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Predominant contributions through lichen and fine litter to litterfall mercury deposition in a subalpine forest

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ABSTRACT

Litterfall, typically referring to needles/leaves, may stand for >50% of the total mercury (Hg) deposition in forest ecosystems. By detailed categorisation, we reveal for the first time that the contributions through lichens and fine litter, together 9.98 μg Hg m^{-2} yr^{-1}, could be as high as that in needle litter (9.96 μg m^{-2} yr^{-1}) to the annual total Hg deposition (44.6 μg m^{-2} yr^{-1}) in a subalpine forest in Switzerland. Noticeably, needle litter had the highest contribution (53%) to total Hg in the autumn litterfall but lichens and fine litter together predominated in other seasons (47–59%). Such a seasonal pattern is caused by the high ability of lichens and fine litter to accumulate Hg and the high needle litterfall in autumn, which is related to a good rainfall in summer followed by a dry period in autumn. The constantly higher Hg levels in lichens and fine litter than in needle litter together with similar seasonal patterns of litterfall during 2009–2019 and rainfall during 1980–2019 suggest that our finding can be generally valid. Here, we highlight not only the considerable role of non-needle litterfall in Hg deposition but also the association with weather for seasonal Hg dynamics in different litterfall components.

1. Introduction

Mercury (Hg) in the environment has drawn large public concerns not only due to its high toxicity but also because of its unique and physicochemical behaviour, which is different from that of the other metal(loid)s (Ariya et al., 2015; Beckers and Rinklebe, 2017). The high predominance and stability of Hg as Hg(0) in the atmosphere benefits its uptake by tree leaves, twig and bark (Assad et al., 2016; Berdonces et al., 2017; Osuna-Vallejo et al., 2019; Sensen and Richardson, 2002; Vannini et al., 2014), leading to the specially high contribution through litterfall, typically >50% of the total Hg deposition in forested ecosystems (Blackwell et al., 2014; Fu et al., 2010; Grigal et al., 2000; Ma et al., 2015; Munthe et al., 1995; Wright et al., 2016). Nevertheless, the contribution of Hg in litterfall to total deposition varies largely among different sites, namely from 29% in a German coniferous forest (Schwesig and Matzner, 2000) to 77% in a Chinese broadleaf forest (Wang et al., 2016). Still today, any potential explanation is lacking. In earlier studies, the litterfall composition has not been well defined and has mostly referred to foliar litter (e.g. Berg and McClaugherty, 2014; Liu et al., 2004). Noticeably, terrestrial Hg emissions deposited back onto land predominantly via Hg uptake in the vegetation (Zhou et al., 2021) and the non-foliar Hg uptake accounts for 51% of the total Hg assimilation by vegetation (Zhou and Obrist, 2021). Navratil et al. (2019) were the first to highlight the potential importance of other litter components than the foliar one for total Hg deposition. In Norway spruce forests (Czech republic), needles are responsible for 40–50% and twigs and fine litter stand each for 20% of annual total Hg in litterfall. Recently, leaves were shown to contribute the most to the annual total Hg in litterfall (73%) in a birch forest in SW Europe, whereas the miscellaneous litter fraction contributed 13–21% and twigs as well as

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reproductive structures <10% (Mendez-Lopez et al., 2023). To date, Hg deposition via non-foliar litterfall is scarcely quantified and we may conclude that the role of non-foliar litter in terrestrial Hg biogeochemical cycling is still ill defined. It is also unknown what environmental factors that may influence such contributions. Here, for the first time, we aim at not only quantifying the contribution of different litterfall components to total Hg in litterfall but also to reveal some factors governing such a distribution over different seasons. Here, we have used the term litter mass for the dead matter shed as litter.

2. Materials and methods

2.1. Site description

The investigation was carried out in the Davos Seehornwald research site in the middle range of the subalpine belt in the eastern part of the Swiss Alps, located at an elevation of 1639 m a.s.l. at 46°48′55.2″ N, 9°51′21.3″ E. Mean annual air temperature is 4.5 °C, and mean annual precipitation is 1020 mm. The coniferous forest is dominated by Norway spruce (*Picea abies* (L.) Karst.) with an average tree height of 18 m and a leaf area index of approximately 3.9 m^2 m^{-2}. The average tree age is 100 years, with some trees being older than 300 years. The predominant lichens in the forest are *Pseudevernia furfuracea* and *Usnea subfloridana*. Dwarf shrubs and mosses are the prominant components of the patchy ground vegetation, covering around 30% of the forest floor. The soil types are Humic Cambisols and Ferric Podzols (WRB classification). The Davos Seehornwald research site is a part of the Integrated Carbon Observation System initiative (ICOS) and the Long-term Forest Ecosystem Research (LWF), i.e. the Swiss contribution to the International Co-operative Programme on Assessment and Monitoring of Air Pollution Effects on Forests (ICP Forests).

2.2. Litterfall sampling and Hg analysis

Twenty circular traps (ICOS defined design, detailed in Fig. S1) were installed 1 m above ground for sampling of the litterfall. Each such trap had a 0.50 m^2 collecting area, with a PVC ring over which a polyester net with a mesh size of 0.25 mm was fixed. From August 2018 to October 2019, the litterfall was collected fortnightly or monthly in the autumns of 2018 and 2019, once between December 2018 and May 2019 and twice in summer 2019. Earlier litterfall measurements (2009–2019) were performed within the framework of the LWF programme, using eight traps of a design similar to that of ICOS. The collected litter was first dried at 65 °C and then sorted according to the ICP Forests Manual (Gielen et al., 2017; Ukonmaanaho et al., 2016), Part XIII sampling and analysis of litterfall as well as LWF Manual (Brang, 1997) into needle, cone and seed, wood (branch wood, twig and bark) as well as lichen and fine litter fractions (Fig. S2). The litter of different fractions intended to the Hg analysis was ground, homogenised and stored in a moisture-proof box at 4 °C before analysis using a DMA-80 direct Hg analyser (MLS GmbH, Leutkirch, Germany). The quality control of the measurements was conducted with certified reference materials (pine needles) obtained from the National Institute of Standards and Technology (NIST, USA) SRM 1575a: 0.0399 ± 0.0007 mg kg^{-1} with a recovery of 101 ± 1%.

3. Results and discussion

3.1. Mercury level in different litterfall fractions

In Seehornwald, the Hg level in different fractions of litterfall followed the order of lichen and fine litter fraction (174–350 ng g^{-1}) ≫ wood (64.5–240 ng g^{-1}) ≫ needle (29.2–87.0 ng g^{-1}) ≥ cone fraction (10.5–37.3 ng g^{-1}) (Table 1), which was similar to the findings of Navratil et al. (2019). The Hg level in different litterfall fractions may reflect, (1) the Hg(0) level of the ambient atmospheric environment

Table 1
Concentrations of total mercury in different litterfall fractions of Norway spruce from August 2018 to October 2019 and the corresponding annual fluxes calculated from September 2018 to September 2019 in Davos Seehornwald, Switzerland.

Compartment	Hg concentration (ng g^{-1}) [a]			Hg flux (μg m^{-2} yr^{-1})[b]	Litter mass flux (g m^{-2} yr^{-1})
	Median	Average	Min – Max		
Needle	42.9	49.9	29.2–87.0	9.96 ± 0.22 (40)	176 (58)
Cone	19.0	20.8	10.5–37.3	1.00 ± 0.03 (4)	41.4 (14)
Wood	144	145	64.5–240	4.08 ± 0.10 (16)	38.7 (13)
Lichen and fine litter	264	258	174–350	9.98 ± 0.14 (40)	45.2 (15)
Total litterfall				25.0 ± 0.28 (100)	301 (100)

a: n = 10; Values within brackets show the percentage each fraction to the total litterfall and mass flux.
b: standard deviation of three randomly selected replicates from the pooled sample are shown.

(Barquero et al., 2019; Kang et al., 2019; Navratil et al., 2019; Vannini et al., 2014), (2) the ability of the plant tissue to take up and accumulate Hg(0) (Assad et al., 2016; Berdonces et al., 2017; Osuna-Vallejo et al., 2019; Sensen and Richardson, 2002; Vannini et al., 2014; Wohlgemuth et al., 2022), (3) the duration of Hg(0) exposure (i.e. retention time of plant tissues) (Navratil et al., 2019; Vannini et al., 2014) and (4) the potential of Hg(0) to be re-emitted from the plant tissues (Fay and Gustin, 2007; Hanson et al., 1997; Nicolardi et al., 2012; Vannini et al., 2014).

Although the detailed mechanisms may be different, Hg accumulation in needle litter, bark and lichens were quite similar, namely predominately being taken up as atmospheric Hg(0) (Bargagli, 2016; Converse et al., 2010; Stamenkovic and Gustin, 2009), which would assumedly be oxidised to Hg(II) within the tissues for subsequent incorporation (Khwaja et al., 2006; Laacouri et al., 2013). Lichens, one of the major components of the lichen and fine litter fraction (distinguishable parts responsible for 24–63% of the lichen and fine litter fraction, Fig. S2d), are capable of accumulating many metal(loid)s to high levels, which remarkably exceed their metabolic needs (Backor and Loppi, 2009; Bargagli, 2016). In a black spruce (*Picea mariana*) forest in Canada, Hg concentrations in lichens (300–1100 ng g^{-1}) were also much higher than in bark (100–200 ng g^{-1}) and needles (<50 ng g^{-1}) (Zhang et al., 1995). Globally, lichens have been confirmed to have Hg concentrations (interquartile range from 10 to 180 ng g^{-1}) exceeding Hg in all other vegetation tissues across unpolluted areas (Zhou et al., 2021). The higher Hg concentrations in lichens could be attributed i) to a longer life span compared to needles and ii) bioaccumulation since lichens are living tissue in contrast to bark (Zhang et al., 1995). While Hg(0) vapour may be re-emitted from leaves and bark into the atmosphere (Fay and Gustin, 2007; Hanson et al., 1997), Hg desorption by evaporation from the lichens' fraction was negligible (Nicolardi et al., 2012; Vannini et al., 2014), which may add to the explanation of extraordinarily high Hg levels in lichens. Our fine litter fraction was a crude mix of small particles consisting of male flowers, fragments of lichens, bark flakes, pollen, fine dust and a small unidentifiable part. Certain components of the fine litter such as fine dust are known to contain high levels of Hg (Coufalik et al., 2014).

A lower concentration of Hg in needle litter than in bark and twigs may result from shorter exposure time of needles to atmospheric inputs of Hg. Biomass turnover rates of Norway spruce litter components based on both modelling and field measurements show ~10 times higher values for needles than branches (Muukkonen and Lehtonen, 2004).

Bark and woody structures are parts of the long-term basic 'structural biomass' of the tree. In contrast, the needles, being 'energy collectors' need to function from the point of view of access to sunlight essentially to be renewed more frequently and although their life spans may vary, a maximum age of 5.9–8.6 years has been suggested (Kayama et al., 2007). Cones make up the only fraction showing a positive and significant correlation between fluxes of Hg and Hg concentrations (r = 0.82, $p < 0.05$) (Fig. S3a). This reflects the annual regeneration of Norway spruce cones and explains also the lowest Hg concentrations among different litterfall fractions.

3.2. Annual mercury fluxes with different fractions of litterfall

The annual throughfall flux of Hg measured in September 2018–September 2019 in Seehornwald was calculated to be 19.6 μg Hg m^{-2} yr^{-1} and the total Hg in litterfall was 25.0 μg m^{-2} yr^{-1} (Chen et al., 2022). Thus, litterfall contributed to 56% of the annual total Hg deposition (44.6 μg m^{-2} yr^{-1}; throughfall + litterfall), similar to the majority of data reported (Blackwell et al., 2014; Fu et al., 2010; Grigal et al., 2000; Ma et al., 2015; Munthe et al., 1995). The annual Hg flux in two litterfall fractions, namely wood (4.08 μg m^{-2} yr^{-1}) and cones (1.0 μg m^{-2} yr^{-1}) were remarkably smaller, standing for 16 and 4% of the total Hg in litterfall. We were surprised to find that the lichen and fine litter fraction of litter had an annual flux of Hg (9.98 μg m^{-2} yr^{-1}), which was as high as that in the needle litter (9.96 μg m^{-2} yr^{-1}, Table 1), each accounting for 40% of the total deposition of Hg in litterfall. According to Navratil et al. (2019) similar fractions, i.e. lichens and the fraction 'other material' together were responsible for only 18–27% of the annual total Hg in litterfall. The Hg flux in litterfall usually correlated positively and significantly with the flux of litter mass but not with the Hg concentration in litterfall (Navratil et al., 2016, 2019). Here, we observed similarly positive and significant correlations between the flux in litter mass and the Hg flux with each litterfall fraction ($r > 0.87$, $p < 0.05$) (Fig. S3b). In comparison, the Hg flux correlated only with the Hg concentration in shed cones ($r = 0.82$, $p < 0.05$) (Fig. S3a). This suggests that the size of the Hg flux in each litterfall fraction was governed by its amount of litter mass rather than the Hg concentrations. However, needle litterfall in Seehornwald (176 g m^{-2} yr^{-1}) was nearly 4 times higher than those of cones, wood as well as the lichen and fine litter fraction (38.7–45.2 g m^{-2} yr^{-1}, Table 1). Therefore, the Hg level in the lichen and fine litter fraction would explain the high contribution of this fraction to total Hg in litterfall. This finding highlights the contribution of the fractions other than needles to Hg deposition with litterfall, especially the lichen and fine litter fraction, which seldom has been considered in the past.

3.3. Seasonal dynamics of mercury fluxes in different litterfall fractions

Both litterfall and Hg fluxes vary with season, which was observed for needles, cones, wood and the lichen and fine litter fractions as well as for total litterfall (Table 2). Both litterfall and Hg fluxes were highest in autumn with 182 g m^{-2} and 15.2 μg m^{-2}, respectively, and were lowered to ~60 g m^{-2} and 4–5 μg m^{-2}, respectively, for the winter-spring and summer periods. Among the different litter fractions, Hg in needle litter underwent the most marked drop, namely from 8.1 μg m^{-2} in the autumn to ~1 μg m^{-2} in other seasons, whereas the reductions in the other fractions were smaller. Thus, in the lichen and fine litter fraction Hg decreased from 4.77 μg m^{-2} in autumn to 3.14 μg m^{-2} in summer. This together with the much higher Hg concentrations in the wood as well as the lichen and fine litter fractions as compared to in the needle litter fraction (Table 1) have led to a noticeable switch of the predominant contribution to Hg fluxes between the needle as well as lichen and fine litter fractions in different seasons. The Hg flux in the needle litter predominated in autumn (53%), the lichen and fine litter fraction became dominant in winter-spring and summer periods with 47 and 59%, respectively. In parallel, the contribution of the wood fraction increased from 11% in autumn to ~24% in winter-spring and summer. Although with a similar seasonal switch in the litterfall fraction predominance, the Hg flux in the leaf litter in a SW-European birch forest prevailed not only in autumn but also in summer and, moreover, with a relevance up to >90% of the total Hg litterfall deposition (Mendez-Lopez et al., 2023). In comparison, twigs and reproductive structures predominated in winter and spring (40–60%) and the miscellanous (lichen- and moss-containing) fraction only in early winter (~60%). Still, the contribution of non-foliar fractions to the annual total Hg litterfall deposition in SW Europe (27%) was not as high as in Davos Seehornwald (60%), apparently due to the similar Hg levels among leaves, twigs and micellanous litters (Mendez-Lopez et al., 2023). These studies together highlight the potential influence of the climate conditions and forest type on the contribution of non-foliar litter Hg as compared to the total Hg in litterfall deposition. This was as reflected in Zhou and Obrist (2021) that the contribution of each plant tissue to total Hg assimulation in the vegetation depends on the type of forest and of plant tissue. Thus, in the future more comprehensive research is indispensable to better define the role of non-foliar litter in different forest ecosystems, to gain more accurate Hg deposition fluxes in litterfall.

Table 2
Seasonal fluxes of litter mass and mercury in needle, cone, wood as well as lichen and fine litter fractions and in total litterfall in Davos Seehornwald, Switzerland.

Compartment	2018 Fall		2018 Winter-2019 Spring		2019 Summer	
	Hg flux (μg m^{-2})	Litter mass flux (g m^{-2})	Hg flux (μg m^{-2})	Litter mass flux (g m^{-2})	Hg flux (μg m^{-2})	Litter mass flux (g m^{-2})
Needle	8.10 (53)	126 (69)	1.12 (25)	26.0 (43)	0.74 (14)	24.0 (41)
Cone	0.65 (4)	29.1 (16)	0.17 (4)	6.15 (10)	0.18 (3)	6.07 (10)
Wood[a]	1.68 (11)	10.1 (6)	1.08 (24)	16.7 (28)	1.31 (24)	11.8 (20)
Lichen and fine litter	4.77 (31)	17.0 (9)	2.07 (47)	11.1 (19)	3.14 (59)	17.1 (29)
Total litterfall	15.2 (100)	182 (100)	4.44 (100)	60.0 (100)	5.38 (100)	59.0 (100)

Values within brackets show the percentage each fraction to the total litterfall and mass flux.
a: Wood fraction includes branch wood, twig and bark.

Although the litter production in Seehornwald may differ among years, the general tendency of the seasonal dynamics among different litterfall fractions was similar to that observed during 2018 and 2019 (Fig. 1). It is normal that trees shed foliar litter at the end of the growing season (autumn) (Laskowski and Berg, 2006). An increased litterfall may be related to the weather pattern with a good rainfall in summer followed by a dry period in autumn in Seehornwald (Fig. S4). The trees adapt to water as a limiting factor and shed part of their needles to keep their water balance (Cromer et al., 1984). As the weather pattern is repeated annually the trees 'know' or 'remember' when to start shedding. The Hg level in different litterfall fractions in the order of lichen and fine litter ≫ wood ≫ needle ≥ cones' fractions could be accepted as generally valid, namely that the same order is not only independent of season (Fig. S5) but has also been found at other sites (Navratil et al., 2019). These observations allow us to hypothesise that a switch of the major contribution to total Hg litterfall deposition between the needle litter as well as lichen and fine litter fractions occurs every autumn and summer in Seehornwald.

In summary, this study highlights the significant contribution of non-needle litterfall, namely that lichens and fine litter can serve as major players in Hg deposition in litterfall in the periods winter-spring and summer. Thus, to ignore the contribution of non-needle litterfall could possibly give rise to large errors when estimating Hg deposition in litterfall in forested ecosystems. Additionally, the high contri-bution of

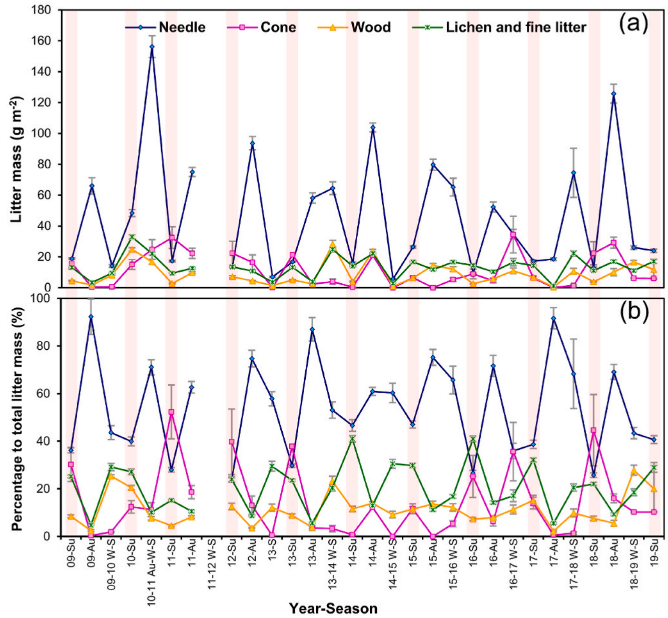

Fig. 1. Seasonal dynamics of (a) litter mass and (b) percentages of different fractions of litter mass in Davos Seehornwald, Switzerland, from June 2009 to October 2019. Su: summer (red shaded areas), Au: autumn, W–S: winter-spring.

lichens and fine litter to total Hg deposition may remind us to include the contribution of epiphytic plants when investigating the atmosphere-forest canopy exchange of Hg. Moreover, we have evidenced that the weather conditions may have a strong influence on the pattern of Hg deposition by different litterfall fractions.

Authorship contribution statement

Jen-How Huang: Funding acquisition, Project administration, Investigation, Methodology, Data processing, Conceptualisation, Writing original draft, Writing – review & editing. Björn Berg: Conceptualisation, Methodology, Writing – review & editing, Chaoyue Chen: Project administration, Writing – review & editing, Anne Thimonier: Resource, Project administration, Maria Schmitt: Resource, Project administration, Stefan Osterwalder: Writing – review & editing, Christine Alewell: Project administration, Writing – review & editing. Jörg Rinklebe: Resources, Writing – review & editing. Xinbin Feng: Supervision, Funding acquisition.

Declaration of competing interest

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

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envres.2023.116005.

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