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Impacts of atmospheric particulate matter pollution on environmental biogeochemistry of trace metals in soil-plant system: a review

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ABSTRACT

Atmospheric particulate matter (PM) pollution and soil trace metal (TM) contamination are binary environmental issues harming ecosystems and human health, especially in the developing China with rapid urbanization and industrialization. Since PMs contain TMs, the air-soil nexus should be investigated synthetically. Although the PMs and airborne TMs are mainly emitted from urban or industrial areas, they can reach the rural and remote mountain areas owing to the ability of long-range transport. After dry or wet deposition, they will participate in the terrestrial biogeochemical cycles of TMs in various soil-plant systems, including urban soil-greening trees, agricultural soil-food crops, and mountain soil-natural forest systems. Besides the well-known root uptake, the pathway of leaf deposition and foliar absorption contribute significantly to the plant TM accumulation. Moreover, the aerosols can also exert climatic effects by absorption and scattering of solar radiation and by the cloud condensation nuclei activity, thereby indirectly impact plant growth and probably crop TM accumulation through photosynthesis, and then threat health. In particular, this systematic review summarizes the interactions of PMs-TMs in soil-plant systems including the deposition, transfer, accumulation, toxicity, and mechanisms among them. Finally, current knowledge gaps and prospective are proposed for future research agendas. These analyses would be conducive to improving urban air quality and managing the agricultural and ecological risks of airborne metals.

Keywords: Aerosol pollution; Trace metals; Terrestrial biogeochemical cycles; Atmospheric dry and wet deposition; Foliar uptake; Crop food safety

Contents

1. Introduction

Atmospheric particulate matters (PMs) or aerosol particles is a central component of the atmospheric chemical and climate system (McNeill, 2017; IPCC, 2014), a component of global biogeochemical cycles with long-range transport (Mahowald et al., 2018; Boyd and Ellwood, 2010), and also a typical environmental pollutant (Fuzzi et al., 2015; Jin et al., 2017; Shiraiwa et al., 2017). Especially in developing China of Asia, the rapid industrialization, urbanization, and associated increases in energy consumption during the last three decades have led to elevated levels of PMs in many regions, and resulted in profound deterioration of both local and regional air quality (Cheng et al., 2013; Luo et al., 2012). Generated from wide sources (Calvo et al., 2013) and composed of numerous hazardous components including toxic or cancerigenic trace metals (TMs) (Luo et al., 2019), they contribute substantially to urban air pollution (Filippelli et al., 2012), and have critical impacts on both ecosystems and human health influenced by PM sizes (Chen et al., 2018). Moreover, the aerosols can also exert climatic and hydrological effects through light absorption and scattering, hygroscopic growth, and cloud condensation nuclei activity (Banerjee et al., 2018). To terrestrial ecosystem, aerosol pollution is significantly altering radiative transfer processes and is thereby potentially affecting plant growth and crop production through photosynthesis. As mobile and suspended environmental media, atmospheric PMs with associated TMs could also be retained by plant leaves or enter soil environments by dry or wet deposition, participating in the TM biogeochemical cycles. Therefore, both the PMs and airborne TMs will influence the environmental behavior and effects of TMs in the soil-plant systems through these multiple ways.

Although some are required as micronutrients or essential elements for living organisms including plants, due to carcinogenic or toxic effects on biota at higher levels and occurrence in the environment (Luo and Wang, 2018), thirteen TMs were considered as priority pollutants by USEPA (2015), including antimony (Sb), arsenic (As), beryllium (Be), cadmium (Cd), chromium (Cr), copper (Cu), lead (Pb), mercury (Hg), nickel (Ni), selenium (Se), silver (Ag), thallium (Tl), and zinc (Zn). Anthropologic activities have changed global biogeochemical cycling of TMs by considerable quantity of diverse emissions into the atmosphere, especially in the urban environment, which will deposit soon or transport long-range to rural or remote area (Kirk and Gleason, 2015). Natural (*e.g.*, crustal minerals originating from wind-eroded bare soils or transported from arid areas by episodic dust storms), road traffic (*e.g.*, vehicle exhaust and re-suspension emissions), and industrial emissions (*e.g.*, fossil fuel combustion and metallurgical processes) are the typical sources of TM-bearing aerosols (Calvo et al., 2013; Pant and Harrison, 2013; Suvarapu and Baek, 2017). Under the global background of air PM pollution and soil heavy metal contamination, it is a significant visual angle to investigate the interactions between the environmental biogeochemistry of TMs in atmospheric and terrestrial systems.

In view of the binary issues of air and soil quality typically in China, the progresses in interactional topics of PMs-TMs for soil-tree in urban environments, soil-crop in agricultural lands, and long-range transport to mountain forest systems, were summarized respectively in this comprehensive review (Fig. 1). It aims to connect the biogeochemical cycles of TMs in various ecosystems by mobile atmospheric PMs. Focusing on sinks of particulate TMs in terrestrial environments, the deposition, foliar/root uptake, transfer, accumulation, toxicity, and mechanisms among them were discussed in detail. These analyses would be conducive to improving urban air quality and managing the agricultural and ecological risks of airborne metals.

2. Background of atmospheric PM pollution and soil TM contamination

2.1. Atmospheric PM pollution and associated TMs

Atmospheric PMs have diverse sizes, sources, chemical compositions, and corresponding effects (Fig. 2). As the main contributors to the urban ambient PM load, anthropogenic particles are generated through combustion of fossil fuels, industrial activities, abrasion, and the re-suspension of natural particles by traffic, construction, and surrounding agricultural activities (Calvo et al., 2013; Li et al., 2013). Although city is a major anthropogenic source to suburban, rural and remote areas, the urban situation may also be exacerbated by the long-range input of naturally occurring particles produced by desert dust, sea spray, volcanoes, grassland fires, and a variety of biological sources. They are usually divided into four size fractions according to the PM aerodynamic diameter (Dp), including total suspended particles (TSP; Dp<100 μm), coarse particulates ($PM_{10-2.5}$; 2.5<Dp<10 μm), fine particles ($PM_{2.5}$; Dp<2.5 μm) and ultrafine particles (UFPs, $PM_{0.1}$; Dp<0.1 μm). The chemical makeup of them can vary tremendously depending on location, meteorology, and source profile (Fuzzi et al., 2015; Jin et al., 2017; Shiraiwa et al., 2017). Typical PM compositions include natural crustal materials (carbonates, silicates), inorganic constituents such as sulfate, nitrate, sodium, potassium, chloride and ammonium, TMs, and organic components. The aerosol sources of metals are dominated by desert dust (mineral aerosols) for Al, Ti, Mn, and Fe, but combustion sources might also contribute to them and may be especially important for Cu, Zn, Pb, while Cd sources may be dominated by volcanoes (Mahowald et al., 2018; Schroeder et al., 1987). Different TMs showed varied enrichment factors in PMs, various size fractions of which have different potential transport ability (Luo et al., 2011; Schulz et al., 2012). Owing to the environmental risks, air quality guideline values for both bulk PMs and associated key harmful TMs were set up globally (Table 1). Specifically, the guidelines are at the range of 10 ~ 35 μg m^{-3} for annual $PM_{2.5}$ and 25 ~ 75 μg m^{-3} for 24h $PM_{2.5}$, 20 ~ 70 μg m^{-3} for annual PM_{10} and 50 ~ 180 μg m^{-3} for 24h PM_{10}, 80 ~ 200 μg m^{-3} for annual TSP and 120 ~ 300 μg m^{-3} for 24h TSP. Meanwhile, the associated TM limits are 6 ng m^{-3} for As, 5 ng m^{-3} for Cd, 0.025 ~ 0.25 ng m^{-3} for Cr(VI), 50 ~ 1000 ng m^{-3} for Hg, 50 ng m^{-3} for Mn, 20 ng m^{-3} for Ni, 1000 ng m^{-3} for 24h V, 500 ng m^{-3} for annual Pb and 150 ~ 1500 ng m^{-3} for seasonal Pb. Nevertheless, different locations are required to develop their individual guideline values since their PMs and associated TMs have different emission sources with different potential risks on human health (Li et al., 2019).

Concentrations of TMs in atmosphere vary greatly between urban and rural areas, as well as with distance from emission sources, such as metal smelters and coal-fired power plants, and TM levels surrounding industrial areas were higher compared with residential and/or commercial areas (Suvarapu and Baek, 2017). For the most concerned Pb, the largest mode of the size distribution of PM-bound Pb has shifted to larger sizes while airborne Pb concentrations have decreased in urban areas of USA and Europe (Cho et al., 2011), influenced by the phase out of tetraethyl Pb additives in gasoline but industrial emissions and re-suspended road dusts became more important sources of Pb. By statistic results of 44 cities in China (Duan and Tan, 2013), the coal burning, iron and steel industry and vehicle emission were important atmospheric TM sources, and the atmospheric TM concentrations were generally high in winter and low in summer, due to meteorological conditions, sources and transportation. The levels of atmospheric TMs in China were much higher than those of developed countries such as the USA and Europe but slightly lower than cities in India and Pakistan, and the pollution of Cr (85.7±111 ng m^{-3}), As (51.0±67.0 ng m^{-3}) and Cd (12.9±19.6 ng m^{-3}) were serious. Showed by the levels and sources of monitored PM_{10} bound TMs (Cd, Co, Ni, Pb, V, and Zn) in seven cities stretching across northern China from west inland to the east coast (Luo et al., 2014), both PM_{10} and the associated TM levels for urban and rural areas were comparable, implying that the current pattern of regional pollution in China differs from the usual decreasing urban-rural-background transect. Judging from weight contents (mg kg^{-1}), multivariate statistical analysis by principal component analysis (PCA), and absolute principal component scores-multiple linear regression analysis (APCS-MLR), the typical "urban metals" (Pb, Zn, and Cd) in northern China were mainly attributable to coal combustion and vehicle emissions with additional industrial sources, and showed higher anthropogenic contribution in eastern cities. However, the airborne Co was mostly of crustal origin, and the V and Ni were mainly from soil/dust in the western region and mostly from the petrochemical industry/oil combustion in the east. As indicated by the Pb isotopic compositions and backward air trajectories, the winter northwestern monsoons and westerly jet streams were the dominant forces in the long-range transport of airborne metals in northern China, with potentially global implications. Luo et al. (2016) also developed the statistics of TMs in 41 major cities and background sites of China over the past decade (Fig. 3), and the average concentrations were 285, 39.9, 75.4, 12.8, 83.4, and 622 ng m^{-3} for Pb, As, Cr, Cd, Cu, and Zn, respectively, higher than many developed countries. Although northwestern China has lower TM concentrations, other regions particularly the mid-eastern China showed higher levels due to the severity of anthropogenic pollution (Fig. 3). This trend is consistent with haze pollution (Zhang et al., 2012), which was almost exclusively concentrated within the regions of North China Plain, Yangtze River Delta (YRD), Pearl River Delta (PRD), and Sichuan Basin. Although China put active and stricter measures to reduce emission rapidly and improve air quality significantly (Luo et al., 2017), typically since the 2013 Action Plan for the Prevention and Control of Air Pollution, the PM levels are still high.

2.2. Soil contamination by TMs related to atmospheric deposition

As a geochemical medium, aerosols carry such TMs and deliver them to ecosystems via atmospheric deposition. Although human health effects are more related to the particles finer than PM_{10} through the inhalation pathway, they can also transport faraway, while the coarser particles will easily deposit into the earth surface system and cause ecological risks. As the major sink of atmospheric TMs, soil contaminated by TMs either in urban or rural areas are also threatening both ecological and human health by various exposure pathways, such as the ingestion of polluted crops/vegetables, and can partially contribute as a re-suspended source to atmospheric TMs simultaneously.

Soil heavy metal pollution has been another serious and widespread environmental issue in China, for which spatial features of pollution levels indicated by pollution index, geo-accumulation index and enrichment factor, and ecological and human health risks were investigated both in urban (Li et al., 2018a; Luo et al., 2012) and agricultural areas. On a national scale statistics employing TM data of the first national soil pollution survey (Chen et al., 2015), Cd, Hg, As, Pb, Cr and Ni were identified as the priority control metals due to their higher concentrations or public risks. Soil metal pollutions were relatively higher in southern provinces than others, that would be related to the higher geochemical background in southwest regions and the intensive human activities in southeast. Through a systematic soil geochemical survey of TMs (As, Cd, Cr, Cu. Hg, Ni, Pb, Sb, Se, Zn) in urban soils of 31 metropolises in China (Cheng et al., 2014), Hg, Cd and Se ranked higher in geo-accumulation likely due to the Hg and Se emissions from fossil fuels, while other reasons for high TM were the numerous hotspots contributed by metallurgical industry and smelt mining, and the naturally high geochemical soil background. Supported by the TM concentrations in soils of 402 industrial sites and 1041 agricultural sites in China (Yang et al., 2018), pollution and associated risks are severer in industrial than agricultural regions, southeast severer than northwest China, and Cd, Pb and As are more serious. In comparison, TM pollution levels in urban soils were medium to high in many European and North American cities, where Pb, Cd and Zn were also generally high in urban soils (Luo et al., 2012). The improvements of soil environment management and heavy metal pollution prevention and control are also underway in China currently.

3. Urban soil-tree system as a sink retaining atmospheric PMs and associated TMs

3.1. Atmospheric PMs captured by tree leaves

Since aerosol pollution are usually popular and harmful in the densely populated urban area, compared with their ecological risks to urban soil-plant system, plants are more often used as biological filters to clean air by accumulating atmospheric PMs on their foliage, although plant materials can also be used as bio-monitors and bio-indicators for air pollution or the atmospheric TM levels (Ram et al., 2015). For example, the quantity and size of PMs deposited on *Platanus acerifolia* leaves across 28 cities in Europe were mainly dependent on the regional background concentration of atmospheric PMs, while the percentage of Fe-based particles emerged as a clear marker of traffic-related pollution in most of the sites (Baldacchini et al., 2017). Facing the worsening air quality around the world, the PM deposition ability of urban greening plant canopies and differences among plant types have been investigated widely (Cai et al., 2017), supporting the plant screening and landscape planning for effective and eco-friendly way to alleviate PM pollution, especially for the re-suspended roadside dusts (Ram et al., 2015).

Attention has been paid to the variations in PM deposition influenced by vegetation factors, the characteristics of leaf deposited PMs, and the physiological impacts of PMs on plants. There are markedly spatio-temporal variations in urban PM leaf deposition, globally meta-analysis results of which suggested that the weekly average value was 1.71 ± 0.05 g m^{-2} wk^{-1}, and the influencing factors include (Cai et al., 2017): (1) vegetation (forest coverage and structure, plant species and types, canopy heights, tree structure and leaf characteristics) determining the PM deposition capacities of vegetation; (2) meteorology (precipitation, wind speed and direction, temperature and relative humidity) influencing the PM deposition processes on leaf surface (Popek et al., 2017); and (3) PM itself (sources, atmospheric concentrations, particle size and chemical components) directly affecting the plant ability of retaining PM (Przybysz et al., 2014; Tomasevic et al., 2005; Zhang et al., 2017a). For instance, fine PMs accounts for the minimum proportion of the total PM mass but its number ratio is maximum and contribute more than 90% of the total number of particles. There was an about 75-fold difference between juniper (*Juniperus rigida*) and Norway maple (*Acer platanoides*). Focused on the plant parameters, PM leaf depositions among various life forms ranked as shrub > tree > herb and liana. By leaf types, the coniferous species was significantly higher than broadleaved species, while by leaf habit, the evergreen species was significantly higher than deciduous species.

3.2. Foliar uptake of TMs in leaf deposition

Unlike root metal uptake that has been investigated comprehensively, TM uptake by plant leaves from the atmosphere was less known (Greger, 2004). Foliar surfaces might uptake deposited TMs through stomata, cuticular cracks, lenticels, aqueous pores, and mainly through ectodesmata which are non-plasmatic channels generally positioned between subsidiary cells and guard cells in the cuticular membrane or epidermal cell wall (Shahid et al., 2017). Furthermore, the cuticle present above the guard cell is more permeable than epidermal cells. Various environmental conditions during plant growth, such as shading, high temperature, humidity, and nutrient deficiency, affect the structure and anatomy of the leaf surface, thereby affecting the TM uptake by the leaves (Marschner, 2012). Besides the morphological characteristics of the plant leaves (Alahabadi et al., 2017; Popek et al., 2017), the chemical speciation of deposited TMs also influences the foliar TM absorption. Similar to root uptake, foliar uptake of TMs may also show a dose dependent pattern (Schreck et al., 2012a). Although this way of penetration might be a major contributor to the TMs in plants, the foliar transfer of TMs and their fate in plant leaves remain unclear (Marschner, 2012).

The foliar accumulation of TMs also varied with both TM and plant species. Results of TMs (Cu, Zn, Pb, Cd) in leaves of 12 plant species from multifarious areas in Shanghai, China showed that (Liang et al., 2017), the background botanical garden site had lower TM concentrations than other sites, and the plants with higher TMs were possibly owing to leaves with higher densities of stomata implied by scanning electron microscopy (SEM). Moreover, these TMs in plant needles potentially originate from soil, while in the leaves of broad-leaved plants might be from bulk atmospheric deposition, that is supported by the relationships between the TM concentrations in soils and the washed new and old leaves. Similarly in Yan′an city of the Loess Plateau, China, tree species with the highest bioaccumulation of a single metal did not have the highest total metal accumulation capacity, and the metal accumulation index (MAI) should be an important indicator for tree species selection in phytoextraction and urban greening (Hu et al., 2014). The high bioaccumulation capacity species can be used for phytoextraction of TMs pollution and green and buffer zone in city (Alahabadi et al., 2017; Hu et al., 2014).

3.3. Deposition of airborne TMs into urban soils and root uptake

The spatial variation in urban TM distribution relates to different functional zones in city (Luo et al., 2012). Roadside green belts, parks and gardens as mainly preserved urban soils are major targets, and atmospheric deposition of TMs from traffic and industry is the key anthropogenic source (Luo et al., 2015). Regarding to traffic, TM levels in urban road dusts were usually higher than the corresponding soils, especially the traffic emissions for Pb, Zn, and Cu (Luo et al., 2012). Generally, the influence of traffic on soil contamination decreased with increasing soil depth and distance to the road (Werkenthin et al., 2014).

The amount of dust deposition on the urban soil surface is huge annually, and is also a soil-forming material (Prokof'eva et al., 2017), which imports organic carbon, salts such as carbonates, pollutants such as oil hydrocarbons and TMs into the soil. The airborne deposits also influence soil physical properties by enriching the soil with clay and coarse silt fractions. Moreover, besides the wet deposition of atmospheric PMs, the temporarily retained PM leaf deposition will also be re-suspended by wind or washed by rain into soils. It was reported that precipitation removed a considerable proportion of particles accumulated on foliage of evergreen vegetation species, and most of the removed PMs were large size fraction, but fine PMs adhere more strongly to foliage (Przybysz et al., 2014). Urban plant roots can then uptake TMs from soils and accumulate them in tissues (Günthardt-Goerg et al., 2019). In the urban environment of Guangzhou, China, the concentrations of TMs were generally in the distribution order of road dust > soil dust ≥ surface soils ≈ top soils > grasses ≥ tree leaves (Bi et al., 2013). Significant correlations between tree leaves and the smallest (<50 μm) fraction of road dust, and between soil dust (50-100 μm) and surface soils, suggested that TMs in them may influence each other.

4. Effects of atmospheric PM pollution on TMs in agricultural soil-crop system

Deferent to the ecological issues of TMs in biota of urban environment, the TMs in agricultural system impact food safety to human health by polluted farmland soil-crop food chain, which is also significantly influenced by the input of TM-rich atmospheric PMs transported from nearby urban and industrial areas, especially for the peri-urban agriculture (Luo et al., 2012). Atmospheric PMs not only increase the TM contents in soils by deposition, but also induce some direct or indirect impacts on crop growth and TM accumulation by foliar uptake or climatic effects.

4.1. Atmospheric dry and wet deposition of TMs into agricultural soils

Atmospheric dry and wet deposition of TMs has been monitored long-term in China (Pan and Wang, 2015; Liu et al., 2019b). Literature statistics of atmospheric TM dry and wet deposition showed that, Cu, Zn, Pb, Cr, Cd, As, Ni and Hg concentrations in atmospheric dust over the last twenty years were higher than the Chinese soil environmental quality standards with multiple of 3.0, 7.4, 7.9, 1.1, 16.5, 1.5, 1.2 and 2.3, respectively; and Pb and Hg concentrations in rainfalls exceeded the surface water standards; thus Cd, Pb and Hg have high priority in preventing atmospheric TMs into soil (Wang et al., 2017). Since the recent decade, the various TM concentrations in atmospheric dusts has decreased by 32~50% than the last decade, and the annual atmospheric dry and wet deposition fluxes of As, Ni and Hg were reduced too; however, the fluxes of Cu, Zn, Pb, Cr and Mn were increased (Wang et al., 2017). Spatially, the Cu, Zn, Pb, Cr, Cd, Ni and Hg concentrations in atmospheric dust of south China were higher than north, but As and Mn concentrations were higher in north China. The annual deposition fluxes of Cu, Zn, Cr, As, Mn and Ni in north were higher than south, but Pb and Cd fluxes were higher in south (Wang et al., 2017). Further supported by the evidences of rainfall TM deposition in Chinese natural terrestrial ecosystems from national-scale network monitoring (Zhu et al., 2016), the atmospheric deposition of soluble Pb, Cd, and Cr was higher in the southwest, central, south, and north China than in the northwest and northeast China, Inner Mongolia, and Qinghai-Tibet. These soluble TM deposition fluxes were significantly correlated with the number of vehicles, and Pb and Cr was positively correlated with oil and coal consumption, while Pb and Cd were positively correlated with their soil contents.

Since soil is the primary terrestrial repository of contaminants, soil compartments have typically been used to determine the deposition of such atmospheric pollutants. Luo et al. (2016) extracted TM datasets from China's Soil Scientific Database (http://www.soil.csdb.cn/) to demonstrate to what extent airborne TMs depositing into agricultural soils, and found that TM patterns in remote farmland topsoil and haze pollution were spatially similar, implying that airborne TMs have caused remote farmland contamination in the mid-eastern region of China as a result of long-range transport and deposition (Xing et al., 2004). Concerning the inventory of TM input into agricultural soils, atmospheric deposition in China was the main pollution source and responsible for 50-93% of the total As, Cd, Cr, Hg, Ni, and Pb inputs in the past decade, that was the most important contributor in north China with its highly developed heavy industry and more coal combustion than south (Peng et al., 2019). Such percentages are higher than the range 25-85% of total inventory inputs in England and Wales (Nicholson et al., 2003).

4.2. Crop accumulation of atmospheric PM-bound TMs through foliar and root uptake

Due to the consumption risks of TMs polluted crops (Schreck et al., 2012a; Wang et al., 2017b), the soil-root and dust-leaf interactions of TMs in plants attract great attention (Harrison and Chirgawi, 1989; Bi et al., 2009; Uzu et al., 2010), both which are possible pathways for atmospheric PM-bound TMs, and these TMs might also be toxic to crop growth and induce physiological and biochemical responses. For instance, exposed to fine process particles enriched with TMs in an industrial area, both foliar and root pathways of TM mixture impact plant leaf fatty acid composition and do not interact (Schreck et al., 2013). Indicated by smelter particles via atmospheric or soil application to various vegetable species, exhibiting different morphologies, use (food or fodder) and life-cycle (lettuce, parsley, ryegrass), the Pb localization and speciation were strongly influenced by the exposure types (root or shoot pathway) and the plant species, and foliar exposure is the main uptake pathway involving the highest concentrations in plant tissues, while root exposure was identified as a minor pathway of Pb transfer (Schreck et al., 2012b). Supported by the soil reciprocal translocation experiments of atmospheric deposition near a Cu smelter in southern China on the soil-pakchoi system (Liu et al., 2019a), atmospheric depositions contributed to 20-85% of shoot Cu and Cd in high deposition site, for which both foliar uptake and atmosphere-soil-root transfer contribute, but 52-62% of shoot Pb from atmospheric depositions was mainly from foliar direct uptake. The newly deposited TMs (Cu, Cd) were preferential retention in topsoil (0-4 cm) and presented as higher bioavailable fractions compared to original soils. To crop growth, the increasing atmospheric TM depositions significantly decreased the photosynthetic parameters of pakchoi; while to human health, potential risks by pakchoi consumption were increased in high deposition site and the TM bioaccessibility were up to 56-81%. Similarly by comparing exposure experiments of two olive orchards in polluted factory area and unpolluted control site (Fourati et al., 2017), the leaves, roots and fruits of atmospheric TM (Cd, Cu, Fe, Mn, Ni, Pb) contaminated plants showed a depression of non-enzymatic and enzymatic antioxidant defences and a disruption of hormonal homeostasis. The anomalous physiological status was also demonstrated by the lower pigments in leaves and fruits, and the chemical and sensory quality of olive oil was also negatively affected by airborne TMs.

Concerning the mechanisms of TM foliar uptake by various plant species exposed to atmospheric PMs fallout, typically for Pb in vegetables, internalization through the cuticle or penetration through stomata openings might be two major mechanisms involved (Schreck et al., 2012b). For instance, the tentative pathways for PM-Pb uptake include: (1) PM deposition on the leaf surface; (2) Chemical transformation on the leaf surface leading to secondary Pb-containing phases and possibly solutes; (3) PM accumulation in stomata and possibly penetration of nanoparticles; Possible solute diffusion via aqueous pores present on cuticular ledges of stomata and anticlinal cell walls of cuticles; (4) Toxicity symptoms (such as necroses) induced by the contaminated PMs on the leaf (Uzu et al., 2010). Many processes can affect foliar transfer, including pollutant interception by plants, TM speciation and bioavailability (Liu et al., 2019a), fixation and penetration through the foliar cuticle, internalization by leaf cells and release into the phloem, leading to transportation and distribution within the plant. Depended on both the plant species and PM characteristics, high quantities of Cd, Sb, Zn and Pb were taken up by the vegetable leaves (cabbage, spinach) exposed to PMs enriched with these TMs frequently observed in the urban atmosphere (Xiong et al., 2014). Atmospheric dust-fall in Nanjing, China significantly inhibited the photosynthetic rates of Chinese cabbage and rice, reduced the leaf chlorophyll contents, and promoted the respiration rates, resulting in decreases of biomass, and significantly increased some TM contents (Suo et al., 2019). Although a maximum of 2 % of the leaf surfaces were covered with PMs, they were enriched in stomatal openings with area occupied up to 12 %. Implied by the crop accumulations of atmospheric As, Cd and Pb deposition in polluted and reference areas (De Temmerman et al., 2015), the atmospheric TM deposition was significantly related to their concentrations in bush bean leaves, stems and pods at green harvest and even As and Pb in seeds at dry harvest though covered by husks. While for the root crops (carrot, celeriac), similar effect was observed in the leaves and storage organs, but the transfer of airborne Pb in the food chain through edible roots might be negligible (De Temmerman et al., 2012).

Overall, plants can accumulate TMs both from soil solution by roots and from deposited PMs by leaves (Marschner, 2012). Nevertheless, for atmospheric TMs, those mechanisms of both foliar and root uptake need better understanding (Fig. 4). Importantly, since the UFPs in atmospheric PMs are nano-scale particles, the recent research progresses and methodology for uptake, translocation, transformation, accumulation, and toxicity of metal-based nanoparticles (NPs) in plants will be referential, such as the direct uptake of insoluble NPs (Lv et al., 2019; Wang et al., 2017a).

4.3. Indirect climate effects of aerosol pollution on plant photosynthesis and TM accumulation

Besides deposition directly contacting with plants, aerosols can also impact plant growth indirectly without contact, by affecting the climate (Myhre et al., 2013) and influencing solar radiance on the earth's surface which is the basis for photosynthesis (Bhagat et al., 2017). Atmospheric PMs can scatter (all aerosols) or absorb (few aerosol types such as black carbon-BC, mineral dust and some organic carbon-OC components) sunlight, reducing the total shortwave (SW= direct + diffuse) light reaching the surface, but light scattering also increases the diffuse fraction (DF= diffuse/SW) of this light (Li et al., 2017; Schiferl and Heald, 2018). Moreover, some aerosols as cloud condensation nuclei (CCN) and ice nuclei (IN) are vital for cloud formation, which cause the cloud albedo effect and the cloud lifetime effect, thereby change the earth's radiation balance and hydrological cycle (Myhre et al., 2013). Therefore, there are results reporting either positive or negative effect of aerosols on plant growth and production, and the overall impacts to crops from those competing effects (SW vs. DF) depend on local light conditions and plant types (Burney and Ramanathan, 2014; Tie et al., 2016). For instance, ozone (O_3) and haze pollution weakens net primary productivity in China (Yue et al., 2017). Local air pollution estimated by aerosol optical depth (AOD) have reduced wheat yields in India (Gupta et al., 2017), and regional haze in China depressed crop yields, and emission controls could enhance crop yields (Chameides et al., 1999). Rice yields in China were estimated to significantly increase by 0.8-2.6% with aerosol concentrations reductions from 20 to 100% (Zhang et al., 2017b). However, researchers could not detect a significantly negative effect of air pollution on grain yields of winter wheat in the North China Plain (Liu et al., 2016). Aerosols can directly impact protein expression in plants and photosynthesis efficiency, and are more advantageous for plant photosynthesis by facilitating diffuse solar radiations (Yan et al., 2014). Considering the beneficial effects of atmospheric PMs to crops given that enhanced light scattering leads to a more even and efficient distribution of photons that could outweigh total incoming radiation loss, air PM pollution may offset the O_3 damage to global crop production (Schiferl and Heald, 2018).

Theoretically, the metal transfer in soil-plant system will sure be influenced by such solar effects (Yruela, 2013) of aerosols, implied by the effects of ultraviolet-B radiation (Shweta and Agrawal, 2006). However, nearly no studies report results. Simulated by shading, reduction of solar radiation decreased the wheat yield but increased the concentrations of mineral metals in grains (Zhang et al., 2019). Therefore, both the impact of aerosols on plant growth and then on TM accumulation are much complicated, and further studies are needed to comprehensively understand the climate change effects and these various plant physiological and biochemical processes responding to aerosol pollution.

5. Long-range effects of atmospheric PMs on TMs in mountain ecosystem

Owing to long-range atmospheric transport, anthropogenic TMs can contaminate natural surface soils (Steinnes and Friedland, 2006), typically enter the remote mountain ecosystems (Achotegui-Castells et al., 2013; Bing et al., 2019). For example, atmospheric Hg deposition significantly contributed to Hg enrichment in remote montane soils (Zhang et al., 2013). The TMs associated with fine PMs suspend for days or weeks, and can travel hundreds to thousands of kilometers. When reaching a rough surface landscape such as mountains with various plants and sharp gap of altitude, the PMs will be removed from the atmosphere. As a result, mountain regions tend to accumulate atmospheric TMs (Bing et al., 2018; Gandois et al., 2010). Meanwhile, remote mountainous areas feature relatively pristine ecosystem with high levels of biodiversity, rare species, and few population centers or tourists. Remote mountains, especially high mountains, are sensitive to global changes and can trap airborne TMs owing to the effects of cold condensation (Bing et al., 2018). Fine PMs in remote mountains are mainly from long-range atmospheric transport, thus their bound TMs can reflect the atmospheric pollution information at a large scale.

Compared with the urban and agricultural ecosystems, the research on atmospheric TMs in the remote mountain ecosystem is relatively less concerned. Such original observation in mountain ecosystem is mainly aimed to obtain a background site where is suitable for continental atmospheric chemical measurements (Adams et al., 1977), and these kinds of sites are still used to compare atmospheric and anthropogenic contribution to TM pollution in industrial or urban areas (Lahd Geagea et al., 2008; Zhang et al., 2013). However, early in the 1970s and 1980s, many reports have showed that some airborne TMs from remote mountains or highlands were subjected to anthropogenic pollution through long-range atmospheric transport (Dams and De Jonge, 1976; Davidson et al., 1981). Since then, an increasing observations and studies have been conducted to focus on the effects of atmospheric PMs on the accumulation of TMs in the remote mountain ranges. The monitoring methods include direct collection of atmospheric PMs with deposition samplers or filters, and through separating the PMs from an archive such as clouds, ice core, and snow/firn (e.g., Bacardit and Camarero, 2010; Carling et al., 2012). According to these studies, human activities have undoubtedly altered global cycles of some TMs by increasing their atmospheric emissions. Thus, the atmospheric input of anthropogenic sourced TMs should be concerned in mountain ecosystem due to their negative effects.

Due to the hard accessibility, the harsh environmental conditions as well as the limitation of measurement techniques, the direct monitoring of airborne TMs in remote mountains is still confronted with a big challenge, especially at a large temporal and spatial scale. In last few decades, many alternative archives have been successfully used to reveal the atmospheric contribution, including bioindicators (e.g., moss, lichen, plant tissues; Bing et al., 2019, 2016c; Chropeňová et al., 2016), peat (Martínez Cortizas et al., 2012), lake sediments (Bacardit et al., 2012; Bing et al., 2016a), soils (Bing et al., 2016b; Wu et al., 2011), etc. The anthropogenic fluxes of TMs in atmosphere have changed over time at a global scale. According to the properties of the environmental archives, the deposition or accumulation of TMs in the mountain ecosystem can reveal various chronological sequences of the metals at different temporal and spatial scales. For example, the bioindicators such as moss and lichen record the metal deposition in recent years due to their short-life duration in the environment, while the peat, sediments and soils can reflect hundreds to thousands of years' deposition of atmospheric metals, which is based on the research resolution. As a result, the deposition history of TMs in these archives may be different due to the effects of regional human activities and air dust transport. Many studies found the earlier Pb pollution in mountain areas than other metals (e.g., Cd, Cu, Zn), but lower Pb deposition at present (Bacardit and Camarero, 2009; Bing et al., 2016a).

The factors influencing the long-range atmospheric TM transport are complex, which depends on the emission sources, particle sizes, meteorological parameters, and mountain surface conditions. As discussed above, the emission sources and distances from them are the main factors determining the input amount of TMs to remote mountains. Before entering mountain ecosystem, the meteorological conditions (e.g., precipitation and wind) regulate the TM deposition (Bacardit and Camarero, 2009), which commonly feature seasonal variation. At a large scale, the monsoon types affect the TM transport (Bing et al., 2019). Many studies in southwestern China have observed that the southwestern and eastern monsoons drive the transport of some TMs (e.g., Cd, Pb) from southern Asia and southwestern China into the high mountains in eastern Tibetan Plateau (Bing et al., 2018; Li et al., 2018b). At a local scale, various deposition patterns of airborne TMs in mountain archives (e.g., soils, forest floor, mosses, and lichens) have been observed due to specific driving factors (Fig. 5). The terrainmodulated precipitation and temperature are important factors causing the TM deposition. With increasing altitude in a mountain, the climate commonly features increasing precipitation and decreasing temperature (Fig. 5a). This is apt to increase the deposition of TMs through mountain condensation effect (Bing et al., 2016b; Xiang et al., 2017). Meanwhile, the different vegetation zones are developed along the altitudes of high mountains, which induce complex mountain surface characteristics. The forest filtering effects through affecting canopy interception of dust and precipitation, altering throughfall and stemfall can markedly regulate the TM deposition in forest floor (Bing et al., 2016b; Gandois et al., 2010). The plant effects also include the direct uptake of trace metals from atmosphere by leaf, which may alter the metal deposition in forest floor (Fig. 5b and c). In addition, the interaction of climate, terrain and vegetation regulated soil properties can shape complex distribution patterns of airborne TMs in mountain floor (Fig. 5d). For instance, the soil acidification to some extent accelerated the runoff and/or leaching of airborne Cd and Pb from the surface soils at the timberline of the eastern slope of Gongga Mountain, which resulted in the marked decrease of metal accumulation (Bing et al., 2018).

To date, the effects of atmospheric sourced TMs on mountain ecosystem such as vegetation succession, animal behavior, and even local human health have few reports. Because the PM surface containing TMs is highly soluble, particularly under the context of global acid deposition, TMs deposited by airborne dust tend to accumulate in biota and threat ecosystem health. Moreover, cloud process can also induce aerosol metal dissolution by enhancing sulfate concentration. Although few studies have concerned the speciation and behavior of TMs (e.g., Cd, Pb, Zn) which were mainly from long-range atmospheric sources in non-contaminated mountain soils (Bing et al., 2016d), there is deficiency concerning the biogeochemical cycle of TMs from atmospheric deposition in soil-forest systems, and the migration of them from land to aquatic system.

6. Conclusions and perspectives

Both aerosol pollution and soil heavy metal pollution are momentous environmental issues in current world, especially in the developing broad China with huge population and rapid urbanization and industrialization. Since TMs in the environment are significant to ecosystems and human health, the terrestrial biogeochemical cycle of TMs has been an important topic for long decades, either the global scale, region scale, local scale, or interface scale. Because of the dry/wet deposition characteristics of atmospheric PMs and associated TMs, they impact soil environments significantly. Although the PMs and airborne TMs are mainly emitted from urban or industrial areas, while owing to the ability of long-range transport, they can also reach the peri-urban, sub-urban, rural, and remote mountain areas, and participate in the TM biogeochemistry of various soil-plant systems, including urban soil-greening tree, agricultural soil-food crops, and mountain soil-natural forest systems. Besides traditional root uptake, the pathway of leaf deposition and foliar absorption also contribute significantly to plant TM accumulation. All these processes result in ecological or health risks. Moreover, indirectly, the aerosol also change solar radiation or climate with cloud, thereby impact plant growth and crop TM accumulation through photosynthesis, and then threat health across food chain.

However, based on the findings summarized in current overview, there are still many research gaps deserved further investigations either for basic knowledge or geochemical/physiological/biochemical/toxicological mechanisms of TMs in urban, agricultural, and remote mountain soil-plant systems:

For data results in various geographical areas of regional or national scale, key perspectives include that, source identification and apportionment of environmental TMs; the detailed and precise emissions and dry/wet depositions of atmospheric PMs and associated TMs at different temporal and spatial scales; inventories and fluxes of airborne TM inputs to various soil environments; the quantitative impacts of aerosols and airborne TMs on crop yields and food safety; ideal landscape and greening vegetation planning for improving urban air quality, etc.

For the biogeochemistry mechanisms in terrestrial processes of different PMs, TMs, and plants, main gaps need to be explored include that, the leaf deposition and foliar uptake of various atmospheric PMs and TMs by plant species and stress response to/accumulation in different organs; the speciation and bioavailability of airborne TMs to plant species; the contribution percentage of foliar TM adsorption compared with root uptake; methods such as stable isotopes in discriminating sources and transportation of TMs in the atmosphere-soil-plant-atmosphere system; plant photosynthesis and related TM transfer responding to aerosol pollution and cloud-climate interactions.

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