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Published in:
Chemosphere

DOI:
10.1016/j.chemosphere.2022.135051

Print publication: 01/09/2022

Document Version
Peer reviewed version

Deposition of trace metals associated with atmospheric particulate matter: Environmental fate and health risk assessment

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Abstract

Anthropogenic and natural sources influence trace metals (TMs) bound to different sized particulate matter (PM) in dry and wet atmospheric deposition, which can create ecosystem and human health issues in the long run. Limited reviews are available summarizing worldwide concentrations in TMs in atmospheric PMs, their sources and pathways. Simultaneously, quantitative assessment of the potential human and ecosystem health risks imposed by the atmospheric particulate matter has not been adequately reviewed. Addressing this gap, here we review, the concentration of TMs in dry deposition mainly varies with the responsible sources, whereas, in wet deposition, it depends on the solubility of TMs. Health risk assessments show that ingestion and dermal contact pathways are more likely to cause health issues, however, the probability of occurring ingestion and dermal contact pathways is limited. The ecological risks are especially significant in urban and industrial regions due to increasing emissions, leading to direct damage to the environment and biological organisms. Attention must be paid to the contribution from non-exhaust and exhaust vehicular emissions for TMs in atmospheric deposition, understanding their impact on stormwater management and urban agriculture. Behaviors of TMs in the atmosphere depends on many complex factors including origin, wind patterns, and weather conditions. Therefore, future research needs to be carried to model and predict the fate and transfer of TMs once they are generated through natural and anthropogenic sources. We believe that such research would allow establishing pollution control policies and measures in urban environments which will be critical to reduce the levels of TMs associated with atmospheric deposition in the environment.

Keywords Atmospheric particulates · Pollutant sources · Risk assessment · Trace metals · Transport pathways

1. Introduction

Natural and synthetic materials released into the atmosphere at levels that have a detrimental effect on human health and the environment are classified as air pollutants (Kampa and Castanas, 2008; Nordberg et al., 2007). Major airborne pollutants include ozone, carbon monoxide, nitrogen dioxide, sulfur dioxide, and particulate matter (PM) (Manisalidis et al., 2020). The latter is particularly concerning because it acts as a carrier for a wide range of pollutants such as trace metals (TMs), hydrocarbons, sulphates, and nitrates, to list but a few (Amodio et al., 2014; Weerasundara and Vithanage, 2015). Furthermore, PM_{10} and $PM_{2.5}$, are mostly related to soils, organic and elemental carbon-bound particles that arise from combustion activities. Atmospheric pollution is usually governed by local climate, geology, and edaphic factors (Karim et al., 2014), natural processes such as wind turbulence, sea spray, volcanic eruptions, dispersion of crustal minerals, and forest fires also play a significant role in generating and spreading air pollutants (Duan and Tan, 2013; Karim et al., 2014; Soriano et al., 2012). However, anthropogenic activity is the primary contributor of atmospheric pollutants, including those known for their elevated toxicity, such as, TMs, volatile organic compounds, and hydrocarbons (Karim et al., 2014). Over the last few decades, TMs have been receiving increasing attention due to their hazardous effects, bioavailability, and non-degradability (Tchounwou et al., 2012).

Atmospheric TM emissions are often related to fast industrialization, increased urbanization, inappropriate agricultural practices, and unsuitable waste disposal methods (Vardhan et al., 2019). The main anthropogenic sources of TMs are industrial activities (including mining operations), weathering of building materials, power generation, and vehicular traffic (Abdellatif and Saleh, 2012; Duruibe and Egwurugwu, 2007). Though trace quantities of metals such as Fe, Cu, Mn, and Zn are critical to human, animal, and plant life, elements like As, Hg, Cd, and Cr can

have a harmful impact on living organisms, even at low concentrations (Duruibe and Egwurugwu, 2007; Khan *et al.*, 2008). Some of the effects of TMs and metalloids on human health comprise respiratory problems, cardiovascular disease, renal damage, neurological disorders, skin lesions, teratogenic effect, and various types of cancer diseases (Nordberg et al., 2007). Therefore, conducting human and ecosystem risk assessment is pivotal to predict the impacts of TM associated with atmospheric deposition (Han et al., 2020; Mama et al., 2020). Non-biodegradable pollutants such as TMs that are persistent in nature are transferred across the different trophic levels through biomagnification, reaching levels that can be toxic for living organisms (Popoola et al., 2018; Sabin et al., 2005).

Airborne TMs associated with particulates eventually deposit through wet and dry deposition processes resulting pollutant build-up on land (Altaf *et al.*, 2021; Zgłobicki et al., 2018). In addition, the TMs deposited on the ground can be flushed by stormwater runoff and contribute to the overall pollution of the receiving water bodies (Davis and Birch, 2010; Weerasundara et al., 2017; Ziyath et al., 2013). Consequently, TM contamination by atmospheric deposition can be a severely limiting factor in improving the quality of water resources (Rocher et al., 2004). Over the last few decades, there have been emerging research studies on air pollutants that have paid close attention to TMs transports and their quantification through the use of different monitoring systems including biological systems, or otherwise, i.e., Lichen, Moss and urban vegetation (Altaf et al., 2021; Demková et al., 2017; Kumar et al., 2021; Santos et al., 2019). Moreover, recent studies have focused on the trace elements in atmospheric deposition in an ecological perspective to assess short-term accumulation which can lead to unhealthy series of events. Therefore, in this review, we describe the status of TMs in the atmosphere; entry pathways into the atmosphere and their behavior, deposition mechanisms, influential factors, and the

behaviors of TMs in different deposition processes, analysis and finally, the risks of deposited atmospheric TMs on human and ecosystem health.

2. Trace metals in the atmosphere

The atmosphere is one of the major carriers for TMs through a suspended mixture of solid and liquid particles (Azimi et al., 2005; Pope and Dockery, 2006; Weerasundara et al., 2017). These particles vary in size, shape, chemical composition, surface area, and solubility, as well as in origin. Yet, PM is generally categorized on the basis of the particle's aerodynamic diameter: (i) coarse particles known as PM_{10}, with an aerodynamic diameter smaller than 10 µm; (ii) fine particles known as $PM_{2.5}$, with an aerodynamic diameter smaller than 2.5 µm; and (iii) ultrafine particles (UFPs), with an aerodynamic diameter smaller than 0.1 µm (Pope and Dockery, 2006). The coarse fraction of PM encompasses suspended or resuspended particles from volcanos, farming, mining, roads, sea spray, windstorms and deserts, amongst others. PM_{10} is also constituted by particles of biological origin such as fungal spores, pollen, other plant and insect parts. The fine fraction of PM is primarily composed of combustion particles, such as those emitted by traffic and domestic fuel combustion, wood burning, mining sites, coal-fired power plants, and industries such as cement, smelters, steelworks, paper mills, etc. (Ghose and Majee, 2020, Kumar et al., 2019). These particulates gain long-range transport ability (LRTA) through mass loaded aerosol (Kumar et al., 2019, 2020). The ultrafine fraction of PM contains organic and elemental carbon, sulfates, and nitrates, typically formed during fossil fuel combustion and condensation of semi- volatile substances (Pope and Dockery, 2006). In comparison to coarse particles, fine and ultrafine particles play a dominant role in human and animal health, as they tend to remain in the atmosphere

for a longer period, penetrate easily into the indoor environments, are transported for long distances, and can even penetrate deeply into the lungs (Pope and Dockery, 2006).

Although PM-bound metals commonly originated from the natural and anthropogenic activities. Studies have shown anthropogenic origins are more frequent (Azimi et al., 2005; Samontha et al., 2007; Weerasundara et al., 2017). Soil, seawater, volcanic dust, and volcanic gases are considered the most common natural sources for atmospheric TMs (Azimi *et al.*, 2005). On the other hand, industrial activities, mining, fossil fuel combustion, traffic-related emissions, incineration of urban solid waste, and wastewater treatments are some of the most widely identified anthropogenic sources of airborne TMs (Azimi et al., 2005; Ghose and Majee, 2020; Li et al., 2019; Samontha et al., 2007). Stationary sources such as mechanical plants, construction worksites and residential heating are linked to the production and emission of particles that contain Fe (Ambade, 2014; Aničić et al., 2009; Fang et al., 2004). Metal-enriched traffic emissions from tailpipe gases and the wear and tear of different vehicle parts and poor road infrastructure are frequently connected to the atmospheric release of a broad range of metals (Novo et al., 2017). Moreover, the quantity and array of metals in the atmosphere surrounding roads are often governed by traffic volume, type of vehicular traffic (e.g. heavyweight vehicles), road age, speed limits and industrial nature of the area (Bernardino et al., 2019; Bernardino et al., 2021; De Silva et al., 2021; Li et al., 2019). Table 1 describes the details of most common sources of traffic-related metal emissions.

Table 1: Traffic-related metal emissions

Source	As	Cd	Cr	Cu	Hg	Ni	Pb	Pd	Pt	Rh	Sb	V	Zn

	Cd	Cr	Cu	Ni	Pb	Sb	Zn
Batteries				■	■	■	
Brakes			■	■		■	■
Catalytic Converters					■	■	■
Fuel	■	■		■	■	■	
Galvanized structures		■		■			■
Lights					■		
Oil		■	■	■			■
Paint					■		
Road abrasion			■	■		■	
Tires		■	■	■		■	■

Note. Based on Weckwerth, 2001, Yan et al., 2018, Bernardino et al., 2019, and De Silva et al., 2020

With the advancement of urbanization, traffic-related pollution and TM expulsion to the environment can be a major threat to ecology and human health. In addition to emission sources, different environmental factors and meteorological conditions (e.g. seasonal variation, wind direction, wind speed, and temperature) have significantly influenced metal concentrations in the atmosphere and deposition pathways as described in the following sections (Cheng et al., 2014; Hovmand et al., 2008; Tasdemir et al., 2006).

3. Deposition of trace metals

The process through which atmospheric pollutants are transferred into the terrestrial and aquatic surfaces is called atmospheric deposition (Azimi et al., 2005; Gunawardena et al., 2013; Han et al., 2014). Bioavailable fractions of TMs in surface environments arose from atmospheric deposition. Around 68-74% of Cd and Zn originated from wet deposition through precipitation and 25-33% through dry deposition based on the literature (Fig. 1). Many studies have shown the characteristics of wet and dry atmospheric deposition through analytical protocols to check the impact of the airborne trace metal load in different areas (Table 2) which are vital to assess the

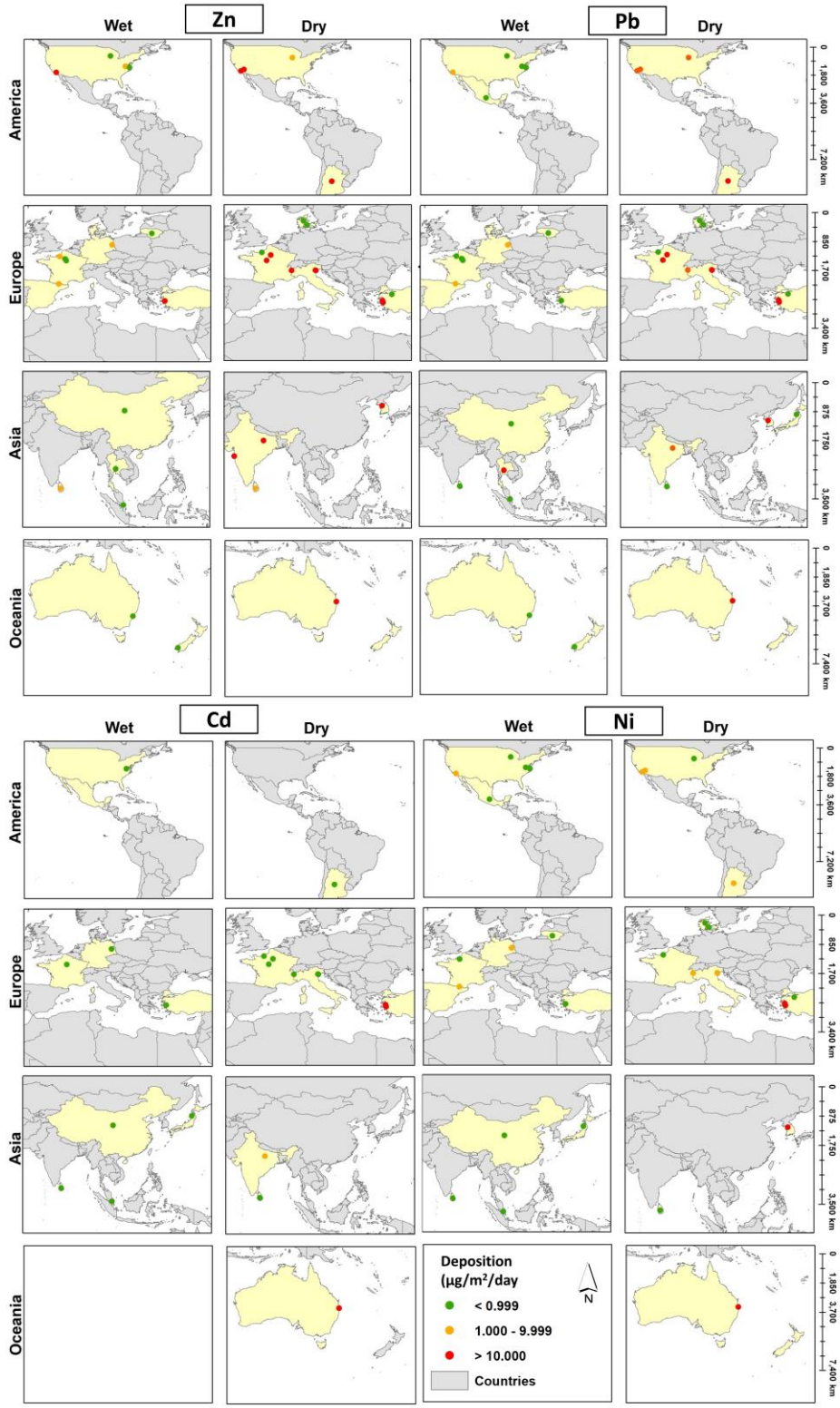
overall environmental quality. While atmospheric deposition operates as a mechanism for the removal of trace metals that are bound with the PM from the atmosphere, simultaneously, it acts as a pathway for loading TMs into the terrestrial and aquatic ecosystems through wet and dry deposition (Kara et al., 2014; Mijić et al., 2010; Yun et al., 2002). Evaluating and determining the presence or absence of these elements in the different bioavailable forms through varying solubility for both wet and dry deposition is a challenging task. Numerous studies are being published over the years with the mechanisms that determine the pathway of pollutant movement in the ecosystem (Ma et al., 2016; Morselli et al., 2003).

3.1 Dry deposition

Dry deposition is defined as the direct deposition of atmospheric particles on ground surfaces without snowfall or rainfall events (Duruibe and Egwurugwu, 2007; Ziyath et al., 2013). As the dimension of TM bound particulates vary from few nanometers to several tenths of micrometers, their residential time in the atmosphere largely depends on the size (Deshmukh et al., 2013). Consequently, dry deposition rates are higher for coarse particulates than for fine PM (Fang et al., 2004).

Dry deposition is particularly concerning in urban and industrialized areas (Shahin et al., 2000). Relatively high TM emission rates and the lack of natural recovery methods, such as vegetation and microbial communities that promote the accumulation and degradation of the deposited PM, contribute to the increment of pollution rates in urban and industrialized environments (Fang et al., 2004; Shahin et al., 2000; Weerasundara et al., 2017). Therefore, worldwide monitoring of dry deposition of atmospheric TMs, especially in urban and

industrialized areas, is essential to assess its risk to biotic and abiotic environments (Fang et al., 2004).



1 **Fig. 1** Map of selected trace metals (Zn, Pb, Cd and Ni) in dry and wet deposition in different

2 countries.

Numerous studies have reported dry atmospheric deposition worldwide (Bermudez et al., 2012; Fang et al., 2004; Theodosi et al., 2010). Table 2 denotes the concentrations of TMs via dry deposition in the world.

Table 2 Dry deposition of trace metals found in different regions of the world (μg m^{-2} day^{-1})

Location	Metal									Remarks	Ref.
	Cr	Cu	Pb	Ni	Zn	Fe	Mn	Cd	Hg		
Guangzhou, China	17.9	9.8	-	3.0	54.1	-	13.1	0.097	-	Urban area	(Huang et al., 2014)
Zhengzhou, China	0.7	0.7	0.8	0.2	1.0	13.0	0.7	1.9	-	Near and urban, industrial area	(Wang et al., 2019)
Saraburi Province, Thailand			10.26[a] 0.92[b] 0.0008[c] 2.42[d]		0.389[a] 0.23[b] 0.17[c] 0.78[d]			0.003[a] 0.001[b] 0.001[c] 0.0006[d]		a-0.03 km, b-1 km and c-1 km away from a smelter, d-heavy traffic area	(Samontha et al., 2007)
Northwest France			0.05	0.085	1.56			0.004	0.003		(Wang et al., 2019)
Creteil, France		0.073	0.093		0.268			0.002		Urban city area	(Garnaud et al., 1999)

Paris, France		0.048	0.116		0.211			0.001		Urban city area	(Garnaud et al., 1999)
Fontainebleau, France		0.022	0.034		0.298			0.014		48km away from an urban city area	(Garnaud et al., 1999)
Aukštaitija, Lithuania	0.004	0.024	0.035	0.007	0.153	1.06	0.047	0.001	0.0001		(Kvietkus et al., 2011)
Žemaitija, Lithuania	0.005	0.056	0.065	0.022	0.384	1.32		0.002	0.0001		(Kvietkus et al., 2011)
Lake Michigan, USA			0.06	0.05	0.21				0.003		(Landis and Keeler, 2002)
Mexico City, USA			0.005	0.007				0.001			(Báez et al., 2007)
Virginia, USA			0.123	0.067	1.14			0.015			(Conko et al., 2004)
Chesapeake, USA			0.012	0.013	0.068			0.001			(Kim et al., 2000)
Eastern Ohio, USA									13.5		(Keeler et al., 2006)
Eastern Ohio, USA									19.7		(Keeler et al., 2006)

Los Angeles, USA	0.84	9.6	1.4	1.8	73			Urban area	(Sabin et al., 2005)
Izmir, Turkey			1.053	1.36	12.610		0.360		(Keeler et al., 2006)
Izmir, Turkey	0.172	0.197	0.07	0.074	1.864		0.031		(Keeler et al., 2006)
Balearic Islands, Spain		0.002	0.0005	0.0008	0.0096	0.56	0.008	Remote area	(Cerro et al., 2020)
Algiers, Algeria		0.06	0.16	0.02		0.35	0.03	0.01	10 km away from the city center	(Oucher et al., 2015)
Fiordland, New Zealand			0.01		0.019		0.0002		(Halstead et al., 2000)
Sydney, Australia		0.005	0.018		0.022				(Davis and Birch, 2010)

Road vehicles and infrastructure are responsible for the emission of a broad array of metals, including Hg, Cd, Cr, Cu, Ni, Pb, Zn, and V, as well as platinum group elements Pd, Pt, and Rh (Bernardino et al., 2019; Bernardino et al., 2021; Novo et al., 2017). The rates of deposition are found to be higher in urban and industrial areas than in non-urban sites, despite the velocities of the wind being subjective to the area being studied. Strong co-relations were achieved in the aerosol mass loaded concentrations with respect to the particle size that determines the fluxes in the given area (Rocher et al., 2004). However, heavy traffic-oriented sites are known to have heavy

turbulence in the air, showing severe dry deposition, as studied reported by Mohan (2016) (Azimi et al., 2005; Tasdemir et al., 2006). Therefore, it can be concluded that traffic activities are major contributor to the atmospheric heavy metal dry deposition (Gunawardena et al., 2015; Weerasundara et al., 2017; Werkenthin et al., 2014).

Dry deposition is also related to meteorological parameters such as, wind direction, seasonality, and topographic profiles (Negral et al., 2021). It has been reported that the dry deposition in day time was higher than night time fluxes which were reported at Sha-Lu, a traffic island in central Taiwan, mainly due to the considerable influence of wind speed. The daytime average wind speed was recorded 2.5 times of night time wind speed. This was also further confirmed by the Pearson's correlation coefficient values (0.63 and 0.74 for day time and night time respectively) (Fang et al., 2004).

The influence of particle size on the long range transport of mass loaded air and the deposition of atmospheric TMs was clearly demonstrated by dust storm events (Yi et al., 2001; Han et al., 2004). The yellow sand event occurred in Korea was well-known dust storm responsible for transporting both crustal and anthropogenic TMs in Korea from the Gobi desert, the Alashan semi-desert, the central China loess plateau, and from the major industrial areas of China (Han et al., 2004). The Al deposition, reported in the yellow sand event was 1.5-3 times higher than the normal levels (Han et al., 2004; Yi et al., 2001;). Dry deposition of PM and size distribution were simultaneously studied. The migration of yellow sand was the trailing effect of TM sources (Han et al., 2004). The concentrations of Pb, Zn, Cu, and Ni originated by the anthropogenic activities were about 4.7 times higher than the normal levels (Han et al., 2004; Yi et al., 2001;). These findings confirmed a significant load of anthropogenic TMs in the yellow sand event, as these are associated with fine PMs exhibiting longer range transport ability than coarse particulates.

It has been proven impact of dry deposition on the aquatic ecosystems as it provides significant pathways for loading the trace metals from the atmosphere onto the water bodies (Lim et al., 2006; Pope and Dockery, 2006). Proximity to the source of the elements along with the size distribution of the pollutant, plays a major role in dry deposition fluxes on receptor surfaces (Sakata and Asakura, 2011). Diurnal changes in the deposition was found inconsistent due to the concentration variations and size distribution, especially in winter and spring seasons with the reduced precipitations and the minimum emission sources. For instance, Guo et al. (2017) has reported that the Chromium levels were found 19.7 and 5.42 g/TM^2/month in winter and summer in Miyun water reservoir of Beijing. Moreover, meteorological factors such as the wind speed influences the mass loaded aerosol transport from the source to the sampling point significantly affecting the dry deposition rates. As studied in Lim et al. (2006), TM deposition in the coastal transect of California was primarily caused by the offshore advection of contaminated air from the nearby industrial sources in the day time (Lim et al., 2006; Pope and Dockery, 2006).

Studies have indicated that the emission control of PM-bound metals is an effective way to reduce TM emissions into the atmosphere (Weerasundara et al., 2017). For example, an eight years long study in Paris was able to observe a significant decrease of Cd and Pb, a trend that can be attributed to the utilization of unleaded gasoline and a better flue gas treatment of point sources of contaminants (Azimi et al., 2005).

3.2 Wet deposition

Wet deposition is a process involving the combination of water droplets and particles scavenged in and below the clouds, due to precipitation (Ambade, 2014; Samontha et al., 2007). During precipitation, fine particles of anthropogenic origin get wiped away due to nucleation where aerosols or droplets get in the way. In contrast coarse particles of crustal origin get scavenged

below the clouds during precipitation (Aničić et al., 2009; Samontha et al., 2007). In summary, atmospheric particles get deposited on ground surfaces during rainfall or snow events, after being dissolved in the clouds and attached with precipitation droplets (Azimi et al., 2005; Duruibe and Egwurugwu, 2007). This allows TM wet deposition to be quantified by analyzing the precipitation of an area (Zhang et al., 2018).

The concentration of a metal deposited through wet deposition is generally positively correlated with its solubility (Pöschl, 2005). However, in the context of wet deposition, the solubility of metals depends on several factors, such as the pH of rainwater and the origin of particulates scavenged from the air (Wuana and Okieimen, 2011). It has been noted that different metals found in wet deposition samples exhibit different soluble fractions. For example, Al and Fe have soluble fractions of less than 20% in wet deposition. Co, Ni, Pb, Mn, Cu, and Zn exhibit soluble fractions around 50-70%, and Cd exhibits 80% soluble fraction when present in wet deposition (Hsu et al., 2010; Wuana and Okieimen, 2011). Moreover, it is important to compare the "excess number" of each metal to determine its origin. The "excess number" (expressed in percentual terms), is the equivalent of the enrichment factor and denotes the fraction of non-crustal origin of a metal 1997). A lower or near 0 "excess number" suggests a crustal origin, while an "excess number" closer to 100 points to an anthropogenic origin. According to that, the crustal fraction is significant for Al, Co, Mn, and Cr (23- 63%). Ni, Cu, Zn, Cd, and Pb are 90-100% anthropogenic. The low soluble fraction is reported for metals with a crustal origin, and the higher soluble fraction is for anthropogenic elements (Hsu et al., 2010; Samontha et al., 2007). Samantha *et al*. 2007 used the enrichment factor to assess the origin of metals from a lead smelter and heavy traffic are in Bangkok. Following atmospheric sampling for one year period, authors determined that for both tested locations, Pb, Zn, and Cd exhibited an anthropogenic origin based on

enrichment factors beyond 20 while Fe and Al mainly were of crustal origin, as indicated by their enrichment factor closer to unity (Samontha et al., 2007). Similar results were obtained in a study conducted in Kandy, Sri Lanka, indicating the crustal origins of Al and Fe found in heavy traffic areas. Atmospheric wet deposition was of a crustal origin. The metals with crustal origin were reported in significantly higher concentrations than anthropogenic metals such as Zn, Pb, Cr, Cd, Cu, Ni, and Mn (Weerasundara *et al., 2017)*. Table 3 shows the metal concentrations through wet deposition around the world.

Table 3 Wet deposition of TMs found in different regions of the world (mg m^{-2} $year^{-1}$)

Location	Metal									Remarks	Ref.
	Cr	Cu	Pb	Ni	Zn	Fe	Mn	Cd	Hg		
Guangzhou, China	0.61	9.33	-	4.66	47.9	-	32.7	0.215	-	Urban area	(Huang et al., 2014)
Jiaozhou Bay, China	0.67	-	2.21	-	24.1	14.2	24.2	0.13	-	Area with rapid industrialization	(Xing et al., 2017)
Wanzhou, China	<DL	<DL	0.12	<DL	0.49	0.26	0.64	<DL	-	Urban environment	(Azimi et al., 2004)
Yangtze River Delta, China		10.4	20.6	25.99	25.47			0.64		Industrial area	(Ma et al., 2019)
Taiyuan, China	14.4	24.4	7.3	3.6	59.1	655.0	17.1	0.3	-	Urban area	(Tian et al., 2020)
Jomson, Nepal	0.96	1.32	0.86	1.07	9.60	385.5	10.8	0.02	0.01		(Tripathee et al., 2020)
Balearic Islands, Spain		0.3	0.2	0.2	1.9	77	1.1			Remote area	(Cerro We thank the reviewer

							for his comments We thank the reviewer for his comments, 2020)
Northwest France		0.05	0.085	1.56	0.004	0.003	(Connan et al., 2013)
Creteil, France	0.073	0.093		0.268	0.002		(Garnaud et al., 1999)
Paris, France	0.048	0.116		0.211	0.001		(Garnaud et al., 1999)
Fontainebleau, France	0.022	0.034		0.298	0.014		(Garnaud et al., 1999)

The origin of the metal is not the only factor deciding the solubility of a particular metal (Hsu et al., 2010; Soriano et al., 2012; Wuana and Okieimen, 2011). Alike dry deposition, wet deposition is also influenced by the preceding weather (Muezzinoglu and Cizmecioglu, 2006). Wet deposition will have increased metal concentrations when a rainfall event occurs after a long dry period (Muezzinoglu and Cizmecioglu, 2006; Weerasundara et al., 2017). Apart from the preceding weather, metal concentrations in wet deposition also hang on the precipitation duration (Kvietkus et al., 2011; Weerasundara et al., 2017). It has been noted that the concentration of TMs in wet deposition is higher in short-term than in long-term precipitation, as many metals are washed during the first minutes of the event (Šakalys et al., 2003; Kvietkus et al., 2011). Usually, rainfall

events occur irregularly. Therefore, metal concentration patterns in wet deposition are also irregular (Kvietkus et al., 2011) (Table 3). The influence of seasonal variability is shown to cause varieties in recorded TM concentrations. For example, higher concentrations are recorded during non-monsoon rain events, which can be explained by the effect of dilution occurring during heavy monsoon rain events (Cong et al., 2010; Tripathee et al., 2014). Lower concentrations during monsoon can also be attributed to the impact of heavy and relatively cleaner marine airflow (Hovmand et al., 2008).

3.3 Deposition on biological agents

The dust accumulated on impervious surfaces may not be retained for an extended period because stormwater runoff and wind can carry the particles to nearby water bodies or resuspend them into the atmosphere (Harmens et al., 2010). Biological agents such as mosses, lichens, and higher plants can biologically fix, absorb, and immobilize deposited atmospheric metals (Cloquet et al., 2006; Holt and Miller, 2011). However, higher plants cannot absorb the deposited particles through their cuticle layer. Hence, higher plants also act as impervious surfaces for deposition. Mosses and lichens can act as bio-indicators for atmospheric deposition (Geiser et al., 2010; Giordano et al., 2005). Since they have a high surface/volume ratio, they accumulate a higher amount of dust and TMs from the atmosphere (Giordano et al., 2005; Kularatne and Freitas, 2013). Additionally, a higher ion exchange capacity of mosses and lichens leads to efficient absorption of TMs and other ions. Also, the lack of cuticle and morphological variability make them suitable sites for atmospheric deposition (Giordano et al., 2005). In fact, atmospheric deposition is one of the major mineral nutrition sources for lichens. The lichens are rather prone to deposition and absorption of

TMs through wet and dry deposition processes as they lack a root system (Kularatne and Freitas, 2013).

In urban environments, the metals deposited on lichens and mosses are predominantly of anthropogenic origin, However, depending on the geological location, metals identified in lichens and mosses can be attributed not only to anthropogenic origins but also to natural geochemical sources such as volcanic rock, soil and sea salts (Giordano et al., 2005). Table 4 presents the different types of lichen species that have been used to study the atmospheric TM concentrations.

Table 4 Different species of mosses and lichens that used as biomonitors in different studies

Organism	Species	Region	Reference
Epiphytic lichen	*Usnea, Bryoria*	Northwestern North America	(Simonetti et al., 2003)
Lichens	*Evernia prunastri* *Ramalina farinacea* *Usnea* sp. *Hypogymnia physodes*	France	(Cloquet et al., 2006)
Epiphytic lichens	*Parmotrema reticulatum*	New Zealand	(Kularatne and Freitas, 2013)
Lichens	*Parmelia sulcata*	Netherlands	(Sloof, 1995)
Mosses	*Sphagnum girgensohnii*	Belgrade, Serbia	(Vallero, 2014)
Terrestrial moss	*Pseudoscleropodium purum*	Galicia,Spain	(Boquete et al., 2014)
Mosses	*H. splendens, P. cschreberi*	Finland	(Poikolainen et al., 2004)
Mosses	*Hylocomium splendens* *Pleurozium schreberi*	Norway	(Berg and Steinnes, 1997)
Mosses	*Pleurochaete squarrosa* *Hypnum cupressiforme*	Urban Taranto	(Buonocore et al., 2010)

Mosses	*Hypnum cupressiforme* *Brachythecium*	Europe	(Ghio, 2006)
Mosses	*Sphagnum capillifolium* *Pseudevernia furfuracea*	Naples, Italy	(Giordano *et al.*, 2005)

A study conducted in Metz, France, employed three different lichen species (*Evernia purunastri, Ramalina farinacea*, and *Hypogymnia physodes*) to assess the variation of metal concentrations in urban and rural areas. At a 4 km radius area from the city center, metal concentrations in lichen samples were 2-3 times lower whereas in rural areas, metal levels decreased by ten-fold. Smaller incidence of anthropogenic activities in rural areas is the likely explanation for such variation while these observations are supported by the enrichment factor calculations. Median concentrations of TM in collected lichen samples were in the order of Zn > Pb > Cu > Cr > Ni > As > Cd and for all of these metals, the enrichment factor was found to be around 9, even at as farthest as 15.5 km from the city center indicating their anthropogenic origin. As the vehicular activities and other industrial activities are high in the city center, and reported concentrations were also higher than other sampling sites. Furthermore, there can be other factors such as wind speed and direction contributing to the contaminant transfer. For example, Pb concentrations have shown a systematic increase along the south-north cross-section where industrial activities are high while the frequent and strong wind blows from south to north (Cloquet et al., 2006). In a study carried out across Europe, mosses were used as bioindicators to find the spatial patterns and temporal trends of atmospheric metal deposition. Due to the existence of rhizoids in mosses, these differ from lichens in accepting TMs. These rhizoids can absorb metals, while some metals can be absorbed by the surface of the aboveground parts (Harmens et al., 2010).

4. Impacts of trace metals

The emitted TMs do not deposit on surfaces right after the emission, they disperse and get transported. Throughout this retention period, the TMs can cause direct and indirect harmful impacts on human and ecosystem health (Laguionie et *al.*, 2014). Especially, TMs emissions that have anthropogenic origins are known to cause significant surface TMs depositions on the biosphere components such as water bodies, soil, and man-made surfaces, as studied by many researchers. This section summarizes recent studies about the impacts of TMs on the ecosystem and human health.

4.1 Impacts on ecosystem health

In the aquatic environments, metals are available for biogeochemical cycling in their dissolved forms (Muezzinoglu and Cizmecioglu, 2006; Soriano et al., 2012). Dissolved forms of TMs are dispersed and discharged into nearby groundwater environment sources, as well as agricultural lands and other soil environments. These transfer pathways are extremely important and dangerous as they eventually allow TMs to enter the food chain and drinking water sources increasing the exposure to humans (Pandey et al., 2010).

TMs can enter water bodies directly via deposition and indirectly via surface runoff. Dry deposition on the aquatic ecosystem has shown to be impactful, as it provides significant contribution to the TM loading from the atmosphere onto the water bodies (Rocher et al., 2004). Areal dispersion of TMs can spread in a wider range, hence, TMs deposited onto water bodies are more dominant than the TMs from surface runoff (Pandey et al., 2010; Zhang et al., 2018).

A study conducted in Jiaozhou Bay, China, a rapidly industrializing area of the region with increasing vehicle traffic due to the newly constructed bridge and tunnel, revealed moderate

amounts of Se, Cd, Zn, Pb, Mn, Cr, and Co due to the wet deposition thorough rain and snow. Also, the study concluded short, yet heavy rain events resulted in a surge of TM content in the surface water polluting the marine aquatic environment (Xing et al., 2017). In a study conducted to investigate the TM contamination of the Ganga river at Varanasi, Zn > Ni > Cr > Pb > Cu > Cd elements were found at concentrations mostly below the permissible drinking water standards. All the tested TMs were increasing downstream, while some farthest sampling sites detected heavy metals above permitted limits (Pandey et al., 2010).

The atmospheric deposition of TMs can easily contaminate the soil. Difficulties related to metal detection can occur, when the sampling site is considerably distant from anthropogenic sources or heavily fertilized over the years, making soil analysis for TMs from atmospheric deposition a problematic task (Azimi et al., 2004; Chen and Lu, 2018; Hu et al., 2018). A study in France found out that the atmospheric deposition is a significant input pathway for Cu, Ni, Pb, and Zn in soils. The deposition of none of these TMs agreed with the total precipitation. In other words, dry deposition resulted in higher depositions while wet deposition during more significant rain events was low; however, the peak concentrations for each metal were recorded in different periods during the study (Azimi et al., 2004; Chen and Lu, 2018).

It has been reported that the anthropogenic activities led to heavy metal accumulation in the peri-urban agricultural soils of China threatening soil environmental quality and agricultural product security (Peng et al., 2019). In the literature study of China from 2008-2018 about heavy metal inputs from various pollution sources to agricultural soils, it has been reported that the atmospheric deposition was the main pollution source which was responsible for 50–93% of the total As, Cd, Cr, Hg, Ni, and Pb inputs, with livestock manures contributed to approximately 76% of total Cu inputs (Shi et al., 2019). Input and output inventory of heavy metals in the agricultural

soil of Zhejiang showed that atmospheric deposition was responsible for 47.88% and 76.87% of the total Cr and Pb inputs, respectively. Livestock manures accounted for approximately 54- 85% of the total As, Cu, and Zn inputs. Livestock manure and irrigation were the main sources of Hg, contributing 50.25% and 38.63% of the total inputs, respectively. Ni was derived mainly from atmospheric deposition (57.86%), followed by irrigation (22.69%). As for Cd, the relative contributions of atmospheric deposition, irrigation, and livestock manure were similar. Crop harvesting and leaching were found to be the dominant output pathways of the soil elements Cd, Cu, Hg, and Zn, being responsible for 74.43–83.62% of the total outputs. Surface runoff was the dominant output pathway for As, Cr, Ni, and Pb, accounting for approximately 73.36%, 46.32%, 54.16%, and 48.11% of the total outputs, respectively (Shi et al., 2019). In a study conducted in China, atmospheric dry deposition was found to be a crucial source of trace metal available to plants. The deposition fluxes of Cu, Cd, and Pb were studied over a year and speciation studies of soil analyzed in those areas revealed a rise of 8-19% for Cu, 36- 42% Cd, and 7-14% Pb as compared to their control counterpart (Khan et al., 2008).

Association of TMs with atmospheric PM can result in the long-range transport of TMs due to environmental conditions (Siudek and Frankowski, 2017; Zang et al., 2021). In a recent study, Zang et al. (2021) reported evidence of long-range transport of anthropogenic TMs and their impact on remote forest ecosystems. The study was set up in the Qilian Mountains, China, and the authors collected wet atmospheric deposition samples for about two months representing low, middle, and high forest coverage. As a summary, the order of TM concentrations was observed to be high coverage sites> middle coverage sites> low coverage sites, while the order of TM concentrations was Cu > Pb > Ni > Cr > Cd > As > Hg. They also found traces of more bioavailable Hg and Cd in rainwater, which can cause various serious problems to the ecosystem (Zang et al.,

2021). Another study conducted in a remote forest site in Poland reported depositions in the order of Zn < Cu < As < Pb < Cr < Ni < Cd indicating long-range transport of TMs of anthropogenic origins such as coal combustion and vehicular emissions (Siudek and Frankowski, 2017).

Barks of the trees are known to shelter long-term deposition of atmospheric TMs. Frequent rainfall events run over these dry deposits to remove TMs from the bark and transfer them to the soil beneath the stemflow or even to the surface run-off. However, the same rainfall events can contaminate some parts of the tree bark which are unharmed by dry deposition previously, while some are vacated for further deposition (Catinon et al., 2012; Türtscher et al., 2017). In a long-running study conducted in Austria, it was found that a reduction in vehicular and industrial emissions resulted in a decline in the atmospheric deposition as demonstrated by the TM content in foliage and soil. Researchers have re-sampled soil within the stemflow area and foliage of beech (*Fagus sylvatica*) stands to find that TM pollution no longer poses a threat compared to the permissible limits. For example, Pb concentration in the soil in stemflow area was reduced by 78% over three decades. This indicates that the reduction in anthropogenic origins of TMs could result in a reduced impact on terrestrial ecosystems (Türtscher et al., 2017). Similar observations can be seen with man-made impervious surfaces such as concrete structures, pavements, and asphalt roads. Dry depositions of TMs enter the water cycle during rain events as rainfall and surface runoff wash out such depositions. In general, the efficiency of the wash-out processes depends on the surface roughness and the chemical properties such as pH of the rain and surface run-off volume (Murphy et al., 2015; Wicke et al, 2012;).

Ecological health risk assessment is an efficient tool that can be used to determine the risk accompanied by metal deposition through atmospheric deposition (Ma et al., 2016). In ecological

risk assessment, metals with a longer accumulation history tend to represent an extreme ecological risk (Mugoša et al., 2016). If TMs exhibits possibility of accumulation, ecological risks may increase. Furthermore, as many studies have indicated, industrial regions are rich in atmospheric pollution, compared to commercial and residential areas, resulting in higher ecological risks (Ma et al., 2016; Sun et al., 2010).

With different TMs exhibiting various toxicity levels at different concentrations, the risk assessment tools can be used efficiently to get a better idea about the impact and risk imposed by a particular TMs on ecosystem health rather than just looking at their measured concentrations. For example, TMs such as Cd, Cu, Pb, and Zn have been the focus of most studies as their impact on ecosystem health is very significant (Sun et al., 2010; Zhu et al., 2012). Among these metals, Cd can pose a severe health risk even at minor concentrations compared to other TMs, as Cd exhibits higher toxicity and percentages in the exchangeable and carbonate fractions. (Sun et al., 2010; Zhu et al., 2012). Therefore, even if Cd is present in low concentrations in atmospheric deposition, due to its high hazard index, it tends to pose a significant risk on ecosystem health (Ma et al., 2016; Ntakirutimana et al., 2013; Sun et al., 2010; Zhu et al., 2012).

4.2 Impacts of trace metals on human health

TMs can be severely detrimental to human health. Due to their toxicity, TMs can cause deleterious effects on the renal, neurological, gastrointestinal, reproductive, cardiovascular, and hematological systems (Nordberg et al., 2007). Regarding health risk by atmospheric metal deposition, three major pathways can affect human health: ingestion, inhalation, and dermal contact (Cai et al., 2019; Peng et al., 2017). Generally, strategies of evaluating the potential health effect of metals in atmospheric dust are based on either assessing metal toxicity by exposure routes or measuring the

levels of exposure to those metals for the potential receptors (Ma et al., 2016; Xu et al., 2015). Risk indices are defined in this prospect to evaluate the risk of TMs on human health at a certain study location. In terms of exposure routes, indices such as long-term daily exposure from hand-mouth intake, inhalation, and skin contact are some important indices that are defined and evaluated in experimental studies (Cai et al., 2019; Peng et al., 2017; Weerasundara *et al.*, 2018). Often, human health assessments miss the effect of atmospheric contaminants when compared to the well-documented focus on aqueous contaminants. However, Zhang et al. (2020) highlighted that if the TM concentration is high and the population densities are high in a certain area, it is important to understand the increased risk on human health. Their comments were mainly based on the higher risks associated with atmospheric As in populated regions such as China, India, and Bangladesh (Zhang et al., 2020).

The detailed symptoms, disorders, and disease conditions linked to metal bioaccumulation are categorized in Table S1, while a comparison of risk assessment indices from worldwide studies are summarized in Table S2. In summary, most research studies have shown that the ingestion pathway and ingestion are the major exposure routes that can affect human health due to the atmospheric dust (Cai et al., 2019; Lu et al., 2014; Xu et al., 2015).

Hazard quotient (HQ) and hazard index (HI) are used in assessing the risk due to each metal (Ma et al., 2016; Sun et al., 2014). Here, HQ is calculated by the ratio between the average daily dose (mg/kg/day) and a specific reference dose. Average daily doses can be calculated for different exposure pathways such as ingestion, inhalation, and dermal contact. Hence HQ can be calculated for each of these pathways. The specific reference dose is a comprehensive estimation of maximum allowable risk due to daily exposure. Therefore, if HQ>1, it indicates that the considered exposure pathway can cause problems to human health. On the other hand, HI is

defined for a single or many metals using the sum of relevant HQs calculated for various exposure pathways. Furthermore, values of HI>1 indicate that the considered element causes risk to human health while the probability increases with the value of HI (Du *et al.*, 2013; Ferreira-Baptista and De Miguel, 2005; Ma *et al.*, 2016).

In summary, the highest HQ due to Pb was observed in the adult exposure pathway (Figure S1a). Mn, Cd, Ni, and Cr were observed to be highly associated with the considerable hazard quotient due to the inhalation pathway in humans (Figure S1b). However dermal contact pathway has the most sensitivity for the most range of TMs in both children and adults (Figure S1c). The cities with mining activities and massive industrial activities are at higher risk due to atmospheric TM deposition. Although traffic activities have a leading role in the TM composition of atmospheric deposition, most of the time, the traffic activities themselves cannot create significant risk on human health (Ma et al., 2016; Sun et al., 2014).

Cai et al., 2019 reported that the ingestion of TMs in children is much more significant than that of adults. They observed Pb and Cr be main elements to be uptake via hand–mouth pathway; however, overall carcinogenic and noncarcinogenic risks were below the safe thresholds (Cai et al., 2019). Peng et al., 2016 also indicated that the ingestion and inhalation were the major pathways for TMs extracted from PM2.5. Pb was again the major contributor to the calculated risk even though overall risks were below the permissible limits (Peng et al., 2017). Similar results were found in a study conducted in Kandy where nine TMs were tested to pose a below-limit risk on its inhabitants. In this study, Fe, Cu, and Al were shown to have a higher risk of ingestion in children, indicating it could pose a significant threat to them in the future, if corrective measures are not taken to reduce these pollutants (Weerasundara et al., 2018).

Most importantly, though one metal has increased concentrations in metal loads, the risk may be lower than metal with a comparatively lower concentration. This is mainly due to the risk potential of the type of metal as well as its origin. For example, the metals having crustal origin will be deposited with higher concentration but their risk to human health is low, and anthropogenic metals create a comparatively higher health risk (Weerasundara et al., 2018). The reason is the differences in reference dosages (acceptable limits) between two particular metals. Generally, crustal metals have comparatively higher reference dosages and vice-versa.

Many studies agree that children have higher sensitivity to trace metals in deposition loads (Lu et al., 2014; Mugoša et al., 2016). They are exposed to more atmospheric dust as children have a higher body surface area to volume ratio than adults (Table S2). Literature corroborates with the fact that the importance of body surface area to volume in health risk assessments. This also implies that in a polluted environment, the younger population has a higher risk on their health. Aligning with that, many findings suggest that the children should adhere to hygienic practices, such as washing hands after outdoor activities as a precaution to reduce TM intake through digestion, especially if they live in urban areas (Cai et al., 2019).

5. Conclusion

It is well known that the atmosphere directly contacts with humans and ecosystems. Therefore, the changes in the atmospheric conditions can adversely impact health, environmental quality, and water quality. In summary, we have reviewed the characteristics of TMs bound to PM in dry and wet atmospheric deposition. These TMs are originated from both natural and anthropogenic sources, and concentrations of TMs changes depends on the nature of the source, weather patterns and many other parameters fueling a complex situation to deal with. These contaminants can also

travel long distances due to the wind patterns, making sensitive ecosystems vulnerable. In another branch of studies, potential human and ecosystem health risks of atmospheric TMs are being identified which is appreciative in reestablishing the quality of air in regions with significant anthropogenic activities.

Through this review, we understand that TMs emitted into the atmosphere do not deposit on surfaces right after the emissions. Thus, continuous monitoring of trace elements is essential to determine the chemical budget of atmospheric TMs and their biological effects on the environment and human lives.

The involvement of particulate matter has been shown to cause long-range transportation of TMs. However, its effect on indigenous flora and fauna found in remote areas are understudied now. Furthermore, assessment of the status of heavy metal contamination of the atmosphere and the urban water environments has an important role when considering the implementation of management strategies for atmospheric emission and deposition of TMs. Regional risk assessments of TMs in atmospheric deposition needs to be encouraged and improved since the metal concentrations in the environment may not give a definite direction nor guideline to plan the pollution control strategies. Therefore, strong attention should be paid to the assessment of human and ecosystem health risk assessment as well. Therefore many research studies needs to be conducted in these areas immediately.

Although many developed countries have daily monitoring stations to assess the heavy metal concentrations in the atmosphere, most developing countries do not have such infrastructure nor capability due to the high cost of test equipment and monitoring station management. Furthermore, most countries target to investigate air pollution but not the atmospheric deposition and water pollution through atmospheric deposition of TMs. Therefore, the passive monitoring of

atmospheric deposition is required to understand the quality of the atmosphere and atmospheric deposition. The cost of sensors and test procedures seems to be expensive and unavailable in some regions in the world, as we see most of the studies are concentrated on a limited number of areas. This pattern can be seen around the world. Although it is complicated to determine the most accurate method for digestion and extraction of TMs in atmospheric deposition in the laboratories, it would be good to develop low-cost and reliable sensor technologies to track the air quality and TM depositions in real-time. This will allow authorities to act promptly in densely polluted areas that require immediate attention.

Novel computer tools can be used to conduct more efficient and important research on air quality and TM depositions. For example, modeling studies on the transport pathways, the influence of deposition on stormwater pollution, and relationship to health will also be interesting pathways for future research approaches. Cloud computing applications can be useful to share knowledge with researchers in real-time as well as to conduct studies in remote areas without lower human involvement. Multidisciplinary research involving environmental science and engineering disciplines, computer science and information technology disciplines would be able to achieve many leads in these attempts.

On the other hand, governments bares the reasonability of imposing and updating the rules and regulations against pollution conditions which are essential to control the emission and deposition of TMs. Fruitful findings in the areas mentioned above will allow enforcing stricter, more precise, more scientific, and rational laws and regulations for pollution and emission control, especially in urbanized, densely populated areas.

Acknowledgments The financial support from National Science Foundation of Sri Lanka through a research grant (No. NSF/2014/EB/03) is greatly acknowledged.

Declarations

Funding National Science Foundation of Sri Lanka research grant (No. NSF/2014/EB/03)

Conflicts of interest/Competing interests The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this review paper.

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