



Leaf Age Affects Mercury Accumulation in Evergreen Plants

Håkan Pleijel · Jenny Klingberg ·
Henrik Sjöman · Göran Wallin

Received: 18 September 2024 / Accepted: 31 December 2024 / Published online: 18 January 2025
© The Author(s) 2025

Abstract We investigated the mercury (Hg) concentration of the full range of needle age classes (NACs) in two conifers, nine NACs in *Picea abies* and fourteen in *Abies pinsapo* var. *marocana*, as well as three leaf age classes (LACs) in two broadleaved evergreen species, *Trochodendron aralioides* and *Rhododendron catawbiense*. Additionally, the Hg concentration of the wooden branch segments to which the NACs were attached in the two conifers was studied. *Picea abies* showed a continued Hg accumulation over all NACs, but with an age-dependent decline in the accumulation rate. In *Abies pinsapo* var. *marocana*, maximum needle concentrations of Hg were reached after

eight years. The concentration remained constant for NACs 9–14, indicating that needles had become saturated with Hg. The Hg concentrations of the branch segments were much lower than those of the needles in the older NACs. Over the three LACs of *Trochodendron aralioides* and *Rhododendron catawbiense* there was a steady increase in concentration with a weak indication of a declining Hg uptake rate in older leaves. The average needle/leaf lifetime Hg uptake rate per year was only half that of broadleaved species across all NACs and LACs. We conclude that in conifers maintaining a larger number of NACs there is a decline of the Hg accumulation rate in older NACs. In future biogeochemical research (empirical and modelling) and biomonitoring studies, the age of sampled leaves needs to be considered to account for the age dependence of leaf Hg concentration and accumulation rate.

Supplementary Information The online version contains supplementary material available at <https://doi.org/10.1007/s11270-025-07752-2>.

H. Pleijel (✉) · J. Klingberg · G. Wallin
Biological and Environmental Sciences, University
of Gothenburg, P.O. Box 463, 40530 Göteborg, Sweden
e-mail: hakan.pleijel@bioenv.gu.se

J. Klingberg · H. Sjöman
Gothenburg Botanical Garden, Carl Skottsbergs Gata 22A,
41319 Göteborg, Sweden

J. Klingberg
Gothenburg Global Biodiversity Centre, P.O. Box 463,
40530 Gothenburg, Sweden

H. Sjöman
Department of Landscape Architecture, Planning
and Management, Swedish University of Agricultural
Science, 23053 Alnarp, Sweden

Keywords Broadleaved · Conifer · Needle age ·
Leaf age · Hg · Saturation

1 Introduction

Mercury (Hg) is a highly problematic neurotoxic metal (UN Environment, 2019) with large-scale, substantial atmospheric transport (Gworek et al., 2017). Uptake by plants is of critical importance for the biogeochemical cycling of Hg (Obriest et al., 2018). In forest ecosystems, 60–90% of Hg has been estimated

to originate from vegetation uptake (Zhou et al., 2021). Mostly this takes place in the form of gaseous elemental Hg (Hg^0) uptake through stomata to leaves or needles (Laacouri et al., 2013; Risch et al., 2017). Deposition of Hg^0 to the exterior surfaces of leaves seems to be of limited importance (Frescholtz et al., 2003). Vegetation substantially affects the lifetime of Hg in the atmosphere, which typically ranges from approximately 6 to 18 months (Gworek et al., 2017), as well as the seasonal fluctuations in atmospheric concentrations in the Northern Hemisphere (Jiskra et al., 2018). Since stomatal conductance is affected by environmental factors such as vapour pressure deficit and temperature, the Hg uptake by vegetation can be influenced by climate change (Wohlgemuth et al., 2023).

Highly time-resolved isotope techniques to study Hg ecosystem fluxes have shown that the accumulation of Hg in leaves or needles is the net result of bidirectional fluxes to and from vegetation (Bishop et al., 2020; Sommar et al., 2020; Yuan et al., 2019). Over longer time periods (months-years) net uptake was generally observed (Demers et al., 2013). This net accumulation by plants can be investigated by studying the development of Hg concentrations of leaves and needles over their full life span (Pleijel et al., 2021).

Several studies have shown the Hg concentration to increase as deciduous leaves (Assad et al., 2016; Laacouri et al., 2013) and evergreen conifer needles get older (e.g., Hutnik et al., 2014; Navrátil et al., 2019; Wohlgemuth et al., 2020; Pleijel et al., 2021). In some earlier studies, only the youngest 1–3 needle age classes (NACs) were investigated, or all NACs were pooled (e.g. Yang et al., 2018), and in meta-analyses of Hg concentrations in different types of vegetation, the aspect of needle or leaf age was not always analysed (Zhou et al., 2021). Many conifers can retain their needles for several years (Reich et al., 1996, 2014), such as up to 6 years in *Pinus sylvestris*, 12 years in *Picea abies* and 16 years in *Abies balsamea* (Niinemets & Lukjanova, 2003). Navrátil et al. (2019), in an investigation of the effect of a severe bark beetle infestation on litterfall mercury deposition in the Czech Republic, studied four NACs in Norway spruce (*Picea abies*) and observed a continued accumulation during that range of NACs. There is consequently a need for information on Hg accumulation over the full range of NACs in

biogeochemistry, but also in biomonitoring based on needle accumulation of metals such as Hg (Bertolotti & Gialanella, 2014).

There are observations suggesting that the rate of Hg uptake declines as needles get older. For example, calculations by Wohlgemuth et al., (2020, 2022) accounted for a successive reduction in the rate of Hg uptake as *Picea abies* needles became older, based on observations in Bavaria. Pleijel et al. (2021), combining data from several sources for different conifer species, also found an indication of a reduction in the Hg accumulation rate in up to five-year-old needles. A declining Hg uptake rate with increasing needle age can depend on physiological aging with lower concentrations of important nutrients like nitrogen and phosphorus (Manghabati et al., 2019; Oren et al., 1988), but also on older needles being shaded to a larger extent than younger, which will promote stomatal closure (Zimmermann et al., 1988). A further possibility is that the leaf Hg concentration approaches saturation in terms of a dynamic equilibrium where the rate of Hg evasion equals the rate of deposition of Hg as suggested to be the case for semi-volatile polycyclic organic compounds (McLachlan, 1999) and was discussed also for Hg in earlier literature (Ericksen & Gustin, 2004; Hanson et al., 1995).

The development of leaf Hg uptake rate over time is highly relevant also for evergreen broadleaved trees, which have been investigated to a much lesser extent than evergreen conifers and broadleaved deciduous species. A recent exception is the study of the evergreen olive tree (*Olea europaea*) in olive groves in Lebanon (Tabaja et al., 2023), where a continued leaf accumulation of Hg over several years was observed. In many tropical and Mediterranean evergreen broadleaved species, leaf age is hard to assess accurately due to continuous or recurrent leaf flushing over the whole season (Kozłowski, 1964). However, in cooler climates, many evergreen broadleaved species, such as *Trochodendron aralioides*, and many species in the genus *Rhododendron*, carry several leaf age classes (LACs) which can easily be distinguished as distinct annual cohorts.

If the Hg uptake rates observed for the youngest needles are not representative for the needles over their full life span it means that data for young needles, or needles of unspecified age, cannot be used directly to estimate the flux of Hg with litterfall of

needles. Niinemets and Lukjanova (2003) have pointed out that the bias in limiting measurements to, and basing ecophysiological modelling of processes at the ecosystem level on data for the youngest needles, can have strong implications, e.g. in assessments of stand level carbon gain. This concern for a bias resulting from a selective focus on younger needles or leaves, applies also to Hg, since conifers predominantly shed their oldest needles as litter (Muukkonen, 2005; Nebel & Matile, 1992; Wyttenbach & Tobler, 2000). Concentrations of younger needles or a mix of needle age classes are thus not representative for the flux of Hg and other elements with needle litter.

To address the knowledge gap represented by the current lack in the data on Hg concentration, and consideration of the development of Hg accumulation over needle or leaf lifetime, we investigated the Hg concentration of the full range of NACs, and associated wooden branch segments, in two conifers: *Picea abies* (nine NACs) and *Abies pinsapo* ssp. *marocana* (fourteen NACs). The latter species was selected to represent conifers carrying a large number of NACs. Furthermore, the Hg concentration of three LACs was investigated for two evergreen broadleaved species: *Trochodendron aralioides* and *Rhododendron catawbiense*. Using this methodological approach, we add novel information on Hg concentration and accumulation rate in evergreen needles and leaves, promoting the understanding and supporting modelling of a significant aspect of the biogeochemical cycling of Hg in the environment. Our research questions were:

- Do evergreen conifer needles continuously accumulate mercury (Hg) across all needle age classes (NACs), or is there a saturation or even a decline in older NACs?
- What is the concentration of Hg in the wooden branch segments, to which the different NACs in conifers are attached, compared with needle Hg?
- Do leaves of evergreen broadleaved species exhibit an accumulation pattern of Hg similar to that of evergreen conifers?
- How large is the average life-time Hg accumulation rate in conifer needles and leaves of evergreen broadleaved trees when grown under the same ambient conditions?

2 Materials and Methods

2.1 Plant Material

Norway spruce (*Picea abies*) is the most common tree species in Sweden, where the study was conducted. Three *Picea abies* trees in a semi-closed forest stand of the Änggårdssbergen nature reserve next to the Gothenburg Botanical Garden were sampled (N57.66, E11.96). In addition, three trees of Moroccan fir (*Abies pinsapo* var. *marocana*) were sampled in the nearby arboretum situated in the forest landscape next to the botanical garden (N57.67, E11.96). *Abies pinsapo* var. *marocana* was selected because it maintains a broad range of complete NACs, at our site fourteen. The number of complete NACs in *Picea abies* was nine. The Wheel tree (*Trochodendron aralioides*) and the Catawba rhododendron (*Rhododendron catawbiense*) are evergreen broadleaved species. Three individuals of each species were sampled in the Gothenburg Botanical Garden (N 57.68, E11.95). Both species had three complete LACs. The fourth LAC showed senescence and was largely shed in both species. Determination of the age class of needles and leaves in the four species is described in detail in Supplementary Fig. 1.

2.2 Sampling, Preparation and Analysis of Samples

For each sampled *Picea abies* or *Abies pinsapo* var. *marocana* tree, a minimum of three branches were used. Sampling was made on 22 September 2020. For these two conifers, both the needles and the different annual branch segments to which the different NACs were attached were sampled and analysed. In the case of *Trochodendron aralioides* and *Rhododendron catawbiense*, sampling was made on 21 September 2020. For each sampled tree a minimum of three branches were sampled. One sample for the second LAC of *Trochodendron* was lost during the handling of samples.

Branches with leaves or needles in the outer, light-exposed part of the crown were selected and cut down with a pruner. The samples were packed in polyethylene plastic bags, transported in a cool bag and stored in a fridge until further handling. Samples were carefully collected to avoid contamination. Needles were separated from the wooden

branch segments at the laboratory while the leaves of the broadleaved species were separated from the branches in the field. Leaf, needle and branch samples were dried in 70 °C (Pleijel et al., 2021) in aluminium envelopes until constant weight, at least 48 h. Thereafter the samples (1–2 g each) were ground to a fine powder using a ball mill (Model MM 301, Retsch, Haan, Germany) equipped with grinding jars and balls made of wolfram carbide. The grinding equipment was carefully vacuum cleaned between each sample. Samples were analysed for the content of 37 elements including Hg using inductively coupled plasma mass spectrometry (Dry Vegetation ICP-MS, 37 elements after digestion in HNO₃ and then aqua regia: Method VG101 by Bureau Veritas Mineral laboratories, Vancouver, BC, Canada). In this paper, only the Hg data are used. The laboratory has implemented an analytical quality management system meeting the requirements of ISO/IEC 17025 and ISO 9001. Further details on the analysis as well as on Quality Assurance and Quality Control which includes the use of blanks, internal standards and repeated measurements of some samples can be obtained here: https://publications.gc.ca/site/archiv-ee-archived.html?url=https://publications.gc.ca/collections/collection_2024/rncan-nrcan/m183-2/M183-2-8837-1A-eng.pdf. The method detection limit for Hg was 1 ng g⁻¹. No sample had a Hg concentration below the detection limit.

2.3 Statistical Analysis

Differences between species, leaf or needle age, as well as the interaction of species with leaf or needle age, were investigated using a mixed design analysis of variance (ANOVA) with leaf or needle age set as the within-subjects variable (repeated measures). This analysis was made separately for conifers (9 NACs) and broadleaved species (3 LACs). One-way ANOVA was used to analyse differences in Hg lifetime uptake between species with Tukey's honestly significant difference (HSD) as the post-hoc test. IBM SPSS Statistics (version 25) was used for the ANOVA statistical analyses.

3 Results

The needle concentration of Hg initially increased over time in both *Picea abies* and *Abies pinsapo* var. *marocana* (Fig. 1a). While the Hg concentration continued to increase over all nine NACs in *Picea abies*, the concentration increase ceased completely in NACs older than eight years in *Abies pinsapo* var. *marocana* (Fig. 1a). In *Abies* the maximum needle concentrations reached ~110 ng g⁻¹ while the corresponding value for *Picea* was 75 ng g⁻¹. The mixed design ANOVA showed that needle age and the needle age*species interaction both were strongly significant ($p < 0.001$), while the species difference as such was non-significant ($p = 0.14$).

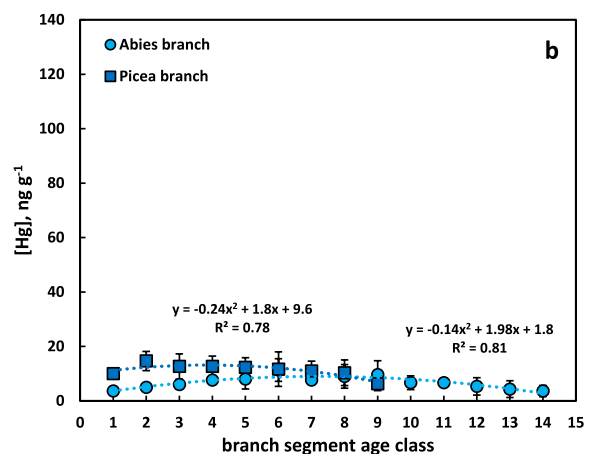
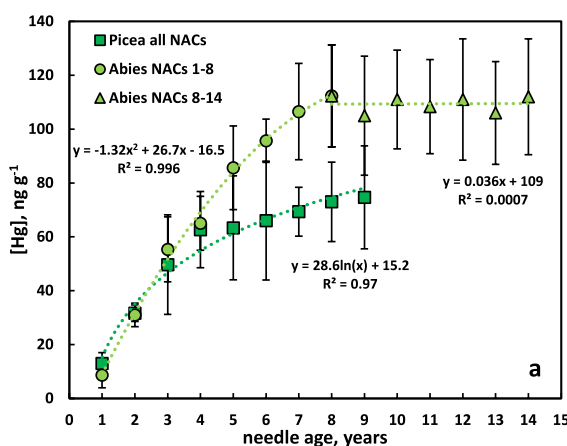


Fig. 1 Development of the Hg concentration, [Hg], of (a) needles, and (b) associated branch segments, of *Picea abies* and *Abies pinsapo* var. *marocana* over the full range of NACs. For

Abies pinsapo var. *marocana* separate curve fittings were used for needle age classes up to 8 and needle age classes above 8

Hg concentrations of the branch segments to which the different NACs were attached (Fig. 1b) were on average 5 and 12.8 times lower than needle concentrations, respectively, in *Picea abies* and *Abies pinsapo* var. *marocana*. Maximum branch concentrations were 13 ng g^{-1} and 11 ng g^{-1} in *Picea abies* and *Abies pinsapo* var. *marocana*, respectively. Only the first NAC had Hg concentrations of the same magnitude as those of the corresponding branch segments. In both species an initial

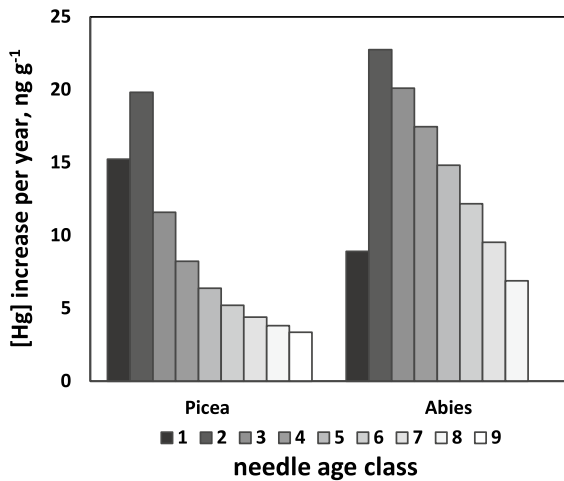
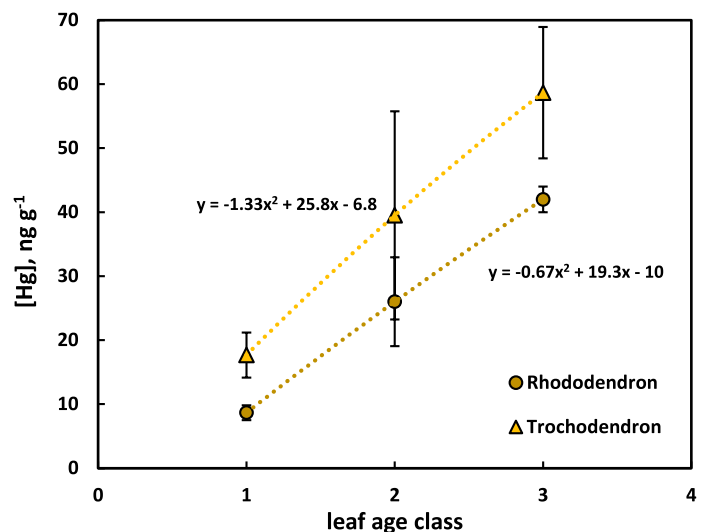


Fig. 2 Estimated mean annual increase in needle Hg concentration, [Hg], for all nine needle age classes (NACs) in *Picea abies* and the first eight NACs (the concentration remained constant in older NACs) of *Abies pinsapo* var. *marocana* based on the curves fitted to the data in Fig. 1a

Fig. 3 Development of the Hg concentration, [Hg], in leaves of *Trochodendron aralioides* and *Rhododendron catawbiense* over three LACs



increase in branch segment Hg concentration followed by a decrease was indicated. However, the mixed design ANOVA was non-significant for the Hg concentrations of branch segments with respect to species, needle age and their interaction.

Based on the regression curves fitted to the observed needle Hg concentrations of the two conifers displayed in Fig. 1a, the annual rate of concentration increase was estimated (Fig. 2). For all except the first NAC a steady decline in annual Hg accumulation rate was obtained. The smaller uptake in the first NAC can be explained by the shorter growing season of the current year needles, being fully developed by mid-June, while older needles start their physiological activity around two months earlier. Also, some autumn Hg uptake after the harvest date (22 September) can be expected for current year needles, which will in this analysis be included in the Hg accumulation of the second NAC.

A steady increase in leaf Hg concentration over the three LACs of *Trochodendron aralioides* and *Rhododendron catawbiense* was observed (Fig. 3). The mixed design ANOVA showed that the age effect was highly significant ($p < 0.001$), while the difference between the species was non-significant ($p = 0.127$). The negative quadratic terms of the regression functions represent an indication that there was some levelling off in the accumulation rate of Hg as leaves age, but this effect was very small in relation the rate of uptake.

Since the flux of elements with leaves and needles dropped as litter will represent averaged net accumulation over their full life span, the average leaf or needle lifetime Hg accumulation per growing season (by dividing the Hg concentration of the oldest LAC or NAC with its age expressed in years) of the different species is compared in Fig. 4. In addition, the average annual net uptake after three years for the conifer species was calculated and shown in the figure for comparison. After three years, the accumulation rate was of similar magnitude in all four species. However, when the full range of LACs (three years in both broadleaved species) and NACs (nine years in *Picea abies* and fourteen years in *Abies pinsapo* var. *marocana*) the average life-time annual Hg accumulation rate was only about half in the two conifer species compared to the broadleaved species. In this case the ANOVA was significant ($p < 0.001$) and the post-hoc test showed that the Hg uptake rate of the two broadleaved species was significantly different from that of the two conifers, while neither the two broadleaved, nor the two conifer species, were significantly different from each other.

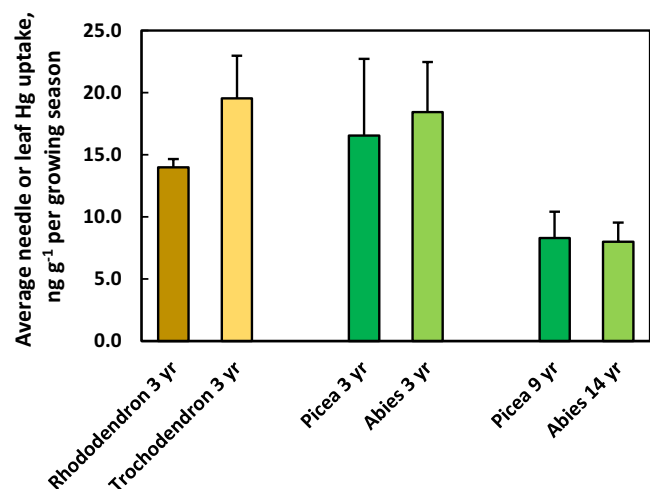
4 Discussion

Despite prior observations of mercury (Hg) accumulation with increasing needle age (e.g. Navrátil et al., 2019; Wohlgemuth et al., 2020), this study is the first to demonstrate continued Hg accumulation in conifer needles up to the 9th NAC in *Picea abies*. This has significant implications for the biogeochemical

cycling of Hg compared to the results from earlier studies as the oldest age classes produces most of needle litter (Flower-Ellis, 1996). The continued increase in Hg content in the conifer needles resulted in maximum concentrations that are higher than concentrations reported in earlier studies (e.g. Zhou et al., 2021), the likely reason being that predominantly younger needles have previously been studied. Navrátil et al. (2019), however, observed average Hg concentrations of four-year old *Picea abies* needles above 50 ng g^{-1} which is comparable to 63 ng g^{-1} for this NAC in our study. Similarly, Hg concentrations in the range of $\sim 50\text{--}65 \text{ ng g}^{-1}$ were observed for four-year old *Picea abies* needles in the data set used by Wohlgemuth et al. (2020). In both these studies, like ours, Hg concentrations of the youngest NACs were much lower. Our findings suggest that there is a risk of underestimating the concentration of Hg in the needles that produce litter by up to approximately 70–80%, depending on species and needle longevity, if first year needle concentrations are used instead of observations for the oldest NAC. This was also emphasized by Méndez-López et al. (2022) studying Hg accumulation in *Pinus pinaster*. The genus *Pinus* has shorter lived needles than *Picea* and *Abies* but over the range of four NACs these authors found a pattern of continued Hg accumulation over time and NAC 3–4 to make the main contribution to the flux of Hg with litterfall.

As indicated also in some previous studies (e.g. Wohlgemuth et al., 2020), the rate of Hg accumulation declined over time and in the case of *Abies* in our study it completely ceased for NACs older than

Fig. 4 Average annual rate of accumulation of Hg in all four investigated species for the first three years as well as for the full needle life span of the two conifers (9 years in *Picea* and 14 years in *Abies*)



eight years. This can be explained by a declining physiological activity of older needles and shading of older needles in the canopy resulting in a lower stomatal conductance (Zimmermann et al., 1988; Robakowski & Bielini, 2017) limiting uptake of Hg^0 from the atmosphere. In addition, there is evidence that the mass of needles increases as they age (Flower-Ellis, 1996), which could, if of sufficient magnitude, dilute Hg and other mineral components, thus partly masking a continued uptake of Hg. However, the most likely interpretation is that the needles successively become saturated with Hg as a dynamic equilibrium between uptake and evasion of Hg is reached. Such an equilibrium is likely to have been achieved in *Abies*, which also had a faster initial uptake rate of Hg leading to higher maximum Hg concentration promoting the establishment of an equilibrium. This would explain the lack of concentration change over NACs 9–14. From our data it seems to take several years to reach such an equilibrium at background atmospheric concentrations of Hg. It was not completely reached in the 9th NAC of *Picea abies*, which could be associated with lower maximum concentrations in this species compared to *Abies pinsapo* var. *marocana*. The theoretical foundation of this kind of equilibrium has been presented by McLachlan (1999) for organic semi-volatile compounds. That Hg has similar semi-volatile properties is evidenced by the bi-directional fluxes of this element observed in highly temporally resolved measurements using stable isotope techniques (Yuan et al., 2019).

In deciduous broadleaved species, an essentially linear increase in Hg over time has been observed during the growing season (e.g. Assad et al., 2016; Bushey et al., 2008; Pleijel et al., 2021), and even in the three LACs of *Trochodendron* and *Rhododendron* of our study, the accumulation rate of Hg was almost linearly related to age. In these cases, the decline in the Hg accumulation rate with increasing leaf age seems to be of limited importance. It becomes significant only for more long-lived leaves or needles. In the literature there exists one earlier study of Hg accumulation in leaves of Japanese azalea, *Rhododendron japonicum* (syn. *Azalea japonica*, Gaggi et al., 1991). Although this was a short-term laboratory chamber study with comparatively high atmospheric Hg exposure, it showed a continued accumulation of Hg by the leaves during the exposure period.

Our study does not directly address the details of the mechanisms by which Hg is taken up by leaves and needles. Although existing literature suggests that stomatal uptake of gaseous elemental mercury (Hg^0) is the quantitatively most important process (Frescholtz et al., 2003; Laacouri et al., 2013; Risch et al., 2017) it cannot be excluded that some of the deposition to leaves, especially in more polluted areas, takes place as particle-bound Hg, which has a much shorter lifetime in the atmosphere than Hg^0 (Gworek et al., 2017) and thus deposits more quickly. Depending on the size of the stomatal aperture, smaller particles carrying metals, may partly enter leaves through the stomata (Gao et al., 2021; Kwak et al., 2020). However, elemental Hg has over the last decades been established as the dominant component of dry deposition in remote areas (Bishop et al., 2020).

Conifer needles have been used in biomonitoring as accumulators of metals and other chemical species. Our study highlights the necessity of carefully taking account of needle age, as has been pointed out also by Bertolotti and Gialanella (2014), older needles providing a stronger chemical signal for Hg due to the continued accumulation over several years. In principle, these considerations apply also to both deciduous and evergreen broadleaved trees. This is relevant also for many other metals than Hg, such as lead (Pb), zinc (Zn), chromium (Cr) and vanadium (V), which are investigated in urban, industrial or rural environments using conifer needles for biomonitoring. For example, Sulhan et al. (2023) investigated the content in of Cr and Zn in needles of *Picea pungens* in Ankara, Türkiye, and found this species to be an appropriate biomonitor for these metals with, like Hg in our investigation, in general rising concentrations over the seven NACs investigated. Likewise, Pleijel et al. (2023) observed rising concentrations of Pb and antimony (Sb) with rising needle age in *Picea abies*. Sardans and Peñuelas (2005) studied a broader range of eleven metals, including Hg, in differently polluted areas of the Barcelona region, Spain, and found clear associations with general pollution level and metal concentrations in needles of *Pinus halepensis*, but also a number of other species used for biomonitoring. It can be noted in this context that also other plant fraction than needles or leaves, such as bark and wood, can be useful in biomonitoring for some metals as exemplified by the study of Cr concentrations in

Picea orientalis of Cetin (2024) and the investigation presented by Isinkaralar et al. (2024) on the accumulation of a broad range of metals in five conifers.

The substantially lower Hg concentrations of the essentially woody branch segments, compared to needles (except the first NAC) are expected in the light of earlier findings by e.g. Yang et al. (2018) and the data summarized in the meta-analysis by Zhou et al. (2021). This strongly indicates that most of the Hg taken up by needles remains in this compartment and that wood, on a per mass basis, is a small sink for Hg. However, one needs to keep in mind that the amount of wood in a forested ecosystem can be very substantial and represent a large, combined pool of Hg although the concentration is low. Furthermore, the much lower Hg concentrations observed for the essentially woody branch segments compared to needles do not suggest wood to be a better plant fraction to use in biomonitoring of Hg compared to needles.

The evergreen leaf habit of needles and leaves represents a resource conservation strategy (Aerts, 1995). Further, the longevity of foliage is influenced both by genetic constitution and acclimation to the environment (Pornon et al., 2011). For example, Reich et al. (1996) found that *Picea abies* and *Pinus sylvestris* retained their needles longer at higher latitude and high altitude. In addition, nitrogen availability can affect needle life span (Reich et al., 2014). An important implication of the life-time average Hg uptake rate presented in Fig. 4 is that the higher maximum Hg concentrations of long-lived needles does not necessarily translate into a higher long-term average uptake rate.

Climate as such can influence Hg accumulation in leaves/needles as exemplified by the larger uptake of Hg by *Pinus pinaster* needles in more oceanic climatic conditions with higher precipitation, promoting higher stomatal conductance and thus Hg absorption, in Galicia, North-West Spain (Méndez-López et al., 2022). In addition, Hg accumulation could be affected by climate change in several ways. Firstly, as pointed out by Wohlgemuth et al. (2023), an altered climate can influence stomatal conductance and thus leaf uptake of Hg. Secondly, in line with the argument provided earlier in the previous paragraph, the average needle life span can be shortened at higher temperatures, leading to a larger average annual needle lifetime uptake of Hg. Finally, on the longer-term climate change is likely to lead to a shift in the

abundance and geographical distribution of species with different leaf habits (Ma et al., 2023), e.g. with an increased occurrence of deciduous broadleaved forests, having a faster leaf Hg accumulation rate (Zhou et al., 2023), replacing evergreen conifers.

We compared the accumulation of Hg in needles of the most common tree species, the conifer *Picea abies*, in Sweden and several other areas of northern Europe, with that of three other evergreen species grown under the same ambient conditions. A general pattern of continued accumulation over several years was observed in all investigated species, but with considerably higher lifetime average uptake in the two broadleaved species compared to the two conifers because of the decline in the rate of net Hg uptake in older needles.

5 Conclusions

The most important conclusions from this study are:

- 1) Net Hg accumulation in evergreen conifers and broadleaved species continues over several years.
- 2) In the conifers, the rate of Hg uptake by the needles declined over time and ceased completely after eight years in *Abies pinsapo* var. *marocana*, indicating that a saturation point had been reached with no further net accumulation of Hg.
- 3) In the more short-lived evergreen leaves of *Trochodendron* and *Rhododendron*, saturation seems to be of limited significance.
- 4) When analysing the role of litterfall as an ecosystem flux of Hg, the average lifetime Hg uptake rate of needles and leaves, which was smaller in conifers compared to broadleaved species in this study, should be accounted for.
- 5) The largely woody branch segments to which the different needle age classes were attached had much lower Hg concentrations than the needles except for the youngest needle age class.
- 6) In future biogeochemical research (empirical and modelling) and biomonitoring studies, the age of sampled leaves and needles needs to be considered to account for the age dependence of Hg concentration and accumulation rate. This applies also to other metals monitored with this approach.

Our study highlights that the leaf and needle life span is of critical importance in the assessment of Hg in plant samples and their interpretation in the context of Hg circulation in the environment as well as in bio-monitoring based on metal accumulation in evergreen foliage.

Acknowledgements Planning, sampling and preparation of the manuscript was funded by the research council Formas (grant: 2017-00696). Chemical analyses were financially supported by the strategic research area BECC (Biodiversity and Ecosystem Services in a Changing Climate, www.becc.lu.se).

Authors Contributions *Håkan Pleijel*: Conceptualization, Field Sampling, Data Curation, Writing-Original Draft, Visualisation, Investigation, Funding Acquisition; *Jenny Klingberg*: Field Sampling, Laboratory Analysis, Statistical Analysis, Writing-Review and Editing; *Henrik Sjöman*: Field Sampling, Writing-Review and Editing; *Göran Wallin*: Field Sampling, Laboratory Analysis, Writing-Review and Editing.

Funding Open access funding provided by University of Gothenburg.

Data Availability Data used in this study are available here: <https://doi.org/10.5878/flqk-px15>.

Declaration

Competing Interests The authors declare that they have no financial or non-financial competing interests of relevance to the content of this article.

Open Access This article is licensed under a Creative Commons Attribution 4.0 International License, which permits use, sharing, adaptation, distribution and reproduction in any medium or format, as long as you give appropriate credit to the original author(s) and the source, provide a link to the Creative Commons licence, and indicate if changes were made. The images or other third party material in this article are included in the article's Creative Commons licence, unless indicated otherwise in a credit line to the material. If material is not included in the article's Creative Commons licence and your intended use is not permitted by statutory regulation or exceeds the permitted use, you will need to obtain permission directly from the copyright holder. To view a copy of this licence, visit <http://creativecommons.org/licenses/by/4.0/>.

References

- Aerts, R. (1995). The advantage of being evergreen. *Trends in Ecology & Evolution*, 10, 402–407. [https://doi.org/10.1016/S0169-5347\(00\)89156-9](https://doi.org/10.1016/S0169-5347(00)89156-9)
- Assad, M., Parelle, J., Cazaux, D., Gimbert, F., Chalot, M., & Tatit-Froux, F. (2016). Mercury uptake into poplar leaves. *Chemosphere*, 146, 1–7. <https://doi.org/10.1016/j.chemosphere.2015.11.103>
- Bertolotti, G., & Gialanella, S. (2014). Review: Use of conifer needles as passive samplers of inorganic pollutants in air quality monitoring. *Analytical Methods*, 6, 6208–6222. <https://doi.org/10.1039/c4ay00172a>
- Bishop, K., Shanley, J. B., Riscassi, A., de Wit, H. A., Eklöf, K., Meng, B., Mitchell, C., Osterwalder, S., Schuster, P. F., Webster, J., & Zhu, W. (2020). Recent advances in understanding and measurement of mercury in the environment: Terrestrial Hg cycling. *Science of the Total Environment*, 721, 137647. <https://doi.org/10.1016/j.scitotenv.2020.137647>
- Bushey, J. T., Nallana, A. G., Montesdeoca, M. R., & Driscoll, C. T. (2008). Mercury dynamics of a northern hardwood canopy. *Atmospheric Environment*, 42, 6905–6914. <https://doi.org/10.1016/j.atmosenv.2008.05.043>
- Cetin, I. Z. (2024). Used in urban area for landscape planning and design spatial and temporal variations in chromium (Cr) concentrations in *Picea orientalis* L. *Turkish Journal of Agriculture - Food Science and Technology*, 12, 1730–1738. <https://doi.org/10.24925/turjaf.v12i10.1730-1738.7093>
- Demers, J. D., Blum, J. D., & Zak, D. R. (2013). Mercury isotopes in a forested ecosystem: Implications for air-surface exchange dynamics and the global mercury cycle. *Global Biogeochemical Cycles*, 27, 222–238. <https://doi.org/10.1002/gbc.20021>
- Eriksen, J. A., & Gustin, M. S. (2004). Foliar exchange of mercury as a function of soil and air mercury concentrations. *Science of the Total Environment*, 324, 271–279. <https://doi.org/10.1016/j.scitotenv.2003.10.034>
- Flower-Ellis, J. G. K. (1996). *Crown structure and phytomass distribution in Scots pine and Norway spruce trees: 1. Computer-based field sampling routines*. Department for Production Ecology, Faculty of Forestry, Swedish University of Agricultural Science Report No. 2, ISSN 1401–5625.
- Frescholtz, T., Gustin, M. S., Schorran, D. E., & Fernandez, C. J. (2003). Assessing the source of mercury in foliar tissue of quaking aspen. *Environmental Toxicology and Chemistry*, 22, 2114–2119. <https://doi.org/10.1002/etc.5620220922>
- Gaggi, C., Chemello, G., & Bacci, E. (1991). Mercury vapour accumulation in azalea leaves. *Chemosphere*, 22, 869–872. [https://doi.org/10.1016/0045-6535\(91\)90244-8](https://doi.org/10.1016/0045-6535(91)90244-8)
- Gao, P. P., Xue, P. Y., Dong, J. W., Zhang, X. M., Sun, H. X., Geng, L. P., Luo, S. X., Zhao, J. J., & Liu, W. J. (2021). Contribution of PM_{2.5}-Pb in atmospheric fallout Pb accumulation in Chinese cabbage leaves via stomata. *Journal of Hazardous Materials*, 407, 124356. <https://doi.org/10.1016/j.jhazmat.2020.124356>
- Gworek, B., Dmuchowski, W., Baczewska, A. H., Bragozevska, P., Bemowska-Kalabun, O., & Wrosek-Jakubowska, J. (2017). Air contamination by mercury, emissions and transformations – a review. *Water, Air, and Soil Pollution*, 228, 123. <https://doi.org/10.1007/s11270-017-3311-y>
- Hanson, P. J., Lindberg, S. E., Tabberer, T. A., Owens, J. G., & Kim, K. H. (1995). Foliar exchange of mercury vapor: Evidence for a compensation point. *Water, Air, and Soil*

- Pollution*, 80, 373–382. <https://doi.org/10.1007/BF01189687>
- Hutnik, R. J., McClenahan, J. R., Long, R. P., & Davis, D. D. (2014). Mercury Accumulation in *Pinus nigra* (Austrian Pine). *Northeastern Naturalist*, 21, 529–540. <https://doi.org/10.1656/045.021.0402>
- Jiskra, M., Sonke, J. E., Obrist, D., Bieser, J., Ebinghaus, R., Myhre, C. L., Pfaffhuber, K. A., Wängberg, I., Kyllönen, K., Worthy, D., Martin, L. G., Labuschagne, C., Mkololo, T., Ramonet, M., Magand, O., & Dommergue, A. (2018). A vegetation control on seasonal variations in global atmospheric mercury concentrations. *Nature Geoscience*, 11, 244–250. <https://doi.org/10.1038/s41561-018-0078-8>
- Isinkaralar, K., Isinkaralar, O., Koc, I., Cobanoglu, H., & Canturk, U. (2024). Accumulation analysis and overall measurement to represent airborne toxic metals with passive tree bark biomonitoring technique in urban areas. *Environmental Monitoring and Assessment*, 196, 689. <https://doi.org/10.1007/s10661-024-12879-6>
- Kozłowski, T. T. (1964). Shoot growth in woody plants. *Botanical Review*, 30, 335–392.
- Laacouri, A., Nater, E. A., & Kolka, R. K. (2013). Distribution and uptake dynamics of mercury in leaves of common deciduous tree species in Minnesota, USA. *Environmental Science & Technology*, 47, 10462–10470. <https://doi.org/10.1021/es401357z>
- Kwak, N. J., Lee, J. K., Park, S., Kim, H., Lim, Y. J., Lee, K., Son, J., Oh, C., Kim, I., & Woo, S. Y. (2020). Surface-based analysis of leaf microstructures for adsorbing and retaining capability of airborne particulate matter in ten woody species. *Forests*, 11, 946. <https://doi.org/10.3390/f11090946>
- Ma, H., Crowther, T. W., Mo, L., Maynard, D. S., Renner, S. S., van den Hoogen, J., Zou, Y., Liang, J., de-Miguel, S., Nabuurs, G.-J., Reich, P. B., Niinemets, Ü., Abegg, M., Adou Yao, Y. C., Alberti, G., Almeyda Zambrano, A. M. V., Alvarado, B., Alvarez-Dávila, E., Alvarez-Loayza, ..., Zohner, C. M. (2023). The global biogeography of tree leaf form and habit. *Nature Plants* 9, 1795–1809. <https://doi.org/10.1038/s41477-023-01543-5>
- Manghabati, H., Weis, W., & Göttelein, A. (2019). Changes in phosphorus concentration in needles of adult Norway spruce - nutrient re-translocation or dilution effect? *European Journal of Forest Research*, 138, 539–546. <https://doi.org/10.1007/s10342-019-01188-0>
- McLachlan, M. S. (1999). Framework for the interpretation of measurements of SOCs in plants. *Environmental Science and Technology*, 33, 1799–1804. <https://doi.org/10.1021/es980831t>
- Méndez-López, M., Gómez-Armesto, A., Eimil-Fraga, C., Alonso-Vega, F., Rodríguez-Soalleiro, R., Arias-Estévez, M., & Nóvoa-Muñoz, J. C. (2022). Needle age and precipitation as drivers of Hg accumulation and deposition in coniferous forests from a southwestern European Atlantic region. *Environmental Research*, 215, 114223. <https://doi.org/10.1016/j.envres.2022.114223>
- Muukkonen, P. (2005). Needle biomass turnover rates of Scots pine (*Pinus sylvestris* L.) derived from the needle-shed dynamics. *Trees*, 19, 273–279. <https://doi.org/10.1007/s00468-004-0381-4>
- Navrátil, T., Nováková, T., Roll, M., Shanley, J. B., Koáček, J., Kaňa, J., & Cudlín, P. (2019). Decreasing litterfall mercury deposition in central European coniferous forests and effects of bark beetle infestation. *Science of the Total Environment*, 682, 213–225. <https://doi.org/10.1016/j.apgeochem.2016.10.005>
- Nebel, B., & Matile, P. (1992). Longevity and senescence of needles in *Pinus cembra* L. *Trees*, 6, 156–161. <https://doi.org/10.1007/BF00202431>
- Niinemets, Ü., & Lukjanova, A. (2003). Total foliar area and average leaf age may be more strongly associated with branching frequency than with leaf longevity in temperate conifers. *New Phytologist*, 158, 75–89. <https://doi.org/10.1046/j.1469-8137.2003.00712.x>
- Obrist, D., Kirk, J. L., Zhang, L., Sunderland, E. M., Jiskra, M., & Selin, N. E. (2018). A review of global environmental mercury processes in response to human and natural perturbations: Changes of emission, climate, and land use. *Ambio*, 47, 116–140. <https://doi.org/10.1007/s13280-017-1004-9>
- Oren, R., Werk, K. S., Schulze, E.-D., Meyer, J., Schneider, B. U., & Schramel, P. (1988). Performance of two *Picea abies* (L.) Karst. stands at different stages of decline – VI. Nutrient Concentration. *Oecologia*, 77, 151–162. <https://doi.org/10.1007/BF00379181>
- Pleijel, H., Klingberg, J., Nerentorp, M., Broberg, M. C., Nyirambangutse, B., Munthe, J., & Wallin, G. (2021). Mercury accumulation in leaves of different plant types – the significance of tissue age and specific leaf area. *Biogeosciences*, 18, 6313–6328. <https://doi.org/10.5194/bg-18-6313-2021>
- Pleijel, H., Klingberg, J., Strandberg, B., Sjöman, H., & Wallin, G. (2023). Accumulation of antimony and lead in leaves and needles of trees: The role of traffic emissions. *Heliyon*, 9, e13548. <https://doi.org/10.1016/j.heliyon.2023.e13548>
- Pornon, A., Marty, C., Winterton, P., & Lamaze, T. (2011). The intriguing paradox of leaf lifespan responses to nitrogen availability. *Functional Ecology*, 25, 796–801. <https://doi.org/10.1111/j.1365-2435.2011.01849.x>
- Reich, P. B., Oleksyn, J., Modrzyński, J., & Tjoelker, M. (1996). Evidence that longer needle retention of spruce and pine populations at high elevations and high latitudes is largely a phenotypic response. *Tree Physiology*, 16, 643–647. <https://doi.org/10.1093/treephys/16.7.643>
- Reich, P. B., Richa, R. L., Luc, X., Wang, Y. P., & Oleksyn, J. (2014). Biogeographic variation in evergreen conifer needle longevity and impacts on boreal forest carbon cycle projections. *PNAS*, 111, 13703–13708. <https://doi.org/10.1073/pnas.1216054110>
- Risch, M. R., DeWild, J. F., Gay, D. A., Zhang, L., Boyer, E. W., & Krabbenhoft, D. P. (2017). 747 Atmospheric mercury deposition to forests in the eastern USA. *Environmental Pollution*, 228, 8–18. <https://doi.org/10.1016/j.envpol.2017.05.004>
- Robakowski, P., & Bielini, E. (2017). Needle age dependence of photosynthesis along a light gradient within an *Abies alba* crown. *Acta Physiologiae Plantarum*, 39(83), 1–12. <https://doi.org/10.1007/s11738-017-2376-y>
- Sardans, J., & Peñuelas, J. (2005). Trace element accumulation in the moss *Hypnum cupressiforme* Hedw. and the trees

- Quercus ilex* L. and *Pinus halepensis* Mill. in Catalonia. *Chemosphere*, 60, 1293–1307. <https://doi.org/10.1016/j.chemosphere.2005.01.059>
- Sommar, J., Osterwalder, S., & Zhu, W. (2020). Recent advances in understanding and measurement of Hg in the environment: Surface-atmosphere exchange of gaseous elemental mercury (Hg^0). *Science of the Total Environment*, 721, 137648. <https://doi.org/10.1016/j.scitotenv.2020.137648>
- Sulhan, O. F., Sevik, H., & Isinkaralar, K. (2023). Assessment of Cr and Zn deposition on *Picea pungens* Engelm. In urban air of Ankara Türkiye. *Environment, Development and Sustainability*, 25, 4365–4384. <https://doi.org/10.1007/s10668-022-02647-2>
- Tabaja, N., Ameroux, D., Chalak, L., Fourel, F., Tessier, E., Jomaa, I., El Riachy, M., & Bentaleb, I. (2023). Seasonal variation of mercury concentration of ancient olive groves of Lebanon. *Biogeosciences*, 20, 619–633. <https://doi.org/10.5194/bg-20-619-2023>
- UN Environment. (2019). *Global Mercury Assessment Report 2018*. UN Environmental Programme, Chemicals and Health Branch Geneva.
- Wohlgemuth, L., Osterwalder, S., Joseph, C., Kahmen, A., Hoch, G., Alewell, C., & Jiskra, M. (2020). A bottom-up quantification of foliar mercury uptake fluxes across Europe. *Biogeoscience*, 17, 6441–6456. <https://doi.org/10.5194/bg-17-6441-2020>
- Wohlgemuth, L., Rautio, P., Ahrends, B., Russ, A., Vesterdal, L., Waldner, P., Timmerman, V., Eickenscheidt, N., Fürst, A., Greve, M., Roskams, P., Thimonier, A., Nicolas, M., Kowalska, A., Ingerslev, M., Merilä, P., Benham, S., Iacoban, C., Hoch, G., ... Jiskra, M. (2022). Physiological and climate controls on foliar mercury uptake by European tree species. *Biogeoscience*, 19, 1335–1353. <https://doi.org/10.5194/bg-19-1335-2022>
- Wohlgemuth, L., Feinberg, A., Buras, A., & Jiskra, M. (2023). A spatial assessment of current and future foliar Hg uptake fluxes across European forests. *Global Biogeochemical Cycles*, 37, e2023GB007833. <https://doi.org/10.1029/2023GB007833>
- Wytenbach, A., & Tobler, L. (2000). The concentrations of Fe, Zn and Co in successive needle age classes of Norway spruce [*Picea abies* (L.) Karst.]. *Trees*, 14, 198–205. <https://doi.org/10.1007/PL00009763>
- Yang, Y., Yanai, R. D., Driscoll, C. T., Montesdeoca, M., & Smith, K. T. (2018). Concentration and content of mercury in bark, wood, and leaves in hardwoods and conifers in four forested sites in the northeastern USA. *PLoS ONE*, 13(4). <https://doi.org/10.1371/journal.pone.0196293>
- Yuan, W., Sommar, J., Lin, C. J., Wang, X., Li, K., Liu, Y., Zhang, H., Lu, Z., Wu, C., & Feng, X. (2019). Stable isotope evidence shows re-emission of elemental mercury vapour occurring after reductive loss from foliage. *Environmental Science and Technology*, 53, 651–660. <https://doi.org/10.1021/acs.est.8b04865>
- Zhou, J., Obrist, D., Dastoor, A., Jiskra, M., & Ryjkov, A. (2021). Vegetation uptake of mercury and impacts on global cycling. *Nature Reviews Earth & Environment*, 2, 269–284. <https://doi.org/10.1038/s43017-021-00146-y>
- Zhou, J., Bollen, S. W., Roy, E. M., Hollinger, D. Y., Wang, T., Lee, J. T., & Obrist, D. (2023). Comparing ecosystem gaseous elemental mercury fluxes over a deciduous and coniferous forest. *Nature Communications*, 14, 2722. <https://doi.org/10.1038/s41467-023-38225-x>
- Zimmermann, R., Oren, R., Schulze, E.-D., & Werk, K. S. (1988). Performance of two *Picea abies* (L.) Karst. stands at different stages of decline – II. Photosynthesis and Leaf Conductance. *Oecologia*, 76, 513–518. <https://doi.org/10.1007/BF00397862>

Publisher's Note Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.