

Review Article

Atmospheric Deposition: Sampling Procedures, Analytical Methods, and Main Recent Findings from the Scientific Literature

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The atmosphere is a carrier on which some natural and anthropogenic organic and inorganic chemicals are transported, and the wet and dry deposition events are the most important processes that remove those chemicals, depositing it on soil and water. A wide variety of different collectors were tested to evaluate site-specificity, seasonality and daily variability of settleable particle concentrations. Deposition fluxes of POPs showed spatial and seasonal variations, diagnostic ratios of PAHs on deposited particles, allowed the discrimination between pyrolytic or petrogenic sources. Congener pattern analysis and bulk deposition fluxes in rural sites confirmed long-range atmospheric transport of PCDDs/Fs. More and more sophisticated and newly designed deposition samplers have been used for characterization of deposited mercury, demonstrating the importance of rain scavenging and the relatively higher magnitude of Hg deposition from Chinese anthropogenic sources. Recently biological monitors demonstrated that PAH concentrations in lichens were comparable with concentrations measured in a conventional active sampler in an outdoor environment. In this review the authors explore the methodological approaches used for the assessment of atmospheric deposition, from the analysis of the sampling methods, the analytical procedures for chemical characterization of pollutants and the main results from the scientific literature.

1. Introduction

Atmospheric deposition is the transfer of atmospheric pollutants (dust, particulate matter containing heavy metals, polycyclic aromatic hydrocarbons, dioxins, furans, sulphates, nitrates, etc.) to terrestrial and aquatic ecosystems and nowadays is receiving more and more attention by the scientific community, becoming the subject of a specific research area in the environmental sciences. The research in atmospheric deposition has increased a great deal over the past years, because of its increasing significant contribution to the explanation of pollution phenomena in many environmental compartments along with the possibility to evaluate the impacts of pollution sources at long and short distance (as

in fugitive emissions) and the possibility to carry out long-term studies aimed at performing health impact assessment on exposed population. The atmosphere is the carrier on which some natural and anthropogenic organic and inorganic chemicals are transported, and deposition events are the most important processes that remove those chemicals, depositing it on soil and water surfaces.

The prominent source of aerosols in the atmosphere, at the global scale, is the dust injected from arid regions, followed by soil and marine erosion and the anthropogenic sources [1].

Aerosol deposition occurs through three mechanisms depicted in Table 1 [2, 3]. In-cloud scavenging removes more than 70% of aerosols in number and more than 99% in mass,

TABLE 1: Mechanisms of aerosol deposition [2–5].

Wet deposition	Rain washout	Within a cloud	In-cloud scavenging or rainout, or droplets nucleation around particles
		Below a cloud	Droplets-particle collision
Dry deposition	Deposition processes which are not influenced by precipitation		
Occult deposition	Water droplets deposited by the interception of fog, mist, or clouds that play a significant role only in the case of frequently cloud-covered zone, or by adjective fog, but are negligible in urban areas		

whereas weak precipitation, having less than 0.1 mm h^{-1} intensity, can remove 50–80% of the below-cloud aerosol, in both number and mass within 4 h [4, 5].

Dry deposition occurs with several mechanisms like turbulent diffusion, sedimentation, Brownian diffusion, interception, inertial forces, electrical migration, thermophoresis, and diffusio-phoresis [6].

Deposition rates are governed by meteorological factors (wind velocity, relative humidity), particle characteristics (size and shape), and surface characteristics (friction velocity, microscale roughness, and temperature) [7].

The samplers used to evaluate atmospheric deposition can be differentiated into various categories depending on which deposition is collected: dry (only dry deposition is collected, when there is no precipitation), wet (the sampler collects only during rain), and bulk (wet and dry deposition are collected together). Mosses and lichens are used as “biomonitors” for an indirect assay of atmospheric deposition.

Wet deposition flux is conventionally calculated using the concentration measured in precipitation samples and the amount of precipitation recorded in the analysis period.

Under the approximation that the concentrations of pollutants in precipitation (C) depend on the concentrations in the air (K) within which precipitation is formed, the scavenging ratio (W) is defined as

$$W = \frac{C}{K}. \quad (1)$$

When the amount of precipitation is expressed as P , the wet deposition flux (F_w) of the pollutant is related to K , W , and P by

$$F_w = WKP. \quad (2)$$

Differences in the wet deposition fluxes of the pollutant between two sites may be due to different atmospheric concentrations and scavenging ratios.

In rainy areas dry deposition can be neglected compared with wet deposition [8] or does not basically modify the chemical characteristics of the wet deposition but can be the dominant fraction in arid and semiarid regions where intense dust loadings take place [9–12] and it is necessary to separate wet and dry deposition.

Dry deposition dominates the atmospheric delivery of particulate matter, total phosphorous, Ca^{2+} , Mg^{2+} , and K^+ , whereas wet deposition dominates the atmospheric delivery

of Na^+ , total nitrogen, NO_3^- , and SO_4^{2-} [13]. Conversely in higher precipitation regime areas, wet deposition reflects long-range transport phenomena, while dry deposition is more linked to local pollution levels [14] and dominates deposition processes of micropollutants in the highly industrialized areas [15].

Dry deposition of organic micropollutants (polycyclic aromatic hydrocarbons (PAHs), polychlorobiphenyls (PCBs), polybromodiphenyl ethers (PBDEs), and dibenzofurans) dominates atmospheric deposition during no raining periods in some polluted areas [16, 17].

Presence of particulate SO_4^{2-} , NO_3^- , and NH_4^+ , in the so called “secondary aerosol,” is the result of the reaction between gaseous precursors SO_2 and NO_x of anthropogenic origin, with oxidants such as O_3 and OH radicals, toward the formation of H_2SO_4 -containing aerosol and gaseous HNO_3 , and then to the reaction with NH_3 precursor. This “gas-to-particle” reaction is accompanied by a droplet-to-particle conversion, in which SO_2 and NH_3 produce secondary stable $(\text{NH}_4)_2\text{SO}_4$ aerosol, whereas NH_4NO_3 aerosols tend to dissociate under low NH_3 concentrations. These aerosols remain in the atmosphere until removed by wet or dry deposition and due to the residence time (about a week) are responsible for long-range transport of sulphur and nitrogen [18].

Speciation of deposited particle gives the opportunity to study mass balance of some metals as in the case of mercury in Lake Michigan and Lake Superior, which showed that atmospheric deposition contributes largely to the total annual input of mercury [19, 20].

Study of atmospheric bulk deposition of PAHs in an urban area reveals the presence of a plume of highest concentrations in zone with heavy vehicular traffic and favourable topography for the concentration of emitted pollutants, like in the active sampling in air, and also the diagnostic ratio analyses apportioned the major source of emissions [21].

Due to the extreme versatility of the analytical tools recently developed in the study of the atmospheric deposition, the existence of numerous methodological approaches, there is the necessity in the studying of the state of the art of this new environmental subject.

In this review, the authors explore the methodological approaches used for the assessment of atmospheric deposition starting from the analysis of the sampling methods, the analytical procedures for chemical characterization of pollutants, and the main results from the scientific literature, dividing pollutants into four major classes, starting from

TABLE 2: List of different collectors used in sampling atmospheric deposition of organic pollutants (PAHs, PCBs, PCDDs/Fs).

Collectors	PAHs	PCBs	PCDDs/Fs	References
Glass funnel-bottle bulk collector	X		X	[21, 23–25]
Stainless steel bucket	X	X	X	[23, 32]
Stainless steel platter	X			[22, 26]
Stainless steel funnel attached to a glass filter setup	X			[27]
Funnel connected to absorber cartridge (Amberlites IRA-743)	X	X	X	[23, 28, 29]
Funnel connected to absorber cartridge (XAD-2)		X	X	[30]
Automatic wet-only collectors	X			[31]
Two vessels equipped with a rain sensor	X			[15]



FIGURE 1: Bulk deposition sampler positioned round the borders of an industrial site in Taranto (Italy).

organics, followed by inorganics (metals other than mercury, ions), mercury (on which particular focus has been dedicated by researches due to its peculiarity), and biomonitors.

2. Sampling and Analytical Techniques

2.1. Organics. The atmospheric deposition of organics pollutants, such as polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), polybrominated diphenyl ethers (PBDEs), and polychlorinated dibenzo-p-dioxins and furans (PCDDs/Fs), can be estimated using different suitable collectors, as listed in Table 2 [21–25]. You can use wet-only collectors that are designed to collect only sedimenting wet particles or bulk ones to collect all sedimenting wet and dry particles, depending both on the aim of the study and on the sampling site (rural, industrial, or urban areas). Glass funnel-bottle bulk collectors, consisting of a cylindrical funnel and a sample collection vessel, were widely used for bulk determinations of PAHs and PCDDs/Fs [21, 23–25]. The cylindrical vertical section should be of sufficient height to avoid sampling losses resulting from splashing and the diameter for the opening area and the volume of the collector should be selected, in order to collect all the precipitation for the required sampling duration. Sampling period ranges from one week to one month. The height of the opening area of the collector shall be at least 1.5 m above ground, in order to avoid

sample contamination due to ground during heavy rains (Figure 1). The sampled rainwater was stored in a refrigerator at 2°C until analysis. When the volume reached 2 L, the bottles were immediately transported to the laboratory. After that, samples were filtered with precleaned (heated at 450°C for 24 h) Whatman GF/F filters (0.7 μm, 47 mm i.d.).

Stainless steel buckets were used to collect PAHs bulk deposition in remote, rural village, and urban areas [23]. Distilled water was added into the buckets before sampling, and the amount of distilled water was determined according to the evaporation and precipitation situation, generally 50 mL in summer and winter, and 100 mL in the other seasons. About 60 mL glycol was also added into each bucket to avoid freezing of water in winter and to reduce the effects of biodegradation.

Alternatively to previous collector, bulk deposition samples can be collected with a stainless steel platter whose diameter and depth were 60–76 cm and 19 cm, respectively [22, 26].

The collection of bulk deposition of PAHs was achieved by Li et al. [27] using a stainless steel funnel with an area of 0.049 m² attached to a glass filter setup. The funnel was placed horizontally, 1.2 m above the ground level. After about 30 days, the inner surfaces of the stainless steel funnels were wiped with precleaned cotton. The cotton and filter (Whatman, Grade GF/F, pore size 0.7 mm, diameter 90 mm,

and thickness 420 mm) were combined together as particle-bounded deposition fluxes of PAHs.

A passive sampling technique using a funnel-absorber-cartridge device was adopted and validated in the field by Gocht et al. to monitor the atmospheric deposition of PAHs in rural regions of Southern Germany [28]. The sampling system consists of a borosilicate glass funnel and a large adsorption cartridge packed with Amberlite IRA-743 (15 g of the absorber material which was fixed on top and at the bottom with glass wool plugs). While bulk deposition percolates through the funnel and cartridge, PAHs from both the water and particle phases are collected from the wet and dry deposition by adsorption and filtration, respectively. In the field, the sampling systems were housed in an aluminum box. After each sampling period, funnels were purged with 200 mL acetone in order to collect adsorbed and deposited PAHs from the glass surfaces. The cartridges were sequentially solvent extracted in four steps (50 mL for each) with the same acetone used before for the cleaning of the funnels (i.e., the purge solution). The IRA-743 resin bulk sampler was also used for the monitoring of long-term bulk deposition of PCBs, PBDEs, and PCDD/Fs [23, 29]. Alternatively to the previous collector, Hovmand et al. have used a tube filled with XAD-2 and connected to a borosilicate glass funnel for monitoring bulk deposition flux of PCDDs/Fs and PCBs [30].

Automatic wet-only collectors can be used to collect PAHs only during the precipitation events [31]. The sampler contains a humidity sensor which controls the lid of wet and dry collector compartments automatically. During the wet deposition events, the sensor moves the lid onto the dry collector and after the sensor surface becomes dry, the lid on the dry collector goes onto the wet collector. The aluminum cylindrical container installed into the dry collector compartment was filled with 3 L of distilled water to collect both particulate and gas-phase PAHs. In addition, it is possible to collect dry and wet depositions separately, using two sampling devices consisting of two vessels equipped with a rain sensor capable of triggering the cover, so as to protect the dry sample and collect the wet deposition in the other container [15].

Different extraction and analytical methods were used to detect the sixteen EPA priority PAHs (naphthalene (NAP), acenaphthene (ACE), acenaphthylene (ACY), fluorene (FLO), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLA), pyrene (PYR), benzo[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), dibenzo[a,h]anthracene (DahA), indeno[1,2,3-cd]pyrene (IcdP), and benzo[ghi]perylene (BghiP)): Soxhlet extraction clean-up using a silica gel column and GC-MS analysis [27, 32]; solid-phase extraction using speed C18 cartridges (acetone:tetrahydrofuran, 1:1 v/v) and analysis of the extracts by GC-FID; liquid-liquid extraction (MeOH) followed by GC-MS analysis [22] or UV/VIS HPLC analysis [30] or HPLC-FL analysis [33]; accelerated solvent extraction (ASE) followed by GC-MS analysis of the extract [27].

The sample extraction, purification, and analysis of PCDDs/Fs were performed following the EPA method 1613,

developed for isomer specific determination of the 2,3,7,8-substituted, dibenzo-p-dioxins, and dibenzofurans in aqueous, solid, and tissue matrices by isotope dilution, high resolution capillary column gas chromatography (HRGC), high resolution mass spectrometry (HRMS) [23, 28, 29, 34]. About PBDEs concentrations, the deposition samples were Soxhlet extracted and the extracts were cleaned up by sulfuric acid, multilayer silica gel, and gel permeation chromatography (GPC) columns and analyzed using a HRGC/HRMS [23]. Finally, the determination of deposition fluxes of PCBs was performed according to EPA method 1668B developed for chlorinated biphenyl congeners in water, soil, sediment, biosolids, and tissue by HRGC/HRMS [23–35]. Most of the literature data on atmospheric depositions of PCDDs/Fs and PCBs are given in International Toxic Equivalents (I-TEQ) scale.

2.2. Inorganics. The atmospheric deposition of inorganics pollutants, such as ions and metals, has to be estimated using suitable collectors, listed in Table 3. Three different types of collectors can be used: wet-only, bulk, and Bargerhoff. The wet-only collector is designed to collect only sedimenting wet particles, while the bulk and Bargerhoff ones [36] are designed to collect all sedimenting wet and dry particles. The wet-only and bulk collectors are bottle and funnel combinations [37, 38] while the Bargerhoff collector is an open bucket. Moreover, there are automatic wet-only collectors which allow collecting only during precipitation events. They consist of a lid that opens and closes over the sample container orifice, a precipitation sensor and a motorized drive mechanism with associated electronic controls. In addition, it is possible to collect dry and wet depositions separately using the samplers equipped with two polyethylene buckets and a lid controlled by a rain sensor, which moves depending on the beginning and the end of the rain event [39]. All collectors shall have a cylindrical vertical section of sufficient height to avoid sampling losses resulting from splashing and the diameter for the opening area and the volume of the collector need to be selected in order to collect all the precipitation for the required sampling duration. Typical sampling periods in fact vary from one week to one month, depending on meteorological condition. The height of the opening area of the collector shall be at least 1.5 m above ground in order to avoid the sample contamination due to ground during heavy rains. All parts of collectors shall be made in inert material such as HDPE, in order to avoid metals contaminations and the sample containers must be cleaned prior to sampling with distilled water and a 1% nitric acid solution to eliminate particles deposited or adsorbed onto container walls during prior collections [17, 38, 40]. After the deposition collecting, the sample is transferred to the laboratory in the sampling bottle (wet-only and bulk) or bucket (Bargerhoff), filtered and analyzed. Metals are digested by nitric acid at 200–250°C for 2 h in microwave system while ions are extracted by deionized water, in sonication system. The digested or extracted samples are finally analyzed by ICP-MS and GF-AAS for metals and by IC for ions quantification [41, 42]. The sulphate concentrations in atmospheric deposition could be also determined gravimetrically or by using barium chloride

TABLE 3: List of different collectors used in sampling atmospheric deposition of inorganic pollutants (metals and ions).

Collectors	Metals	Ions	References
HDPE funnel-bottle collector	X	X	[38, 42, 59]
HDPE bucket collector	X	X	[37]
HDPE automatic wet-only collectors	X		[17]
HDPE automatic wet-dry collectors		X	[39]
PVC dry deposition plate	X	X	[43–58, 223]
Water surface sampler (WSS)	X	X	[17, 59–62]
PE sheets and boxes	X		[36]

(BaCl₂). Collected particulate samples are coated with barium chloride (BaCl₂) in a vacuum evaporator, whose pressure is maintained below 106 mm Hg. The coated samples were then exposed in a desiccator cabinet to a relative humidity of 85% for 2 h to allow the sulfate present to react with BaCl₂ to form distinctive products (BaSO₄) identifiable by SEM [43–48]. In addition to bulk and wet-only depositions, several techniques of measuring dry deposited material have been reported in literature. These techniques employ a smooth surface plate with a sharp leading edge which is fitted to a galvanized iron stand. This plate made of polyvinyl chloride, and similar to those used in wind tunnel studies, is mounted on a wind vane and is 21.5 cm long, 8 cm wide, and 0.8 cm thick with a sharp leading edge (<10° angle) pointing toward the wind direction [17, 49–57]. Alternatively, a 33 cm diameter circular PVC plate has been used [53]. The sharp leading edge and smooth horizontal surface are supposed to cause the minimum airflow disruption and thus provide an estimation of the lower limit estimation of deposition [49, 58]. This technique is advantageous to control the sampling time, sample extraction, and parameters estimation. However, there are some limitations for the smooth surface plates. Particles with diameter less than 10 μm do not settle well under the influence of gravity, and bounce-off and resuspension might occur. In order to prevent the bouncing-off and resuspension of particles, the plates can be covered with two Mylar strips (7:6 × 5 cm²) coated by Apezion L grease (thickness B5 mm) or with projection films [57] coated with approximately 20 mg of silicon grease [52–56]. As an alternative, the plain surface can be covered using a double-sided adhesive tape affixed to the Mylar strip [43]. Anyway, Mylar strip or overhead projection films have to be cleaned by deionized water or by wiping with methanol-soaked wipes and then immersed in 10% nitric acid followed by methanol, five minutes each. Moreover, before and after exposure, they have to be maintained at a relative humidity of 50% at 25°C for more than 48 h and then weighed to a precision of 0.0001 g, in order to determine the total mass of collected particles. After the deposition collecting, and before the extraction and analysis of deposited pollutants of interest, the grease on the strip or films has to be eliminated. For this purpose, they are washed with distilled ultrapure hexane which is subsequently evaporated by directing a stream of ultrapure nitrogen on its surface [38, 53, 54]. Dry deposition samples were collected in the water surface sampler (WSS) [59], having a circular open surface area continuously refreshed with recirculated water that enters from the center, overflowed from the weirs at the

sides, and collected in the WSS holding tank, thanks to an adjustable liquid pump [60–62]. At the end of dry deposition sampling, all of the water was transferred into a clean plastic bottle with a plastic stopper, carried to the laboratory, and analyzed.

Collecting settled airborne particles using polyethylene sheets and boxes (1–1.5 m²) as samplers and a clean brush can be useful [36]. The sample bags were sealed in the field with plastic clips, double bagged in zip-sealed bags, and placed in nylon stuff sacks to limit abrasion during transport from the collection sites. All the samples were divided into two size fractions: >30 mm (coarse particle) and <30 mm (fine particle) using a copper sieve, pressed in order to obtain a tablet, and finally an elemental analysis was performed using wavelength dispersive X-ray fluorescence spectrometry.

2.3. Mercury. Mercury is released in to the atmosphere through both natural and anthropogenic activities [63–71] in three main forms: elemental vapour Hg(0), gaseous divalent Hg(II), and particulate phase mercury Hg(p). Temporal and spatial scales of mercury transport in the atmosphere and its transfer to aquatic and terrestrial receptors depend primarily on the chemical and physical interactions with atmospheric contaminants [72]. Atmospheric deposition is widely recognized as the primary mechanism by which Hg enters terrestrial and aquatic ecosystems, where it can be converted to the organic, bioaccumulative form: methylmercury (MeHg) which resides in the soils for long periods [19, 73–76]. It has been widely accepted that mercury is never removed from the environment; it just moves to other locations and eventually ends up in soils and sediments [77–79]. Adequate techniques for sampling and analysis of mercury in precipitation have grown considerably during the last decade. Sampling of atmospheric depositions of mercury is carried out with several collectors described in Table 4. The materials used (funnels and bottles of collection) according to European directive 2008/50/CE [80] should be made of borosilicate glass, PTFE, or PFA and are “wet-only” or “bulk” type samplers.

There are samplers that have two containers for collecting wet and dry deposition, respectively, with one barrelhead (dust preventing cover) that can switch between the two containers automatically depending on the weather conditions [81]. During a precipitation event (rain or snow), the barrelhead switches to the dry deposition container, thereby exposing the wet container to precipitation, and vice versa. In this case, wet precipitations are stored in a high density

TABLE 4: Collectors used in atmospheric depositions of mercury.

Sampler type	Main components	References
Bulk samplers		
IVL/Sweden	Borosilicate glass funnel with glass filter, capillary tube, and glass bottle	[80–82]
WDNR modified IVL/USA	Glass funnel, glass bottles, and heating system to prevent freezing in winter	
GKSS/Germany	Teflon funnel, brown-glass bottles	
Wet-only samplers		
NSA 181 KD/Germany	Quartz glass funnel, Teflon tube, Teflon bottles, funnel heating in winter, and thermostatic system for samples (maintained at 4°C)	[80–82]
ARS 721/Germany	Borosilicate glass funnel, PFA bottle (2 L) heating system	
MDN 1 sampler modified Aerochem Metric Sampler/USA-NADP	Double system for Hg—glass funnel, glass capillary, and bottle and for trace metals—PE or Teflon funnel and PE or Teflon bottle	

polyethylene (HDPE) plastic bag, whereas the snow samples during the cold seasons were melted at room temperature. Figure 2 shows a scheme of “bulk” sampler.

Several institutes belonging to monitoring networks throughout Europe and North America have provided sampling and analysis procedures [82]. The bulk samplers based on the IVL/Sweden design are composed of a borosilicate glass funnel (diameter 8.2 cm, area 52.8 cm²) with glass filter and capillary tube (diameter 4 mm, length 0.5 cm), while the WDNR sampler modified IVL/USA has a heating system to prevent freezing in winter, glass funnel (diameter 9.7 cm), and glass bottle (500 mL). Finally, GKSS/Germany model sampler has a Teflon funnel (diameter 35 cm) and brown-glass bottles. On the contrary, the wet-only samplers are made of quartz glass funnel (diameter 25 cm, area 490 cm²), Teflon tube (length 25 cm), and Teflon bottles (1L), and the NSA 181 KD/Germany sampler also has a thermostatic system for samples maintained at 4°C. Furthermore the ARS 721/Germany samplers have a borosilicate glass funnel (diameter 25.6 cm, area 500 cm²), PFA bottle (2 L), and heating system to prevent the formation of ice. The MDN sampler modified Aerochem Metric Sampler/USA-NADP is equipped with double system for Hg (glass funnel, glass capillary, and bottle) and trace metals (PE or Teflon funnel and PE or Teflon bottle). Furthermore, wet-only samplers have the advantage of avoiding particle dry deposition, although the contribution of gaseous or particulate mercury species to the wet deposition fluxes in nonindustrialized or nonurban areas is probably not large. Both samplers can be designed for sampling during all seasons and all climatic conditions. However, the sampling times range from 1 week to 1 month and the samples are monthly preserved with HCl (5 mL/L) prior to sampling. Relevant instruments for the analysis of mercury are the AFS and the AAS; the latter requires larger sample volumes due to the lower detection limit. All the material required for a sample must be handled carefully during transit and manipulation of the samples, to avoid contamination. Samples should be stabilized immediately after sampling and analysis must be carried out within 6

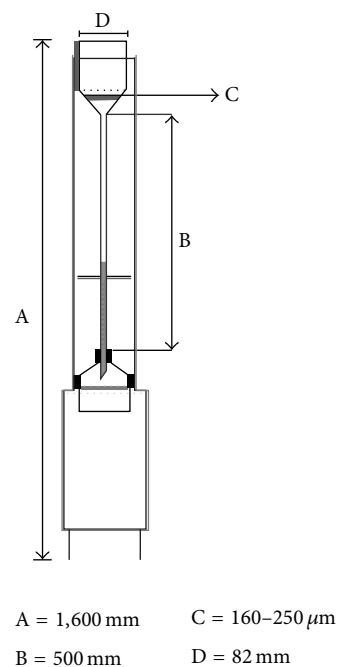


FIGURE 2: Scheme of a bulk type sampler for Hg [80].

months. Determination of total Hg involves the following steps (see Table 5): Hg, present in its various forms, is oxidized to Hg²⁺ by Bromine monochloride (BrCl); so all the Hg²⁺ is reduced to Hg(0) using tin chloride (SnCl₂). Hg vapors, using the purge and trap technique, are concentrated on a gold trap; by thermal desorption, Hg passes into a second trap from which, after a new thermal desorption, it is sent into the measuring cell, where it is detected by CVAFS [83].

In ambient air the particulate fraction of mercury is usually <5% [84], but this increases the risk of gas to particulate conversions during sampling. Therefore, CVAFS technique [85, 86] is used to analyze the Hg(p) which is collected on quartz fiber disc held in a miniaturized

TABLE 5: Analysis of total mercury and particulate phase mercury.

Total mercury, THg		References
Sample pretreatment	Digestion in HNO ₃ , BrCl oxidation, and NH ₂ OH·HCl prereduction	
Preconcentration	SnCl ₂ reduction, purging, and collection on gold traps	[83]
Detection	CVAFS	
Detection limit	3s of total blank at typical sample volumes	
Quality assurance/control	Determination of procedural blanks and analytical recovery	
Particulate phase mercury, Hg(p)		
Miniaturized device combines quartz fiber filter and the quartz wool plug technologies and serves as both particulate trap (during sampling) and pyrolyzer (during analysis) for airborne particulate-phase mercury species		[85–92]
Detection	CVAFS	
Graphite plate collection technique coupled with Zeeman electrothermal atomic absorption spectroscopy		
Design	Plate for atomization in PdCl ₂ -coated pyrolytic graphite tube with chemical modifier consisting of a 5% (NH ₄) ₂ S solution	[85–92]
Collection efficiency	99%	
Detection	ETAAS	
Quality assurance/control	A better time-resolved procedure with the higher accuracy (±8%)	

device [87]. This device combines the inherent advantages of the quartz fiber filter and the quartz wool plug technologies and serves as both particulate trap (during sampling) and pyrolyzer (during analysis) for airborne particulate-phase mercury species [88, 89]. A new analytical method was developed for direct analysis of Hg(p) by a graphite plate collection technique coupled with ETAAS [90] using the plate for atomization in PdCl₂-coated pyrolytic graphite tube with chemical modifier consisting of a 5% (NH₄)₂S solution, which increases the collection efficiency of particle-bound mercury to 99%. Compared to traditional techniques [91], the proposed method is a better time-resolved procedure with the higher accuracy (±8%) [92].

2.4. Biodeposition. Biomonitoring is regarded, in a general sense, as a means to assess trace element concentrations in aerosols and deposition. This implies that the monitor should concentrate the element of interest and quantitatively reflect its ambient conditions [93]. In addition, environmental impact on the biomonitor's behaviour is viewed as resulting in changes in the dose-response relationships [94].

With proper selection of organisms, the general advantage of the biomonitoring approach is related primarily to the permanent and common occurrence of the organism in the field, even in remote areas, the ease of sampling, and the absence of any necessary expensive technical equipment; moreover, it allows long-term monitoring with a large number of sampling sites and also the simultaneous determination of several pollutants within the same matrix [95].

Of all biological species used in biomonitoring, lichens and mosses are the most used in the literature as lack of any roots in comparison with higher plants allows mineral supplies from aerial sources and not from the substratum, so they are strongly dependent for their mineral nutrients on wet and dry deposition on the plant surface (of either

nutrients or pollutants) [96–98]. The morphology of lichens and mosses does not vary with seasons; thus, accumulation can occur throughout the year. Lichens and mosses usually have considerable longevity, which led to their use as long-term integrators of atmospheric deposition [99–101]. Natural variabilities in ambient macro- and microclimate conditions, such as acidity, temperature, humidity, light, altitude, or ambient elemental (nutritional) occurrences, may cause the biomonitor to exhibit variable behaviour. Part of this variance is shown as local variance, but this behaviour becomes a problem when it seriously affects the biomonitor in its accumulative responses [94].

Different strategies might be followed when lichens are used in biomonitoring studies: the use of in situ lichens (observation or chemical analysis of indigenous plants) and the use of lichen transplants (collection of biomonitors from a nonpolluted site and transplantation to sites to be monitored). Both can give information about spatial and temporal effects of pollutants deposition. Because lichen growth is very slow, the sampling of in situ lichens gives information about air pollution long-term effects. However, sometimes there is a need to study the patterns of the pollutants deposition throughout time, particularly with climate variables. In this case, lichen transplants should be used to provide information about air pollution short-term effects [102]. Table 6 shows the main features of the different strategies of lichens biomonitoring. In general, the passive biomonitoring method has one major disadvantage: all processes and all sources act at the same time and there is no possibility of separating them and looking for a particular one [103]. This method has been widely used to monitor metal pollution in terrestrial and aquatic environments [104]. The spatial impact integrated over time was evaluated from the concentrations of Ca, Fe, and Mg in in situ *Xanthoria parietina*. While, for temporal sampling, the fruticose lichen *Ramalina canariensis* Steiner

TABLE 6: Main features of the different strategies of lichens biomonitoring.

Strategies	In situ lichens	Transplants
Definition	Observation or chemical analysis of indigenous plants	Collection of biomonitors from a nonpolluted site and transplantation to sites to be monitored
Time effects	Air pollution long-term effects	Air pollution short-term effects
Major impact	Spatial impact	Temporal impact
Common species	<i>Xanthoria parietina</i>	<i>Ramalina canariensis</i> Steiner

was chosen because it is relatively easy to collect, clean, and transplant, its fruticose growth form facilitates particle interception and tolerates some sea influence [105]. After being cleaned from debris, each sample was dried for 24 h at 80°C and weighed. An acid digestion was then made with HNO₃ at 200°C. The total concentration of Ca, Fe, Mg, and K was determined by AAS using an air/acetylene flame, with CsCl and LaCl₃ added to samples and standards, as suppressants of ionization and refractory compound formation, respectively. For N and S concentrations, different thalli from each sampling point were dried for 24 h at 30°C and then homogenized in a mill; replicates were separated and the total concentrations determined by elemental analysis (CHNS/O Analyser). More recently, lichens have been used to monitor PCDDs/Fs and PAHs [106–109]. The lichen species selected for biomonitoring of PCDDs/Fs atmospheric deposition was *Xanthoria parietina* growing on house roof tiles [110].

Lichen species *R. canariensis* Steiner (epiphytic, fruticose, and bushy-like structured lichen) were collected mainly from *Pinus pinea* Aiton, on a minimum of five to ten trees at each sampling point, and always at a 1–3 m height. At each sampling site, collection of lichens was restricted to a square area not larger than 50 × 50 m and not smaller than 10 × 10 m. Lichen sampling was performed after a dry period of about 70–80 days (precipitation below 7 mm) and during a meteorologically stable period [111]. For lichen analyses of PAHs, approximately each sample was extracted in a Soxhlet with acetonitrile for 24 h. After extraction, all extracts were concentrated by rotary vacuum evaporation and cleaned up in a florisil column with acetonitrile as eluting solvent. Subsequently, the extracts were again evaporated and concentrated with a gentle stream of purified N₂. The samples were analyzed by HPLC, coupled to both ultraviolet/visible and ultraviolet fluorescence detector [112]. Also the measurement of dioxins was performed using ground and dried lichen samples, extracted using Soxhlet method (toluene), cleanup on mixed silica column, aluminum oxide column, and gel-chromatography (Bio-Beads S-X3A) method, and quantified with HRGC/HRMS. Pine needles from *P. pinea* are classified as a good biomonitor of toxic organic compounds including PAHs. Samples were collected from the terminal part of branches, always at the same position on the tree, packed in brown glass bottles, protected from sunlight, and immediately stored at 4°C. All samples were extracted and analyzed for the 16 EPA-PAHs within two months [112]. *Tillandsia usneoides*, from Bromeliaceae, is an epiphyte that captures all its nutrients from the atmosphere and concomitantly accumulates heavy metals. Its morphology, with millimetric

dimensions of the leaves and no roots, makes it ideal for handling and preparation of transplanting systems, and due to its high relation between surface area and mass, it has a high efficiency, for example, in Hg accumulation. Systems were hung at 2–20 m height in open areas, close to and in the surroundings of the gold shops as well as in control areas. Mineralisation of the plants is carried out for Hg analysis and determination of Hg was done by cold vapour atomic absorption spectrophotometry [113]. As shown in Table 14, the most commonly used techniques to obtain the elemental concentration of heavy metals are AAS, AFS, AMA, CVAAS, CVAFS, ETAAS, FAAS, GFAAS, ICP-ES, ICP-MS, and ICP-OES XRF [114–116].

3. An Overview of Findings from Scientific Literature

3.1. Particulate Matter. Many studies were carried out in order to determine the concentration of settled particles. Cao et al. compared deposition rate of atmospheric particles in Xi'an Shaanxi Province, Central China, with those observed in the world [36, 117–124]. The annual average dust deposition fluxes in China (Table 7) ranks among the first observed throughout the world, with values ranging from 0.05 (remote location) to 450 g m⁻² y⁻¹ (desert). The deposition fluxes in the Chinese cities are generally high due to a great number of sources impacting on air quality: presence of coal-burning power plant, sand transport from deserts, drought climate, and heavy traffic. In addition, several studies aimed to evaluate site-specificity, seasonality, and daily variability of settleable particle concentrations. In particular, Wu et al. [125] measured dry deposition flux at Taichung Harbor (Taiwan) and they found levels (μg m⁻² sec⁻¹) 15 times lower than those determined at urban site in Taiwan (86 μg m⁻² sec⁻¹) by Fang et al. [55]. Moreover, dry deposition flux at harbor site was 3 times higher in winter than in summer. The same findings were obtained by Soriano et al. for industrial site in Spain [38]. Probably the high dry deposition fluxes in winter were due to the lower dispersive capacity of atmosphere in that season that determines an accumulation of pollutants emitted into the air because of transport phenomena and of diffusion of pollutants that occur too slowly [126]. On the contrary, the same authors observed opposite features at costal site where highest dry deposition levels were determined in summer, when the ambient dryness and low precipitation increase the atmospheric dust content. In this season in fact, erosion and transport of common soil minerals by means of

TABLE 7: Annual average dust deposition rates ($\text{g m}^{-2} \text{yr}^{-1}$) in different sites throughout the world.

Location	Deposition rate	Reference
Miami, Florida, USA	1.26	[123]
Sapporo, Japan	5.20	[124]
Tel Aviv, Israel	11.0	[122]
Jiuquan, Gansu Province, China	20	[117]
New South Wales, Australia	37.6	[121]
Qingdao, Shandong Province, China	47.45	[4]
Phoenix, Arizona, USA	54.5	[120]
Tianshui, Gansu Province, China	65	[117]
Xi'an, Shaanxi Province, China	76.7	[36]
Lanzhou, Gansu Province, China	110	[117]
Beijing, China	138.9	[119]
Desert regions, China	450	[224]
Namoi Valley, Australia	31.4	[225]
German, Alps	49.3	[226]
Penny Ice Cap, Canada	0.05	[227]

wind and African dust episodes are highly probable in the Mediterranean area [127]. On a daily scale, Fang et al. and Yun et al. found that the average dry deposition fluxes measured for the daytime period were higher than those determined for nighttime period [56]. Moreover, these values strongly correlate with wind speed because, beyond traffic vehicles, wind speed variation between daytime and nighttime period plays an important role in influencing the dry deposition fluxes.

The size distribution of settleable particles has been also evaluated in some papers. Cao et al. showed on average about 10% of the settled dusts having size $<2 \mu\text{m}$, about 80% having size between 2 and $50 \mu\text{m}$, and $<10\%$ having size $>50 \mu\text{m}$ [36]. Despite the concentrations in small particles being high, the dry deposition fluxes are mainly determined by larger particles because of their higher dry deposition velocity.

Finally, it is important to consider that several studies estimate the deposition flux of particle using a multistep model that it takes into account the ambient concentrations and deposition velocities. Despite the approximations used, the estimated fluxes result in good agreement with those measured [42, 53, 57, 125, 128–130].

3.2. Organics. It was demonstrated that wet and dry deposition events are the major processes that remove POPs from the atmosphere causing soil contamination. Hence, there is a need for monitoring the atmospheric deposition of POPs for early recognition of trends and early development of environmental action plans. Researches on atmospheric deposition of POPs, such as PAHs, PCDDs/Fs, and PCBs, have been carried out widely in different parts of the world in order to determine their deposition fluxes, investigating its spatial and seasonal variations, and assess the influence of emission sources, local population distribution, and atmospheric concentrations [21, 32, 131, 132].

3.2.1. Polycyclic Aromatic Hydrocarbons. The comparison among different collector types, sampling workup procedures, and analysis methods showed that the highest deposition rates of PAHs and low measurement uncertainty were obtained with a funnel-bottle collector [24, 133]. The deposition rates obtained with the wet-only collectors were the lowest at industrial sites and under dry weather conditions. For the open-jar collectors, the measurement uncertainty was high [133]. Several monitoring campaigns were conducted in different types of sampling sites and in different seasons in order to measure PAHs deposition fluxes and to study the effect of the Meteorological parameters on monitored levels (see Table 8). Wang et al. in 2011 detected the bulk deposition of 15 PAHs in remote, rural village, and urban areas of Beijing Tianjin region, North China, in spring, summer, fall, and winter [32]. The annually averaged PAHs concentration and deposition fluxes were $11.81 \pm 4.61 \mu\text{g/g}$ and $5.2 \pm 3.89 \mu\text{g/m}^2/\text{day}$, respectively. These values were comparable to those reported previously in North China, where deposition fluxes of 15 PAHs were $4.88 \mu\text{g/m}^2/\text{day}$ in Tianjin and $5.14 \mu\text{g/m}^2/\text{day}$ in rural area of Beijing [125, 131]. Moreover, PAHs flux depositions were similar to those monitored in Seoul ($5.5 \mu\text{g/m}^2/\text{day}$ and $12 \mu\text{g/m}^2/\text{day}$ in spring and winter [134]), in Tampa Bay, USA, ($6.8 \mu\text{g/m}^2/\text{day}$ [135]), in Izmit Bay, Turkey ($8.3 \mu\text{g/m}^2/\text{day}$ [31]), and in Manchester and Cardiff (UK) ($5.2 \mu\text{g/m}^2/\text{day}$ and $4.1 \mu\text{g/m}^2/\text{day}$, resp., [136]). However, the values were significantly higher than that in rural area of Southern Germany ($0.55 \mu\text{g/m}^2/\text{day}$ [28]), in Eastern Mediterranean ($0.46 \mu\text{g/m}^2/\text{day}$ [137]), in rural area of France during heating time period ($0.13 \mu\text{g/m}^2/\text{day}$ [138]), in urban area of western Greece ($0.19 \mu\text{g/m}^2/\text{day}$ [139]), in southern Ontario, Canada ($0.07 \mu\text{g/m}^2/\text{day}$ [140]), and in New England coast ($0.22 \mu\text{g/m}^2/\text{day}$ [141]). The deposition profile was dominated by fluorene (17–28%), phenanthrene (22–35%), pyrene (12%), and chrysene (9–11%)

TABLE 8: Annually averaged PAHs deposition fluxes in different monitored sites.

Annually averaged PAHs deposition fluxes ($\mu\text{g}/\text{m}^2/\text{day}$)			References
Beijing Tianjin region (North China)	Remote, rural, and urban area	5.2 ± 3.89	[32]
Tianjin region (North China)	Rural, urban, and industrialized areas	4.88	[125]
Beijing Tianjin region (North China)	Rural area	5.16	[131]
Seoul (Korea)	Urban area	5.5–12	[134]
Inchon (Korea)	Industrialized area	8.2–22	[134]
Yangsuri (Korea)	Rural background areas	4.1–10	[134]
Tampa Bay (USA)	Urban areas	6.8	[135]
Izmit Bay (Turkey)	Urban/industrialized area	8.3	[31]
Manchester (UK)	Urban areas	5.2	[136]
Cardiff (UK)	Urban areas	4.1	[136]
Southern Germany	Rural areas	0.55	[28]
Eastern Mediterranean	Rural area	0.46	[137]
Paris (France)	Coastal area	0.05–0.62	[138]
Paris (France)	Rural area	0.002–0.63	[138]
Paris (France)	Urban area	0.16–1.3	[138]
Paris (France)	Forest area	0.01–0.5	[138]
Western Greece	Urban areas	0.19	[139]
Southern Ontario (Canada)	Forest areas	0.07	[140]
New England	Coast areas	0.22	[141]

[15, 21, 22, 27, 32]. Moving from wet season (from April to September) to dry season (from October to March), the deposition profile shows a significant increase in the contribution of the lighter compounds, while the atmospheric profile remains essentially unchanged [27, 28, 31]. The deposition fluxes of particle-bound PAHs were not correlated with particulate PAHs in air, but positively correlated with rainfall during the wet season (daily precipitation more than 8.4 mm/day). On the contrary, in dry season, with precipitation less than 3.0 mm/day, the deposition fluxes were well correlated with the atmospheric concentration of the particulate PAHs ($r^2 = 0.62$, $P < 0.1$) but not correlated with rainfall. This finding should be linked to the different distribution patterns of PAHs profile among the particle size fractions in wet and dry seasons; the different size particles have the different precipitation scavenging ratios and deposition velocities [27]. Impact of the ambient temperature on the deposition fluxes was shown by the significant correlations between the deposition fluxes and temperature for PAHs with higher concentrations in winter and lower values in summer [142]. Pekey et al. [31] verified that, while the wet deposition fluxes of 3-ringed PAHs were higher in cold period than in the warm period, the fluxes of 4- or 6-ringed PAHs did not show any significant variation. However, the contributions of 5-ringed PAHs to the total deposition fluxes were very high in warm period when compared with the contributions of 3- and 4-ringed PAH compounds. The spatial distribution of PAHs deposition flux was also assessed in several studies [21, 32, 142].

The monitoring campaign of PAHs in bulk deposition samples conducted by Wang et al. [32], in different areas of Beijing Tianjin region, North China, allowed examining the spatial distribution and emission sources in remote, rural,

and urban areas. The obtained results showed that PAHs bulk deposition flux in rural villages ($3.91 \mu\text{g}/\text{m}^2/\text{day}$) and urban areas ($8.28 \mu\text{g}/\text{m}^2/\text{day}$) was 3.8 and 9.1 times higher than that in background area ($0.82 \mu\text{g}/\text{m}^2/\text{day}$), respectively. This spatial variation of deposition fluxes of PAHs was related to the PAHs emission sources, local population density, and air concentration of PAHs [32]. The different emission sources were qualitatively identified on the basis of PAHs composition profiles represented by diagnostic ratios, according to Motelay-Massei et al. [33]. The FLA/PYR ratio allowed separating the pyrolytic and petrogenic PAH sources: FLA/PYR ratio higher than 1 is a characteristic of a pyrolytic origin, whereas values lower than 1 are typical of a petrogenic origin, such as coal combustion in Europe and North America [33]. This ratio can provide some specific information about the PAHs origin and the change in PAHs sources during the year. Values of IcdP/IcdP + BghiP and FLA/(FLA + PYR) allowed us to confirm PAHs sources: an FLA/(PYR + FLA) ratio < 0.4 implies petroleum, 0.4–0.5 implies petroleum combustion (liquid fossil fuel and vehicle and crude oil), and > 0.5 implies combustion of coal, grass, and wood [33]. The judgment rule for IcdP/(IcdP + BghiP) is the same with that for FLA/(PYR + FLA). Most samples collected in Beijing Tianjin region had ratios > 0.5 , indicating a predominant influence of coal/biofuel combustion. In addition, a significant difference for these ratios between winter and the other three seasons was observed confirming the coal and biomass (straw, firewood) combustion as the prevalent source in winter (the wintertime consumption of these fuels was 1.5–2 times higher than in summer due to residential heating). Cavalcante et al. [21] studied the influence of urban activities on the depositional fluxes of PAHs in bulk precipitation collected in the Fortaleza

metropolitan area, Ceará, Brazil. The highest concentrations and depositional fluxes of PAHs were detected in a zone with heavy automobile traffic and favorable topography for the concentration of emitted pollutants. Diagnostic ratio analyses of PAH samples showed that the major source of emissions is gasoline exhaust, with a small percentage originating from diesel fuel. Contributions from coal and wood combustion were also found [21].

The study conducted by Li et al. [142] in 15 sites in the Pearl River Delta (PRD) from December 2003 to November 2004 showed higher spatial variations in the center of the PRD than at the coastal sites for all PAHs. Levels were higher in the center and lower in the edge of the PRD, confirming that PAHs in the PRD region were predominantly originated from vehicle emission and coal combustion. PAHs two-year monitoring campaign conducted in three rural regions of Southern Germany using passive deposition samplers showed that spatial variability due to topography was negligible and differences between open-field and forest deposition were within a factor of 2. Based on correlations with ambient temperature, advection was identified as the most important factor that controls the atmospheric deposition of PAHs. Gas adsorption significantly contributes to the deposition of the semivolatile compounds in forests, but particle deposition seems to be the major pathway for all PAHs [28].

3.2.2. Polychlorinated Dibenzo-*p*-dioxins and Dibenzofurans. PCDDs/Fs were measured in bulk deposition at three Danish rural forest sites with a mutual maximum distance of 450 km [30]. Mean bulk deposition fluxes were $1 \text{ ng m}^{-2} \text{ y}^{-1}$ I-TEQ and deviated less than 30% among the sites; there was a relatively small geographical variation in PCDDs/Fs bulk deposition over Denmark and at the neighboring stations in Northern Germany. Similar congener patterns, but lower bulk deposition fluxes, were found in a semirural area in Lake Maggiore in Northern Italy ($140 \text{ pg TEQ m}^{-2} \text{ d}^{-1}$) and a suburban area near the Adriatic sea (Rimini, Italy) ($0.75\text{--}3.73 \text{ pg TEQ m}^{-2} \text{ d}^{-1}$) [25, 143]. This finding confirms a long-range atmospheric transport of PCDDs/Fs in areas characterized by low level contamination. Bulk deposition of PCDDs/Fs collected in Danish rural forest sites also showed a moderate seasonal variation with mean winter fluxes that were twice the fluxes during summertime. In particular, it was found that the winter/summer ratios of deposition values for 17 congeners were centered on a mean value of $2.1 \text{ pg TEQ m}^{-2} \text{ d}^{-1}$ with no systematic differences between the congeners [30]. Time series of PCDDs/Fs depositions in a large conurbation in northwestern Germany were analyzed by Bruckmann et al. [144]. Data analysis highlighted a decreasing trend of PCDDs/Fs depositions within the last 2 decades (about a factor of 5) at the urban background sites. At the sites in the near surroundings of the industrial sources, the decrease was even more pronounced (about one order of magnitude). The decrease of depositions at the remote site was smaller, as important local sources were lacking and the levels were dominated by long-range transport. Depositions have reached background levels since about 2005 ($5\text{--}10 \text{ pg TEQ m}^{-2} \text{ d}^{-1}$ in remote site) and there

was no further decrease [144]. Fang et al. [29] used resin deposition samplers to monitor PCDDs/Fs deposition at four sites around steel complexes in Pohang, South Korea. The bulk deposition within the steelwork complexes showed the highest $\Sigma 4\text{--}8$ PCDDs/Fs (tetra-octa) fluxes, ranging from 204 to $608 \text{ pg m}^{-2} \text{ d}^{-1}$, confirming steelwork complexes as major sources of PCDDs/Fs. The deposition around the steelwork complexes was characteristic of high amount of lower chlorinated PCDDs. Apparent seasonal variations of the bulk deposition at each site and seasonal homologue patterns of PCDDs/Fs were observed, probably resulting from temperature-dependent gas/particle partitioning [29].

3.2.3. Polychlorinated Biphenyls. Several studies conducted in industrial, urban, and rural areas showed that particle phase depositions dominated the dry deposition processes for the removal of PCBs from the atmosphere, and the atmospheric deposition fluxes in the cold season were higher than those detected during the warm season [143, 145–147]. Average dry deposition fluxes of total PCBs in industrial, urban, and rural areas of southern Taiwan ranged from 289 to $1010 \text{ pg m}^{-2} \text{ d}^{-1}$ ($0.540\text{--}1.94 \text{ pg WHO-TEQ m}^{-2} \text{ d}^{-1}$) [147]. These values were higher than those measured in Turkey near an industrial site, where average particulate $\Sigma 41$ -PCBs dry deposition fluxes were 349 ± 183 and $469 \pm 328 \text{ ng m}^{-2} \text{ d}^{-1}$ in summer and winter [146] and, at a suburban site, where PCBs flux ranged from 2 to $160 \text{ ng m}^{-2} \text{ d}^{-1}$ [148]. Mi et al. in 2012 [147] verified that the dry deposition velocity of individual PCBs ($0.069\text{--}3.38 \text{ cm/s}$) increased as the number of chlorinated substitutes increased. This evidence is due to the fact that low chlorinated PCBs are predominant in gas phase and have lower deposition velocities.

3.3. Inorganics

3.3.1. Metals. Besides environmental consequences, particulate matter was linked to health damage as it contains a wide range of toxic metals and organic contaminants. Some metals like Cu and Zn are essential nutrients but they can cause harmful effects depending on their concentration levels. Cd is considered very toxic, classified as potential carcinogenic [149]. Pb is also recognized by the World Health Organization as one of the most dangerous chemical elements for the human health [150, 151]. In order to describe these issues a lot of scientific work was carried out. Fang et al. [55] measured downward and upward dry deposition fluxes of particulate heavy metals (Fe, Pb, Zn, Cu, Mg, and Mn) in daytime period (from 6:30 a.m. to 6:30 p.m.) and nighttime period (from 6:30 p.m. to 6:30 a.m.) in a traffic sampling site in Sha-Lu, a small city in the central Taiwan. The average downward and upward dry deposition fluxes found in the daytime period (54.1 and $26.5 \text{ g/m}^2 \text{ s}$, resp.) were all higher than average downward and upward dry deposition fluxes found in the nighttime period (26.2 and $12.1 \text{ g/m}^2 \text{ s}$, resp.). This fact is probably due to the vehicular traffic and wind speed variation between daytime and nighttime period. In addition, the average downward dry deposition fluxes are greater than the upward dry deposition fluxes for all the heavy metals

in either daytime or nighttime period, and the proposed reasons are that the wind speed and concentration difference between daytime and nighttime period lead to these results. Daytime and nighttime dry deposition fluxes of particulate heavy metals were measured also in Kunpo, a small city in the Seoul metropolitan area in Korea. Also in this site, the fluxes of nighttime were lower than daytime in all periods due to the lower wind speeds during the nighttime (the average wind speed of 1.47 m s^{-1} in nighttime versus 2.23 m s^{-1} in daytime) and the lower atmospheric concentrations than daytime. The average fluxes of Al and Ca, typical crustal species, were 1–2 orders of magnitude higher than Mn and anthropogenic elements such as As, Cd, Cu, Ni, Pb, and Zn.

Fang et al. [56] studied the dry deposition fluxes of metallic elements at Taichung Airport in central Taiwan in relationship with the size of particulate matter. At this site, the average downward dry deposition fluxes of metallic elements (Fe, Zn, Mn, Cu, Pb, Cr, and Mg) were 218.0, 109.0, 194.3, 21.9, 56.0, 11.2, and $105.7 \mu\text{g m}^{-2} \text{ d}^{-1}$, respectively. The most of these fluxes are due to particles larger than $2.5 \mu\text{m}$ in size due to their high deposition velocities. The results also indicate that the best approach to estimate overall dry deposition is by using either coarse particle or total particle concentrations. The same authors [57] also investigated seasonal concentrations of metallic elements Mn, Zn, Cr, Cu, and Pb in dry deposit particulates at five characteristic sampling sites in central Taiwan. Average seasonal concentrations of Mn, Zn, Cr, Cu, and Pb in dry deposits were the highest in fall and winter and lowest in summer at all five sampling sites. In addition, the average concentrations of dry deposited Mn, Zn, Cr, Cu, and Pb were the highest at Quan-Xing, an industrial site, in which many industrial processes take place. Also Soriano et al. in 2012 [38] performed a study of heavy metal concentrations in the settleable particulate matter in two locations (Almazora and Vila-real) which were positioned within the ceramic cluster of Castellón (Spain) and had high industrial density. The results related to the heavy metal concentrations over the observation period show a high seasonal variability for heavy metal content in settleable particulate and a strong dependence on the rainfall in the study area. The maximum values of heavy metals were obtained in spring or autumn coinciding with the highest rainfall, while the lowest values were generally obtained during periods of low rainfall (summer). This fact was associated with the scavenging ability of the rain in removing pollutants from the atmosphere. In the atmosphere, the gases are absorbed and the particulate matter is trapped in raindrops falling on the ground [152]. In falling, the drops of rain (much larger than drops of water) drag solid particles in their path (dust or contaminants). Other authors have made the same observation. Melaku et al. [153] found a clear decrease in the concentrations of heavy metals in the samples collected in Washington, DC (precipitation samples and ambient air samples), during periods of high rainfall. These data provide strong evidence that rain affects the concentrations of heavy metals in any type of sample [153]. The importance of dependence of deposition of heavy metals from climatic characteristics was discussed also by Muezzinoglu and Cizmecioglu [59]. In this

study, deposition samples were collected and analyzed for selected heavy metals in Izmir, Turkey. The average total heavy metal concentrations for Cr, Cd, Pb, Cu, Zn, and Ni were generally higher than the values previously measured at different sites around the world [154–158]. In particular, the dry deposition for Izmir was 10–50 times higher for Cd, 4–20 times higher for Pb, and 5–30 times higher for Zn than in Tokyo, Japan [154]. Although the sampling methods may differ, statistical analysis showed that the sticky deposition plate and water surface sampler methods give the same dry deposition results [58, 154].

Sakata et al. [17] measured wet and dry deposition fluxes of As, Cd, Cr, Cu, Hg, Mn, Ni, Pb, and V in the Tokyo Bay area, which is highly affected by anthropogenic sources. The order of the average ratios of dry/wet deposition fluxes at three sites was $\text{Cr}, 6.9 > \text{As}, 5.9 > \text{Mn}, 4.7 > \text{V}, 4.5 > \text{Cu}, 4.4 > \text{Ni}, 3.7 > \text{Cd}, 2.9 > \text{Pb}, 2.0 > \text{Hg}, 1.0$. Thus, the atmospheric deposition of the trace elements except Hg in Tokyo Bay is predominantly dry deposition. For such trace elements, the wet and dry deposition fluxes within the bay were higher than those inland. The mean deposition fluxes (wet + dry) of the trace elements in the Tokyo Bay area can be compared with those estimated, or directly measured, in other aquatic regions [74, 141, 159–164]. The fluxes of Cd, Cr, Cu, Ni, Pb, and V in the Tokyo Bay area were similar to those in the Seine Estuary [159] or the Pearl River Delta [160], where highly industrialized regions are present. However, the Tokyo Bay area had much higher atmospheric deposition fluxes of trace elements than other aquatic regions in the US and Europe [74, 141, 161–164], as shown in Table 9.

Sabin et al. [53] analyzed the role of major roadways, such as a freeway, as a significant source of localized metal deposition to urban surfaces and the role of resuspension in the net deposition and dispersion of particulate matter near roadways. In particular, the authors characterized dry deposition patterns of Cr, Cu, Pb, Ni, and Zn upwind and at increasing distances downwind of the freeway in coastal Los Angeles. The dry deposition fluxes of metals were higher at short distances from the freeway and quickly reduced to urban background fluxes within 150 m, especially for Cu, Pb, and Zn. These results were similar to the observations of Zhu et al. [165] for ultrafine particle concentrations ($d < 0.1 \mu\text{m}$) measured downwind of the same freeway in which high concentrations near the freeway reduced to urban background within 300 m. The fluxes of Cu, Pb, and Zn were significantly higher close to the freeway suggesting that the freeway acts as a significant source of these metals especially concentrated on larger particles, which are expected to deposit close to their source. The freeway likely represents a source of large particles containing Cu, Pb, and Zn because of resuspension of road dust, as vehicles travel on the freeway at high velocities [166, 167], and from tire and brake wear from vehicles [168–170].

3.3.2. Ions. The chemical characterization of wet and dry depositions plays an important role in sources identification. In particular, ions determination in wet and dry depositions is essential for understanding regional variations, local influences by anthropogenic or natural sources, and long-range

TABLE 9: Comparison of mean wet + dry deposition fluxes ($\text{mg m}^{-2} \text{yr}^{-1}$) of trace elements in aquatic regions.

Location	Period	As	Cd	Cr	Cu	Mn	Ni	Pb	V	References
Tokyo Bay, Japan	2004-2005	2.9	0.39	6.2	16	87	6.8	9.9	6.9	[17]
Seine Estuary, France	2001-2002		0.36		9.4		4.1	18		[159]
Pearl River delta, China	2001-2002		0.07	6.4	19	9.0	8.4	13	2.1	[160]
Massachusetts Bay, USA	1992-1993	0.02	0.27	2.7	2.5	3.4	1.5	1.8	0.60	[141]
Lake Superior, USA	1993-1994	0.17	0.46	0.21	3.1	4.2	0.80	1.5	0.34	[161]
Lake Michigan, USA	1993-1994	0.14	0.45	0.20	1.9	2.8	0.61	1.6	0.14	[161]
Lake Erie, USA	1993-1994	0.18	0.49	1.1	4.2	4.5	0.74	1.8	0.60	[161]
North Sea	1992-1994	0.25		2.4	1.3	2.6	1.2	3.7	1.1	[162]
Ligurian Sea	1997-1998		0.06	0.62	1.3		1.1	1.2		[163]

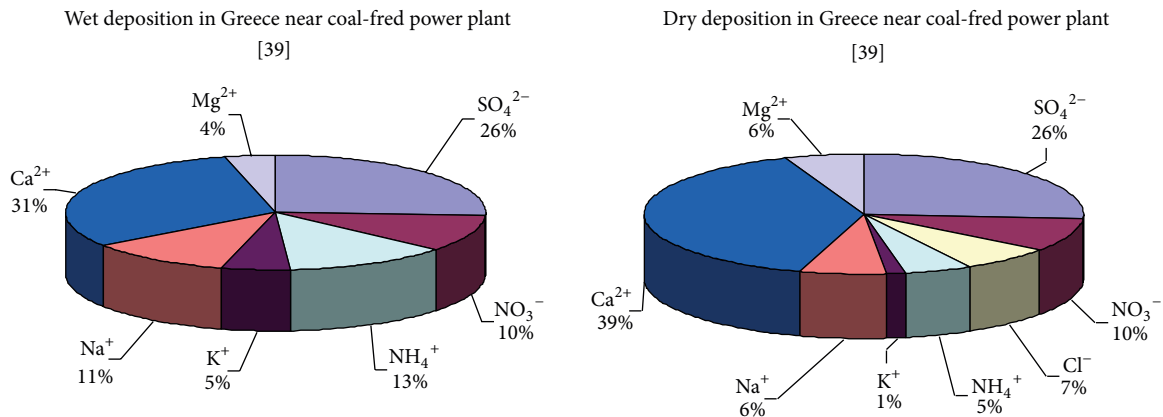


FIGURE 3: Wet and dry deposition fluxes percentage of ions at a site downwind to coal-fired power plants in Northwestern Greece [39].

transport phenomena of air pollutants. Among the ions, more attention is paid to the atmospheric deposition of nitrogen and sulfur species because of drawbacks that they could lead to for the ecosystems such as acidification and accumulation of excess nutrients [171]. The deposition flux of nitrate in rural and suburban region is controlled by the imported NO_x from motor vehicles, the deposition of sulphate by power plants and factories [172], and deposition of ammonium mainly originated from the ammonia volatilization loss from N-fertilization in agricultural fields and animal production [173]. Tsitouridou and Anatolaki [39] also found high sulphate ($27 \text{ meq m}^{-2} \text{ y}^{-1}$) and calcium ($31 \text{ meq m}^{-2} \text{ y}^{-1}$) content in wet and dry deposition samples at two sites in northwestern Greece (Figure 3), where four coal-burning power plants were present, highlighting the strong influence of fly ash and SO_2 emissions as also described by other authors [174].

The dry and wet deposition fluxes for these species are well correlated with air pollutions, and their decreasing in the recent two decades in Europe and USA is substantially due to a more stringent legislation for the reduction of atmospheric concentrations of sulphur dioxide and nitrogen oxides [175]. Nevertheless, the sulphur and nitrogen deposition values in many areas still remain far from satisfactory and they substantially change among the different sampling sites [176]. Rossini et al., for example, found deposition fluxes in industrial area, about three times higher (10.4, 8.9, and

$20.1 \text{ mg m}^{-2} \text{ d}^{-1}$ for sulphate, ammonium, and nitrate, in average, resp.) than in urban site (5.9, 2.3, and $8.9 \text{ mg m}^{-2} \text{ d}^{-1}$ in average, resp.) in Venetian lagoon (Italy) and yearly deposition fluxes at least twice higher than those determined at coastal (Castelporziano and Pula) and remote (Tessa) sites in Italy [177]. Qi et al. in 2013 [178] also reported an overview of nitrogen dry deposition fluxes over the oceans. In Yellow Sea [179, 180] and the East China Sea [181] were determined similar values for the dry deposition fluxes. However, these values were higher than the values of coastal regions in Japan [182], US [183], the West Baltic Sea [184], and the Atlantic Ocean [185]. In particular, the higher concentrations of inorganic nitrogen were most likely related to the fossil fuel combustion [186], animal waste [187], and large-scale utilization of nitrogenous fertilizers in China [188, 189]. In addition, the values of dry deposition flux of nitrate and ammonium over Yellow sea were similar to Singapore [137, 190] but lower than that of inland China [191, 192] due to high emission intensities of NH_3 , NO_x , and SO_2 in inland regions. Moreover, several papers highlighted also the differences in dry and wet contribution to the total depositions relating to different site typologies. Tsitouridou and Anatolaki [39] observed that the contribution of dry deposition to the total (wet + dry) at urban site and for the site closed the power plant in Greece is higher (60–70%) than wet deposition for ions, while an opposite pattern characterized by higher contribution for wet than dry deposition was observed for

TABLE 10: Mercury emissions from natural and anthropogenic sources [193].

Emissions from natural sources	Mg y ⁻¹	% global emission budget	Emissions from anthropogenic sources	Mg y ⁻¹	% global emission budget
Oceans	2682	36	Combustion of fossil fuel	810	11
Biomass burning	675	9	Artisanal small scale gold mining	400	5
Desert, metalliferous, and nonvegetated zones	546	7	Nonferrous metal production	310	4
Tundra and grassland/Savannah/Prairie/Chaparral	448	6	Cement production	236	3
Lakes	96	1	Caustic soda production	163	2
Forests	342	5	Waste disposal	187	2
Agricultural areas	128	2	Pig iron production	43	1
Evasion after mercury depletion events	200	3	Mercury production	50	1
	90	1	Coal bed fires	32	0
Volcanoes and geothermal areas			VCM production	24	0
			Other	65	1
Total	5,207	70	Total	2,320	30

the remote site. These findings suggest that dry deposition of sulphate, nitrate, and ammonium, which are due to gaseous species deposition for 70–90%, are generally more connected with local emissions with respect to wet deposition which better reflects transport phenomena and other gas-particle reaction formations. Anyway, the wet contribution to total deposition of ions increases also at local levels when high rainfall was observed. In fact, the monthly variation of the wet deposition is mainly related to rainfall, showing the significance of the precipitation effect on the prevailing scavenging mechanism on the ions [176, 178].

3.4. Mercury. Mercury emissions from natural processes (primary mercury + reemissions), including mercury depletion events, were estimated to be 5,207 Mg y⁻¹ which represent more than 70% of the global mercury emission budget (GEb) (see Table 10). Oceans were the most important sources followed by biomass burning, desert, metalliferous and nonvegetated zones, tundra, and grassland. Anthropogenic source, which includes a large number of industrial point sources, was estimated to account for 2,320 Mg of mercury emissions annually. The majority of mercury emissions originate from combustion of fossil fuel, followed by artisanal small scale gold mining, nonferrous metal production, cement production, caustic soda production, waste disposal, pig iron production, and other processes [193]. However, the anthropogenic Hg emissions in Europe were still higher (341.8 ty⁻¹ in 1995) than the natural emissions, estimated to be about 250–300 ty⁻¹, although a decrease of 45% results in comparison with the emissions registered in 1990. Coal combustion has been the major source of anthropogenic emissions contributing to more than half of the total anthropogenic emissions [194]. As in most of the industrialized areas, the anthropogenic emission of Hg represents a particular concern; atmospheric deposition of Hg is gaining importance in the scientific community in evaluating the biogeochemical fate of this metal.



FIGURE 4: Newly designed wet-deposition sampler, Wet N-con System [195].

Ann Chalmers et al. [195] described reliability and collection efficiency of a newly designed wet-deposition sampler (Figure 4) and presented THg and MeHg wet-deposition data and analysis from four sites around the Boston area. Wet-deposition samples were collected from January 2002 to August 2004 and analyzed for THg, and a subset of samples from September 2003 to August 2004 was analyzed for MeHg. Concentrations of THg in precipitation ranged from 0.73 to 24.6 ng L⁻¹ at the four sites, whereas MeHg concentrations at all sites were below the detection level of 0.04 ng L⁻¹. The Manchester site, in the most urban environment, had the highest precipitation-weighted THg concentration (8.31 ng L⁻¹), and Blue Hill, the closest site to Boston, had the highest deposition rate (9.98 μg m⁻² y⁻¹). The regional background site, in Laconia, NH, had the lowest precipitation-weighted THg concentration (6.87 ng L⁻¹) and the lowest deposition rate (6.56 μg m⁻² y⁻¹). Finally, they

found that the average annual Hg wet-deposition rate in metropolitan Boston (Manchester, Beverly, and Blue Hill) was $9.16 \mu\text{g m}^{-2} \text{y}^{-1}$, 28 percent higher than the Hg deposition rate at the regional background site.

Fulkerson et al. [196] attested that the total flux of Hg to the earth's surface involves both wet and dry processes. However, direct measurements of dry deposition are technically difficult to make and have very large uncertainties [197–200]. Consequently, inferential methods have been used to estimate dry deposition, but these methods require knowledge of ambient concentrations of each species of Hg in the atmosphere.

The importance of wet scavenging of mercury has been demonstrated by Huang et al. [81], studying the wet deposition of mercury at a remote site in the Tibetan Plateau, collecting deposition samples (both rain and snow) over a 2-year period from July 2009 to 2011, and showing concentrations of mercury from 3.8 to 5.3 ng L^{-1} during the monsoon season and from 6.9 to 8.2 ng L^{-1} during the non-monsoon season. Monsoon season (June through September) is typically the rainy season in the Tibetan Plateau; approximately 90.6% of the annual total precipitation fell in the monsoon season. As a consequence of the strong seasonality of precipitation at study site, total mercury wet deposition fluxes were also seasonal with 83% of the wet deposition fluxes occurring during the monsoon season. Further works have been carried out on the atmospheric mercury deposition, and many of these ones were treated in a review of studies of 2012 [201] in which the current understanding on atmospheric Hg emissions, distribution, and transport in China was reported. The magnitude of Hg emissions to the atmosphere from Chinese anthropogenic sources has been estimated to be in the range $500\text{--}700 \text{ t y}^{-1}$, whereby comprising a significant proportion of the globe total anthropogenic emissions. According to the studies by Pirrone et al. and Wu et al. [193, 202], coal combustion in China released approximately 256–268 t of Hg to the atmosphere in 2003, accounting for about 40% of the total anthropogenic emissions in that country. Furthermore, Hg(p) concentrations in Chinese urban air are generally significantly high, where the observations fell in a range from 109 to 1180 pg m^{-3} .

Rose et al. [203] studied the MeHg concentrations by bulk deposition during winter in a remote Scottish mountain lake, Lochnagar, covering a 7-year period from 2001 to 2008 and showing an unusual seasonal pattern as elevated MeHg concentrations occur each winter while concentrations fall below the limit of detection each summer. Concentrations above detection limit occurred each year between autumn and spring; summer deposition concentrations were lower and usually below detection limit. In particular, peak concentrations occurred in the first few months of each year (late February to early April) and ranged between 0.27 ng L^{-1} (April 2004 and 2005) and 0.88 ng L^{-1} (March 2006). In addition, in October 2006 and October 2007, peaks were also recorded reaching 1.2 ng L^{-1} and 0.31 ng L^{-1} , respectively. Finally, the authors concluded that the observed winter inputs of MeHg to the Lochnagar catchment may therefore represent a significant input to the loch of this biologically

important pollutant, especially during snowmelt when winter accumulation enters the loch in a short period of time.

Drevnick et al. [204] used lake sediments to derive estimates of net atmospheric Hg deposition to Svalbard, Norwegian Arctic. Sedimentary Hg accumulation in these lakes is a linear function of the ratio of catchment area to lake area, and they used this relationship to model net atmospheric Hg flux using a DMA-80 Direct Mercury Analyzer; preindustrial and modern estimates are $2.5 \pm 3.3 \text{ mg m}^{-2} \text{y}^{-1}$ and $7.0 \pm 3.0 \text{ mg m}^{-2} \text{y}^{-1}$, respectively. Hg concentrations and accumulation rates in lake sediments increase from relatively low values in preindustrial sediments to peak values in recent sediments. Concentrations increase up-core from 20– 50 ng g^{-1} dry wt. to 60– 90 ng g^{-1} dry wt. These results are consistent with other studies of lake sediments throughout the Arctic [205–207]. In Tables 11 and 12 is reported a collection data of THg and MeHg deposition fluxes.

3.5. Biomonitors. The most commonly used organisms for air pollution and atmospheric deposition assessment, as biomonitors, are lichens, mosses, pine needles, and plants. Table 13 shows the most common biological species used in biomonitoring with main properties and references. Lichens were recognized as potential indicators of air pollution as early as the 1860s in Europe and elsewhere [100, 208]. Lichens were used to monitor metals, sulphur, nitrogen, fluoride, radionuclides, and a variety of organic compounds, such as dioxins and furans, PCBs, and substances originated from organochlorine pesticides [110–112, 209, 210]. There are only a few studies using lichens as biomonitors of PAHs [109, 211, 212]. The majority of these studies were conducted in natural and forested ecosystems or in urban environments [111, 112].

In later works, averaged elemental contents of filter-trapped air particulate materials or deposition were compared with biomonitor's averaged metal concentrations: Andersen [213] reported parallelisms and linear relations in lichens, Sloof [99] and Jeran et al. [214] found positive correlations between metals in air particulates and transplanted lichens, and Berg and Steinnes [215] observed significant correlations between wet deposition and metal concentrations in mosses.

Goyal and Seaward [216] demonstrated possible metal uptake by lichen's rhizine, Prussia and Killingbeck [217] explained differences in lichen metal content associated with differences in substrata, and De Bruin and Hackenitz [218] found metal concentrations which did not differ between lichens and their bark substrata.

Concentrations of PCDDs/Fs in lichens tend to decrease after a wet deposition period. This decrease is the greatest for the highest molecular weight compounds (the most chlorinated PCDDs/Fs, such as octa-chloro-dibenzo-dioxin), meaning that these compounds are probably associated with the surface of lichens. This association may be mainly due to a slower diffusion rate of the highest molecular weight compounds through the lichen thallus; the slower rate will cause a higher concentration of these compounds at lichen surface and thus will make them more susceptible to mechanical wash-off. As POPs are not soluble in water, rain is not

TABLE 11: Collection of some data of THg deposition fluxes.

Location	Period	Classification site	THg deposition flux ($\mu\text{g m}^{-2} \text{y}^{-1}$)	References
Boston (Manchester site)	January 2002 to August 2004	Urban	9.38	[195]
Boston (Laconia)	January 2002 to August 2004	Background	6.56	[195]
Nam Co Station, China	2009–2011	Alpine	1.75	[81]
Chongqing	March 2003 to Feb. 2006	Suburban	77.6	[228]
Mt. Leigong, China	2008-2009	Alpine	6.1	[229]
Mt. Changbai, China	2005-2006	Alpine	8.4	[230]
Svalbard, Norwegian Arctic	Preindustrial time	Lake sediments	2.5	[204]
Svalbard, Norwegian Arctic	Postindustrial time	Lake sediments	7.0	[204]
Changchun	Jul. 1999 to Jul. 2000	Urban	152	[55]
Mt. Gonggaa, Sichuan	Jan. to Dec. 2006	Remote	9.1	[231]

TABLE 12: Collection of some data of MeHg deposition fluxes.

Location	Period	Classification site	MeHg deposition flux ($\mu\text{g m}^{-2} \text{y}^{-1}$)	Ref.
Nam Co Station, China	2009–2011	Alpine	0.11	[81]
Mt. Leigong, China	2008-2009	Alpine	0.06	[229]
Experimental Lakes	1992–1994	Boreal	0.04	[232]
Wujiang, Guizhou	Jan. to Dec. 2006	Semiremote	0.18	[233]
Mt. Leigong, Guizhou	May 2008 to May 2009	Remote	0.06	[234]
Mt. Gonggaa, Sichuan	Jan. to Dec. 2006	Remote	0.11	[231]

TABLE 13: Most common biological species used in biomonitoring with main properties and references.

Biological species	Properties	Pollutants	References	
Lichens	Lack of roots	Ions	[110]	
	Aerial resource supply	N and S	[110]	
	Same morphology throughout the seasons	PCDDs/Fs	[110, 111]	
	Longevity	PAHs	[94, 106, 107, 109, 112, 211–218]	
	Slow growth	Heavy metals	[98, 100]	
Mosses	Lack of roots	Heavy metals	[95, 96, 103]	
	Aerial resource supply			
	High surface area/mass ratio			
Pine needles	High efficiency in accumulation	PAHs, POPs	[97, 104]	
	One conifer tree branch has several year-classes of needles, which makes it possible to obtain a pollution profile for more than one year	PAHs	[112]	
		PCDDs/Fs	[111]	
Plants	No roots	Heavy metals	[113]	
	<i>Tillandsia usneoides</i>			
	Aerial resource supply			
	Mill metric dimensions of leaves			
	High surface area/mass ratio			
	<i>Taraxacum officinale</i> L. <i>Trifolium pratense</i> L.	High efficiency in accumulation	Heavy metals, in particular Cu	[115]
	<i>Urtica dioica</i> L.	Versatile and diffuse evergreen ornamental specie	Heavy metals, in particular Pb	[115]
<i>P. tobira</i>	Useful to assess levels and patterns of pollutants in urban areas	Heavy metals	[221]	

TABLE 14: List of the main analytical techniques and classes used in deposition studies, with acronyms.

Class technique	Analytical technique	Acronym
Atomic absorption spectroscopy	Atomic absorption spectroscopy	AAS
	Cold vapour atomic absorption spectroscopy	CVAAS
	Electrothermal atomic absorption spectroscopy	ETAAS
	Flame atomic absorption spectroscopy	FAAS
	Graphite furnace atomic absorption spectroscopy	GFAAS
Atomic fluorescence spectroscopy	Atomic fluorescence spectroscopy	AFS
	Cold vapour atomic fluorescence spectroscopy	CVAFS
Inductively coupled plasma spectrometry	Inductively coupled plasma emission spectrometry	ICP-ES
	Inductively coupled plasma mass spectrometry	ICP-MS
	Inductively coupled plasma optical emission spectrometry	ICP-OES
X-ray fluorescence	X-ray fluorescence	XRF
Mercury analyser	Advanced mercury analyser	AMA
Chromatography	High performance liquid chromatography-mass spectrometry, coupled to both ultraviolet/visible and ultraviolet fluorescence detector	HPLC-MS/UV-Vis-FL
	Gel permeation chromatography and quantification with high resolution gas chromatography—high resolution mass spectrometry	GPC and HRGC-HRMS
	Gas chromatography—mass spectrometry	GC-MS
	Gas chromatography flame-ionization detectors	GC-FID
	High performance liquid chromatography-fluorescence detection	HPLC-FL
	Ion chromatography	IC
Accelerated solvent extraction	Accelerated solvent extraction	ASE
Elemental analysis	Carbon, hydrogen, nitrogen, sulphur, and oxygen-elemental analyzer	CHNS/O
Electron microscopy	Scanning electron microscopy	SEM

likely to act as vehicle to lead POPs from the surface to the bulk of lichens. Also, after a given volume of rain, the levels of PCDDs/Fs in lichens remain relatively constant, meaning that a fraction of POPs will be captured inside the lichen thallus or associated with insoluble particles trapped by the fungus and not accessible to further wash-off [107].

PAH concentrations in lichens were compared with PAH concentrations measured in a conventional active sampler in an outdoor environment. Significant positive correlations between high molecular weight PAHs, sixteen EPA-PAHs, and BaP equivalent concentrations in lichens and those in air were found. Concentrations of sixteen EPA-PAHs in lichens and air showed a seasonal variation, with highest values during winter and lowest values during summer [94].

Blasco et al. [208] used lichens and found that the road traffic was the main source of PAHs in the Pyrenees Mountains region. These authors found that PAHs in lichens reflected the atmospheric particulates when they studied the PAH pollution caused by vehicle emissions in a tunnel but they did not make a calibration between lichens and air. When comparing lichens to soil and air, it was shown that profile of PAHs in lichens was substantially different from the one in soil but similar to air; it was also revealed that lichens intercept PAHs from both the vapor and particulate-phases of air [112]. More recently, using spatial models of

PAHs accumulated in lichens, it was possible to fingerprint multiple sources of atmospheric PAHs in a regional area. Lichens seem to be an excellent candidate for biomonitoring PAHs in the atmosphere [94].

PCDDs/Fs profiles in *R. canariensis* were more similar to the ones found for air samples rather than the ones found for soil, showing that they are not reflecting soil particle resuspension or soil vaporization. Compared to other biomonitors (pine needles, fruits, and vegetables), lichens have appeared to accumulate greater concentrations of PCDDs/Fs, meaning that they may provide useful data, especially in areas where levels are below the detection limit for other monitors [111].

Lichens monitoring allowed the integration of PCDDs/Fs atmospheric deposition for much longer periods, allowing relating low levels with long-term chronic effects on health. Thus, the production of high-resolution data on environmental exposure essential to perform reliable environmental health studies was possible. It was argued that PCDDs/Fs in lichens may be used as spatial estimators of the potential risk of inhalation by the population present in the area [110].

Numerous biomonitors have been used to monitor PCDDs/Fs, including vegetation (pine needles, leaves, grass, vegetables, etc.), birds, fishes, and mollusks. To date, pine needles are the most used biomonitors to evaluate air deposition of PCDDs/Fs, as they can be found worldwide, allowing

comparisons between countries. While vegetation is mainly used to provide information on the short-term exposure to PCDDs/Fs, soil samples are also commonly analyzed in order to describe long-term exposure to PCDDs/Fs, since soil is a sink for these compounds [111].

For mosses, Kuik and Wolterbeek [219] found relatively high crustal contributions to the moss levels of elements such as Al, Sc, La, and further lanthanides; D. H. Brown and R. M. Brown [96] suggested that the increase in cation exchange capacity from moss apex to base is part of its natural balance of elements which, in turn, is affected by the proximity of the soil.

Mosses accumulate metals in a passive way, acting like ion exchangers; most of the metals in mosses show a correlation between dry bulk amount and wet deposition concentration [93, 215].

Pleurozium schreberi, *Hylocomium splendens* [103, 215], and *Hypnum cupressiforme* [93] are the moss species more often chosen in world biomonitoring.

Although mosses and lichens receive elements from the atmosphere through wet and dry deposition in areas with widespread geochemical natural and anthropogenic sources of metals, they cannot be used interchangeably as biomonitors, as element compositions of the moss were affected more than that of the lichen by the geochemical features of the region [220]. In these environments, epiphytic lichens seem to be more reliable than biomonitors of atmospheric deposition of trace elements, though more sensitive to atmospheric pollution by sulphur compounds.

Malizia et al. [115] employed inductively coupled plasma atomic emission spectroscopy (ICP-AES) to assess the concentration of selected heavy metals (Cu, Zn, Mn, Pb, Cr, and Pd) in soil and plants and found that the leaves of *Taraxacum officinale* L. and *Trifolium pratense* L. can accumulate Cu and *Urtica dioica* L. representing the vegetal species can accumulate the highest fraction of Pb.

Atmospheric deposition of heavy metals, using the epiphytic moss genera *Fabronia ciliaris* collected from six urban sites in the Metropolitan Zone of Toluca Valley in Mexico, revealed that the concentrations of K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn, Rb, Sr, and Pb (determined by total reflection X-ray fluorescence technique) showed an evidence of anthropogenic impact in the industrial and urban areas, mainly due to the intense vehicular traffic and fossil fuel combustion [116]. Concentration of Hg in samples of lichen *Hypogymnia physodes*, the moss *Pleurozium schreberi*, and the soil humus collected in Polish and Czech Euroregions Praded and Glacensis showed a statistical similarity to the concentrations determined for other areas located far from significant sources of Hg, with some different surface distributions in the considered regions [114]. Introducing a new way of analyzing the results of biomonitoring, based on the comparison factor (CF) (defined as a ratio of a difference between the concentrations of an analyte in lichens and in mosses, to the arithmetic means of these concentrations), they were able to indicate that a deposited bioavailable analyte was amenable to the primary emission and not to the secondary enrichment of the atmospheric aerosol with the local soil pollutants.

An effective air pollution control strategy requires source apportionment of airborne pollutant emission sources and plants may contribute to depict emission scenarios of PM₁₀. *P. tobira*, a versatile and diffuse evergreen ornamental species in the Mediterranean urban environment, is a suitable passive biomonitor, useful to assess levels and distribution patterns of inorganic solid pollutants in urban areas. The ICP-MS analysis carried out for several elements (Al, Ba, Be, Bi, Br, Ca, Cd, Cl, Co, Cr, Cs, Cu, Fe, K, Li, Mg, Mn, Mo, Na, Ni, Pb, Si, Ti, V, Zn) and factor analysis allowed identifying three main source groups of elements (crustal components, sea-salt spray, and anthropogenic sources), useful to elucidate source-receptor relationships [221].

Due to their features, very often biospecies are employed in survey aimed at describing the distribution of selected elements in some extended areas. For example, the European moss survey has an important role in identifying spatial and temporal trends in atmospheric heavy metal pollution across Europe [222]. Recent decline in emission and subsequent deposition of heavy metals across Europe has resulted in a decrease in the heavy metal concentration in mosses for the majority of metals (As, Cd, Fe, Pb, V, Cu, Ni, and Zn), except Hg and Cr, for whom no significant reduction was observed. This route appears essential for monitoring future trends at a high spatial resolution and provides a useful tool for additional validation of modeled atmospheric deposition fluxes. Biospecies, such as mosses, lichens, and plants, provide a cheap, effective alternative to deposition analysis. The goals of these experimental approaches, due to their low cost and spatial distribution, are often focused on the potential role of vegetation for the removal of particulate pollution.

4. Conclusions

Atmospheric deposition processes, which are important to understand the fate and distribution of organic and inorganic pollutants, have been, for a long time, a topic of minor importance with respect to direct air monitoring in the evaluation of environmental air quality and human health risk assessment. The extreme versatility of the analytical tools recently developed in the study of the atmospheric deposition, the development of new collectors, and new analytical and statistical tools have enabled the use of atmospheric deposition information for assessing the impact of different pollutants on many environmental compartments and to perform the source apportionment to receptor sites at long and short distance from the sources. Different collectors were used to evaluate site-specificity, seasonality, and daily variability of settleable particle concentrations. Monitoring over long periods has revealed that erosion and transport of common soil minerals by means of wind and African dust episodes are highly probable in the Mediterranean area. Atmospheric deposition of POPs (PAHs, PCDDs/Fs, and PCBs) in different parts of the world allowed determining their deposition fluxes, investigating their spatial and seasonal variations, and assessing the influence of emission sources, local population distribution, and Meteorological parameters on monitored levels. The different PAH emission

sources were qualitatively identified on the basis of composition profiles represented by different diagnostic ratios. It was found that FLA/PYR ratio higher than 1 is a characteristic of a pyrolytic origin, whereas values lower than 1 are typical of a petrogenic origin, such as coal combustion in Europe and North America. Congener pattern analysis and bulk deposition fluxes in rural sites confirmed a long-range atmospheric transport of PCDDs/Fs in areas characterized by low level contamination and their moderate seasonal variation with higher winter fluxes than during summertime. Metal flux deposition monitoring allowed confirming the role of major roadways as significant sources of localized metal deposition to urban surfaces and of resuspension in the net deposition and dispersion of particulate matter near roadways. Newly designed wet-deposition samplers were also used for the characterization of deposited mercury, which plays an important role due to its toxicological proprieties. Results demonstrated the importance of rain scavenging in the deposition of mercury and the relatively higher magnitude of Hg deposition from Chinese anthropogenic sources, where a significant proportion of the globe total anthropogenic emissions of this metal is located. In recent years, more attention was paid to biological monitors for the detection of metals, sulphur, nitrogen, fluoride, radionuclides, metabolites originated from organochlorine pesticides, and a variety of organic compounds like those aforementioned. The most commonly used organisms for air pollution and atmospheric deposition assessment, conducted in natural and forested ecosystems or in urban environments, were lichens, mosses, pine needles, and plants. Positive correlations were observed between metals in air particulates and transplanted lichens and between wet deposition and metal concentrations in mosses. Moreover, it was demonstrated that PAH concentrations in lichens were comparable with concentrations measured in a conventional active sampler in an outdoor environment. In conclusion, the importance of atmospheric deposition is growing more and more within the scientific community not only for the possibility it offers to analyze the biogeochemical cycle of elements, at both local and global scale, but also for the assessment of the environmental impact of pollutants on soil and water compartments, following the uptake and the fate, from the sources to the receptors.

Abbreviations

PAHs:	Polycyclic aromatic hydrocarbons
PCBs:	Polychlorobiphenyls
PBDEs:	Polybromodiphenyl ethers
NO _x :	Nitrogen oxides
OH:	Hydroxyl radical
PCDDs/Fs:	Polychlorinated dibenzo-p-dioxins and dibenzo furans
EPA:	Environmental Protection Agency
PVC:	Polyvinyl chloride
VCM:	Vinyl chloride monomer
NAP:	Naphthalene
ACE:	Acenaphthene
ACY:	Acenaphthylene
FLO:	Fluorene

PHE:	Phenanthrene
ANT:	Anthracene
FLA:	Fluoranthene
PYR:	Pyrene
BaA:	Benzo[a]anthracene
CHR:	Chrysene
BbF:	Benzo[b]fluoranthene
BkF:	Benzo[k]fluoranthene
BaP:	Benzo[a]pyrene
DahA:	Dibenzo[a,h]anthracene
IcdP:	Indeno[1,2,3-cd]pyrene
BghiP:	Benzo[ghi]perylene
MeOH:	Methanol
Hg(0):	Elemental vapour mercury
Hg(II):	Gaseous divalent mercury
Hg(p):	Particulate phase mercury
THg:	Total mercury
MeHg:	Methyl mercury
PTFE:	Polytetrafluoroethylene
PFA:	Perfluoroalkoxy alkanes
HDPE:	High density polyethylene
POPs:	Persistent organic pollutants
PCDFs:	Polychlorinated dibenzofurans
I-TEQ:	International toxic equivalents
WHO-TEQ:	World Health Organization-Toxic Equivalents
GEb:	Global mercury emission budget.

Conflict of Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

References

- [1] W. H. Schlesinger, *Biogeochemistry: An Analysis of Global Change*, Academic Press, New York, NY, USA, 1997.
- [2] G. J. Dollard, M. H. Unsworth, and M. J. Harve, "Pollutant transfer in upland regions by occult precipitation," *Nature*, vol. 302, no. 5905, pp. 241–242, 1983.
- [3] J.H. Seinfeld and S. N. Pandis, *Atmospheric Chemistry and Physics*, John Wiley & Sons, Hoboken, NJ, USA, 2nd edition, 2006.
- [4] L. Zhang, D. V. Michelangeli, and P. A. Taylor, "Numerical studies of aerosol scavenging by low-level, warm stratiform clouds and precipitation," *Atmospheric Environment*, vol. 38, no. 28, pp. 4653–4665, 2004.
- [5] C. Andronache, "Estimates of sulfate aerosol wet scavenging coefficient for locations in the Eastern United States," *Atmospheric Environment*, vol. 38, no. 6, pp. 795–804, 2004.
- [6] M. J. Zufall and C. I. Davidson, "Dry deposition of particles," in *Atmospheric Particles*, R. M. Harrison and R. van Grieken, Eds., IUPAC Series on Analytical and Physical Chemistry of Environmental Systems 5, pp. 425–473, John Wiley & Sons, Chichester, UK, 1998.
- [7] M. Langner, T. Draheim, and W. Endlicher, "Particulate matter in the urban atmosphere: concentration, distribution, reduction—results of studies in the Berlin metropolitan area,"

- in *Perspective in Urban Ecology, Ecosystems and Interactions Between Humans and Nature in the Metropolis of Berlin*, W. Endlicher, Ed., pp. 15–41, Springer, Berlin, Germany, 2011.
- [8] M. D. Loye-Pilot and J. M. Martin, *The Impact of Desert Dust Across the Mediterranean*, Edited by S. Guerzoni and S. Chester, Kluwer Academic Publishers, Dordrecht, The Netherlands, 1996.
 - [9] A. Avila and M. Alarcón, “Relationship between precipitation chemistry and meteorological situations at a rural site in NE Spain,” *Atmospheric Environment*, vol. 33, no. 11, pp. 1663–1677, 1999.
 - [10] N. Kubilay, S. Nickovic, C. Moulin, and F. Dulac, “An illustration of the transport and deposition of mineral dust onto the eastern Mediterranean,” *Atmospheric Environment*, vol. 34, no. 8, pp. 1293–1303, 2000.
 - [11] R. Morales-Baquero, E. Pulido-Villena, and I. Reche, “Atmospheric inputs of phosphorus and nitrogen to the southwest Mediterranean region: biogeochemical responses of high mountain lakes,” *Limnology and Oceanography*, vol. 51, no. 2, pp. 830–837, 2006.
 - [12] E. Pulido-Villena, I. Reche, and R. Morales-Baquero, “Significance of atmospheric inputs of calcium over the southwestern Mediterranean region: high mountain lakes as tools for detection,” *Global Biogeochemical Cycles*, vol. 20, no. 2, 2006.
 - [13] R. Morales-Baquero, E. Pulido-Villena, and I. Reche, “Chemical signature of saharan dust on dry and wet atmospheric deposition in the south-western mediterranean region,” *Tellus B: Chemical and Physical Meteorology*, vol. 65, no. 1, 2013.
 - [14] R. Balestrini, L. Galli, and G. Tartari, “Wet and dry atmospheric deposition at prealpine and alpine sites in Northern Italy,” *Atmospheric Environment*, vol. 34, no. 9, pp. 1455–1470, 2000.
 - [15] A. Gambaro, M. Radaelli, R. Piazza et al., “Organic micropollutants in wet and dry depositions in the Venice Lagoon,” *Chemosphere*, vol. 76, no. 8, pp. 1017–1022, 2009.
 - [16] Y.-L. Wu, L.-F. Lin, L.-T. Hsieh, L.-C. Wang, and G.-P. Chang-Chien, “Atmospheric dry deposition of polychlorinated dibenzo-p-dioxins and dibenzofurans in the vicinity of municipal solid waste incinerators,” *Journal of Hazardous Materials*, vol. 162, no. 1, pp. 521–529, 2009.
 - [17] M. Sakata, Y. Tani, and T. Takagi, “Wet and dry deposition fluxes of trace elements in Tokyo Bay,” *Atmospheric Environment*, vol. 42, no. 23, pp. 5913–5922, 2008.
 - [18] W. Ruijgrok, C. I. Davidson, and K. W. Nicholson, “Dry deposition of particles: implications and recommendations for mapping of deposition over Europe,” *Tellus B*, vol. 47, no. 5, pp. 587–601, 1995.
 - [19] M. S. Landis and G. J. Keeler, “Atmospheric mercury deposition to Lake Michigan during the Lake Michigan Mass Balance Study,” *Environmental Science and Technology*, vol. 36, no. 21, pp. 4518–4524, 2002.
 - [20] K. R. Rolffhus, H. E. Sakamoto, L. B. Cleckner et al., “Distribution and fluxes of total and methylmercury in Lake Superior,” *Environmental Science and Technology*, vol. 37, no. 5, pp. 865–872, 2003.
 - [21] R. M. Cavalcante, F. W. Sousa, R. F. Nascimento, E. R. Silveira, and R. B. Viana, “Influence of urban activities on polycyclic aromatic hydrocarbons in precipitation: distribution, sources and depositional flux in a developing metropolis, Fortaleza, Brazil,” *Science of the Total Environment*, vol. 414, pp. 287–292, 2012.
 - [22] F. Esen, S. Siddik Cindoruk, and Y. Tasdemir, “Bulk deposition of polycyclic aromatic hydrocarbons (PAHs) in an industrial site of Turkey,” *Environmental Pollution*, vol. 152, no. 2, pp. 461–467, 2008.
 - [23] M. Bergknut, H. Laudon, S. Jansson, A. Larsson, T. Gocht, and K. Wiberg, “Atmospheric deposition, retention, and stream export of dioxins and PCBs in a pristine boreal catchment,” *Environmental Pollution*, vol. 159, no. 6, pp. 1592–1598, 2011.
 - [24] UNI EN 15980, “Air quality—determination of the deposition of benz[a]anthracene, benzo[b]fluoranthene, benzo[j]fluoranthene, benzo[k]fluoranthene, benzo[a]pyrene, dibenz[a,h]anthracene and indeno[1,2,3-cd]pyrene,” 2011.
 - [25] I. Vassura, F. Passarini, L. Ferroni, E. Bernardi, and L. Morselli, “PCDD/Fs atmospheric deposition fluxes and soil contamination close to a municipal solid waste incinerator,” *Chemosphere*, vol. 83, no. 10, pp. 1366–1373, 2011.
 - [26] L.-C. Guo, L.-J. Bao, J.-W. She, and E. Y. Zeng, “Significance of wet deposition to removal of atmospheric particulate matter and polycyclic aromatic hydrocarbons: a case study in Guangzhou, China,” *Atmospheric Environment*, vol. 83, pp. 136–144, 2014.
 - [27] J. Li, H. Cheng, G. Zhang, S. Qi, and X. Li, “Polycyclic aromatic hydrocarbon (PAH) deposition to and exchange at the air-water interface of Luhua, an urban lake in Guangzhou, China,” *Environmental Pollution*, vol. 157, no. 1, pp. 273–279, 2009.
 - [28] T. Gocht, O. Klemm, and P. Grathwohl, “Long-term atmospheric bulk deposition of polycyclic aromatic hydrocarbons (PAHs) in rural areas of Southern Germany,” *Atmospheric Environment*, vol. 41, no. 6, pp. 1315–1327, 2007.
 - [29] M. Fang, S.-D. Choi, S.-Y. Baek, H. Park, and Y.-S. Chang, “Atmospheric bulk deposition of polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs) in the vicinity of an iron and steel making plant,” *Chemosphere*, vol. 84, no. 7, pp. 894–899, 2011.
 - [30] M. F. Hovmand, J. Vikelsøe, and H. V. Andersen, “Atmospheric bulk deposition of dioxin and furans to Danish background areas,” *Atmospheric Environment*, vol. 41, no. 11, pp. 2400–2411, 2007.
 - [31] B. Pekey, D. Karakaş, and S. Ayberk, “Atmospheric deposition of polycyclic aromatic hydrocarbons to Izmit Bay, Turkey,” *Chemosphere*, vol. 67, no. 3, pp. 537–547, 2007.
 - [32] W. Wang, S. L. Massey Simonich, B. Giri et al., “Spatial distribution and seasonal variation of atmospheric bulk deposition of polycyclic aromatic hydrocarbons in Beijing-Tianjin region, North China,” *Environmental Pollution*, vol. 159, no. 1, pp. 287–293, 2011.
 - [33] A. Motelay-Massei, D. Ollivon, B. Garban, K. Tiphagne-Larcher, I. Zimmerlin, and M. Chevreuil, “PAHs in the bulk atmospheric deposition of the Seine river basin: source identification and apportionment by ratios, multivariate statistical techniques and scanning electron microscopy,” *Chemosphere*, vol. 67, no. 2, pp. 312–321, 2007.
 - [34] EPA. Method 1613, “Tetra-through Octa-Chlorinated Dioxins and Furans by Isotope Dilution HRGC/HRMS,” U.S. Environmental Protection Agency, 1994.
 - [35] EPA. Method 1668B, “Chlorinated Biphenyl Congeners in Water, Soil, Sediment, Biosolids, and Tissue by HRGC/HRMS,” 2008.
 - [36] Z. Cao, Y. Yang, J. Lu, and C. Zhang, “Atmospheric particle characterization, distribution, and deposition in Xi’an, Shaanxi Province, Central China,” *Environmental Pollution*, vol. 159, no. 2, pp. 577–584, 2011.
 - [37] S. Guerzoni, G. Rampazzo, E. Molinaroli, and P. Rossini, “Atmospheric bulk deposition to the Lagoon of Venice—part

- II. Source apportionment analysis near the industrial zone of Porto Marghera, Italy,” *Environment International*, vol. 31, no. 7, pp. 975–982, 2005.
- [38] A. Soriano, S. Pallarés, F. Pardo, A. B. Vicente, T. Sanfeliu, and J. Bech, “Deposition of heavy metals from particulate settleable matter in soils of an industrialised area,” *Journal of Geochemical Exploration*, vol. 113, pp. 36–44, 2012.
- [39] R. Tsitouridou and C. Anatolaki, “On the wet and dry deposition of ionic species in the vicinity of coal-fired power plants, Northwestern Greece,” *Atmospheric Research*, vol. 83, no. 1, pp. 93–105, 2007.
- [40] European Standard EN 15841, “Ambient air quality-Standard method for determination of arsenic, cadmium, lead and nickel in atmospheric deposition,” 2009.
- [41] M. Odabasi, A. Muezzinoglu, and A. Bozlaker, “Ambient concentrations and dry deposition fluxes of trace elements in Izmir, Turkey,” *Atmospheric Environment*, vol. 36, no. 38, pp. 5841–5851, 2002.
- [42] T. Endo, H. Yagoh, K. Sato et al., “Regional characteristics of dry deposition of sulfur and nitrogen compounds at EANET sites in Japan from 2003 to 2008,” *Atmospheric Environment*, vol. 45, no. 6, pp. 1259–1267, 2011.
- [43] H.-H. Yang, L.-T. Hsieh, M.-C. Lin, H.-H. Mi, and P.-C. Chen, “Dry deposition of sulfate-containing particulate at the highway intersection, coastal and suburban areas,” *Chemosphere*, vol. 54, no. 3, pp. 369–378, 2004.
- [44] E. K. Bigg, A. Ono, and J. A. Williams, “Chemical tests for individual submicron aerosol particles,” *Atmospheric Environment*, vol. 8, no. 1, pp. 1–13, 1974.
- [45] G. P. Ayers, “An improved thin film sulphate test for submicron particles,” *Atmospheric Environment*, vol. 11, no. 4, pp. 391–395, 1977.
- [46] G. P. Ayers, “On the use of nitron for detection of nitrate in individual aerosol particles,” *Atmospheric Environment*, vol. 12, no. 5, pp. 1227–1230, 1978.
- [47] Y. Mamane and R. G. de Pena, “A quantitative method for the detection of individual submicrometer size sulfate particles,” *Atmospheric Environment*, vol. 12, no. 1–3, pp. 69–82, 1978.
- [48] Y. MaMane and R. F. Pueschel, “A method for the detection of individual nitrate particles,” *Atmospheric Environment*, vol. 14, no. 6, pp. 629–639, 1980.
- [49] T. M. Holsen, K. E. Noll, S.-P. Liu, and W.-J. Lee, “Dry deposition of polychlorinated biphenyls in urban areas,” *Environmental Science and Technology*, vol. 25, no. 6, pp. 1075–1081, 1991.
- [50] H.-S. Tai, J. J. Lin, and K. E. Noll, “Characterization of atmospheric dry deposited particles at urban and non-urban locations,” *Journal of Aerosol Science*, vol. 30, no. 8, pp. 1057–1068, 1999.
- [51] Y. Tasdemir, C. Kural, S. S. Cindoruk, and N. Vardar, “Assessment of trace element concentrations and their estimated dry deposition fluxes in an urban atmosphere,” *Atmospheric Research*, vol. 81, no. 1, pp. 17–35, 2006.
- [52] H.-J. Yun, S.-M. Yi, and Y. P. Kim, “Dry deposition fluxes of ambient particulate heavy metals in a small city, Korea,” *Atmospheric Environment*, vol. 36, no. 35, pp. 5449–5458, 2002.
- [53] L. D. Sabin, J. Hee Lim, M. Teresa Venezia, A. M. Winer, K. C. Schiff, and K. D. Stolzenbach, “Dry deposition and resuspension of particle-associated metals near a freeway in Los Angeles,” *Atmospheric Environment*, vol. 40, no. 39, pp. 7528–7538, 2006.
- [54] Y.-S. Wu, G.-C. Fang, J.-C. Chen et al., “Ambient air particulate dry deposition, concentrations and metallic elements at Taichung Harbor near Taiwan Strait,” *Atmospheric Research*, vol. 79, no. 1, pp. 52–66, 2006.
- [55] G.-C. Fang, Y.-S. Wu, S.-H. Huang, and J.-Y. Rau, “Dry deposition (downward, upward) concentration study of particulates and heavy metals during daytime, nighttime period at the traffic sampling site of Sha-Lu, Taiwan,” *Chemosphere*, vol. 56, no. 6, pp. 509–518, 2004.
- [56] G.-C. Fang, Y.-S. Wu, W.-J. Lee, T.-Y. Chou, and I.-C. Lin, “Ambient air particulates, metallic elements, dry deposition and concentrations at Taichung Airport, Taiwan,” *Atmospheric Research*, vol. 84, no. 3, pp. 280–289, 2007.
- [57] G.-C. Fang, Y.-L. Huang, J.-H. Huang, and C.-K. Liu, “Dry deposition of Mn, Zn, Cr, Cu and Pb in particles of sizes of 3 m, 5.6 m and 10 m in central Taiwan,” *Journal of Hazardous Materials*, vol. 203–204, pp. 158–168, 2012.
- [58] C. I. Davidson, S. E. Lindberg, J. A. Schmidt, L. G. Cartwright, and L. R. Landis, “Dry deposition of sulfate onto surrogate surfaces,” *Journal of Geophysical Research*, vol. 90, no. 1, pp. 2123–2130, 1985.
- [59] A. Muezzinoglu and S. C. Cizmecioglu, “Deposition of heavy metals in a Mediterranean climate area,” *Atmospheric Research*, vol. 81, no. 1, pp. 1–16, 2006.
- [60] U. M. Shahin, X. Zhu, and T. M. Holsen, “Dry deposition of reduced and reactive nitrogen: a surrogate surfaces approach,” *Environmental Science and Technology*, vol. 33, no. 12, pp. 2113–2117, 1999.
- [61] M. Odabasi, A. Sofuoglu, N. Vardar, Y. Tasdemir, and T. M. Holsen, “Measurement of dry deposition and air-water exchange of polycyclic aromatic hydrocarbons with the water surface sampler,” *Environmental Science and Technology*, vol. 33, no. 3, pp. 426–434, 1999.
- [62] S.-M. Yi, T. M. Holsen, and K. E. Noll, “Comparison of dry deposition predicted from models and measured with a water surface sampler,” *Environmental Science and Technology*, vol. 31, no. 1, pp. 272–278, 1997.
- [63] N. Pirrone, P. Costa, J. M. Pacyna, and R. Ferrara, “Mercury emissions to the atmosphere from natural and anthropogenic sources in the Mediterranean region,” *Atmospheric Environment*, vol. 35, no. 17, pp. 2997–3006, 2001.
- [64] N. Pirrone, J. M. Pacyna, and H. Barth, “Atmospheric mercury research in Europe,” *Atmospheric Environment*, vol. 35, pp. 2997–3006, 2001.
- [65] I. M. Hedgecock, N. Pirrone, G. A. Trunfio, and F. Sprovieri, “Integrated mercury cycling, transport, and air-water exchange (MECAWEx) model,” *Journal of Geophysical Research D: Atmospheres*, vol. 111, no. 20, Article ID D20302, 2006.
- [66] A. P. Dastoor and D. Davignon, “Global mercury modeling at Environment Canada,” in *Mercury Fate and Transport in the Global Atmosphere*, Ch 17, pp. 519–532, Springer, New York, NY, USA, 2009.
- [67] L. Jaeglé, S. A. Strode, N. E. Selin, and D. J. Jacob, “The geochem model,” in *Mercury Fate and Transport in the Global Atmosphere*, Ch 18, pp. 533–545, Springer, New York, NY, USA, 2009.
- [68] G. Jung, I. M. Hedgecock, and N. Pirrone, “The ECHMERT model,” in *Mercury Fate and Transport in the Global Atmosphere*, Ch 19, pp. 547–569, Springer, New York, NY, USA, 2009.
- [69] C. Seigneur, K. Vijayaraghavan, K. Lohman, and L. Levin, “The AER/EPRI global chemical transport model for mercury (CTM-HG),” in *Mercury Fate and Transport in the Global Atmosphere*, Ch 21, pp. 589–602, Springer, New York, NY, USA, 2009.

- [70] O. Travnikov and I. Ilyin, "The EMEP/MSC-E mercury modeling system," in *Mercury Fate and Transport in the Global Atmosphere*, Ch 20, pp. 571–587, Springer, New York, NY, USA, 2009.
- [71] O. R. Bullock and L. Jaeglé, "Importance of a global scale approach to using regional models in the assessment of source-receptor relationships for mercury," in *Mercury Fate and Transport in the Global Atmosphere*, Ch 16, pp. 503–517, Springer, New York, NY, USA, 2009.
- [72] W. H. Schroeder and J. Munthe, "Atmospheric mercury—an overview," *Atmospheric Environment*, vol. 32, no. 5, pp. 809–822, 1998.
- [73] M. S. Landis, A. F. Vette, and G. J. Keeler, "Atmospheric mercury in the Lake Michigan basin: influence of the Chicago/Gary urban area," *Environmental Science and Technology*, vol. 36, no. 21, pp. 4508–4517, 2002.
- [74] C. R. Hammerschmidt and W. F. Fitzgerald, "Methylmercury in freshwater fish linked to atmospheric mercury deposition," *Environmental Science and Technology*, vol. 40, no. 24, pp. 7764–7770, 2006.
- [75] U.S. Environmental Protection Agency (U.S. EPA), "Mercury study report to congress," EPA-452/r-97-003, Office of Air Quality Planning and Standards, Office of Research and Development, Washington, DC, USA, 1997.
- [76] L. E. Gratz, G. J. Keeler, M. Morishita, J. A. Barres, and J. T. Dvonch, "Assessing the emission sources of atmospheric mercury in wet deposition across Illinois," *Science of the Total Environment*, vol. 448, pp. 120–131, 2013.
- [77] National Atmospheric Deposition Program Office, "Monitoring Mercury Deposition. A key tool understanding the link between emissions and effects," Brochure 2005-01, 2008.
- [78] A. Mudroch, "Lake Ontario sediments in monitoring pollution," *Environmental Monitoring and Assessment*, vol. 28, no. 2, pp. 117–129, 1993.
- [79] D. R. Engstrom, E. B. Swain, and M. E. Brigham, "Abstracts with Program, International Conference on Mercury as a Global Pollutant," British Columbia, 1994.
- [80] Legislative Decree no. 155, "Attuazione della direttiva 2008/50/CE relative alla qualità dell'aria più pulita in Europa," Official Gazette of the Republic, 216, Supplement 217, 2010.
- [81] J. Huang, S. Kang, Q. Zhang et al., "Wet deposition of mercury at a remote site in the Tibetan Plateau: concentrations, speciation, and fluxes," *Atmospheric Environment*, vol. 62, pp. 540–550, 2012.
- [82] Norwegian Institute for Air Research, "EMEP manual for sampling and chemical analysis," EMEP/CCC-Report 1/95, 2001.
- [83] OSPAR Commission, "JAMP Guidelines for the Sampling and Analysis of Mercury in Air and Precipitation," Monitoring guidelines, Ref. No Agreement 1997-8, 1997.
- [84] M. Horvat, "Mercury analysis and speciation in environmental samples," in *Global and Regional Mercury Cycles. Sources, Fluxes and Mass Balances*, vol. 21 of NATO-ASI-Series 2, *Environment*, pp. 1–31, 1996.
- [85] K. Kvietkus, Z. Xiao, and O. Lindqvist, "Denuder-based techniques for sampling, separation and analysis of gaseous and particulate mercury in air," *Water, Air, and Soil Pollution*, vol. 80, no. 1–4, pp. 1209–1216, 1995.
- [86] J. Munthe, *The Redox Cycling of Mercury in the Atmosphere [Ph.D. thesis]*, Department of Inorganic Chemistry, University of Göteborg, 1991.
- [87] Z. F. Xiao, J. Munthe, and O. Lindqvist, "Sampling and determination of gaseous and particulate mercury in the atmosphere using cold-coated denuders," *Water, Air, and Soil Pollution*, vol. 56, pp. 141–151, 1991.
- [88] J. Y. Lu, W. H. Schroeder, T. Berg, J. Munthe, D. Schneeberger, and F. Schaedlich, "A device for sampling and determination of total particulate mercury in ambient air," *Analytical Chemistry*, vol. 70, no. 11, pp. 2403–2408, 1998.
- [89] J. Y. Lu and W. H. Schroeder, "Comparison of conventional filtration and a denuder-based methodology for sampling of particulate-phase mercury in ambient air," *Talanta*, vol. 49, no. 1, pp. 15–24, 1999.
- [90] G. Strubel, V. Rzepka-Glinder, K. H. Grobecker, and K. Jarrar, "Heavy metals in human urinary calculi: determination of chromium, lead, cadmium, nickel and mercury in human urinary calculi by direct solid sampling atomic absorption spectrometry using Zeeman background correction," *Fresenius Journal of Analytical Chemistry*, vol. 337, no. 3, pp. 316–319, 1990.
- [91] H. Morita, H. Tanaka, and S. Shimomura, "Atomic fluorescence spectrometry of mercury: principles and developments," *Spectrochimica Acta B: Atomic Spectroscopy*, vol. 50, no. 1, pp. 69–84, 1995.
- [92] J. C. Yu, B. Zhang, and Y.-K. Lai, "Direct determination of mercury in atmospheric particulate matter by graphite plate filtration-electrothermal atomic absorption spectrometry with Zeeman background correction," *Spectrochimica Acta B: Atomic spectroscopy*, vol. 55, no. 4, pp. 395–402, 2000.
- [93] K. Szczepaniak and M. Biziuk, "Aspects of the biomonitoring studies using mosses and lichens as indicators of metal pollution," *Environmental Research*, vol. 93, no. 3, pp. 221–230, 2003.
- [94] B. Wolterbeek, "Biomonitoring of trace element air pollution: principles, possibilities and perspectives," *Environmental Pollution*, vol. 120, no. 1, pp. 11–21, 2002.
- [95] J. A. Fernández and A. Carballeira, "Differences in the responses of native and transplanted mosses to atmospheric pollution: a possible role of selenium," *Environmental Pollution*, vol. 110, no. 1, pp. 73–78, 2000.
- [96] D. H. Brown and R. M. Brown, "Reproducibility of sampling for element analysis using bryophytes," in *Element Concentration Cadasters in Ecosystems*, H. Lieth and B. Markert, Eds., pp. 55–62, VCH Publishers, Weinheim, Germany, 1990.
- [97] S. Augusto, C. Máguas, and C. Branquinho, "Guidelines for biomonitoring persistent organic pollutants (POPs), using lichens and aquatic mosses—a review," *Environmental Research*, vol. 118, pp. 330–338, 2013.
- [98] J. Garty, "Biomonitoring atmospheric heavy metals with lichens: theory and application," *Critical Reviews in Plant Sciences*, vol. 20, no. 4, pp. 309–371, 2001.
- [99] J. E. Sloof, *Environmental lichenology: biomonitoring trace-element air pollution [Ph.D. thesis]*, Delft University of Technology, Interfaculty Reactor Institute, Delft, The Netherlands, 1993.
- [100] J. Garty, "Lichens as biomonitors for heavy metal pollution," in *Plants As Biomonitors: Indicators for Heavy Metals in the Terrestrial Environment*, B. Markert, Ed., pp. 193–263, VCH, Weinheim, Germany, 1993.
- [101] D. H. S. Richardson, *Pollution Monitoring with Lichens*, Richmond, Slough, UK, 1992.
- [102] M. E. Conti and G. Cecchetti, "Biological monitoring: lichens as bioindicators of air pollution assessment—a review," *Environmental Pollution*, vol. 114, no. 3, pp. 471–492, 2001.

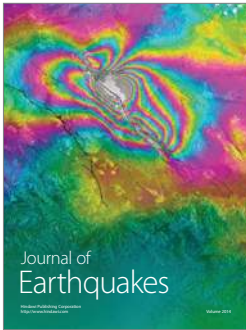
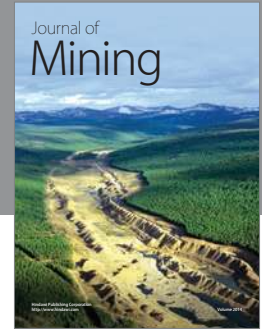
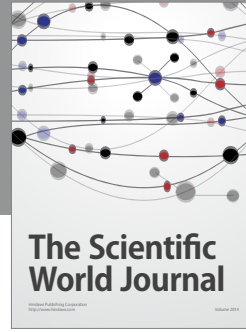
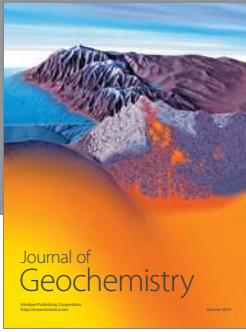
- [103] D. Čeburnis and D. Valiulis, "Investigation of absolute metal uptake efficiency from precipitation in moss," *Science of the Total Environment*, vol. 226, no. 2-3, pp. 247–253, 1999.
- [104] S. Roy, C. K. Sen, and O. Hänninen, "Monitoring of polycyclic aromatic hydrocarbons using "moss bags": bioaccumulation and responses of antioxidant enzymes in *Fontinalis antipyretica* Hedw," *Chemosphere*, vol. 32, no. 12, pp. 2305–2315, 1996.
- [105] C. Branquinho, G. Gaio-Oliveira, S. Augusto, P. Pinho, C. Máguas, and O. Correia, "Biomonitoring spatial and temporal impact of atmospheric dust from a cement industry," *Environmental Pollution*, vol. 151, no. 2, pp. 292–299, 2008.
- [106] W. B. Studabaker, S. Krupa, R. K. M. Jayanty, and J. H. Raymer, "Measurement of polynuclear aromatic hydrocarbons (PAHs) in epiphytic lichens for receptor modelling in Athabasca Oil Sands Region (AOSR): a pilot study," in *Developments in Environmental Science*, K. E. Percy, Ed., vol. 11, pp. 391–425, 2012.
- [107] S. Augusto, M. J. Pereira, C. Máguas, A. Soares, and C. Branquinho, "Assessing human exposure to polycyclic aromatic hydrocarbons (PAH) in a petrochemical region utilizing data from environmental biomonitors," *Journal of Toxicology and Environmental Health A*, vol. 75, pp. 819–830, 2012.
- [108] J. E. Schrlau, L. Geiser, K. J. Hageman, D. H. Landers, and S. M. Simonich, "Comparison of lichen, conifer needles, passive air sampling devices, and snowpack as passive sampling media to measure semi-volatile organic compounds in remote atmospheres," *Environmental Science and Technology*, vol. 45, no. 24, pp. 10354–10361, 2011.
- [109] V. Shukla, D. K. Upreti, D. K. Patel, and M. Yunus, "Lichens reveal air PAH fractionation in the Himalaya," *Environmental Chemistry Letters*, vol. 11, no. 1, pp. 19–23, 2013.
- [110] S. Augusto, M. J. Pereira, A. Soares, and C. Branquinho, "The contribution of environmental biomonitoring with lichens to assess human exposure to dioxins," *International Journal of Hygiene and Environmental Health*, vol. 210, no. 3-4, pp. 433–438, 2007.
- [111] S. Augusto, F. Catarino, and C. Branquinho, "Interpreting the dioxin and furan profiles in the lichen *Ramalina canariensis* Steiner for monitoring air pollution," *Science of the Total Environment*, vol. 377, no. 1, pp. 114–123, 2007.
- [112] S. Augusto, C. Máguas, J. Matos, M. J. Pereira, and C. Branquinho, "Lichens as an integrating tool for monitoring PAH atmospheric deposition: a comparison with soil, air and pine needles," *Environmental Pollution*, vol. 158, no. 2, pp. 483–489, 2010.
- [113] O. Malm, M. B. Castro, W. R. Bastos et al., "An assessment of Hg pollution in different goldmining areas, Amazon Brazil," *Science of the Total Environment*, vol. 175, no. 2, pp. 127–140, 1995.
- [114] A. Klos, M. Rajfur, I. Šrámek, and M. Waclawek, "Mercury concentration in lichen, moss and soil samples collected from the forest areas of Praded and Glacensis Euroregions (Poland and Czech Republic)," *Environmental Monitoring and Assessment*, vol. 184, no. 11, pp. 6765–6774, 2012.
- [115] D. Malizia, A. Giuliano, G. Ortaggi, and A. Masotti, "Common plants as alternative analytical tools to monitor heavy metals in soil," *Chemistry Central Journal*, vol. 6, no. 2, article S6, 2012.
- [116] G. Zarazúa-Ortega, J. Poblano-Bata, S. Tejada-Vega et al., "Assessment of spatial variability of heavy metals in metropolitan zone of toluca valley, Mexico, using the biomonitoring technique in mosses and TXRF analysis," *The Scientific World Journal*, vol. 2013, Article ID 426492, 7 pages, 2013.
- [117] E. Derbyshire, X. Meng, and R. A. Kemp, "Provenance, transport and characteristics of modern aeolian dust in western Gansu Province, China, and interpretation of the Quaternary loess record," *Journal of Arid Environments*, vol. 39, no. 3, pp. 497–516, 1998.
- [118] C. R. Lawrence and J. C. Neff, "The contemporary physical and chemical flux of aeolian dust: a synthesis of direct measurements of dust deposition," *Chemical Geology*, vol. 267, no. 1-2, pp. 46–63, 2009.
- [119] R. Zhang, M. Wang, L. Sheng, Y. Kanai, and A. Ohta, "Seasonal characterization of dust days, mass concentration and dry deposition of atmospheric aerosols over Qindao, China," *China Particuology*, vol. 2, no. 5, pp. 196–199, 2004.
- [120] T. L. Pewe, "Desert dust: an overview," *Special Paper of the Geological Society of America*, vol. 186, pp. 1–10, 1981.
- [121] G. H. McTainsh and A. W. Lynch, "Quantitative estimates of the effect of climate change on dust storm activity in Australia during the last glacial maximum," *Geomorphology*, vol. 17, no. 1-3, pp. 263–271, 1996.
- [122] E. Ganor and Y. Mamane, "Transport of Saharan dust across the eastern Mediterranean," *Atmospheric Environment*, vol. 16, no. 3, pp. 581–587, 1982.
- [123] J. M. Prospero, R. T. Nees, and M. Uematsu, "Deposition rate of particulate and dissolved aluminum derived from Saharan dust in precipitation at Miami, Florida," *Journal of Geophysical Research-Atmospheres*, vol. 92, no. 12, pp. 14723–14731, 1987.
- [124] M. Uematsu, Z. Wang, and I. Uno, "Atmospheric input of mineral dust to the western North Pacific region based on direct measurements and a regional chemical transport model," *Geophysical Research Letters*, vol. 30, no. 6, article 1342, 2003.
- [125] S. P. Wu, S. Tao, F. L. Xu et al., "Polycyclic aromatic hydrocarbons in dustfall in Tianjin, China," *Science of the Total Environment*, vol. 345, no. 1-3, pp. 115–126, 2005.
- [126] C. Monn, O. Braenli, G. Schaeppi, C. Schindler, U. Ackermann-Liebrich, and P. Leuenberger, "Sapaldian team, particulate matter b10 μm (PM10) and total suspended particulate (TPS) in urban rural and alpine air in Switzerland," *Atmospheric Environment*, vol. 29, pp. 2565–2573, 1995.
- [127] X. Querol, A. Alastuey, T. Moreno et al., "Spatial and temporal variations in airborne particulate matter (PM10 and PM2.5) across Spain 1999–2005," *Atmospheric Environment*, vol. 42, no. 17, pp. 3964–3979, 2008.
- [128] K. Baker and P. Scheff, "Assessing meteorological variable and process relationships to modeled PM2.5 ammonium nitrate and ammonium sulfate in the central United States," *Journal of Applied Meteorology and Climatology*, vol. 47, no. 9, pp. 2395–2404, 2008.
- [129] O. Hertel, C. Geels, L. M. Frohn et al., "Assessing atmospheric nitrogen deposition to natural and semi-natural ecosystems—experience from Danish studies using the DAMOS," *Atmospheric Environment*, vol. 66, pp. 151–160, 2013.
- [130] U. Im, S. Christodoulaki, K. Violaki et al., "Atmospheric deposition of nitrogen and sulfur over southern Europe with focus on the Mediterranean and the Black Sea," *Atmospheric Environment*, vol. 81, pp. 660–670, 2013.
- [131] S. Zhang, W. Zhang, Y. Shen, K. Wang, L. Hu, and X. Wang, "Dry deposition of atmospheric polycyclic aromatic hydrocarbons (PAHs) in the southeast suburb of Beijing, China," *Atmospheric Research*, vol. 89, no. 1-2, pp. 138–148, 2008.
- [132] Y. Tasdemir and F. Esen, "Dry deposition fluxes and deposition velocities of PAHs at an urban site in Turkey," *Atmospheric Environment*, vol. 41, no. 6, pp. 1288–1301, 2007.

- [133] D. Gladtko, F. Bakker, H. Biaudet et al., "Different collector types for sampling deposition of polycyclic aromatic hydrocarbons—comparison of measurement results and their uncertainty," *Journal of Environmental Monitoring*, vol. 14, no. 8, pp. 2054–2062, 2012.
- [134] S. Y. Bae, S. M. Yi, and Y. P. Kim, "Temporal and spatial variations of the particle size distribution of PAHs and their dry deposition fluxes in Korea," *Atmospheric Environment*, vol. 36, no. 35, pp. 5491–5500, 2002.
- [135] N. Poor, R. Tremblay, H. Kay et al., "Atmospheric concentrations and dry deposition rates of polycyclic aromatic hydrocarbons (PAHs) for Tampa Bay, Florida, USA," *Atmospheric Environment*, vol. 38, no. 35, pp. 6005–6015, 2004.
- [136] C. J. Halsall, P. J. Coleman, and K. C. Jones, "Atmospheric deposition of polychlorinated dibenzo-p-dioxins/dibenzofurans (PCDD/Fs) and polycyclic aromatic hydrocarbons (PAHs) in two UK cities," *Chemosphere*, vol. 35, no. 9, pp. 1919–1931, 1997.
- [137] S. Karthikeyan, J. He, S. Palani, R. Balasubramanian, and D. Burger, "Determination of total nitrogen in atmospheric wet and dry deposition samples," *Talanta*, vol. 77, no. 3, pp. 979–984, 2009.
- [138] B. Garban, H. Blanchoud, A. Motelay-Massei, M. Chevreuil, and D. Ollivon, "Atmospheric bulk deposition of PAHs onto France: trends from urban to remote sites," *Atmospheric Environment*, vol. 36, no. 34, pp. 5395–5403, 2002.
- [139] E. Terzi and C. Samara, "Dry deposition of polycyclic aromatic hydrocarbons in urban and rural sites of Western Greece," *Atmospheric Environment*, vol. 39, no. 34, pp. 6261–6270, 2005.
- [140] Y. Su, F. Wania, T. Harner, and Y. D. Lei, "Deposition of polybrominated diphenyl ethers, polychlorinated biphenyls, and polycyclic aromatic hydrocarbons to a boreal deciduous forest," *Environmental Science and Technology*, vol. 41, no. 2, pp. 534–540, 2007.
- [141] D. Golomb, D. Ryan, N. Eby, J. Underhill, and S. Zemba, "Atmospheric deposition of toxics onto Massachusetts Bay—I. Metals," *Atmospheric Environment*, vol. 31, no. 9, pp. 1349–1359, 1997.
- [142] J. Li, X. Liu, G. Zhang, and X.-D. Li, "Particle deposition fluxes of BDE-209, PAHs, DDTs and chlordane in the Pearl River Delta, South China," *Science of the Total Environment*, vol. 408, no. 17, pp. 3664–3670, 2010.
- [143] J. Castro-Jiménez, S. J. Eisenreich, G. Mariani, H. Skejo, and G. Umlauf, "Monitoring atmospheric levels and deposition of dioxin-like pollutants in sub-alpine Northern Italy," *Atmospheric Environment*, vol. 56, pp. 194–202, 2012.
- [144] P. Bruckmann, E. Hiester, M. Klees, and C. Zetzsch, "Trends of PCDD/F and PCB concentrations and depositions in ambient air in Northwestern Germany," *Chemosphere*, vol. 93, no. 8, pp. 1471–1478, 2013.
- [145] M.-J. Teil, M. Blanchard, and M. Chevreuil, "Atmospheric deposition of organochlorines (PCBs and pesticides) in northern France," *Chemosphere*, vol. 55, no. 4, pp. 501–514, 2004.
- [146] A. Bozlaker, M. Odabasi, and A. Muezzinoglu, "Dry deposition and soil-air gas exchange of polychlorinated biphenyls (PCBs) in an industrial area," *Environmental Pollution*, vol. 156, no. 3, pp. 784–793, 2008.
- [147] H.-H. Mi, Z.-S. Wu, L.-F. Lin et al., "Atmospheric dry deposition of polychlorinated dibenzo-p-dioxins/dibenzofurans (PCDD/Fs) and polychlorinated biphenyls (PCBs) in southern Taiwan," *Aerosol and Air Quality Research*, vol. 12, no. 5, pp. 1016–1029, 2012.
- [148] S. S. Cindoruk and Y. Tasdemir, "Deposition of atmospheric particulate PCBs in suburban site of Turkey," *Atmospheric Research*, vol. 85, no. 3-4, pp. 300–309, 2007.
- [149] C. G. Fraga, "Relevance, essentiality and toxicity of trace elements in human health," *Molecular Aspects of Medicine*, vol. 26, no. 4-5, pp. 235–244, 2005.
- [150] E. K. Silbergeld, M. Waalkes, and J. M. Rice, "Lead as a Carcinogen: experimental evidence and mechanism of action," *American Journal of Industrial Medicine*, vol. 38, pp. 316–323, 2000.
- [151] L. Fewtrell, R. Kaufman, and A. Prüss-Üstün, "Lead: assessing the environmental burden of diseases at national and local levels," WHO, Geneva, Switzerland, 2003.
- [152] J. B. Shukla, A. K. Misra, S. Sundar, and R. Naresh, "Effect of rain on removal of a gaseous pollutant and two different particulate matters from the atmosphere of a city," *Mathematical and Computer Modelling*, vol. 48, no. 5-6, pp. 832–844, 2008.
- [153] S. Melaku, V. Morris, D. Raghavan, and C. Hosten, "Seasonal variation of heavy metals in ambient air and precipitation at a single site in Washington, DC, USA," *Environmental Pollution*, vol. 155, no. 1, pp. 88–98, 2008.
- [154] M. Sakata and K. Marumoto, "Dry deposition fluxes and deposition velocities of trace metals in the Tokyo Metropolitan Area measured with a water surface sampler," *Environmental Science and Technology*, vol. 38, no. 7, pp. 2190–2197, 2004.
- [155] L. Morselli, B. Brusori, F. Passarini et al., "Heavy metals monitoring at a Mediterranean natural ecosystem of Central Italy. Trends in different environmental matrixes," *Environment International*, vol. 30, no. 2, pp. 173–181, 2004.
- [156] S.-M. Yi, E.-Y. Lee, and T. M. Holsen, "Dry deposition fluxes and size distributions of heavy metals in Seoul, Korea during yellow-sand events," *Aerosol Science and Technology*, vol. 35, no. 1, pp. 569–576, 2001.
- [157] T. M. Holsen, K. E. Noll, and G. J. Keeler, "Dry deposition and particle size distributions measured during the Lake Michigan Urban Air Toxics Study," *Environmental Science & Technology*, vol. 27, no. 7, pp. 1327–1333, 1993.
- [158] R. D. Paode, S. C. Sofuoglu, J. Sivadechathep, K. E. Noll, T. M. Holsen, and G. J. Keeler, "Deposition fluxes and mass size distributions of Pb, Cu, and Zn measured in southern Lake Michigan during AEOLOS," *Environmental Science and Technology*, vol. 32, no. 11, pp. 1629–1635, 1998.
- [159] A. Motelay-Massei, D. Ollivon, K. Tiphagne, and B. Garban, "Atmospheric bulk deposition of trace metals to the Seine river Basin, France: concentrations, sources and evolution from 1988 to 2001 in Paris," *Water, Air, and Soil Pollution*, vol. 164, no. 1-4, pp. 119–135, 2005.
- [160] C. S. C. Wong, X. D. Li, G. Zhang, S. H. Qi, and X. Z. Peng, "Atmospheric deposition of heavy metals in the Pearl River Delta, China," *Atmospheric Environment*, vol. 37, no. 6, pp. 767–776, 2003.
- [161] C. W. Sweet, A. Weiss, and S. J. Vermette, "Atmospheric deposition of trace metals at three sites near the great lakes," *Water, Air, and Soil Pollution*, vol. 103, no. 1-4, pp. 423–439, 1998.
- [162] J. Injuk, R. V. Grieken, and G. D. Leeuw, "Deposition of atmospheric trace elements into the North Sea: coastal, ship, platform measurements and model predictions," *Atmospheric Environment*, vol. 32, no. 17, pp. 3011–3025, 1998.
- [163] V. Sandroni and C. Migon, "Atmospheric deposition of metallic pollutants over the Ligurian Sea: labile and residual inputs," *Chemosphere*, vol. 47, no. 7, pp. 753–764, 2002.

- [164] W. F. Fitzgerald, R. P. Mason, and G. M. Vandal, "Atmospheric cycling and air-water exchange of mercury over mid-continental lacustrine regions," *Water, Air, and Soil Pollution*, vol. 56, pp. 745–767, 1991.
- [165] Y. Zhu, W. C. Hinds, S. Kim, and C. Sioutas, "Concentration and size distribution of ultrafine particles near a major highway," *Journal of the Air and Waste Management Association*, vol. 52, no. 9, pp. 1032–1042, 2002.
- [166] G. A. Sehmel, "Particle resuspension: a review," *Environment International*, vol. 4, no. 2, pp. 107–127, 1980.
- [167] K. W. Nicholson, J. R. Branson, P. Giess, and R. J. Cannell, "The effects of vehicle activity on particle resuspension," *Journal of Aerosol Science*, vol. 20, no. 8, pp. 1425–1428, 1989.
- [168] J. C. Chow, J. G. Watson, R. T. Egami et al., "Evaluation of regenerative-air vacuum street sweeping on geological contributions to PM₁₀," *Journal of the Air and Waste Management Association*, vol. 40, no. 8, pp. 1134–1142, 1990.
- [169] J. C. Chow and J. G. Watson, "Fugitive emissions add to air pollution," *Environmental Protection*, vol. 3, pp. 26–31, 1992.
- [170] T. B. Councell, K. U. Duckenfield, E. R. Landa, and E. Callender, "Tire-wear particles as a source of zinc to the environment," *Environmental Science and Technology*, vol. 38, no. 15, pp. 4206–4214, 2004.
- [171] C. T. Driscoll, D. Whittall, J. Aber et al., "Nitrogen pollution in the northeastern United States: sources, effects, and management options," *BioScience*, vol. 53, no. 4, pp. 357–374, 2003.
- [172] M. Glasius, M. F. Carlsen, T. S. Hansen, and C. Lohse, "Measurements of nitrogen dioxide on Funen using diffusion tubes," *Atmospheric Environment*, vol. 33, no. 8, pp. 1177–1185, 1999.
- [173] J. Zhou, J. Cui, G. Q. Wang, Y. Q. He, and Y. H. Ma, "Ammonia volatilization in relation to N application rate and climate factors in upland red soil in spring and autumn," *Acta Pedologica Sinica*, vol. 44, no. 3, pp. 499–507, 2007.
- [174] M. Flues, P. Hama, M. J. L. Lemes, E. S. K. Dantas, and A. Fornaro, "Evaluation of the rainwater acidity of a rural region due to a coal-fired power plant in Brazil," *Atmospheric Environment*, vol. 36, no. 14, pp. 2397–2404, 2002.
- [175] I. Hunová, J. Maznová, and P. Kurfürst, "Trends in atmospheric deposition fluxes of sulphur and nitrogen in Czech forests," *Environmental Pollution*, vol. 184, pp. 668–675, 2014.
- [176] C. Anatolaki and R. Tsitouridou, "Atmospheric deposition of nitrogen, sulfur and chloride in Thessaloniki, Greece," *Atmospheric Research*, vol. 85, no. 3–4, pp. 413–428, 2007.
- [177] P. Rossini, S. Guerzoni, E. Molinaroli, G. Rampazzo, A. De Lazzari, and A. Zancanaro, "Atmospheric bulk deposition to the lagoon of Venice—part I. Fluxes of metals, nutrients and organic contaminants," *Environment International*, vol. 31, no. 7, pp. 959–974, 2005.
- [178] J. H. Qi, J. H. Shi, H. W. Gao, and Z. Sun, "Atmospheric dry and wet deposition of nitrogen species and its implication for primary productivity in coastal region of the Yellow Sea, China," *Atmospheric Environment*, vol. 81, pp. 600–608, 2013.
- [179] G. Zhang, J. Zhang, and S. Liu, "Characterization of nutrients in the atmospheric wet and dry deposition observed at the two monitoring sites over Yellow Sea and East China Sea," *Journal of Atmospheric Chemistry*, vol. 57, no. 1, pp. 41–57, 2007.
- [180] J. Zhang, G. S. Zhang, Y. F. Bi, and S. M. Liu, "Nitrogen species in rainwater and aerosols of the Yellow and East China seas: effects of the East Asian monsoon and anthropogenic emissions and relevance for the NW Pacific Ocean," *Global Biogeochemical Cycles*, vol. 25, no. 3, Article ID GB3020, 2011.
- [181] T. Nakamura, K. Matsumoto, and M. Uematsu, "Chemical characteristics of aerosols transported from Asia to the East China Sea: an evaluation of anthropogenic combined nitrogen deposition in autumn," *Atmospheric Environment*, vol. 39, no. 9, pp. 1749–1758, 2005.
- [182] K. Matsumoto, H. Minami, Y. Uyama, and M. Uematsu, "Size partitioning of particulate inorganic nitrogen species between the fine and coarse mode ranges and its implication to their deposition on the surface ocean," *Atmospheric Environment*, vol. 43, no. 28, pp. 4259–4265, 2009.
- [183] Y. Gao, "Atmospheric nitrogen deposition to Barnegat Bay," *Atmospheric Environment*, vol. 36, no. 38, pp. 5783–5794, 2002.
- [184] S. C. Pryor, R. J. Barthelmie, L. L. S. Geernaert, T. Ellermann, and K. D. Perry, "Speciated particle dry deposition to the sea surface: results from ASEPS '97," *Atmospheric Environment*, vol. 33, no. 13, pp. 2045–2058, 1999.
- [185] A. R. Baker, T. Lesworth, C. Adams, T. D. Jickells, and L. Ganzeveld, "Estimation of atmospheric nutrient inputs to the Atlantic ocean from 50 N to 50 S based on large-scale field sampling: fixed nitrogen and dry deposition of phosphorus," *Global Biogeochemical Cycles*, vol. 24, no. 7, Article ID GB3006, 2010.
- [186] R. Atkinson, "Atmospheric chemistry of VOCs and NO_x," *Atmospheric Environment*, vol. 34, no. 12–14, pp. 2063–2101, 2000.
- [187] Q. Zhang, D. G. Streets, K. He et al., "NO_x emission trends for China, 1995–2004: the view from the ground and the view from space," *Journal of Geophysical Research D: Atmospheres*, vol. 112, no. 22, Article ID D22306, 2007.
- [188] C. G. Su, B. Yin, Z. L. Zhu, and Q. R. Shen, "Gaseous loss of nitrogen from fields and wet deposition of atmospheric nitrogen and their environmental effects," *Soils*, vol. 37, no. 2, pp. 113–120, 2005.
- [189] R. Yang, K. Hayashi, B. Zhu, F. Li, and X. Yan, "Atmospheric NH₃ and NO₂ concentration and nitrogen deposition in an agricultural catchment of Eastern China," *Science of the Total Environment*, vol. 408, no. 20, pp. 4624–4632, 2010.
- [190] J. He, R. Balasubramanian, D. F. Burger, K. Hicks, J. C. I. Kuylenstierna, and S. Palani, "Dry and wet atmospheric deposition of nitrogen and phosphorus in Singapore," *Atmospheric Environment*, vol. 45, no. 16, pp. 2760–2768, 2011.
- [191] J. L. Shen, A. H. Tang, X. J. Liu, A. Fangmeier, K. T. W. Goulding, and F. S. Zhang, "High concentrations and dry deposition of reactive nitrogen species at two sites in the North China Plain," *Environmental Pollution*, vol. 157, no. 11, pp. 3106–3113, 2009.
- [192] P. Huang, J. Zhang, A. Zhu, X. Xin, C. Zhang, and D. Ma, "Atmospheric deposition as an important nitrogen load to a typical agroecosystem in the Huang-Huai-Hai Plain—1. Measurement and preliminary results," *Atmospheric Environment*, vol. 45, no. 20, pp. 3400–3405, 2011.
- [193] N. Pirrone, S. Cinnirella, X. Feng et al., "Global mercury emissions to the atmosphere from anthropogenic and natural sources," *Atmospheric Chemistry and Physics*, vol. 10, no. 13, pp. 5951–5964, 2010.
- [194] E. G. Pacyna, J. M. Pacyna, and N. Pirrone, "European emissions of atmospheric mercury from anthropogenic sources in 1995," *Atmospheric Environment*, vol. 35, no. 17, pp. 2987–2996, 2001.
- [195] A. Chalmers, M. A. Nilles, D. P. Krabbenhoft, and E. Prestbo, "Analysis of Mercury Wet-Deposition Data Collected with a Newly Designed Sampler, Boston, Massachusetts Metropolitan Area, 2002–2004," USGS Science for a changing world. Open-File Report, 2005.

- [196] M. Fulkerson, F. N. Nnadi, and L. S. Chasar, "Characterizing dry deposition of mercury in urban runoff," *Water, Air, and Soil Pollution*, vol. 185, no. 1-4, pp. 21-32, 2007.
- [197] S. N. Lyman, M. S. Gustin, E. M. Prestbo, and F. J. Marsiks, "Estimation of dry deposition of atmospheric mercury in Nevada by direct and indirect methods," *Environmental Science and Technology*, vol. 41, no. 6, pp. 1970-1976, 2007.
- [198] J. A. Graydon, V. L. St. Louis, H. Hintelmann et al., "Long-term wet and dry deposition of total and methyl mercury in the remote boreal ecoregion of Canada," *Environmental Science and Technology*, vol. 42, no. 22, pp. 8345-8351, 2008.
- [199] L. Zhang, L. P. Wright, and P. Blanchard, "A review of current knowledge concerning dry deposition of atmospheric mercury," *Atmospheric Environment*, vol. 43, no. 37, pp. 5853-5864, 2009.
- [200] S.-O. Lai, J. Huang, P. K. Hopke, and T. M. Holsen, "An evaluation of direct measurement techniques for mercury dry deposition," *Science of the Total Environment*, vol. 409, no. 7, pp. 1320-1327, 2011.
- [201] X. Fu, X. Feng, J. Sommar, and S. Wang, "A review of studies on atmospheric mercury in China," *Science of the Total Environment*, vol. 421-422, pp. 73-81, 2012.
- [202] Y. Wu, S. Wang, D. G. Streets, J. Hao, M. Chan, and J. Jiang, "Trends in anthropogenic mercury emissions in China from 1995 to 2003," *Environmental Science and Technology*, vol. 40, no. 17, pp. 5312-5318, 2006.
- [203] N. L. Rose, J. Munthe, and A. McCartney, "Winter peaks of methylmercury in deposition to a remote Scottish mountain lake," *Chemosphere*, vol. 90, no. 2, pp. 805-811, 2013.
- [204] P. E. Drevnick, H. Yang, C. H. Lamborg, and N. L. Rose, "Net atmospheric mercury deposition to Svalbard: estimates from lacustrine sediments," *Atmospheric Environment*, vol. 59, pp. 509-513, 2012.
- [205] W. F. Fitzgerald, D. R. Engstrom, C. H. Lamborg, C.-M. Tseng, P. H. Balcom, and C. R. Hammerschmidt, "Modern and historic atmospheric mercury fluxes in northern Alaska: global sources and arctic depletion," *Environmental Science and Technology*, vol. 39, no. 2, pp. 557-568, 2005.
- [206] S. Jiang, X. Liu, and Q. Chen, "Distribution of total mercury and methylmercury in lake sediments in Arctic Ny-Ålesund," *Chemosphere*, vol. 83, no. 8, pp. 1108-1116, 2011.
- [207] D. C. G. Muir, X. Wang, F. Yang et al., "Spatial trends and historical deposition of mercury in eastern and northern Canada inferred from lake sediment cores," *Environmental Science and Technology*, vol. 43, no. 13, pp. 4802-4809, 2009.
- [208] H. A. Carreras, E. D. Wannaz, and M. L. Pignata, "Assessment of human health risk related to metals by the use of biomonitors in the province of Córdoba, Argentina," *Environmental Pollution*, vol. 157, no. 1, pp. 117-122, 2009.
- [209] D. Calamari, E. Bacci, S. Focardi, C. Gaggi, M. Morosini, and M. Vighi, "Role of plant biomass in the global environmental partitioning of chlorinated hydrocarbons," *Environmental Science and Technology*, vol. 25, no. 8, pp. 1489-1495, 1991.
- [210] J. Garty, "Environmental and element content in lichens," in *Trace Elements—Their Distribution and Effects in the Environment*, pp. 245-276, Elsevier Science, 2000.
- [211] C. Domeño, M. Blasco, C. Sánchez, and C. Nerín, "A fast extraction technique for extracting polycyclic aromatic hydrocarbons (PAHs) from lichens samples used as biomonitors of air pollution: dynamic sonication versus other methods," *Analytica Chimica Acta*, vol. 569, no. 1-2, pp. 103-112, 2006.
- [212] M. Blasco, C. Domeño, and C. Nerín, "Use of lichens as pollution biomonitors in remote areas: comparison of PAHs extracted from lichens and atmospheric particles sampled in and around the Somport tunnel (pyrenees)," *Environmental Science and Technology*, vol. 40, no. 20, pp. 6384-6391, 2006.
- [213] A. Andersen, M. F. Hovmand, and I. Johnsen, "Atmospheric heavy metal deposition in the Copenhagen area," *Environmental Pollution*, vol. 17, no. 2, pp. 133-151, 1978.
- [214] Z. Jeran, J. Jacimovic, B. Smodis, and F. Batic, "Epiphytic lichens as quantitative biomonitors for atmospheric element deposition," in *Proceedings of the International Workshop on Biomonitoring of Atmospheric Pollution (with Emphasis on Trace Elements) (BioMAP '00)*, pp. 22-28, IAEA, Lisbon, Portugal, September 1997.
- [215] T. Berg and E. Steinnes, "Use of mosses (*Hylocomium splendens* and *Pleurozium schreberi*) as biomonitors of heavy metal deposition: from relative to absolute deposition values," *Environmental Pollution*, vol. 98, no. 1, pp. 61-71, 1997.
- [216] R. Goyal and M. R. D. Seaward, "Metal uptake in terricolous lichens—I. Metal localization within the thallus," *New Phytologist*, vol. 89, no. 4, pp. 631-645, 1981.
- [217] C. M. Prussia and K. T. Killingbeck, "Concentrations of ten elements in two common foliose lichens: leachability, seasonality, and the influence of rock and tree bark substrates," *Bryologist*, vol. 94, no. 2, pp. 135-142, 1991.
- [218] M. de Bruin and E. Hackenitz, "Trace element concentrations in epiphytic lichens and bark substrate," *Environmental Pollution B: Chemical and Physical*, vol. 11, no. 2, pp. 153-160, 1986.
- [219] P. Kuik and H. Wolterbeek Th., "Factor analysis of atmospheric trace-element deposition data in the netherlands obtained by moss monitoring," *Water, Air, and Soil Pollution*, vol. 84, no. 3-4, pp. 323-346, 1995.
- [220] R. Bargagli, F. F. Monaci, F. Borghini, F. F. Bravi, and C. Agnorelli, "Mosses and lichens as biomonitors of trace metals. A comparison study on *Hypnum cupressiforme* and *Parmelia caperata* in a former mining district in Italy," *Environmental Pollution*, vol. 116, no. 2, pp. 279-287, 2002.
- [221] G. Lorenzini, C. Grassi, C. Nali, A. Petiti, S. Loppi, and L. Tognotti, "Leaves of *Pittosporum tobira* as indicators of airborne trace element and PM10 distribution in central Italy," *Atmospheric Environment*, vol. 40, no. 22, pp. 4025-4036, 2006.
- [222] H. Harmens, D. A. Norris, E. Steinnes et al., "Mosses as biomonitors of atmospheric heavy metal deposition: spatial patterns and temporal trends in Europe," *Environmental Pollution*, vol. 158, no. 10, pp. 3144-3156, 2010.
- [223] H.-J. Yun, S.-M. Yi, and Y. P. Kim, "Dry deposition fluxes of ambient particulate heavy metals in a small city, Korea," *Atmospheric Environment*, vol. 36, no. 35, pp. 5449-5458, 2002.
- [224] X. Y. Zhang, R. Arimoto, G. H. Zhu, T. Chen, and G. Y. Zhang, "Concentration, size-distribution and deposition of mineral aerosol over Chinese desert regions," *Tellus B. Chemical and Physical Meteorology*, vol. 50, no. 4, pp. 317-330, 1998.
- [225] S. R. Cattle, G. H. McTainsh, and S. Wagner, "Aeolian dust contributions to soil of the Namoi Valley, Northern NSW, Australia," *Catena*, vol. 47, no. 3, pp. 245-264, 2002.
- [226] C. Kufmann, "Measurement and climatic control of eolian sedimentation on snow cover surface in the northern Calcareous Alps (Wetterstein-Karwendel and Berchtesgadener Alps, Germany)," *Zeitschrift für Geomorphologie*, vol. 50, no. 2, pp. 245-268, 2006.
- [227] C. M. Zdanowicz, G. A. Zielinski, and C. P. Wake, "Characteristics of modern atmospheric dust deposition in snow on the

- Penny Ice Cap, Baffin Island, Arctic Canada,” *Tellus B. Chemical and Physical Meteorology*, vol. 50, no. 5, pp. 506–520, 1998.
- [228] Z. W. Wang, X. S. Zhang, J. S. Xiao, Z. J. Ci, and P. Z. Yu, “Mercury fluxes and pools in three subtropical forested catchments, Southwest China,” *Environmental Pollution*, vol. 157, pp. 801–808, 2008.
- [229] X. Fu, X. Feng, W. Zhu, S. Rothenberg, H. Yao, and H. Zhang, “Elevated atmospheric deposition and dynamics of mercury in a remote upland forest of Southwestern China,” *Environmental Pollution*, vol. 158, pp. 2324–2333, 2010.
- [230] Q. Wan, X. Feng, J. Lu et al., “Atmospheric mercury in Changbai Mountain area, Northeastern China—II. The distribution of reactive gaseous mercury and particulate mercury and mercury deposition fluxes,” *Environmental Research*, vol. 109, pp. 721–727, 2009.
- [231] X. Fu, X. B. Feng, W. Z. Zhu, W. Zheng, S. F. Wang, and J. Y. Lu, “Total particulate and reactive gaseous mercury in ambient air on the eastern slope of the Mt. Gongga area, China,” *Applied Geochemistry*, vol. 23, pp. 408–418, 2008.
- [232] V. L. St. Louis, J. W. M. Rudd, C. A. Kelly, and L. A. Barrie, “Wet deposition of methyl mercury in Northwestern Ontario compared to other geographic locations,” *Water, Air and Soil Pollution*, vol. 80, pp. 405–414, 1995.
- [233] Y. N. Guo, X. B. Feng, Z. G. Li et al., “Distribution and wet deposition fluxes of total and methyl mercury in Wujiang reservoir Basin, Guizhou, China,” *Atmospheric Environment*, vol. 42, pp. 7096–7103, 2008.
- [234] X. Fu, X. Feng, Z. Q. Dong et al., “Atmospheric gaseous elemental mercury (GEM) concentrations and wet and dry deposition of mercury at a high-altitude mountain peak in south China,” *Atmospheric Chemical Physics*, vol. 10, pp. 2425–2437, 2010.



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