms, plant growth and productivity, and human health are threatened by elevated accumulation and transport of metal uptake in plants that in the long run serve as food (Liang et al. 2010). Changes that occur in soil chemistry may therefore put entire ecosystems in danger (Enviropaedia 2011). Chemical pollution is one of the major threats to soil biodiversity, which ultimately causes a decline in the biodiversity of soil (Orgiazzi et al. 2016). Such a decline, due to contamination, among others, causes major stress on the healthy functioning of soils, its response to perturbation and ability to recover (Stolte et al. 2016). A decline in all life forms in soils, with regard to quantity and variety (Jones et al. 2005) and associated soil functions (Huber et al. 2008) is therefore considered one of the eight main soil threats in the Thematic Strategy for Soil Protection (COM 231 2006).

All three forests share similar characteristics by way of large parking areas and gravel roads leading up to the forests' entrances, subjecting them to emissions from traffic behavior associated with driving styles, such as frequent braking and acceleration (Apeagyei et al. 2011). Higher metal emissions in air samples in prominent stop-and-start areas have been reported in many studies (Johansson et al. 2009). Orange Kloof and Newlands are additionally impacted by traffic volumes and density (Abdul-Wahab and Yaghi 2004), arising from the adjacent highways. Road design and vehicle speed have also shown to impact metal concentrations (Legret and Pagotto 1999). Non-exhaust emissions release copper and zinc from brake wear (Johansson et al. 2009) and galvanized structures and car tire rubber fragments (Huber et al. 2016), which in addition to manganese in the petrol additive methylcyclopentadienyl manganese tricarbonyl (MMT), may same be emitted from exhaust processes (Nogueira and Röllin 2011). Site 1 of each forest was located close to the entrance of the forest and sites 2 and 3 higher up the mountain. Road dust can nevertheless be resuspended into the air by turbulence from passing vehicles, shearing stress of the tires or wind (Kupiainen 2007) and carry mobilized dust for hundreds of meters (Auerbach et al. 1997), thereby increasing metal concentrations in the nearby or elevated ecosystems (Adachia and Tainoshob 2004). Generally, the highest metal concentrations in soil, leaf litter, and sentinel organisms at Platbos were found at site 1, Orange Kloof at sites 1 and 2, and Newlands at sites 2 and 3 (Kluge et al. 2014). It was hypothesized that site 1 at each of the forests would reveal the highest metal concentrations, due to the higher rate of vehicle activity found at the entrances. However, the two forests closest to the Cape Town CBD presented higher metal concentrations at the sites located higher up the mountain. In fact, the closer the forest to the CBD, as in the case with Newlands, the higher the metal concentrations were at the highest sites on the mountain. This may suggest that those sites were impacted more so by air pollution from the brown haze infiltrating the forests from above (Piketh and Walton 2004), than the immediate emissions arising from the traffic at the foot of the mountain.

Platbos

At site 2, close to a minor gravel road leading up to the campsite's vehicle, traffic emissions were found sufficient to have caused the highest mean manganese (76.28 ± 17.29 mg/kg), copper (1.13 ± 0.57 mg/kg), and zinc (12.6 ± 4.52 mg/kg) concentrations in soil. This road would have endured heavier, but slower traffic due to the Easter holiday visitors in April before the samples were taken in May, subjecting this site to manganese from exhaust emissions containing the petrol additive, MMT (Nogueira and Röllin 2011), as well as copper from brake abrasion and zinc from tire wear, both from braking and acceleration (Thorpe and Harrison 2008). Tire wear may have been more excessive, taking into account the additional tires from caravans and trailers along with the vehicles and may explain why zinc concentrations were higher than copper concentrations. Site 3 in close proximity to the campsites located at the end of the gravel road and screened by the forest produced the lowest metal concentrations possibly due to very slow moving or parked vehicles and low brake abrasion and tire wear rates (Thorpe and Harrison 2008). Zinc showed the overall lowest mean concentrations at site 3 (2.68 ± 4.24 mg/kg), which differed significantly from the concentrations found at sites 1 and 2 ($P < 0.05$) and could also be due to zinc's tendency to stick to the road surface and settle in their immediate area upon emission, because of its larger particle size and weight in tire wear (Budai and Clement 2011) (Table 1).

Vehicle volumes at low speed utilizing the graveled main forest entrance (Kupiainen 2007) in the area of site 1 produced the highest mean concentrations of manganese (70.72 ± 41.34 mg/kg), copper (3.95 ± 1.39 mg/kg), and zinc (16.04 ± 6.77 mg/kg) in leaf litter with significant differences measured in mean concentrations of copper for comparisons: site 1 vs 2 and 1 vs 3, and zinc site comparisons between sites 1 vs 3 and 2 vs 3 ($P < 0.05$). Resuspension of manganese particles from natural soil (Li et al. 2013) and finer copper particles into the air by passing vehicles and wind could have caused the higher metal concentrations in leaf litter (Jacques et al. 2008; Sacharová and Suchara 1998). The elevated zinc concentrations may have been generated from tire wear (Thorpe and Harrison 2008) derived from the vehicles and the trailers and caravans. A decline in manganese and zinc concentrations were observed towards the furthest site, site 3, although copper showed slightly higher concentrations at this site, which may have resulted from copper's high affinity for organic matter (Alloway 1990) (Table 1).

Site 1 subjected to slow, stop-start vehicle traffic from the main forest entrance presented higher mean concentrations (Schauer et al. 2006) of manganese (55.19 ± 7.91 mg/kg), copper (2.88 ± 1.24 mg/kg), and zinc (16.14 ± 3.37 mg/kg) in moss. Enrichment of manganese in forest moss is generally a reflection of manganese cycling and recretion by the canopy (Gandois et al. 2010). Higher copper concentrations may have resulted from background concentrations due to internal cycling from old to new growing tissue (Harmens et al. 2015). Zinc was more than likely enhanced by moss absorbing metals directly from the atmosphere through its dense carpet (Sacharová and Suchara 1998) in an area already impacted by zinc through excessive tire wear (Thorpe and Harrison 2008) (Table 1).

Site 3, in close proximity of the campsites, showed the highest mean manganese (22.81 ± 20.16 mg/kg) concentrations in lichen. Lichens in this ancient forest and site are more protected from high vehicle activities and subsequent emissions and most probably showed higher concentrations due to their age, longer lifespan, and accumulation time (Armstrong and Bradwell 2010). Site 1, associated with slow-moving traffic, braking, and acceleration, on the other hand showed enhanced concentrations of copper (1.86 ± 1.43 mg/kg) and zinc (16.59 ± 7.1 mg/kg) (Apeagyei et al. 2011) in lichen samples. This may have occurred by means of resuspended soil particles (Jacques et al. 2008), as lichens are susceptible to metal accumulation from the atmosphere (Loppi et al. 1997). The lowest mean copper concentrations found at site 2 (0.26 ± 0.51 mg/kg) and zinc concentrations at site 3 mirrored the same pattern found with leaf litter and moss (Table 1).

The unavailability of pill millipedes may be by cause of the extended dry periods and low soil moisture faced with at the time (4.16 to 9.99%) (SANBI 2016) (Table 1).

Orange Kloof

The entrance of this forest is subjected to emissions from high traffic volumes at slower speed, due to the major traffic circle and the large parking area, which goes hand in hand with emissions from brake abrasion and tire wear as a result of constant braking and acceleration (Apeagyei et al. 2011). As a result, the mean copper (34.30 ± 62.56 mg/kg) and zinc (18.19 ± 3.37 mg/kg) concentrations were elevated in soil at site 1 close to the forest entrance. Zinc concentrations showed statistically significant differences between sites 1 and 2 and 1 and 3 ($P < 0.05$), of which the lowest concentrations were measured at site 2. The densely vegetated site 2, in close enough range of the traffic emissions showed the highest mean manganese (258.26 ± 298.35 mg/kg) concentrations in soil. Exhaust emissions containing the petrol additive, MMT (Nogueira and Röllin 2011) could have caused elevated manganese concentrations. However, its crustal origin (Li et al. 2013) could have also contributed, due to the small deposits located on the mountain (Jones 2010). Vegetation may further have elevated the surface soil concentrations (Herndon et al. 2011) by means of biological cycling through litterfall, throughfall, and uptake (Herndon et al. 2015) (Table 1).

Site 1 in close proximity to the traffic circle and carpark showed the highest mean zinc (30.34 ± 17.47 mg/kg) concentrations in leaf litter. Soil also produced the highest zinc concentrations, which may suggest the metal enrichment in leaf litter was caused by microbial translocation and immobilization of metals from underlying contaminated soil layers, by fungi, although this was not directly investigated in this study (Tyler 2005). Significant differences were measured between sites 1 and 2, as well as between 1 and 3 ($P < 0.05$) and zinc concentrations were the lowest at site 3. Leaf litter at site 2 displayed the highest mean concentrations of manganese (583.51 ± 223.2 mg/kg) and copper concentrations (5.48 ± 1.64 mg/kg). Significant differences in manganese concentrations were measured between all of the sites ($P < 0.05$). Increased manganese concentrations in leaf litter can be caused by physico-chemical processes in the tree canopy (Avila and Rodrigo 2004) and copper has a high affinity for organic matter, which may explain the higher copper concentrations (Alloway 1990) (Table 1).

Mean zinc concentrations (22.38 ± 6.8 mg/kg) were also elevated in moss at site 1. The wide spread carpet of the moss absorbs metals directly from atmospheric deposition (Sacharová and Suchara 1998), which in this case would have been emitted from the traffic activities (Kupiainen 2007) at the forest entrance. The highest mean manganese (633.32 ± 241.13 mg/kg) and copper (6.79 ± 1.74 mg/kg) concentrations in moss were found at site 2, of which manganese enrichment could have occurred by cycling of manganese and recretion by the canopy (Gandois et al. 2010) at this densely canopied site. Copper enrichment in moss may have been brought on by resuspended soil particles in the ambient air (Jacques et al. 2008) through wind (Thorpe and Harrison 2008) and subsequently absorbed by moss (Sacharová and Suchara 1998). Both manganese (153.62 ± 66.45 mg/kg) and copper (3.67 ± 1.36 mg/kg) concentrations in moss were the lowest at site 1 (Table 1).

Copper (6.11 ± 1.29 mg/kg) and zinc (41.46 ± 14.76 mg/kg) concentrations in lichen were the highest at site 1, with significant differences found in mean zinc concentrations between sites 1 and 3, as well as between 1 and 2 ($P < 0.05$), of which site 3 presented the lowest zinc concentrations. Lichens are sensitive to atmospheric contaminants (Wolterbeek 2002), rendering them susceptible to metal accumulation from the atmosphere (Loppi et al. 1997), which in all likelihood originated from the traffic circle and parking area (Kupiainen 2007). The highest mean manganese (459.67 ± 279.1 mg/kg) concentration in lichens was found at site 2 with significant differences in mean manganese concentrations measured between sites 2 and 3, as well as between 1 and 3 ($P < 0.05$). Older lichens may have been sampled at the densely vegetated site 2 in this ancient forest, meaning that their longer lifespan and accumulation time may shed some light on their higher concentrations (Armstrong and Bradwell 2010). Resuspended soil particles may also have been functional in causing higher manganese concentrations in lichen (Sacharová and Suchara 1998) (Table 1).

Mean concentrations of manganese (133.21 ± 113.18 mg/kg) and zinc (102.47 ± 18.89 mg/kg) in millipedes were the highest at site 2, with significant differences found in mean concentrations of manganese between sites 1, 2, and 3 ($P < 0.05$). Mean copper (10.84 ± 6.63 mg/kg) concentrations in millipedes were elevated at site 3, in close range of the entrance overlooking Hout Bay and impacted by wind carrying particulate emissions from industries and vehicle traffic (Abdul-Wahab and Yaghi 2004) and the lowest concentrations were found at site 1. Significant differences in copper concentrations were measured between sites 1 and 2, as well as between 1 and 3 ($P < 0.05$). The quantities of metals found in decaying leaf litter expose millipedes to much higher metal concentrations (Hopkin 1989), because they digest just about half of the bacterium and fungal hyphae in the litter (Anderson and Bignell 1982). The metal accumulation observed in millipedes is thus an indication of the millipedes consuming polluted litter at contaminated sites (Da Silva Souza et al. 2014) (Table 1).

Newlands

Site 3 is the more densely vegetated site, situated the highest and furthest from the forest entrance and highway at the foot of Newlands forest. Small manganese deposits are present on Table Mountain (Jones 2010) and soil naturally contains manganese (Yang et al. 2011a), which may have enhanced the manganese (997.06 ± 511.37 mg/kg) concentrations in soil measured at this site. The concentrations may further have been elevated by vegetation enhancing soil surface concentrations through biological cycling (Herndon et al. 2011). However, zinc (29.99 ± 23.59 mg/kg) concentrations in soil were also the highest at site 3, similar to findings in Poznań city, which was attributed to traffic emissions resulting from volumes, vehicle component wear, as well as domestic heating (Gunawardena et al. 2013). Newlands forest is impacted by atmospheric pollution from the Cape Town CBD due its position and it may therefore be suggested that site 3 may in part have been contaminated by the city's anthropogenic activities and concentrated pollutants, also from the brown haze (Wang et al. 2014) that covers Cape Town during this very season of sampling (CMA 2015). Site 2 situated lower on the mountain in the area of the contour path in clear view of the Cape Town CBD may same have been impacted by strong winds and particulate emissions from the brown haze (Wang et al. 2014), as this site produced the highest mean copper (34.57 ± 46.00 mg/kg) concentrations. Such findings were also reported in an urban forest impacted by both copper and zinc, located far from heavy traffic and point source emissions and was ascribed to influence from local winds (Siudek et al. 2015) (Table 1).

As in the case with soil, site 3 displayed the highest mean concentrations of manganese (1142.39 ± 350.81 mg/kg) and zinc (33.77 ± 14.22 mg/kg) in leaf litter. Manganese concentrations could have been enhanced by physico-chemical processes in the tree canopy (Avila and Rodrigo 2004). Significant differences in mean manganese concentrations were found between sites 1 and 3, as well as between 1 and 2 ($P < 0.05$), of which site 1 showed the lowest concentrations (564.88 ± 82.71 mg/kg). The elevated zinc concentrations may have occurred through the metals moving upward from the polluted soil into the leaf litter via microbiota (Kaila et al. 2012), as soil at this site also contained the highest zinc concentrations. Site 1, located at the foot of the forest and in close proximity of the forest entrance, parking and highway is characterized by volumes of vehicles at different speed, brake, use and industrial emissions (Duong and Lee 2011), which may clarify the highest mean copper concentrations (8.39 ± 3.35 mg/kg) in leaf litter at this site in addition to copper's high affinity for organic matter (Alloway 1990) (Table 1).

Manganese enriched in moss at site 3 (611.13 ± 206.33 mg/kg) is generally a reflection of manganese cycling in forest ecosystems and recretion by the canopy (Gandois et al. 2010). Site 2, close to the contour path, presented the highest mean copper (14.62 ± 3.76 mg/kg) and zinc (33.04 ± 9.15 mg/kg) concentrations in moss with significant differences found between sites 2 and 3, as well as between 1 and 2 ($P < 0.05$). Studies have shown forest sites to be impacted by copper from brake abrasion and zinc from tire wear and domestic heating even at long distances from heavy traffic and point source emissions, due to wind (Gunawardena et al. 2013; Siudek et al. 2015) and could well have been the case at this site. Copper concentrations may further have been enhanced by background concentrations occurring from internal cycling from old to new growing tissue (Harmens et al. 2015) (Table 1).

Once again, site 3 showed the highest mean manganese (348.45 ± 198.36 mg/kg) concentrations in lichen, which could be by cause of resuspended soil particles in the ambient air (Jacques et al. 2008), as manganese was found constantly higher in soil, leaf litter, and moss at this site. Mean copper (8.15 ± 5.69 mg/kg) and zinc (41.27 ± 10.43 mg/kg) concentrations in lichen were the highest at site 2 impacted by air emissions in all likelihood transported from the industrial areas and traffic activities by wind (Duong and Lee 2011), as well as the pollutant-laden brown haze (Wang et al. 2014). Lichens being sensitive to atmospheric contaminants (Wolterbeek 2002) are known to accumulate metals from the atmosphere (Loppi et al. 1997) (Table 1).

Millipedes at site 2 presented the highest mean concentrations of manganese (903.95 ± 127.44 mg/kg), copper (15.09 ± 5.55 mg/kg), and zinc (148.41 ± 48.1 mg/kg). Mean copper concentrations displayed significant differences between sites 2 and 3, as well as between sites 1 vs 3 ($P < 0.05$) and site 3 showed the lowest concentrations. Leaf litter is known to accumulate an exceeding amount of metals (Bengtsson 1986) and millipedes ingest the polluted litter from these contaminated sites (Da Silva Souza et al. 2014). Site 2, situated lower on the mountain than site 3, was especially exposed to anthropogenic emissions (Abdul-Wahab and Yaghi 2004) from the Cape Town CBD and the recurring brown haze episodes (Piketh and Walton 2004). This is quite possibly due to the brown haze occurring as low as 30 m above ground (CMA 2015), causing site 2 to be more impacted by the haze than site 3 (Table 1).

Comparisons of manganese, copper, and zinc concentrations between the three forests

Platbos presented the lowest manganese, copper, and zinc concentrations in the soil, leaf litter, and sentinel organisms.

Due to the absence of industrial activities in this area, contamination more than likely originated from the immediate vehicle-related traffic from visitors, owners, and general maintenance (Lee et al. 2012). These findings concur with other findings of emissions decreasing swiftly within kilometers of emission hotspots like cities additionally impacted by industries and urbanization (Aznar et al. 2008), since the overall highest metal concentrations were measured at Newlands, closest to the Cape Town CBD, followed by Orange Kloof. Contamination of these forests may thus have occurred from particulate emissions containing these metals, derived from the many industries functioning in this city, but exhaust and non-exhaust emissions from vehicles and vehicle wear (Adachia and Tainoshob 2004) have been earmarked as one of the principal sources of metal contamination in urban environments, due to the vast amounts of metals they emit into the air, water, and soil (Christoforidis and Stamatis 2009). Manganese at all three forests was measured in higher concentrations than zinc and copper, of which the origin may be from parent material (Li et al. 2013) and exhaust emissions containing the petrol additive methylcyclopentadienyl manganese tricarbonyl (MMT) (Nogueira and Röllin 2011). Zinc concentrations were constantly higher at all three forests than copper concentrations, suggesting that tire wear played more of a role than brake wear, of which copper is emitted. It is also possible that the copper concentrations were lower, due to its finer particle size, which is easily emitted into the atmosphere and is deposited over extended areas, as opposed to zinc that tends to remain closer to its source (Thorpe and Harrison 2008). Higher zinc concentrations were, however, also found in Poznań city from domestic heating in addition to traffic emissions resulting from volumes and vehicle component wear (Gunawardena et al. 2013).

The brown haze hovering over Cape Town is laden with complex mixtures of these and other pollutants (Cheng et al. 2013; CMA 2015) that would have made several appearances during this sampling occasion and in all probability contributed significantly to the metal contamination in these forests. The notably higher metal concentrations at Newlands, closest to the city center during the period of constant haze episodes, signified haze impact on urban forests, threatening not only the above ground biodiversity, but also the biodiversity of its soil, which is closely linked. Soil biodiversity is fundamental in providing services to sustain the above ground biodiversity (Sylvain and Wall 2011). It is thus evident in these results that secluded forest ecosystems, located in industrialized areas are infiltrated by pollutants by long-range transport through the atmosphere and emphasizes that air pollution is not restricted to the city (Steinnes and Friedland 2006). Chemical contamination from industrialization and urbanization are thus the main culprits (Zhang et al. 2003) threatening soil, which should be able to enhance plant and biological productivity and preserve biodiversity (Vrscaj et al. 2008). Even though the metal concentrations measured in these forests were not as high as have been cited in many other countries, the measured concentrations and patterns at the very least should serve as a warning sign that urban forests, generally used recreationally and as food sources, poses a possible ecological risk to the food chain (Desaules et al. 2001) and the Cape Town forests are indeed impacted by the increasing urbanization, industries, vehicle traffic, and prolonged brown haze episodes.

Newlands presented the highest mean concentrations of manganese (707.94 ± 376.14 mg/kg) in soil that was three times higher than were measured at Orange Kloof and 12 times higher than were found at Platbos. Manganese deposits present on Table Mountain (Jones 2010) may indeed clarify in part some of the concentrations measured in the soil at all three forests (Yang et al. 2011b). However, the increasing metal concentrations towards Newlands, closest to the CBD may suggest that manganese concentrations were enhanced by exhaust fumes containing the petrol additive, MMT (Nogueira and Röllin 2011) from volumes of vehicles driving at different speeds on the heavily traveled adjacent highway, as well as from the CBD (Thorpe and Harrison 2008). Mean zinc (25.92 ± 14.05 mg/kg) concentrations at Newlands were also twice as high as were measured at Orange kloof and three times higher than at Platbos, reiterating a pattern of pollution (Aznar et al. 2008). Zinc contributions were most likely from traffic emissions due to volumes, vehicle component wear, but also from domestic heating in this colder season (Gunawardena et al. 2013). The brown haze, containing metals, also from surrounding industries (Amato et al. 2014) may further have caused an escalation in metal concentrations (Cheng et al. 2013). Copper mean concentrations were the highest at Orange Kloof (18.62 ± 40.66 mg/kg), with statistical differences found between all the forests ($P < 0.05$). The fine copper particles from brake abrasion emitted into the air upon its release can travel far, due to wind (Siudek et al. 2015) and impact the forest. Platbos metal concentrations in soil were significantly lower when compared to the following general back ground concentrations for soil: manganese (40–900 mg/kg) (Nogueira and Röllin 2011), copper (5–70 mg/kg) (ATSDR 2004), and zinc (10–300 mg/kg) (IZA 2015). Manganese and copper concentrations found at Orange and Newlands were comparable to the above concentrations, but zinc concentrations were significantly lower (Table 2).

Newlands forest under anthropogenic pressure from Cape Town's vehicle traffic, related to volumes, speed, brake, and tire abrasion, various metal industries (Adachia and Tainoshob 2004), natural soil (Yang et al. 2011a), and brown haze (Piketh and Walton 2004) presented the highest mean manganese (854.34 ± 321.51 mg/kg), copper (7.67 ± 2.52 mg/kg), and zinc (29.21 ± 11.34 mg/kg) concentrations in leaf litter. All three metals displayed significant differences in mean concentrations between all the forests ($P < 0.05$). Manganese, enhanced by physico-chemical processes in the tree canopy (Avila and Rodrigo 2004), copper's high affinity for organic matter (Alloway 1990), and metals moving upward from the polluted soil into the leaf litter via microbiota (Kaila et al. 2012) may have been functional in causing higher metal concentrations in a forest already highly impacted by anthropogenic activities (Amato et al. 2014). The metal concentrations in leaf litter measured at Platbos were significantly lower than were cited in the literature for: Manganese in a Medvednica Nature Park forest (385–1070 mg/kg) (Jelaska et al. 2007) and copper (4.08 mg/kg) and zinc in a Finnish forest (69.63 mg/kg) (Hristovskia et al. 2014). Orange Kloof and Newlands manganese and copper concentrations were comparable, but zinc concentrations were lower in comparison with the abovementioned concentrations (Table 2).

Mean manganese (432.97 ± 219.47 mg/kg), copper (10.62 ± 3.92 mg/kg), and zinc (31.32 ± 7.54 mg/kg) concentrations in moss showed the highest results at Newlands, adjacent to the large city (Li 2006; Kluge et al. 2014) and higher rates of anthropogenic activities (Abdul-Wahab and Yaghi 2004). Significant differences in mean manganese, copper, and zinc concentrations were found in moss between all three forests ($P < 0.05$). Elevated concentrations of manganese could have resulted from manganese cycling in the forest ecosystem and recretion by the canopy (Gandois et al. 2010), copper from background concentrations caused by internal cycling from old to new growing tissue (Harmens et al. 2015), and zinc from absorbing these metals directly from atmospheric deposition (Sacharová and Suchara 1998). The mean metal concentrations measured at Platbos were significantly lower than the following concentrations found for: manganese (97.40–172.81 mg/kg), copper (3.78–8.20 mg/kg), and zinc (24.31–34.72 mg/kg) concentrations in French forests (Gandois et al. 2014). Manganese concentrations at Orange Kloof and Newlands forests were significantly higher than the manganese concentrations in the French forest, but copper and zinc concentrations were comparable (Table 2).

The highest mean manganese (234.58 ± 234.52 mg/kg) concentrations in lichen were measured at Orange Kloof, with significant differences found between the forests: Newlands vs Platbos and Orange Kloof vs Platbos ($P < 0.05$). Higher manganese concentrations in lichen at the ancient Orange Kloof forest may be resultant of age due to longer accumulation time (Armstrong and Bradwell 2010), but also resuspension of soil particles containing manganese (Sacharová and Suchara 1998). Newlands forest displayed the highest mean concentrations of copper (10.62 ± 3.92 mg/kg) and zinc in lichen (31.32 ± 7.54 mg/kg). Significant differences in mean concentrations of copper were recorded between Newlands and Platbos and Orange Kloof and Platbos and zinc between all the forests ($P < 0.05$). Lichens are sensitive to atmospheric contaminants (Wolterbeek 2002) causing them to be susceptible to metal accumulation from the atmosphere (Loppi et al. 1997), which may have caused the higher concentrations. Platbos mean concentrations of manganese, copper, and zinc were comparable to unpolluted French forest concentrations of: manganese (25.14–29.29 mg/kg), copper (4.66–6.88 mg/kg), and zinc (22.05–30 mg/kg) (Gandois et al. 2014). Orange Kloof concentrations of all three metals were comparable to the French forest concentrations. Newlands forest manganese concentrations exceeded the French forest manganese concentrations, but the copper and zinc concentrations were (Table 2).

Millipedes at Newlands showed the highest mean concentrations of manganese (159.63 ± 119.47 mg/kg), copper (10.27 ± 5.73 mg/kg), and zinc (116.38 ± 38.38 mg/kg). These millipedes live among metal-contaminated soil and leaf litter, of which they ingest high quantities of (Hopkin 1989) in a forest that is continuously impacted by metal pollution from anthropogenic activities on a greater scale than the other two forests and it shows in the higher metal concentrations found in them (da Silva Souza et al. 2014). It should, however, be noted that several factors have an influence on toxic element accumulation in invertebrates, such as food preferences, breeding, development stage, sex, season and physiological conditions (Jelaska et al. 2007). Copper (306–585 mg/kg) concentrations found in millipedes in unpolluted forests in the Netherlands exceeded the concentrations found in this study, as did the zinc (152–827 mg/kg) concentrations (Hobbelen et al. 2004) (Table 2).

Conclusion

It may be concluded that vehicle traffic, associated with exhaust and non-exhaust emissions, with additional inputs from industries and especially the brown haze to the Cape Town forests, were instrumental in the elevated manganese, copper, and zinc concentrations measured in the forests. The metal concentrations at the various sites and forests are, however, influenced by traffic characteristics, such as road environment, vehicle amounts, speed of vehicles, braking and acceleration, as well the atmospheric dispersion situations. Newlands forest, closest to Cape Town, presented the highest metal concentrations, followed by Orange Kloof and Platbos, demonstrating a polluted related pattern. Similarly, higher metal concentrations were found at the sites in closer proximity of higher traffic areas. The petrol additive, MMT, and manganese cycling in all likelihood elevated manganese concentrations in leaf litter at the Cape Town forests. Leaf litter enriched in copper is generally caused by copper's high affinity for organic matter and in zinc in lichen normally due to polluted soil underneath the leaf litter moving upward via microbiota. Mosses with larger absorption areas showed higher accumulation of manganese and copper and zinc enriched in lichen is generally due to biological recycling. Millipedes consuming polluted litter are evident in the metal concentrations they displayed. The combined use of sentinel organisms was found as reliable indicators of atmospheric pollution. It is also in the interest of human health and well-being to conserve urban forests with the emphasis on soil biodiversity for themselves and the environment as a whole.

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