

# Mercury in the terrestrial environment: a review

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## Review

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# Abstract

Environmental contamination by mercury is and will continue to be a serious risk for human health. Pollution of the terrestrial environment is particularly important as it is a place of human life and food production. Publication presents a review of the literature on issues related to Hg pollution of the terrestrial environment: soil and plants and their transformations. Different forms of atmospheric Hg may be deposited on surfaces by way of wet and dry processes. These forms may be sequestered within terrestrial compartments or emitted back into the atmosphere, and the relative importance of these processes is dependent on the form of Hg, the surface chemistry, and the environmental conditions. On the land surface, Hg deposition mainly occurs in the oxidized form ( $\text{Hg}^{2+}$ ), and its transformations are associated primarily with the oxidation-reduction potential of the environment and the biological and chemical processes of methylation. The deposition of Hg on ground with low vegetation is 3–5 times lower than that in forests. The estimation of Hg emissions from soil and plants, which occur mainly in the Hg<sup>0</sup> form, is very difficult. Generally, the largest amounts of Hg are emitted from tropical regions, and the lowest levels are from the polar regions.

## Background

Mercury (Hg) is considered to be a peculiar chemical element, because it displays particularly strong chemical and biological activity as well as variability in form (liquid and gaseous). Hg compounds with very different chemical and physical properties are included in various cycles of natural circulation [1–2]. Hg is a globally distributed pollutant due to its characteristics such as low melting and boiling points, conversions between chemical forms and participation in biological cycles. As a result of anthropogenic emissions, the global atmospheric Hg deposition rate is approximately three times higher than in preindustrial times and has increased by a factor of 2–10 in and around the most industrialized regions [3].

## Introduction

Hg-contaminated land environments pose a risk to global public health, with Hg being listed as one of the ‘ten leading chemicals of concern’ [4]. In 2013, the UN (United Nations) introduced the ‘Minamata Convention on Mercury’, which aims for a more global effort for managing the risk presented by Hg to human health and the environment. As of November 2017, the Convention had 84 signatories, and more countries are expected to join in the future. This concerted action, if successful, will have great implications for public health for decades to come; however, there are many hurdles on the way to achieving this goal [5–6].

Our understanding of the critical processes driving global Hg cycling, in particular those that affect large-scale exchange of Hg among major environmental compartments, has advanced substantially over the past decade. This progress has been driven by major advances in three interconnected areas: new data, new models, and new analytical tools and techniques [7].

The advances achieved over the last decade for the assessment of Hg emissions from major man-made and natural sources have contributed to the possibility of improving the assessment of the impacts of atmospheric deposition of Hg on the terrestrial environment [8]. The assessment of Hg emissions poses serious methodological problems. In estimating these impacts, state institutions mainly focus on inventories of their sources, while international organizations apply different models, using emission factors and statistical data of the industrial production and consumption of Hg-containing materials. It is particularly difficult to distinguish natural and anthropogenic emissions from re-emissions from the land and from oceans [9–10]. Hg emission sources include both natural processes unfolding in the biosphere and anthropogenic sources. In 2008, the following classification was adopted in a United Nations Environment Programme (UNEP) report [11], which distinguished three emission sources:

- Current emissions from natural sources;
- Current emissions from anthropogenic sources;
- Re-emissions from past deposits from natural and anthropogenic sources.

Various models are applied to estimate Hg emission levels into the atmosphere, However, the emission levels determined by using these models differ substantially. Travnikov et al. [12] compared the global emission levels from natural and anthropogenic sources determined by using four models [13–16]. The models differed significantly in their estimations of global total emissions from 4,000 Mg yr<sup>-1</sup> to 9,230 Mg yr<sup>-1</sup>, of which natural emissions and re-emission ranged from 45–66% of total emissions. A later summary of the four models [12] of global anthropogenic emissions was estimated at 1,870 Mg yr<sup>-1</sup>, but global natural emission and re-emission were already significantly different from each other from 3,995 Mg yr<sup>-1</sup> [16] to 8,600 Mg yr<sup>-1</sup> [17]. The proportions between the amounts of natural emissions and anthropogenic emissions are not precisely determined. The ratios vary by authors and encompass a relatively wide range of 0.8–1.8 [18–21]. The relatively broad emission ranges presented in various studies are caused by the following: volatility of Hg compounds, distribution of sources, low levels of Hg concentrations in the air, concentrations that are much lower than levels of other basic pollutants and difficulty of determinations [22–23].

Despite the many applicable global, regional and national programs and conventions aimed at reducing Hg emissions, the global THg emissions in the years 2000–2015 increased by 1.8% [6]. Emission reductions only took place in North America and in Europe without the former USSR, and the largest increase was in Central America (+ 5.4%) and South Asia (4.0%). Considering the emissions of Hg by source category, the reductions were in the following fields: dental (-5.6%) and electrical equipment (-5.2%), while the greatest increases were from Hg production (+ 7.9%), cement production (+ 6.3%) and caustic soda production (+ 6.3%). In absolute terms, the largest emissions by far in 2015 came from artisanal and small-scale gold mining (775 Mg yr<sup>-1</sup>) and then from coal combustion (558 Mg yr<sup>-1</sup>).

## Mercury Emissions From Natural Sources

The estimate of Hg emissions from natural sources includes contributions from primary natural sources and from the re-emission processes of historically-deposited Hg over land and sea surfaces. Hg emitted from volcanoes, geothermal sources and topsoil enriched in Hg pertains to primary natural sources, whereas the re-emission of previously deposited Hg on vegetation, land or water surfaces is primarily related to land use changes, biomass burning, meteorological conditions and the exchange mechanisms of gaseous Hg at the air-water/topsoil/snow-ice pack interfaces [8, 24].

A characteristic feature of natural Hg emissions is their distributed nature and wide range of distribution, in comparison to anthropogenic sources. Hg from natural sources is introduced on a global scale into the atmosphere, and Hg from anthropogenic sources is deposited mainly locally and regionally, therefore it is difficult to determine emission levels and to apply effective control methods. Natural Hg emission processes also include re-emission of Hg previously deposited from the atmosphere in the process of wet and dry deposition, coming from both natural and anthropogenic sources. This makes it more difficult to estimate Hg emissions from natural sources [21–22, 25].

Annual global Hg emissions from natural sources on land are estimated by various authors and covers a wide range, e.g., total emissions of 1,600–2,500 Mg yr<sup>-1</sup>, including re-emission of 790–2,000 Mg yr<sup>-1</sup>. According to most authors, re-emissions were higher than primary emissions [19, 26–29].

## Volcanoes

Volcanoes and geothermal activities are important sources of Hg pollution in terrestrial environments. Hg is emitted from volcanoes primarily as gaseous Hg<sup>0</sup> and the Hg/SO<sub>2</sub> ratio is generally adopted to estimate Hg emissions. The annual average of Hg release to the atmosphere without episodic strong eruptions for volcanoes and geothermal activities is ~ 75–112 Mg yr<sup>-1</sup> of Hg, accounting for approximately < 2% of the contribution from natural sources [30–33]. In volcanic plumes, Hg is present both in the gas phase, as elemental Hg<sup>0</sup> and reactive Hg<sup>II</sup>, and in the particle phase, as Hg<sub>p</sub> forms. The proportions of these species are highly variable. Hg<sup>II</sup> and Hg<sub>p</sub> typically amount to < 5% of THg, with Hg<sup>0</sup> as the most abundant form [34–36]. Hg levels in volcanic ash nanoparticles (36 ± 4 mg kg<sup>-1</sup>) are dramatically higher than their bulk concentrations (0.08 mg kg<sup>-1</sup>) [37]. Many areas of geothermal activity have long been associated with elevated levels of Hg in the soil and air in places such as Hawaii, Iceland, western parts of the United States and New Zealand [30, 38].

The average annual global Hg emissions estimate from biomass burning (emissions from wildfires: forests, savannas and grasslands) for 1997–2006 was 675 (± 240) Mg yr<sup>-1</sup>. This accounts for 8% of all current anthropogenic and natural emissions. The largest Hg emissions are from tropical and boreal Asia, followed by Africa and South America [39–40]. The important factor for Hg concentrations in forest soils is the time since stand-replacing fires have occurred, and high soil burn severity has the potential to reduce the concentrations of Hg in burned soils for tens to hundreds of years [41–42].

# Exchange Of Mercury Between The Atmospheric And Terrestrial Ecosystems

The different forms of atmospheric Hg may be deposited on surfaces by way of wet and dry processes. These forms may be sequestered within terrestrial compartments or emitted back to the atmosphere, with the relative importance of these processes being dependent on the form of Hg, the surface chemistry, and environmental conditions. Many models assume that the net GEM exchange with soil surfaces is zero; however, as discussed below, some components are assimilated into foliage over the growing season and accumulate in soils [43].

On the land surface, Hg deposition is mainly in the oxidized form ( $\text{Hg}^{2+}$ ), and its transformations are associated primarily with the oxidation-reduction potential of the environment and with the biological and chemical processes of methylation. For soils in which oxidizing conditions predominate, the  $\text{Hg}^{2+}$  and  $\text{Hg}_2^{2+}$  forms dominate, in soils with reducing conditions, mainly Hg and sulfur compounds are present. Methyl-Hg compounds are most commonly found in soils with transient conditions [1].

For GEM (gaseous elemental mercury  $\text{Hg}^0$ ), the residence time is estimated at 6 to 18 months, while GOM (gaseous mercury in oxidized form) and TPM (total particulate mercury) are quickly removed from the air through wet and dry deposition and their residence times are estimated to be hours or days, at most [29, 44–45]. Given the long time for removal from the air, GEM can be transported over large distances [12]. The particular Hg species are characterized by different dry deposition rates which also determine their residence times. The dry residence times of the different Hg species form the following series [46]:

$$\text{GEM } 0.19 \text{ cm s}^{-1} < \text{TPM } 2:1 \text{ cm s}^{-1} < \text{GOM } 7:6 \text{ cm s}^{-1}$$

According to Marsik et al. [47], the dry deposition rates of GOM and GEM are much higher in daytime than during nighttime. As with Lindberg et al. [46], these authors explain this fact by the closure of plant stomata during nighttime. The deposition rates also depend on the type of surface [48]. Caffrey et al. [49] determined that the deposition rates of particulate air pollutants on the ground with low vegetation were 3–5 times lower than those in forests. Deposition is also affected by the weather conditions, air humidity, insolation and atmospheric precipitation. Research Converse et al., [50] an uncontaminated high-elevation wetland meadow in Shenandoah National Park, Virginia (USA) showed that the highest Hg deposition was observed in the spring ( $4.8 \text{ ng m}^{-2} \text{ h}^{-1}$ ); summer emissions were ( $2.5 \text{ ng m}^{-2} \text{ h}^{-1}$ ); near zero flux took place in the fall ( $0.3 \text{ ng m}^{-2} \text{ h}^{-1}$ ); and winter emissions were ( $4.1 \text{ ng m}^{-2} \text{ h}^{-1}$ ). These studies also suggest that stomatal processes are not the dominant mechanism for ecosystem-level GEM exchange. Table 1 shows a summary of biome-level Hg depositions and soil Hg turnover times [51]. The concentration of Hg in soils is therefore a function of the deposition rate and carbon turnover time. High soil concentrations in desert ecosystems are driven by a combination of higher deposition and extremely slow Hg turnover. Tropical and temperate lifetimes are similar despite the faster carbon turnover in tropical systems due to the relative balance between Hg provided by wet deposition versus leaf uptake.

Table 1  
Summary of Biome Level Hg Deposition and Soil Hg Turnover Times [51].

Biome	Mean Hg deposition ( $\text{g m}^{-2}$ )		Mean soil Hg turnover time* (years)	
	Preindustrial	Present day	Preindustrial	Present day
Tropical forest	0.9	3.7	234	126
Temperate forest	0.8	2.9	250	151
Boreal forest	0.5	1.5	998	560
Grassland	1.0	3.5	522	269
Tundra	0.3	0.8	1108	702
Desert	0.5	1.4	2387	1748
*with respect to respiration.				

Emissions from soils have the form of GEM and depend on many factors [52–56]:

- The properties of soils: Hg content, the contents of organic compounds, and saturation
- The concentrations of oxidants, mainly ozone, in the air
- The weather conditions: solar radiation, temperature, humidity and winds

Soil Hg fluxes are significantly lower in dark conditions than in the light for all but grassland sites [57].

It is most difficult to estimate Hg emissions from plants and these emissions mainly occur in the form of  $\text{Hg}^0$  [58–60]. Ericksen et al. [57] suggested the following hierarchy of environmental parameters influencing Hg flux: soil moisture > light > air concentration > relative humidity > temperature. Table 2 shows a summary of total Hg (THg) fluxes from terrestrial regions, and Table 3 shows average fluxes (or in some cases the range of fluxes) for various ecosystems measured by a number of investigators [24].

Table 2  
Summary of mercury fluxes from terrestrial regions [24]

Region	Evasion (average) (Mg yr <sup>-1</sup> )	Ratio <sup>1</sup> (%)
Forest	342	7,5
Tundra/Grassland/Savannah/Prairie/Chaparral	448	9,9
Desert/Metalliferous/Non-vegetated Zones	546	12,0
Agricultural areas	128	2,8
Total	1664	-
Volcanoes and geothermal areas	90	-
Biomass burning	675	-
<sup>1</sup> calculated over the total evasion from natural sources which sum 4532 Mg yr <sup>-1</sup>		
<sup>2</sup> Friedli [40] and Mason [24] distinguish as a natural source of mercury emission to the atmospheric air of regions where there are episodes of sudden decreases in mercury concentrations in the air by deposition to the ground and then reemission. These phenomena occur mainly in the arctic regions and Antarctica, and the emission from this source is estimated at 200 Mg ye <sup>-1</sup>		

When analyzing the data in Table 3, the following generalizations can be made [24, 27]: (i) the largest amounts of Hg are emitted from tropical regions (45%), followed by emissions from the temperate zones (41%), with the lowest emissions from the polar regions (8%). Emissions from volcanoes and geothermal areas account for 5%; (ii) areas with vegetation can be ranked according to the size of their emissions as follows: forests > other areas (tundra, savannas, and chaparral) > agricultural areas > grassland ecosystems; and (iii) land areas devoid of vegetation emit more Hg than those with plants.

Table 3

Average fluxes, or in some cases the range of fluxes, for various ecosystems measured by a number of investigators.

Species/ecosystem	Flux <sup>1</sup> nmol m <sup>-2</sup> mth <sup>-1</sup>	References.
Ground Level Forest Floor Sweden	1.4–1.7	48, 61
Model Estimates Hard wood forest	Max 4	62
Model Estimates Forest Soil	4	62
Agricultural crops	Max 11	62
Temperate forest	5.0	63
Deforested site	50	64
Desert Soils	3.6–9.8	64
High Hg regions	Max 1500	59
Maple	20	48
Oak	16.4	48
Spruce	6.1	48
Prairie grass	12.5	65
Typha sp.	60	66
Average global soil	1.5	67
<sup>1</sup> Values have all been converted to a common flux unit of nmol m <sup>-2</sup> month <sup>-1</sup> . Results from the older literature are combined in estimates given in various review papers.		

The overall background soil Hg flux in the United States is estimated to be  $0.9 \pm 0.2$  ng/m<sup>2</sup>/h [57], and in areas with significant Hg pollution, soil emissions are much larger. In the canton of Valais, Switzerland, elemental Hg (Hg<sup>0</sup>) is undetectable in soil, but it was determined that substantial Hg<sup>0</sup> emission did occur ( $20\text{--}1,392$  ng.m<sup>-2</sup> h<sup>-1</sup>) [68]. Urban areas are of particular concern with respect to the global Hg cycle due to [69]: (i) frequent higher terrestrial Hg concentrations and the physically and chemically diverse nature of urban surface covers; (ii) highly variable time series concentrations of ambient atmospheric Hg as a result of regional and local emissions; and (iii) urban meteorology (i.e., heat island effect). In the city of Tuscaloosa (Al, USA), Hg fluxes on bare, undisturbed soil surfaces were as follows (median):  $4.45$  ng.m<sup>-2</sup> h<sup>-1</sup> (residential site),  $1.40$  ng.m<sup>-2</sup> h<sup>-1</sup> (industrial site),  $2.14$  ng.m<sup>-2</sup> h<sup>-1</sup> (commercial site), and  $0.87$  ng.m<sup>-2</sup> h<sup>-1</sup> (mixed land use site) (69 Gabriel et al., 2005). Areas of land devoid of vegetation emit more Hg than those with plants. The annual averaged fluxes in the subtropical forest zones in China from

soil in the forests were  $14.2 \text{ ng m}^{-2} \text{ h}^{-1}$ , and for open-air sites they were  $20.7 \text{ ng m}^{-2} \text{ h}^{-1}$  [70]. Soil Hg fluxes were significantly lower in dark conditions than in light conditions. In grassland sites, the mean soil Hg flux was  $0.6 \pm 0.9 \text{ ng.m}^{-2} \text{ h}^{-1}$  in darkness,  $1.0 \pm 0.7 \text{ ng.m}^{-2} \text{ h}^{-1}$  in light, and  $0.9 \pm 0.7 \text{ ng.m}^{-2} \text{ h}^{-1}$  overall [57].

## Methyl Mercury

Methyl-Hg can be both biotically and abiotically produced in the environment. Methylation of Hg tends to occur in environments with low oxygen levels, low pH, Hg bioavailability, temperature, redox potential and high levels of dissolved organic compounds, and in environments favored by sulfate-reducing bacteria that are largely responsible for methylation. These conditions are found primarily in deep sea environments, in coastal marine sediments, and in some freshwater lakes as well as soils. These conditions are also characteristic for paddy soil [71]. Organic Hg is much more toxic to living organisms than inorganic Hg [2, 72–73]. The content of Methyl-Hg in soils and plants is significantly lower than THg but due to its much higher toxicity, is particularly dangerous for living organisms. In soils in the coniferous boreal forests of Sweden, the background level of pollution from methyl-Hg accounted for 0.35–0.59% of THg [74]. In the Idrija Hg-mining area of Slovenia, a heavily polluted region, methyl-Hg accounted for 0.003% of THg, and its background presence in controls was 0.17% [75]. The content of methyl-Hg relative to THg in the roots of rice in background conditions was 1.9%, and in the leaves it was 0.55%; in areas of Hg mining sites, the concentrations in roots were 0.07% and in the leaves it was 0.01%; and in areas with artisanal Hg mining sites, the concentrations were 0.63% and 0.02%, respectively [76]. Methyl-Hg in heavily contaminated soil in the Rhône valley (Switzerland) accounted for < 0.8% of THg [77].

The paddy soils in Hg-mining areas have a high ability of methylation, and may eventually result in heavily biological effects on the local residents through the food chains, such as the rice that contained high methyl-Hg [78]. In the Wuchuan Hg-mining areas, (Guizhou, China), THg in soil samples ranges from 0.33 to 320  $\text{mg kg}^{-1}$  and for methyl-Hg, ranges from 0.69 to 20  $\text{ng g}^{-1}$ . The rice grain samples contain elevated methyl-Hg concentrations, ranging from 4.2 to 18  $\text{ng g}^{-1}$  while corn grain only 0.5–2.0  $\text{ng g}^{-1}$  [71]. Research carried out in areas with coal-fired power plants in Hunan (China) [79] shows that in the soil samples, THg varied from 0.068 to 0.220  $\text{mg kg}^{-1}$ , (mean value of  $0.130 \pm 0.040 \text{ mg kg}^{-1}$ ), and Methyl-Hg ranged from 0.30 to 3.5  $\mu\text{g kg}^{-1}$  (mean  $1.6 \pm 1.0 \mu\text{g kg}^{-1}$ ) In rice samples,  $\text{Hg}^{(\text{II})}$  concentrations varied from 0.002 to 0.022  $\text{mg kg}^{-1}$  (mean  $0.057 \text{ mg kg}^{-1}$ ), and methyl-Hg concentrations varied from 1.7 to 3.8  $\text{ng gg}^{-1}$  (mean of  $2.4 \pm 0.72 \text{ ng g}^{-1}$ ). Meng et al. [76] showed high affinity of rice for Methyl-Hg, and they further showed that in Hg mining sites, rice seeds can contain MeHg concentrations 2 to 3 orders of magnitude higher than the methyl-Hg concentrations in other local edible crop plants.

Forest fires cause a significant reduction in Methyl-Hg content in the soil. Burned soils in Northwestern Ontario (Canada) had 82% less methyl-Hg than fresh soils [80].

## Mercury In Soil

Soil plays an important role in biogeochemical Hg circulation, it accumulates this element, and it is a source for other environmental components. Hg occurs naturally in soils from geologic sources [7] or as the result of natural events such as forest fires and volcanic eruptions [37]. The total amount worldwide of Hg accumulated in the soils of terrestrial environments is estimated at 200–300 Gg [81–82]. Smith-Downey et al. [51] suggest that organically-bound Hg in preindustrial soils is 200 Gg and that there has been a 20% increase in organically-bound soil Hg (to 240 Gg) from preindustrial steady state conditions to the present day.

All results for Hg soil content, presented in the next part of this publication, concern the topsoil layer. Generally, the average background concentration of Hg in soil ranges from 0.03 to 0.1 mg kg<sup>-1</sup> with an average value of 0.06 mg kg<sup>-1</sup> [83–84] Kabata Pendias and Pendias [1] defined a narrower range of 0.05–0.3 mg kg<sup>-1</sup> but some volcanic and organic soils, especially in Canada, may contain higher values, and in the vicinity of industrial emission sources, the values can be extremely high. Obrist et al. [85] showed that a dataset with more than 1,900 randomly selected sampling points across the western USA indicated median Hg concentrations of 0.019 mg kg<sup>-1</sup>, an average value of 24 mg kg<sup>-1</sup> and only 1% of soil samples exceeded background values (e.g., >0.10 mg kg<sup>-1</sup>). The LUCAS Topsoil Survey of the European Union organization collected over 23,000 topsoil samples (upper 20 cm) from land in all European Union countries (28) except for Croatia [86]. The average for European topsoil Hg concentrations was 0.04 mg kg<sup>-1</sup>, with a range of 0–159 mg kg<sup>-1</sup>. Studies have identified highly polluted, isolated sites, and the larger historical and recent industrial and Hg mining areas show elevated concentrations of Hg. Historically, mining for gold and Hg leads to high Hg concentrations in these mining areas. This may be the reason for high Hg concentrations in some samples from Central Italy, northwest England and eastern Slovakia. Most soil Hg was found as soil matrix-bound divalent Hg (Hg<sup>II</sup>), and elemental Hg (Hg<sup>0</sup>) was undetectable in soils [68, 77].

Table 4 presents examples of the concentrations of Hg in the soils in the vicinity of industrial emission sources, according to different authors, The highest Hg contents were found in soils near Hg mines: Idrija in Slovenia with 500 years of mining activity (< 2,759 mg kg<sup>-1</sup>) [75, 90]; and in Alaska (5,326 mg kg<sup>-1</sup>) [87]. Chlor-alkali plants are also an important source of environmental Hg pollution. Bernaus et al. [104] estimated that Hg levels in the soil around a chlor-alkali plant in the Netherlands were as high as 1,150 mg kg<sup>-1</sup>.

Table 4

Some examples of surface soil layer contamination with mercury in regions of important sources of emissions by various authors.

Country	Location	source of pollution	period	Total Hg mg kg <sup>-1</sup>	Mean/ Median	Reference	
USA	Alaska	Hg-mining		0.05–5326		87	
USA	Texas	Hg-mining		3.8–11		88	
China	Wuchuan	Hg-mining	2003	0.33–320		71	
China	Wanshan	Hg-mining	2002	5.1–790		78	
China	Xiaoqinling	gold mining		0.04–61.2	mean 2.75	89	
Slovenia	Idrija	Hg-mining	1991- 97	1734–2759	mean 2456	75	
		Hg-mining	2000- 01	24- 1055	median 47	90	
Slovenia	Podljubelj	Hg-mining	2003- 04	0.35–244	median 3.7	91	
Slovakia	Rudnany	Hg-mining		9.1–54.3		92	
Spain	Almaden	Hg-mining		1340–4830		93	
Spain	Caunedo	old Hg-mining		0.09– 50.0	13.1	mean 13.1	94
Italy	Vallalta	old Hg-mining		6–21		95	
Turkey	Halıköy	Hg mining	2004	0.10–33	mean 5.5	96	
China	Tongguan	Artisanal gold mining		0.69–23.7	mean 2.91	97	
China	Wanshan	Hg-mining, artisanal gold	2012	0.5–187	mean 31.0	98	
Italy	Mt. Amiata,	Hg mining, volcano- geothermal		2.4–68		99	
France	Vosges Mountains	Chlor-alkali	2002	0.16–3.99		100	
France	Grenoble	Chlor-alkali		1.3–10		101	

Country	Location	source of pollution	period	Total Hg mg kg <sup>-1</sup>	Mean/ Median	Reference
Spain	Flix	Chlor-alkali		0.04–12.9	mean 0.77	102
Germany		Chlor-alkali		0.5–4.2	mean 1.6	103
Netherlands		Chlor-alkali	2004	4.3–1150		104
Portugal	Estarreja	Chlor-alkali		0.010– 91	5.4 mean 5.4	105
China	Huludao	Chlor-alkali Zn- smelting	2006- 08	0.05–14.6		106
China	An Ning	Chlor-alkali		0.09–1.30	mean 0.40	107
China	Kunming	Chlor-alkali polyvinyl chloride		0.15–4.79		108
Portugal	Caveira	sulfide mine		1.1–76.5		109
China	Lianyuan	coal main, steel industry	2015	1.20–3601	mean 178	110
China	Zhuzhou	Zn/Pb smelter	2012	0.62–2.61	mean 1.54	111
China	Chongqing	thermometer factory		0.06–0.88		112
Poland	Warsaw	thermometer factory	2005	122–393	mean 147	113
Italy	Etna	volcano		0.1–0.4		114
European Union		23000 samples	2009- 12	0-1.59	mean 0.04	86

In the urban environment, the diversity of Hg content in soils is relatively high. It is related to the diversity of land functions in towns. The lowest median results were determined for soils in Changchun, China 0.018 mg kg<sup>-1</sup> with a range of 0.012–0.036 mg kg<sup>-1</sup> [115] and in Oslo, Norway (0.06 mg kg<sup>-1</sup> with a range of 0.01–2.3 mg kg<sup>-1</sup>) [116], while the highest concentrations were in Palermo, Italy (median value of 1.85 mg kg<sup>-1</sup> with a range of 0.004–2.61 mg kg<sup>-1</sup>) [117] and Glasgow, Scotland (1.2 mg kg<sup>-1</sup> with a range of 0.312–5.2 mg kg<sup>-1</sup>) [118]. Of note is the reduction of 270% (median from 0.68 mg kg<sup>-1</sup> to 0.37) in Hg pollution from 1987–2009 for soils from Beijing, China [119].

In agricultural soils, pollution by Hg was relatively low, which was shown by low values of the medians and averages, usually below  $0.1 \text{ mg kg}^{-1}$  [eg. Scandinavia – 120–122]. However, the ranges of results were relatively wide and the maximum values often exceeded  $1 \text{ mg kg}^{-1}$ , which may indicate a threat to food production due to need to protect human health in some areas with higher Hg soil levels. In Europe, pastures were slightly more polluted with Hg than plowed fields [120]. Soils in forest environments contained low levels of Hg. Average and median values did not exceed  $1 \text{ mg kg}^{-1}$  but in contrast to agricultural soils, the maximum values were also lower than  $1 \text{ mg kg}^{-1}$ . Mineral forest soils contained less Hg than organic ones: USA [42]; Czech Republic [123]; Sweden [124]. According to Obrist et al. [85] soil Hg concentrations significantly differed among land covers, following the order: forested upland > planted/cultivated > herbaceous upland/shrubland > barren soils. Concentrations in forests were an average of 2.5 times higher than those in barren locations.

The global distributions of soil Hg storage and emissions for both preindustrial and present-day simulations in different biomes are shown in Table 1 [51]. The relatively low soil Hg concentrations in boreal and arctic ecosystems are driven by extremely low deposition. The high soil concentrations in desert ecosystems are driven by a combination of higher deposition and extremely slow Hg turnover. The concentration of Hg in soils is therefore a function of the deposition rate and carbon turnover time.

Physical and chemical properties of the soil affect the Hg cycle in the environment. The soil aggregate size fractions have significant effects on the Hg content in soil. The concentrations of Hg and other heavy metals in soils and sediments generally tend to increase with decreasing grain size and this is due to the propensity of metals to bind with finer particles [113]. Generally, higher values of Hg in soil are found in the fraction that is  $< 63 \mu\text{m}$  [125]. In the Amazonian areas without anthropogenic sources, the fine fraction ( $< 53 \mu\text{m}$ ) of podzolized soils had higher Hg contents than clayey soils [126]. In a temperate forest podzol, Hg mean values increased as the aggregate sizes decrease: clay ( $170 \text{ ng g}^{-1}$ ) > fine silt ( $130 \text{ ng g}^{-1}$ ) > coarse silt ( $80 \text{ ng g}^{-1}$ ) > fine sand ( $32 \text{ ng g}^{-1}$ ) > coarse sand ( $14 \text{ ng g}^{-1}$ ). Total Hg enrichment in clay-sized aggregates ranged 2 to 11 times higher than the values shown by the bulk soil ( $< 2 \text{ mm}$ ) [127]. In a heavily polluted area near the Wanshan Hg mine (China), fine soil aggregate size fractions  $< 231 \mu\text{m}$  showed higher total Hg concentrations and higher soil organic matter content than for the larger aggregate size fractions (231 to 2,000  $\mu\text{m}$ ) [128].

Humic acid influences Hg transport and transformation in soil-plant systems, especially for soils having low clay content. Humic acid reduces the amount of available Hg in soil and prevents Hg from being transported into plants or leached from the soil. Leaching can result in Hg leaking into natural water systems under normal environmental conditions. In practice, humic acid can be used to control Hg transportation into food chains from soil heavily polluted by Hg [129].

The chemical and mineralogical properties of soil affect oxidation and retention of atmospheric Hg. Abiotic Hg oxidation occurs because organic matter has -SH groups, which have a high affinity for Hg ions, Hg oxidation is favored in the presence of compounds with high affinities for the Hg ion [130]. A microbial contribution to Hg oxidation was first proposed by Smith et al. [131], who demonstrated that

typical soil bacteria (*Bacillus* and *Streptomyces*) can oxidize elemental Hg to Hg<sup>2+</sup> through enzymatic paths. Recent studies have shown that Hg can also be oxidized by anaerobic bacteria [132–133]. The soil microbial community is very sensitive to Hg concentrations and this sensitivity is influenced not only by soil properties but also by the plant species growing in the soil. A level of 0.36.mg.kg<sup>-1</sup> of Hg in soils is proposed to be a critical concentration above which plant and soil organisms will be affected [134].

## Mercury In Plants

Many studies have recognized the essential role of terrestrial plants in the biogeochemical cycling of Hg [e.g., 135–137]. Approximately 80% of total Hg accumulated in the aboveground biomass is found in the leaves, and approximately 1% of that Hg is methylated. The concentrations of Hg in aspen tissue grown in high-Hg soil increases in the order of stems < branches < petioles < roots < leaves [138]. Leonard's et al. [139] research in Nevada (USA) in an area with high levels of Hg contamination has shown that for the plant species *Lepidium latifolium*, 70% of the Hg taken up by the roots during the growing season was emitted to the atmosphere.

The main source of Hg in leaves comes from air pollution with Hg<sup>0</sup> and not from soil contamination [135, 140–141]. The studies by Fleck et al. [142] of *Pinus resinosa* have shown that neither woody tissue Hg nor any amount of Hg in the soil or forest floor were closely related to foliar levels, and for some relationships, the opposite was true. They interpret those data to indicate that Hg in plant tissues is derived directly from the atmosphere, not from the soil. It is estimated that in highly contaminated soils, generally less than 2% of the Hg present is available for plants [93]. Total leaf concentrations of Hg varied among species and were most closely correlated with the number of stomates per sample, supporting the hypothesis that stomatal uptake of atmospheric Hg (most likely Hg<sup>0</sup>) is a potential uptake pathway [143]. Research by Arnold et al. [144] also indicated the importance of the nonstomatal pathway for uptake of total gaseous Hg (TGM).

The plants growing beyond the influence of high Hg emissions contained less than 100 ng g<sup>-1</sup> THg. Plants growing in the vicinity of factories which are large emitters of Hg, such as Hg mining [eg. 145–146], chlor-alkali [105, 147], Au-mining [148–149] may contain extremely high Hg contents. Mushrooms have been identified as organisms accumulating more Hg than plants [150]. A synthesis of published vegetation Hg data in the western United States showed that above ground biomass concentrations followed the order: leaves (26 µg kg<sup>-1</sup>) ~ branches (26 µg kg<sup>-1</sup>) > bark (16 µg kg<sup>-1</sup>) > bole wood (1 µg kg<sup>-1</sup>) [85]. Hg concentrations in leaves were monitored from emergence to senescence and showed a strong positive correlation with leaf age [141, 143, 151].

## Toxic Effects On Plants

Hg does not have any beneficial effects on organisms and is thus regarded as the “main threat”, since it is very harmful to both plants and animals, pollutes the air, water and soil, and is toxic [152]. Hg in plants is strongly bound to sulfhydryl/thiol groups of proteins and forms SHgS. This biochemical property

probably determines the toxic effects on plants [1, 153–154]. Studies of the toxic effects of Hg on soil organisms and native plants in fields are limited. The effects of Hg are usually examined in sterile and much-simplified laboratory conditions, which may differ from field conditions to varying degrees [155].

The field study of Moreno-Jiménez et al. [145] was conducted in the mining district of Almadén (Spain), which is a cinnabar (HgS) enriched zone, from which one third of the total Hg produced worldwide is extracted. Mining activity began more than 2,000 years ago, and no other region in the world has been influenced by Hg for such a long period. The region is considered to be one of the regions most polluted by Hg in the world. Hg concentrations in the field plants, *Rumex induratus* and *Marrubium vulgare*, grown in these soils can be considered phytotoxic, although no symptoms of Hg toxicity have been observed in any of the studied plant species. In most contaminated soils and mine tailings, Hg is not readily available for plant uptake [156]. The absorption of organic and inorganic Hg from soil by plants is low, and there is a barrier to Hg translocation from plant roots to tops. Thus, large increases in soil Hg levels produce only modest increases in plant Hg levels by direct uptake from soil. [155]. In terrestrial vegetation, Hg in the aboveground biomass originates primarily from the atmosphere whereas Hg in the roots comes from the soil [157–158]. The research of Lomonte et al. [159] suggested the existence of Hg-stress-activated defense mechanisms in plants and hypothesized that these were a probable reason for the increased production of sulfur compounds in the tested plant species, which stimulated their growth. Hg has very limited solubility in soil, low availability for plant uptake and has no known biological function. This may explain why Hg-hyperaccumulating plants have not yet been identified, meaning that there is a lack of a method for Hg phytoremediation in soils contaminated with Hg [159]. However, studies suggesting the use of transgenic plants for phytoremediation have been recently published [160–161].

Research in laboratory conditions predominantly test the Hg doses that may occur in the environment only occasionally indicates the toxicity of various activities of Hg. With a transition property, mercuric ions are able to induce oxidative stress by triggering the generation of reactive oxygen species (ROS), e.g., superoxide anion radicals,  $H_2O_2$ , and hydroxyl radicals (OH.) in plants [162–163]. This process is correlated with disruption of biomembrane lipids and cellular metabolism resulting in plant injury [164]. Increasing levels of mercury species in the soil exert a wide range of adverse effects on the growth and metabolism of plants [152, 165–166], such as reduced photosynthesis, transpiration, water uptake, chlorophyll synthesis [164, 167–168], and increased lipid peroxidation [162]. Higher Hg content in plants affects the activity of most enzymes. The total activity of stress indicators such as superoxide dismutase (SOD), peroxidase (POD) and ascorbate peroxidase (APX) increased after Hg treatment, but the vast majority of enzymes were depressed at higher concentrations [e.g., 169–171].

## Conclusions

- Atmospheric contamination by mercury continues to be one of the most important environmental problems in the modern world. The following general conclusions can be drawn from a review of the literature, with authors' critical commentary:

- Models differ significantly in their estimations of global total Hg emissions from 4,000 Mg yr<sup>-1</sup> to 9,230 Mg yr<sup>-1</sup>, of which natural emissions and re-emission ranged from 45% to 66%.
- Many factors contribute to such large differences in assessing the level of global emissions: (i) methodological difficulties in assessing re-emission from heavily polluted areas under the influence of contemporary and historical emissions and areas with background pollution (ii) the transformation of various forms of Hg depends on many difficult to evaluate processes, which makes estimating emissions difficult, (iii) occurrence of unusual phenomena associated with the transformation of various Hg forms e.g. mercury depletion events (MDEs), consisting of episodes of sudden drops in total gaseous mercury concentrations and the air in the Antarctic and Arctic.
- Despite the many applicable global, regional and national programs and conventions aimed at reducing Hg emissions, global total Hg emissions in the years 2000-2015 increased by 1.8%. In many countries, including those with high national income, there is a lack of understanding by societies and politicians about the need to reduce emissions.
- The proportions between the amounts of natural emissions and anthropogenic emissions are not precisely determined. This ratio, depending on the authors, has been estimated to be over a relatively wide range of 0.8-1.8.
- Annual global Hg emissions from natural sources on land are estimated by various authors to be over a wide range, with total emissions of 1,600-2,500 Mg yr<sup>-1</sup>, including re-emission of 790-2,000 Mg yr<sup>-1</sup>. The low share of Hg taken by plants from soil is beneficial from the point of view of protecting food against contamination, but also limits the possibility of using plants in phytoremediation of contaminated soils.
- On the land surface, Hg deposition is mainly in the oxidized form (Hg<sup>2+</sup>), and its transformations are associated primarily with the oxidation-reduction potential of the environment and with the biological and chemical processes of methylation.
- The main source of Hg in plant leaves comes from air pollution with Hg<sup>0</sup> and not from soil contamination. It is very difficult to estimate Hg emissions from plants, which mainly occur in the form of Hg<sup>0</sup>.
- Methyl-Hg can be produced both biotically and abiotically in the environment. Methylation of Hg tends to occur in environments with low oxygen levels, low pH, Hg bioavailability, temperature, redox potential and high levels of dissolved organic compounds; environments favored by sulfate-reducing bacteria are largely responsible for the methylation.
- Rice growing conditions mean that the Hg methylation process is extremely intensive. Consequently, rice may contain significantly more Hg than other crops. This is particularly dangerous because rice in many regions of the world is the basis for feeding the population.
- Hg has very limited solubility in soil, low availability for plant uptake and does not have any known biological function. This may explain why Hg-hyperaccumulating plants have not yet been identified, meaning that there is a lack of effective phytoremediation methods for soil contaminated with Hg. This may explain why Hg hyperaccumulator plants have not found practical use in phytoremediation

of contaminated soils. It seems that this may be more applicable in wetland environments. The possibility of using contaminated soil in phytoremediation by transgenic plants is promising and it's the future

- A huge number of scientific publications have been devoted to the problem of Hg environmental pollution. However, these studies are facing many difficulties: (i) analytical difficulties caused by very low Hg contents in all elements of the environment, (ii) the need to determine a specific form of Hg, giving the result i total Hg is in most cases insufficient, (iii) the form in which Hg occurs depends on many environmental factors, they must be accurately recognized and described. Unfortunately, many scientific publications do not meet these requirements.

## Abbreviations

Hg – mercury

UN - United Nations

UNEP - United Nations Environment Programme

GEM - gaseous elemental mercury Hg<sup>0</sup>

GOM - gaseous mercury in oxidized form

TPM -total particulate mercury

TGM - total gaseous Hg

ROS - reactive oxygen species

OH - hydroxyl radicals

SOD - superoxide dismutase

POD - peroxidase

APX - ascorbate peroxides

## Declarations

Ethics approval and consent to participate

Not applicable.

Consent for publication

Not applicable.

## Availability of data and materials

Not applicable; presented information is based on previously published data only.

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## Authors' contributions

WD has been responsible for the concept of the manuscript and drafted the manuscript. BG and AH B-D helped to further elaborate the manuscript. All authors improved the final manuscript.

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