

Title

**Global warming alters mercury accumulation in trees
Mercury concentration records in tree rings of German sessile oak and
Douglas fir – The role of pollution and climatic changes**

**Mercury concentration records in tree rings of sessile oak and Douglas
fir – The role of pollution and climate**

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Abstract

Stomatal uptake of mercury (Hg) by trees and litterfall ~~is~~ are the major pathways of atmospheric Hg to the terrestrial environment. ~~However, underlying processes and controlling factors of Hg accumulation in tree rings which have been used in numerous~~ Most existing studies on tree-ring Hg records were aiming to reconstruct historical atmospheric Hg concentrations at polluted sites ~~are poorly understood~~. ~~However, Especially~~ the role of changing climatic ~~condition~~se such as rising temperature and frequent drought events ~~has been compared to local Hg contamination levels have been~~ widely ignored. ~~We investigated tree rings of oaks at a former Hg mining site and an unpolluted reference site and compared the oak tree-ring records with tree rings of Douglas fir from unpolluted sites, all located in western Germany.~~ ~~We show~~ Our data indicates that the overall Hg ~~load accumulation~~ in trees ~~rings at the investigated sites~~ is determined by local atmospheric Hg ~~emissions~~ loads in soil, but ~~did not follow the historical record of atmospheric Hg pollution.~~ ~~It~~ In contrast, the long-term evolution of tree ring Hg concentrations can statistically be attributed ~~with~~ ~~hydro~~ ~~predominately~~ controlled by changing hydroclimatic ~~ic~~ conditions. ~~Due to~~ ~~d~~ We suggest that ~~D~~ different physiological heat adaption strategies of the investigated ~~Central European~~ oaks are likely to lead to ~~show~~ continuously increasing tree-ring Hg concentration with rising temperature and precipitation rates during the past century, whereas the investigated ~~Douglas fir~~ ~~coniferous~~ trees show a strongly declining trend in Hg load in the same period. Our findings indicate that climate ~~conditions appear to overwrite changes in atmospheric Hg change and may~~ alters the Hg accumulation in different forest types and likely the related transfer of atmospheric Hg to the soil ~~through litterfall~~. ~~However, Hg in tree rings is triggered by a complex system of different biotic and abiotic factors.~~

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Keywords

60 Tree-ring mercury, Douglas fir, Sessile oak, Northern hemisphere tree-ring mercury record, Climate relationship

1 Introduction

Stomatal mercury (Hg) uptake of atmospheric Hg has been shown to be a major component in the global biogeochemical Hg cycle and litterfall is the major vector of atmospheric Hg fluxes to soils.

Based on the assumption that tree-ring Hg concentrations reflect atmospheric Hg concentrations, tree-ring records have been used to reconstruct atmospheric pollution. At present, it is believed that stomatal Hg uptake and transport through the conductive tissue is the major pathway of atmospheric Hg into tree rings and that Hg uptake through roots or the tree's bark is of minor importance (Liu et al., 2024). However, it is widely unknown to which extent other factors than atmospheric Hg concentrations such as temperature, precipitation or physiological processes affect the Hg accumulation in tree rings. Previous studies suggest that coniferous trees are more suitable than

broadleaf trees e.g. oak trees to reflect recording changes in atmospheric Hg loads by tree-ring Hg concentrations, whereas processes influencing the formation of the Hg signal in tree rings of broadleaved trees are poorly constrained. Pine and Larch are the best investigated tree species regarding historical Hg records. At Hg contaminated sites these tree species show close response to elevated atmospheric Hg concentration in their tree rings caused by anthropogenic or natural Hg emissions (Gustin et al., 2022b; Scanlon et al., 2020; Peckham et al., 2019; Navrátil et al., 2018; Nováková et al., 2022; Peng et al., 2024; Kang et al., 2022; McLagan et al., 2022). Oaks are not suitable for dendrochemical studies, which is concluded by e.g. Scanlon et al. (2020), Siwik et al. (2010) and Gustin et al. (2022b). Their main arguments are, that oak Hg records show no temporal trends, hold much lower Hg concentrations compared to coniferous tree species and are low in high-frequency variability. Radial Hg movement between hardwood and softwood (translocation) is also used as an argument to rule out Hg records of oaks for dendrochemical studies. A scientific proof of these arguments is still missing and may have led to a situation that oaks, and broadleaf species in general, are not in focus for Hg tree-ring investigations. Long-term Hg records of these species are thus rare, but urgently needed to get more insight regarding the dendrochemical potential of oaks.

Up to now most studies on tree-ring Hg records have mostly neglected the influence of changing climatic conditions, specifically the strong increase in temperature and severe drought events in the past decades. This might be attributed to the far higher tree-ring Hg concentrations in polluted trees compared to trees from unpolluted areas which may overwrite climate induced patterns. Moreover, tree-ring Hg records from different locations on hemispheric or global larger scale have only been rarely compared to decipher common patterns in Hg accumulation. Global climate change affects Hg loads in soils through litterfall and Hg reemission (Guo et al., 2024; Wang et al., 2019) and adjacent aquatic ecosystems (Li et al., 2020; Zhang et al., 2016)

and thus are important drivers for Hg accumulation in tree rings. Here we present tree-ring Hg records of oak and Douglas fir from contaminated and uncontaminated sites (soil) covering the past ~150 years to decipher climatic and soil Hg related controlling factors of Hg accumulation in tree rings. In addition, we compared literature data of tree-ring Hg records from different sites in the Northern Hemisphere and compared them to climatic variables and the evolution of atmospheric Hg concentrations to show their geographical-wide consistency and connectivity.

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2 Methods

2.1 Tree species and tree-ring samples

105 Five trees per location at Stahlberg and Leimen (each sessile oak, *Quercus petraea* (Matt.) Liebl.)
as well as at Karlshausen (Douglas fir, *Pseudotsuga menziesii* var. *menziesii* (Mirbel) Franco) were
sampled in Rhineland-Palatinate, Germany. From each tree a wood sample at breast height (1m)
was taken using an increment borer (Jim-Gem, USA). The tree rings were measured with an
accuracy of 0.01 mm (Rinn, 2002-2013) and precisely dated to their specific calendar years. All
110 single tree-ring series were cross-checked to one another to ensure accurate tree-ring dating. Then,
5-year blocks (with mid-point years 2017, 2012, 2007 and so forth) were cut with a scalpel.
Additionally, the sapwood tree rings were marked and counted. The cut wood samples were stored
in plastic tubes, uniquely labeled and sent to the laboratory at Technische Universität Braunschweig
(Germany) for mercury concentration measurements.

115 From each individual tree-ring mercury (Hg) concentration time series, the values were z-score
transformed to ensure equal mean and standard deviation before averaging the series to obtain
mean site Hg records for Stahlberg, Leimen and Karlshausen. For further analysis the two mean site
Hg records of the oak locations (Stahlberg, Leimen) were combined to a mean oak Hg record with
equal weight between the two oak Hg records.

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2.2 Mercury concentration measurements in wood samples

The precisely dated tree-ring samples were freeze-dried to constant weight prior to Hg analyses.
The Hg concentration in each wood sample was determined by means of cold-vapor atomic
absorption spectroscopy after thermal decomposition and pre-concentration of Hg through
125 amalgamation on a gold trap using a DMA-80 EVO III direct mercury analyzer (Milestone, USA; EPA
Method 7473 (U.S. Environmental Protection Agency, (EPA), "Mercury in solids and solutions by
thermal decomposition, amalgamation, and atomic absorption spectrophotometry" (1998)). NIST
1515 (Apple Leaves, 44 ± 4 ng Hg g⁻¹,) was used as a standard reference material.

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2.3 Regional averages of seasonal mean air temperature and precipitation sum

The climate data used were downloaded from the German Weather Service Homepage
(https://opendata.dwd.de/climate_environment/CDC/regional_averages_DE/seasonal/). We used
135 the regional averages of seasonal mean air temperature and precipitation sum to calculate the
spring-summer records for Rhineland-Palatinate (Germany), respectively. Spring and summer air
temperature data were averaged and spring and summer precipitation sum data were summed on
annual basis to calculate the spring-summer records. Thereafter, for each 5-year block (mid-point
year of blocks are 2017, 2012, 2007 etc.) the spring-summer temperature / precipitation sum values
were averaged and z-score transformed.

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2.4 Multi-linear regression modeling and statistical analysis

To account for the specific proportion of spring-summer air temperature and precipitation sum
influencing the tree-ring mercury concentration, multi-linear regression modeling was performed
145 (Montgomery et al., 2001). We used Pearson correlation statistics to calculate associations between
tree-ring mercury concentrations and the climate records. Statistical significance was assumed at α
= 1%.

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2.5 Hemisphere-wide tree-ring mercury concentration records

To analyse the synchronicity between different tree-ring Hg concentration records in the Northern
Hemisphere, we used published and available datasets of different tree species with different tree
age at the continents of North America, Asia and Europe. These Hg records were treated in the
following way: If a specific Hg record represented the mean Hg concentration of several trees, the
155 values were z-score transformed. In the case, when such a mean Hg record was on annual timescale,
the values were averaged for 5-year blocks with mid-point years 2017, 2012, 2007 etc. to reduce
data points for better visibility. If tree-ring Hg concentration series for several trees at a specific site
were available, a mean Hg record was calculated by averaging the Hg data by the corresponding

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160 year or year-blocks. The tree-ring Hg records used are: Scanlon et al., 2020; Chellman et al., 2020;
 165 Ghotra et al., 2020; Nováková et al., 2021; Wang et al., 2021; Baroni et al., 2023; Peng et al., 2024;
 Fornasaro et al., 2023. Thereafter, all values were z-score transformed.

2.6 Global Average surface temperature, atmospheric mercury concentration data

165 Global air temperature data were received from the National Centers for Environmental Information,
 National Oceanic and Atmospheric Administration

(<https://www.ncei.noaa.gov/access/monitoring/climate-at-a-glance/global/time-series>).

The atmospheric mercury concentration data were received from Emissions Database for Global
 Atmospheric Research (EDGAR, https://edgar.jrc.ec.europa.eu/country_profile).

170 Leaf activity (number of days between bud break and leaf discoloration) data were obtained for the
 station Otterberg (station-id: 10238, Rhineland-Palatinate, Germany) from the German Weather
 Service (https://opendata.dwd.de/climate_environment/CDC/observations_germany/phenology/).
 All values were averaged for 5-year blocks and z-score transformed.

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3 Results and Discussion

3.1 Tree-ring mercury concentrations in oak and Douglas fir

180 Measurements of mercury (Hg) accumulation in tree rings of sessile oak (*Quercus petraea* (Matt.)
 Liebl.) and Douglas fir (*Pseudotsuga menziesii* var. *menziesii* (Mirbel) Franco)) are rare and
 controlling factors are poorly investigated. We analysed tree-ring Hg concentration in these two
 species growing in Rhineland-Palatinate, western Germany (Fig. 1). Oak tree-ring samples have
 been taken in a mixed deciduous forest at Stahlberg. The site is located at a former Hg mining area
 185 where cinnabar ore (HgS) has been extracted since medieval age and especially during 1934–42
 common era, but not processed. Ore roasting and related Hg emissions to the atmosphere took
 place at the Moschellandsberg site approx. 6 km far from Stahlberg. A deciduous forest at Leimen
 about 40 km distant from the Stahlberg site was selected as an uncontaminated reference site for
 oak. Douglas fir has been sampled at Karlshausen an uncontaminated reference site for coniferous
 trees. Soil Hg concentration at the Stahlberg site show high heterogeneity with a median of 34 mg
 190 kg⁻¹ (max. 205 mg kg⁻¹) attributed to the widespread occurrence of cinnabar and soil organic matter
 bound Hg in these soils (Siebelt, 2024), whereas Hg concentration in soils at the two reference sites
 are in the range of background values (<0.5 mg kg⁻¹).

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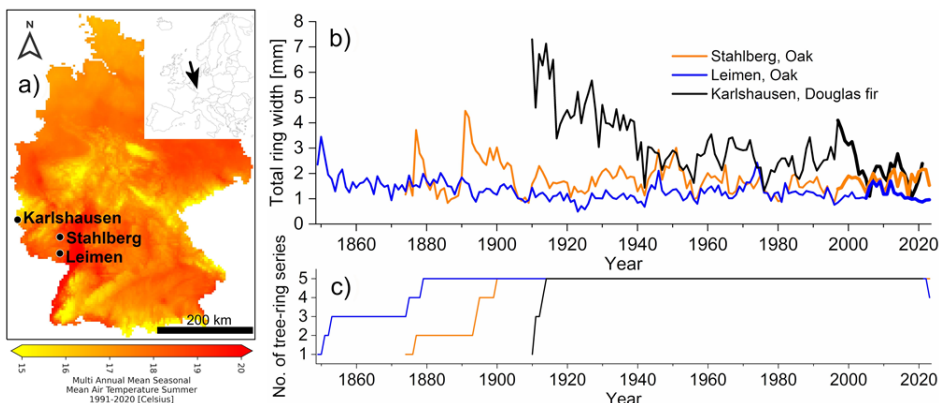
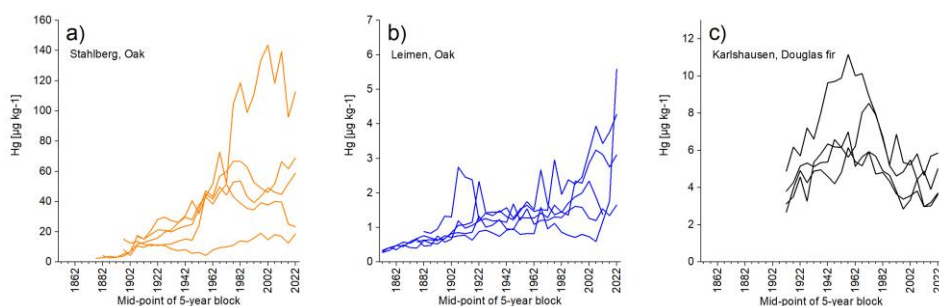


Fig. 1: Area of interest and tree growth dynamic. a) Map color shows the mean air summer temperature in the reference period 1991–2020 and the geographical location of the three sites. b) Mean total ring width of oaks and Douglas firs at the different sites. c) Number of used tree-ring series available per year.

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At the contaminated site (Stahlberg, oak) the mean tree-ring Hg concentration of $50 \mu\text{g kg}^{-1}$ (since 1950) is 25-fold and 10-fold higher than at the uncontaminated sites at Leimen (oak) and Karlshausen (Douglas fir), respectively (Fig. 2, Fig. S4). Tree-ring Hg concentration variability is high among trees at the contaminated site with concentrations ranging from 3–144 $\mu\text{g kg}^{-1}$. At the uncontaminated sites inter-tree-ring Hg concentration variability is much more homogenous with values ranging from <1 –6 $\mu\text{g kg}^{-1}$ (oak, Leimen) and 3–11 $\mu\text{g kg}^{-1}$ (Douglas fir, Karlshausen). Mean annual radial tree growth (total ring width, Fig. 1) of oaks at both sites is lower (1.8 mm at Stahlberg and 1.3 mm at Leimen) compared to Douglas fir (3.1 mm per year). The Douglas fir trees show a pronounced juvenile growth trend, which is suppressed in oak trees. The number of sapwood rings is similar in the investigated trees (~27 in oaks at Stahlberg, ~19 in oaks at Leimen, ~25 in Douglas fir at Karlshausen).
Despite the large difference in tree-ring Hg concentration (mean factor of 10, Fig. 2) between the contaminated and the uncontaminated site, Hg concentrations in tree rings at both oak sites show a clear trend of increasing values and a similar factor of increase (~9.5), although the range was higher at the Stahlberg site (2.8–26.9) than at the Leimen site (3.3–13.8). Hg concentrations in tree rings of Douglas fir are by an average factor of two higher than in oak at the reference site, but do not show a trend of increasing values. Here, Hg concentration in Douglas fir show a maximum in the 1950s–70s and decreasing values thereafter (1980s–2000s).

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225 **Fig. 2:** Tree-ring mercury (Hg) concentration in individual trees through time from three locations in Rhineland-Palatinate, Germany.

230 The high tree-ring Hg concentrations at the Stahlberg site (oak) compared to the Leimen (oak) and Karlshausen (Douglas fir) reference sites indicate a local signal of high atmospheric Hg levels [at this former Hg mining site although no ore processing has taken place at this site](#). We suggest that elevated Hg degassing from the Hg contaminated soil at the Stahlberg site is the main reason for the high tree-ring Hg concentrations. [Interestingly, this signal seems to be local only as the Hg emissions from the roasting site in Moschellandsberg \(6 km away\) during World War II are not recorded in the oak trees from the Stahlberg site.](#)

235 The [strongly](#) differing absolute [tree-ring](#) Hg concentrations at the two oak sites suggest [a strong](#) influence of local atmospheric Hg concentration, especially through soil Hg degassing, [which is largely controlled by temperature as an important forcing factor](#) (Wang et al., 2019). When relative tree-ring Hg variability, including the high among tree variation in factors of concentration increase, is considered, the similar trend of increasing Hg concentrations at both oak sites suggests [an additional](#) common underlying controlling factor different from atmospheric Hg concentrations (Fig. 3). The same assumption is made for relative tree-ring Hg variability in Douglas fir, but [Douglas fir appear to react with a different underlying to this](#) controlling factor.

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245 Here, The Douglas fir tree-ring Hg record does not show a trend of a continuously increasing Hg in tree rings until today, but an increase until 1950s and a pronounced decrease since the 1970s (Fig. 3) opposite to what has been observed in oaks.

250 In contrast, there is consensus regarding the different factors influencing Hg accumulation in tree rings for coniferous species, for broadleaved trees – especially oak – it seems not. In literature, oak has been only rarely investigated and often only a small number of individuals were used. Based on these few studies the impression arose that oak is not suitable to reflect historical atmospheric Hg contamination (Gustin et al., 2022b; Scanlon et al., 2020; Siwik et al., 2010). However in the original literature, no clear evidence has been presented. The main argument is that oak samples show no trend through time or have very low Hg concentration. This may be due to several complex reasons, which we discuss hereafter.

255 Beside other aspects of e.g. climate factors and foliage uptake strategies (isohydric / anisohydric) influencing Hg accumulation in tree rings, radial translocation is another aspect that has to be considered. Radial translocation is basically defined as a movement of an element between softwood and heartwood via living ray cells to detoxify the living tissue in the outermost softwood. Hg is mainly transported radially inwards, i.e. from sapwood into the heartwood, which would severely alter the recorded Hg tree-ring signal (Liu et al., 2023 and references therein). This would automatically lead to a higher Hg concentration in the precisely dated tree rings of past decades. Our investigated oaks at the contaminated and uncontaminated sites do not display any elevating Hg concentration in the heartwood compared to the softwood. Thus, we are convinced that our Hg oak records are biased by radially translocation to a minimal extent amount. Gustin et al. (2022b) stated that much more effort and work is needed to comprehensively understand radial translocation and its potential. We agree with that and want to add that extensive investigation about radial translocation especially in oak is needed to set Hg climate-response studies and their results into the right context. In biological systems, variability of Hg in tree rings is influenced by multiple factors (Boonen et al., 2025). To make robust statements about the potential influence of climate for Hg accumulation in tree rings of oak it is essential to have knowledge about radial translocation. Our here presented results shall be seen as a motivation for studying broadleaved trees and their overlooked potential.

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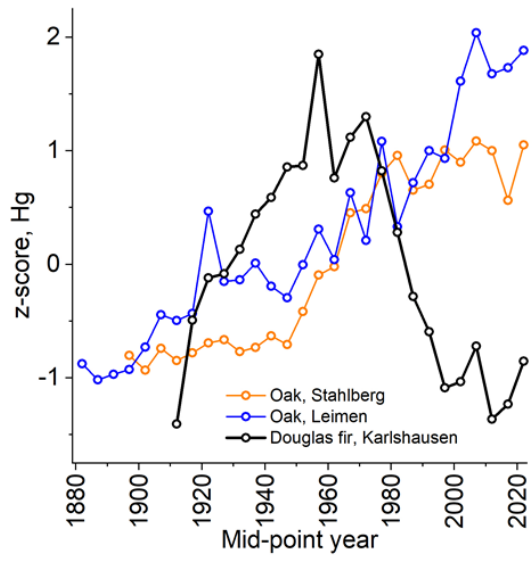


Fig. 3: Tree-ring mercury (Hg) records at the investigated oak (Stahlberg, Leimen) and Douglas fir (Karlshausen) sites in Rhineland-Palatinate, Germany.

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Our Hg oak records from central Europe show a clear elevating trend from past to present. This is a different to what other published Hg oak series show (e.g. Siwik et al., 2010; Scanlon et al., 2020; Gustin et al., 2022b; Perone et al., 2018), namely a low temporal trend as well as low high-frequency variability. The reasons for these differences could be due to the variety of investigated oak species, differences of site and growth factors and local soil Hg concentration. Most Hg oak records are constructed from the Northern American continent and only a viewfew from Europe (Perone et al., 2018), which could lead to significant differences in trend patterns. Another reason that our oak records differ from other studies may come from the fact that records of Red oak, White oak and downy oak are published (Perone et al., 2018; Scanlon et al., 2020; Siwik et al., 2010), whereas our records originate from sessile oak. Sessile oak is very drought tolerant, forms a deep root system and holds a high water-use-efficiency compared to other oak species like Red oak. The number of used trees (3–10) to construct a Hg record also differ between the published studies and may bias a comparison. However, we think that the here constructed Hg tree-ring records are chronologically robust, because the records of the contaminated and uncontaminated sites are similar in regard to the temporal pattern, in their relative variance and in the age of the trees used. The difference of Hg concentrations in oak trees between the sites can be explained by high site-specific Hg loads. The similar trend in both Hg oak records must thus be triggered by other factors, such as climate.

Like in previous studies ~~before~~, we assume that stomatal uptake of Hg is the major pathway of Hg into tree rings and that Hg uptake by roots or bark can be neglected. Moreover, radial translocation of Hg within trees (Arnold et al., 2018; Chellman et al., 2020; Gustin et al., 2022a; Liu et al., 2024; Nováková et al., 2021) can hardly explain our tree-ring Hg records. Recent studies have further argued that besides atmospheric Hg concentration tree physiology may have an effect on the Hg record (Peng et al., 2024).

Moreover, neither our German tree-ring Hg records nor other published European tree-ring records (Fig. 4) match with the modeled Hg emission record for Europe since 1850 (Streets et al., 2017) or emission based modeled atmospheric Hg concentration (Horowitz et al., 2014). Although both, the modeled atmospheric Hg concentration curve and our oak tree-ring records show a steady increase starting at the end of the 19th century, the first increase in modeled global, hemispheric atmospheric Hg concentrations starting from the 1850 until about 1900 as well as the second strong increase between 1930 and 1940 are almost absent in our oak tree-ring records. The peak of atmospheric Hg concentration in the 1960s–70s is poorly visible and occurs only in some trees at the contaminated Stahlberg site, but not at the uncontaminated Leimen site. Here, the highest Hg concentrations are reached significantly later between 2002 and 2010. The transfer function between atmospheric Hg concentrations, the Hg content in leaves and finally in tree rings is not known. It is important to note that modeled global atmospheric Hg concentration increase by a factor of about seven between 1850 and 1970 (European emission increase 4-fold), whereas concentrations in oak tree rings increase by factors of 2 to 17 and 2 to 6 at the Stahlberg and Leimen site, respectively. Hg in Douglas fir tree rings show maximum Hg concentration between the 1955 and 1970 and all other trees show a decline after 1970 by factors between 1.5 and 2 to the Hg levels of the early 20th century. This differs from modeled atmospheric Hg concentrations which show a much weaker decrease of -1.5% and -2.2% per year in the past three decades (Obrist et al., 2018). Similar inconsistency of tree-ring records with hemispheric or global atmospheric Hg concentration or Hg emission records have been reported in previous studies and were explained mainly by local Hg emission sources (Scanlon et al., 2020). The strong deviation between the tree ring Hg data and atmospheric Hg records, along with the considerable variability observed both within and among our study sites and different tree species, indicates that additional local or regional factors—such as temperature, precipitation, air humidity, soil Hg emissions, and the physiological responses of different tree species to these factors—may play a critical role in for governing the accumulation of Hg in tree rings. If local or regional climatic factors determine the accumulation of Hg in tree rings to a considerable amount, in coniferous as well as broadleaves, common trends in tree-ring Hg records should be evident throughout large geographical distances. If this would be true, even when contaminated and uncontaminated sites of the same tree species are compared, the relative trend through time should be similar.

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3.2 Hg tree-ring records of the Northern Hemisphere

Figure 4 gives an overview over published tree-ring Hg records from sampling sites from different regions of all over the Northern Hemisphere. It becomes evident, that our Douglas fir tree-ring Hg record shows the same pattern as those of other coniferous (e.g. Pine, Spruce, Larch) and some broadleaved trees (e.g. Chestnut) from most other locations disregarding local Hg contamination, tree species, forest structure or geographical attributes. However, as seen for the German Douglas fir site, the strong decline in tree-ring Hg concentration by 2–3 standard deviations in the past 50 years do not correspond to the evolution of global/hemisphere atmospheric Hg concentrations, showing a Hg decrease to a much lower extent and a delay in time. This discrepancy suggests that a global or hemispheric underlying factor, or a combination of multiple factors, may exert a bigger larger influence on tree-ring Hg concentrations than atmospheric Hg levels alone. The synchronous widespread spiking of tree-ring Hg records in the ~1970s with their subsequent rapid decline until the year 2000 could needs to be driven by an at least-hemispherical-wide acting factor. Climate Changes in air temperature occurs continental-wide and changes in precipitation air temperature often appears on regional scale, and thus may in particular act as a dominant tree-growth factors, which could may explain their common pattern. The investigated broadleaved oak trees—from contaminated and uncontaminated sites—show a similar relative Hg pattern, which underlines that site-unspecific factors do influence Hg accumulation, like climate, soil type, degree of foliage.

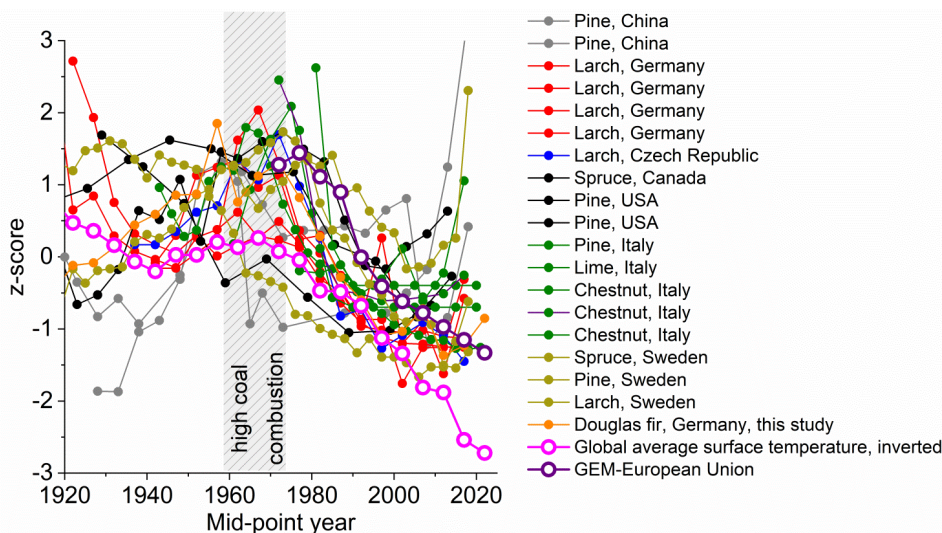


Fig. 4: Records of tree-ring mercury concentration (filled points) at different sites in the Northern Hemisphere (colors refer to countries) Scanlon et al., 2020; Chellman et al., 2020; Ghotra et al., 2020; Nováková et al., 2021; Wang et al., 2021; Baroni et al., 2023; Peng et al., 2024; Fornasaro et al., 2023 and records (circles) of global average surface temperature (inverted) and atmospheric mercury concentration emitted in the European Union.

3.3 The influence of climatic factors on Hg accumulation in oak and Douglas fir tree rings

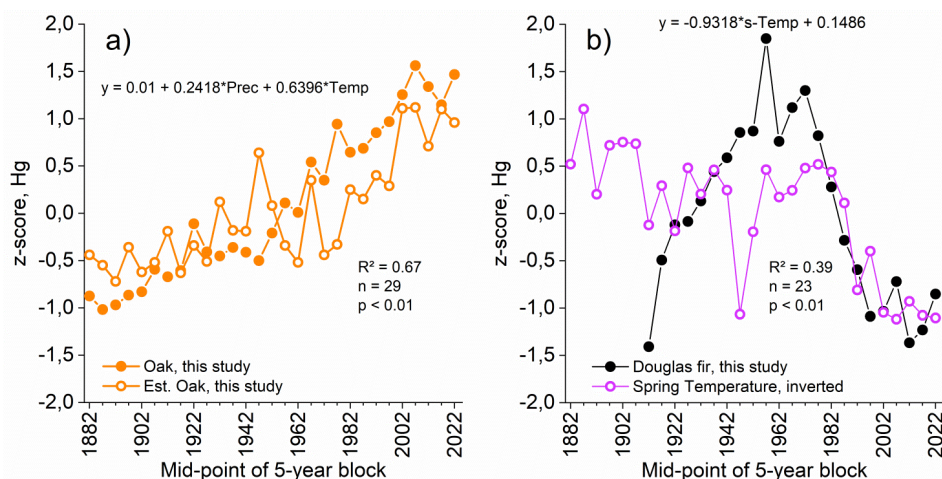
Previous studies have shown that the stomatal uptake of Hg by trees has a physiological component which is controlled by seasonality (Jiskra et al., 2018). Furthermore, temperature and humidity may favor or impede growth and increasing atmospheric CO₂ concentrations will increase plant growth and thus the leaf surface/stomata density and likely the Hg uptake by leaves. Zhang et al. (2016) projected that by 2050 the annual mean Hg(0) dry deposition flux over land will increase by 20% in

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northern mid-latitudes, driven by a combination of increased atmospheric gaseous elemental Hg exposure and increased vegetation and foliage density induced by CO₂ fertilization.

375 In order to investigate the impact of climatic parameters on oak and Douglas fir tree-ring Hg records from this study, spring and summer air temperature and precipitation data were used. The results of these climate-response analyses differ between the investigated coniferous (Douglas fir) and broadleaved (oak) tree species (Fig. 5). Hg in oak tree rings is significantly increasing with rising spring-summer air temperature and spring-summer precipitation sum. Spring-summer temperature influences tree-ring Hg roughly by 2/3 and precipitation sum by 1/3 (for exact weights see formula in Fig. 5a). By weighting these climatic parameters, the tree-ring Hg values can be estimated. The original and estimated oak tree-ring Hg records are significantly related ($R^2 = 0.67$, $p < 0.01$). However, the estimation is slightly off during the 1970s, which may be attributed to an extremely hot and extraordinarily dry year in 1976 and pronounced effects of air pollution (esp. Hg emissions from coal burning).



390 **Fig. 5:** Climate response of tree-ring mercury (Hg) concentration variability for oak (a) and Douglas fir (b). Tree-ring Hg concentration in oak trees can be estimated by spring-summer mean air temperature (Temp) and spring-summer precipitation sum (Prec) and by spring mean air temperature (s-Temp) for Douglas fir trees (b).

395 On the other hand, decreasing Hg in Douglas fir tree rings is also related to climate-change. Here, the concentration of Hg is decreasing with rising spring temperature and is thus showing a statistically significant negative relationship ($R^2 = 0.39$, $p < 0.01$, Fig. 5b). The patterns of Douglas fir tree-ring Hg and spring temperature are much closer related during the past 100 years and almost identical since the past 40 years. The extraordinary warm decade (1945–1954) after the World War II did not result in lower Douglas fir tree-ring Hg, as should be expected by the before mentioned negative relationship. Instead, the Douglas fir tree-ring Hg record is peaking around this decade likely due to the distinct Hg emissions during 1950s–1970s (coal combustion).

405 Site-climatic factors, namely temperature and precipitation, appear to significantly control Hg accumulation in oak and Douglas fir tree rings. In addition, Hg evasion from soil is known to increase with temperature (Obrist et al., 2018). Several studies have shown, that high temperature leads to nearly complete or entire stomatal closure and suppressed sap flow (Samuelson et al., 2019; Niessner et al., 2024; Irvine et al., 1998) blocking foliage Hg uptake and as a consequence Hg accumulation in tree rings in coniferous trees. Rising temperature at a coniferous site should therefore result in lower tree-ring Hg concentration, which is also true for our investigated Douglas

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firs, with exception ~~for~~of the past 20 years when hemisphere-wide tree-ring Hg records are compared to global average surface temperature. Here, a disconnection can be observed which could be explained by an increase of biomass in boreal and temperate forests during the past two decades mirroring the global atmospheric CO₂ growth rate (Yang et al., 2023; Norby et al., 2024) and in consequence leading likely to a net rise in tree-ring Hg accumulation in many tree species. An additional explanation of this observation could be that rising temperature prolong tree's growing season facilitating extended Hg assimilation in relatively humid seasons (i.e. spring and/or autumn). This would also mean that seasonal prolongation by climate change could be an additional driving factor and leads to a disconnection of tree-ring accumulation in coniferous species and local hot temperature periods.

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To put such climate-response results into a broader context it is necessary to mention that not only local climate conditions play a crucial role in regards of tree-ring Hg concentration, but also how ~~the specific tree species do~~ respond physiologically to severe drought (Tardieu and Simonneau, 1998). One strategy (isohydric) to deal with drought is to reduce stomatal conductance (and to fully close the stomata under extraordinary dry conditions) as soil moisture decreases and atmospheric conditions get drier, which appears to be more frequent in mid- to high-latitude conifers. A vast number of tree-ring Hg records are established from conifers, underlining our results that Hg accumulation decreases with temperature-induced lower stomatal conductance. Another strategy (anisohydric) is to maintain stomata open under drought stress, allowing to perform photosynthesis. ~~This~~Such a behavior is often ~~found~~observed for in broadleaved trees.

Performing tree-ring Hg climate-response analysis for the here used oaks and interpreting the results accordingly, we see that tree-ring Hg accumulation in oak is positively correlated to site-specific changes of temperature and precipitation (Fig. 5). This is contrary to how other investigated tree species respond (Fig. 4) and needs to involve all aspects of oak physiology and site conditions to interpret these results. One aspect could be that the increase in tree-ring Hg with rising temperature and precipitation may be attributed to anisohydric eco-physiological features of oaks growing in the mid-latitudes and its temperate climate. These oaks are able to maintain sap flow and continue to operate stomatal conductance under hot and dry summer conditions (Bréda et al., 1993; Kunz et al., 2016; Raftoyannis and Radoglou, 2002; Gauthey et al., 2024), leading to continuous foliage Hg assimilation and tree-ring Hg accumulation.

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A mismatch between most tree-ring Hg records and the evolution of atmospheric Hg concentrations (Fig. 4) can be ~~found~~observed and may indicate that hemispheric or global trends in atmospheric Hg loads appear to be overwritten by (local) climate change effects. Relative changes in tree-ring Hg concentration as well as long-term trends suggest to be predominately controlled by transregional and hemisphere-wide climatic factors ~~such as temperature~~, controlling tree's stomatal conductance, the amounts of needles/leaves, annual growth rates as well as forest coverage.

4 Discussion

Our results show, that Hg concentrations in tree rings are in its magnitude strongly influenced by local anthropogenic emission sources, or natural, temperature-dependent high Hg degassing from soils.

Accordingly, ~~site~~ climatic factors, namely temperature and precipitation, significantly appear to significantly control Hg accumulation in oak and Douglas fir tree rings. A combination of high temperature and precipitation favor Hg uptake in oak whereas high temperature impede it in Douglas fir. In the case of coniferous tree species, such pronounced climate Hg responses are previously mentioned by other authors (Zhang et al., 1995; Eccles et al., 2020). Gustin et al. (Gustin et al., 2022b) and Liu et al. (Liu et al., 2024) mentioned that tree ring Hg in Ponderosa pine (California, USA) and Masson pine (Chongqing, China) are negatively correlated with temperature, respectively. Our data from Douglas fir supports the prevailing assumption that Hg uptake by foliage through stomata is lower when the trees are under drought stress caused by rising temperature or low precipitation. For example, high temperature leads to nearly complete or entire stomatal closure and suppressed sap flow (Samuelson et al., 2019; Niessner et al., 2024; Irvine et al., 1998) blocking foliage Hg uptake and as a consequence Hg accumulation in tree rings. Rising temperature at a coniferous site should therefore result in lower tree ring Hg concentration, which is true for our investigated Douglas firs, with exception for the past 20 years when hemisphere-wide tree-ring Hg

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470 records are compared to global average surface temperature. Here, a disconnection can be
observed which could be explained by an increase of biomass in boreal and temperate forests during
the past two decades mirroring the global atmospheric CO₂ growth rate (Yang et al., 2023; Norby et
al., 2024) and in consequence leading to a net rise tree-ring Hg accumulation in many tree species.
475 An additional explanation of this observation could be that rising temperature prolong tree's growing
season facilitating extended Hg assimilation in relatively humid seasons (i.e. spring and/or autumn).
Not only local climate conditions play a crucial role in regards of tree-ring Hg concentration, but also
how the trees do respond physiologically to severe drought (Tardieu and Simonneau, 1998). One
strategy (isohydric) to deal with drought is to reduce stomatal conductance (and to fully close the
480 stomata under extraordinary dry conditions) as soil water lowers and atmospheric conditions get
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485 contamination. However has been
We found that tree-ring Hg accumulation in oak is positively correlated to site-specific changes of
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(Fig. 4). The increase in tree-ring Hg with rising temperature and precipitation is likely attributed to
anisohydric eco-physiological features of oaks growing in the mid-latitudes and its temperate climate.
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Gauthey et al., 2024), leading to continuous foliage Hg assimilation and tree-ring Hg accumulation.
The mismatch between most tree-ring Hg records and the evolution of atmospheric Hg
concentrations (Fig. 4) indicates that hemispheric or global trends in atmospheric Hg loads appear
495 to be overwritten by climate change effects. Relative changes in tree-ring Hg concentration as well
as long-term trends suggest to be predominately controlled by transregional and hemisphere-wide
climatic factors such as temperature, controlling tree's stomatal conductance, the amount of
needles/leaves, annual growth rates as well as forest coverage.
Our findings imply that climate change, specifically rising temperatures and more frequent drought
500 events as well as increasing CO₂ concentrations, may have altered the role of forests in the global
biogeochemical Hg cycle. Although the relationship between tree-ring and leaf Hg concentrations is
not yet known, it is likely that our observed changes in tree-ring Hg concentrations in different tree
species alter litterfall Hg fluxes to soil which is the most important terrestrial vector of atmospheric
Hg to soil and adjacent aquatic systems (Obrist et al., 2021; Obrist et al., 2018; Zhou et al., 2021).
505 Moreover, global warming is forcing forest management in mid-latitudes to transfer coniferous
monocultural forest into mixed or broadleaf forests, which is likely to further alter Hg fluxes to soils.
During the past decades Europe's forests have increased by up to 9%, with an even ratio between
coniferous and broadleaved trees (Köhl et al., 2020; Palmero-Iniesta et al., 2021). Climate change
capable of affecting future forest productivity in different forest types in temperate and boreal regions
510 and the practice of forestry currently undergoes a transformation process (Park et al., 2014). One
forest management strategy is to expand (non-)native tree species mixture to build climate-resilient
forests (Bohn et al., 2018) and planting deciduous tree species could reduce climate-driven fire
occurrence (Terrier et al., 2013). Native drought-tolerant oak-rich forests, or forests containing tree
species with positive temperature Hg responses in general, can serve as a distinct sink for
515 atmospheric Hg with changing climate. Additionally, such forests are less vulnerable to large-scale
fires, release of Hg into the atmosphere could be reduced. Future forest tree species composition
depends on local to regional climate evolution and forest management strategies (Park et al., 2014).
Overall, this study demonstrates that most tree species in future mid- to high-latitude forest
ecosystems may be less capable to accumulate atmospheric Hg under climate change.

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520 **4 Conclusion and Implications**

525 The here presented results suggest that climatic factors, like temperature and precipitation, have a vast impact on the process of Hg accumulation in tree rings of oak and Douglas fir. Our findings imply that climate change, specifically rising temperatures and more frequent drought events as well as increasing CO₂ concentrations, may have altered the role of forests in the global biogeochemical Hg cycle. Although the relationship between tree ring and leaf Hg concentrations is not yet known, it is likely that our observed changes in tree ring Hg concentrations in different tree species alter litterfall Hg fluxes to soil which is the most important terrestrial vector of atmospheric Hg to soil and adjacent aquatic systems (Obrist et al., 2021; Obrist et al., 2018; Zhou et al., 2021). Moreover, global warming may forcing forest management in mid-latitudes to transfer coniferous monocultural forest into mixed or broadleaf forests, which is likely to further alter Hg fluxes to soils.

530 During the past decades Europe's forests have increased by up to 9%, with an ~even ratio between coniferous and broadleaved trees (Köhl et al., 2020; Palmero-Iniesta et al., 2021). Climate change capable of affecting future forest productivity in different forest types in temperate and boreal regions and the practice of forestry currently undergoes a transformation process (Park et al., 2014). One forest management strategy is to expand (non-)native tree species mixture to build climate resilient forests (Bohn et al., 2018) and planting deciduous tree species could reduce climate-driven fire occurrence (Terrier et al., 2013). Native drought tolerant oak rich forests, or forests containing tree species with positive temperature-Hg responses in general, can serve as a distinct sink for atmospheric Hg with changing climate. Additionally, such forests are less vulnerable to large scale fires, release of Hg into the atmosphere could be reduced. Future forest tree species composition depends on local to regional climate evolution and forest management strategies (Park et al., 2014). Overall, this study demonstrates that most tree species in future mid- to high latitude forest ecosystems may be less capable to accumulate atmospheric Hg under climate change.

545 The presented tree-ring Hg concentrations in oak and Douglas fir from western Germany and the comparison of literature data on Hg tree-ring records from the Northern Hemisphere of the past century and the related evolution of temperature and humidity suggest that Hg concentrations in tree rings are to a large extent determined by climatic conditions, which may partly overwrite pollution effects and the evolution of atmospheric Hg loads. These findings imply that climate change, specifically rising temperatures and more frequent drought events as well as increasing atmospheric CO₂ concentrations, may have altered the role of forests in the global biogeochemical Hg cycle. Although the relationship between tree-ring and leaf Hg concentrations is not yet exactly understood, it is likely that our observed climate related changes in tree-ring Hg concentrations in different tree species alter litterfall Hg fluxes to soil which is the most important terrestrial vector of atmospheric Hg to soil and adjacent aquatic systems (Obrist et al., 2021; Obrist et al., 2018; Zhou et al., 2021). Moreover, global warming may force forest management in mid-latitudes to transfer coniferous monocultural forest into mixed or broadleaf forests, which is likely to further alters the role of forests as a temporal Hg sink. During the past decades Europe's forests have increased by up to 9%, with an ~even ratio between coniferous and broadleaved trees (Köhl et al., 2020; Palmero-Iniesta et al., 2021). Climate change capable of affecting future forest productivity in different forest types in temperate and boreal regions and the practice of forestry currently undergoes a transformation process (Park et al., 2014). One forest management strategy is to expand (non-)native tree species mixture to build climate-resilient forests (Bohn et al., 2018) and planting deciduous tree species could reduce climate-driven fire occurrence (Terrier et al., 2013). Future forest tree species composition depends on local to regional climate evolution and forest management strategies (Park et al., 2014). Native drought-tolerant oak-rich forests, or forests containing tree species with positive temperature-

Hg responses in general, can serve as a distinct sink for atmospheric Hg with changing climate. Additionally, such forests are less vulnerable to large-scale fires, release of Hg into the atmosphere could be reduced.

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5 References

- 575 Arnold, J., Gustin, M. S., and Weisberg, P. J.: Evidence for Nonstomatal Uptake of Hg by Aspen and Translocation of Hg from Foliage to Tree Rings in Austrian Pine, *Environmental science & technology*, 52, 1174–1182, doi:10.1021/acs.est.7b04468, 2018.
- Baroni, D., Ancora, S., Franzaring, J., Loppi, S., and Monaci, F.: Tree-rings analysis to reconstruct atmospheric mercury contamination at a historical mining site, *Front. Plant Sci.*, 14, 1260431, doi:10.3389/fpls.2023.1260431, 2023.
- 580 Bohn, F. J., May, F., and Huth, A.: Species composition and forest structure explain the temperature sensitivity patterns of productivity in temperate forests, *Biogeosciences*, 15, 1795–1813, doi:10.5194/bg-15-1795-2018, 2018.
- Boonen, K., Shetti, R., Navrátil, T., Nováková, T., Rohovec, J., and Lehejšček, J.: Atmospheric mercury pollution recorded in conifer tree rings: Disentangling the effects of tree-ring width, water content, and climate on mercury concentrations, *Dendrochronologia*, 92, 126370, doi:10.1016/j.dendro.2025.126370, 2025.
- 585 Bréda, N., Cochard, H., Dreyer, E., and Granier, A.: Field comparison of transpiration, stomatal conductance and vulnerability to cavitation of *Quercus petraea* and *Quercus robur* under water stress, *Annales Des Sciences Forestieres*, 50, 571–582, 1993.
- 590 Chellman, N., Csank, A., Gustin, M. S., Arienzo, M. M., Vargas Estrada, M., and McConnell, J. R.: Comparison of co-located ice-core and tree-ring mercury records indicates potential radial translocation of mercury in whitebark pine, *The Science of the total environment*, 743, 140695, doi:10.1016/j.scitotenv.2020.140695, 2020.
- Eccles, K. M., Majeed, H., Porter, T. J., and Lehnherr, I.: A Continental and Marine-Influenced Tree-Ring Mercury Record in the Old Crow Flats, Yukon, Canada, *ACS Earth Space Chem.*, 4, 1281–1290, doi:10.1021/acsearthspacechem.0c00081, 2020.
- Fornasaro, S., Ciani, F., Nannoni, A., Morelli, G., Rimondi, V., Lattanzi, P., Coccozza, C., Fioravanti, M., and Costagliola, P.: Tree Rings Record of Long-Term Atmospheric Hg Pollution in the Monte Amiata Mining District (Central Italy): Lessons from the Past for a Better Future, *Minerals*, 13, 688, doi:10.3390/min13050688, 2023.
- 600 Gauthey, A., Kahmen, A., Limousin, J.-M., Vilagrosa, A., Didion-Gency, M., Mas, E., Milano, A., Tunas, A., and Grossiord, C.: High heat tolerance, evaporative cooling, and stomatal decoupling regulate canopy temperature and their safety margins in three European oak species, *Glob Change Biol*, 30, e17439, doi:10.1111/gcb.17439, 2024.
- 605 Ghotra, A., Lehnherr, I., Porter, T. J., and Pizaric, M. F. J.: Tree-Ring Inferred Atmospheric Mercury Concentrations in the Mackenzie Delta (NWT, Canada) Peaked in the 1970s but Are Increasing Once More, *ACS Earth Space Chem.*, 4, 457–466, doi:10.1021/acsearthspacechem.0c00003, 2020.
- Guo, W., Liu, M., Zhang, Q., Deng, Y., Chu, Z., Qin, H., Li, Y., Liu, Y.-R., Zhang, H., Zhang, W., Tao, S., and Wang, X.: Warming-Induced Vegetation Greening May Aggravate Soil Mercury Levels Worldwide, *Environmental science & technology*, doi:10.1021/acs.est.4c01923, 2024.
- Gustin, M. S., Dunham-Cheatham, S. M., Harper, J. F., Choi, W.-G., Blum, J. D., and Johnson, M. W.: Investigation of the biochemical controls on mercury uptake and mobility in trees, *The Science of the total environment*, 851, 158101, doi:10.1016/j.scitotenv.2022.158101, 2022a.
- 615 Gustin, M. S., Ingle, B., and Dunham-Cheatham, S. M.: Further investigations into the use of tree rings as archives of atmospheric mercury concentrations, *Biogeochemistry*, 158, 167–180, doi:10.1007/s10533-022-00892-1, 2022b.
- Horowitz, H. M., Jacob, D. J., Amos, H. M., Streets, D. G., and Sunderland, E. M.: Historical Mercury releases from commercial products: global environmental implications, *Environmental science & technology*, 48, 10242–10250, doi:10.1021/es501337j, 2014.
- 620 Irvine, J., Perks, M. P., Magnani, F., and Grace, J.: The response of *Pinus sylvestris* to drought: stomatal control of transpiration and hydraulic conductance, *Tree Physiology*, 18, 393–402, doi:10.1093/treephys/18.6.393, 1998.

- 625 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., and Dommergue, A.: A vegetation control on seasonal variations in global
atmospheric mercury concentrations, *Nat. Geosci.*, 11, 244–250, doi:10.1038/s41561-018-0078-
8, 2018.
- 630 Kang, H., Liu, X., Guo, J., Zhang, Q., Wang, Y., Huang, J., Xu, G., Wu, G., Ge, W., and Kang, S.:
Long-term mercury variations in tree rings of the permafrost forest, northeastern China, *Sci.*
China Earth Sci., 65, 1328–1338, doi:10.1007/s11430-021-9886-1, 2022.
- Köhl, M., Linser, S., and Prins, K.: *State of Europe's Forests 2020*, 2020.
- 635 Kunz, J., Räder, A., and Bauhus, J.: Effects of Drought and Rewetting on Growth and Gas
Exchange of Minor European Broadleaved Tree Species, *Forests*, 7, 239, doi:10.3390/f7100239,
2016.
- Li, F., Ma, C., and Zhang, P.: Mercury Deposition, Climate Change and Anthropogenic Activities: A
Review, *Front. Earth Sci.*, 8, doi:10.3389/feart.2020.00316, 2020.
- Liu, X., Wang, X., and Wang, D.: Assessment of tree-ring mercury radial translocation and age
effect in Masson pine: Implications for historical atmospheric mercury reconstruction, *Journal*
640 *of environmental sciences (China)*, 138, 266–276, doi:10.1016/j.jes.2022.10.027, 2024.
- Liu, X., Wang, X., Yuan, W., Wang, D., and Feng, X.: Tree rings recording historical atmospheric
mercury: A review of progresses and challenges, *Critical Reviews in Environmental Science and*
Technology, 1–18, doi:10.1080/10643389.2023.2250706, 2023.
- 645 McLagan, D. S., Biester, H., Navrátil, T., Kraemer, S. M., and Schwab, L.: Internal tree cycling and
atmospheric archiving of mercury: examination with concentration and stable isotope analyses,
Biogeosciences, 19, 4415–4429, doi:10.5194/bg-19-4415-2022, 2022.
- Montgomery, D. C., Peck, E. A., and Vining, D. C.: *Introduction to linear regression analysis*, A
Wiley interscience publication, Wiley, New York [u.a.], 641 pp., 2001.
- 650 Navrátil, T., Nováková, T., Shanley, J. B., Rohovec, J., Matoušková, Š., Vaňková, M., and Norton,
S. A.: Larch Tree Rings as a Tool for Reconstructing 20th Century Central European
Atmospheric Mercury Trends, *Environmental science & technology*, 52, 11060–11068,
doi:10.1021/acs.est.8b02117, 2018.
- Niessner, A., Ehekircher, S., Zimmermann, R., Horna, V., Reichle, D., Land, A., Spangenberg, G.,
and Hein, S.: Soil drought sets site specific limits to stem radial growth and sap flow of
655 Douglas-fir across Germany, *Front. Plant Sci.*, 15, doi:10.3389/fpls.2024.1401833, 2024.
- Norby, R. J., Loader, N. J., Mayoral, C., Ullah, S., Curioni, G., Smith, A. R., Reay, M. K., van
Wijngaarden, K., Amjad, M. S., Brettell, D., Crockatt, M. E., Denny, G., Grzesik, R. T.,
Hamilton, R. L., Hart, K. M., Hartley, I. P., Jones, A. G., Kourmouli, A., Larsen, J. R., Shi, Z.,
Thomas, R. M., and MacKenzie, A. R.: Enhanced woody biomass production in a mature
660 temperate forest under elevated CO₂, *Nature Climate change*, 14, 983–988, doi:10.1038/s41558-
024-02090-3, 2024.
- Nováková, T., Navrátil, T., Demers, J. D., Roll, M., and Rohovec, J.: Contrasting tree ring Hg
records in two conifer species: Multi-site evidence of species-specific radial translocation effects
665 in Scots pine versus European larch, *The Science of the total environment*, 762, 144022,
doi:10.1016/j.scitotenv.2020.144022, 2021.
- Nováková, T., Navrátil, T., Schütze, M., Rohovec, J., Matoušková, Š., Hošek, M., and Matys
Grygar, T.: Reconstructing atmospheric Hg levels near the oldest chemical factory in central
Europe using a tree ring archive, *Environmental pollution (Barking, Essex 1987)*, 304, 119215,
doi:10.1016/j.envpol.2022.119215, 2022.
- 670 Obrist, D., Kirk, J. L., Zhang, L., Sunderland, E. M., Jiskra, M., and Selin, N. E.: A review of global
environmental mercury processes in response to human and natural perturbations: Changes of
emissions, climate, and land use, *Ambio*, 47, 116–140, doi:10.1007/s13280-017-1004-9, 2018.
- Obrist, D., Roy, E. M., Harrison, J. L., Kwong, C. F., Munger, J. W., Moosmüller, H., Romero, C.
D., Sun, S., Zhou, J., and Commane, R.: Previously unaccounted atmospheric mercury

- 675 deposition in a midlatitude deciduous forest, *Proceedings of the National Academy of Sciences of the United States of America*, 118, doi:10.1073/pnas.2105477118, 2021.
- Palmero-Iniesta, M., Pino, J., Pesquer, L., and Espelta, J. M.: Recent forest area increase in Europe: expanding and regenerating forests differ in their regional patterns, drivers and productivity trends, *Forstw Cbl*, 140, 793–805, doi:10.1007/s10342-021-01366-z, 2021.
- 680 Park, A., Puettmann, K., Wilson, E., Messier, C., Kames, S., and Dhar, A.: Can Boreal and Temperate Forest Management be Adapted to the Uncertainties of 21st Century Climate Change?, *Critical Reviews in Plant Sciences*, 33, 251–285, doi:10.1080/07352689.2014.858956, 2014.
- Peckham, M. A., Gustin, M. S., and Weisberg, P. J.: Assessment of the Suitability of Tree Rings as Archives of Global and Regional Atmospheric Mercury Pollution, *Environmental science & technology*, 53, 3663–3671, doi:10.1021/acs.est.8b06786, 2019.
- 685 Peng, H., Zhang, X., Bishop, K., Marshall, J., Nilsson, M. B., Li, C., Björn, E., and Zhu, W.: Tree Rings Mercury Controlled by Atmospheric Gaseous Elemental Mercury and Tree Physiology, *Environmental science & technology*, doi:10.1021/acs.est.4c05662, 2024.
- 690 Perone, A., Cocozza, C., Cherubini, P., Bachmann, O., Guillong, M., Lasserre, B., Marchetti, M., and Tognetti, R.: Oak tree-rings record spatial-temporal pollution trends from different sources in Terni (Central Italy), *Environmental pollution (Barking, Essex 1987)*, 233, 278–289, doi:10.1016/j.envpol.2017.10.062, 2018.
- Raftoyannis, Y. and Radoglou, K.: Physiological responses of beech and sessile oak in a natural mixed stand during a dry summer, *Annals of Botany*, 89, 723–730, doi:10.1093/aob/mcf133, 2002.
- 695 Rinn, F.: *TSAP-Win Professional*, RINNTECH, Heidelberg, 2002-2013.
- Samuelson, L. J., Stokes, T. A., Ramirez, M. R., and Mendonca, C. C.: Drought tolerance of a *Pinus palustris* plantation, *Forest Ecology and Management*, 451, 117557, doi:10.1016/j.foreco.2019.117557, 2019.
- 700 Scanlon, T. M., Riscassi, A. L., Demers, J. D., Camper, T. D., Lee, T. R., and Druckenbrod, D. L.: Mercury Accumulation in Tree Rings: Observed Trends in Quantity and Isotopic Composition in Shenandoah National Park, Virginia, *J. Geophys. Res. Biogeosci.*, 125, doi:10.1029/2019JG005445, 2020.
- 705 Siebelt, L.: Investigation of the role of mercury input through litterfall in forest soils at a former mercury mining site (Moschellandsberg), Thesis, Institute of Geoecology, Environmental Geochemistry group, TU Braunschweig, Braunschweig, 2024.
- Siwik, E. I. H., Campbell, L. M., and Mierle, G.: Distribution and trends of mercury in deciduous tree cores, *Environmental pollution (Barking, Essex 1987)*, 158, 2067–2073, doi:10.1016/j.envpol.2010.03.002, 2010.
- 710 Streets, D. G., Horowitz, H. M., Jacob, D. J., Lu, Z., Levin, L., Schure, A. F. H. ter, and Sunderland, E. M.: Total Mercury Released to the Environment by Human Activities, *Environmental science & technology*, 51, 5969–5977, doi:10.1021/acs.est.7b00451, 2017.
- Tardieu, F. and Simonneau, T.: Variability among species of stomatal control under fluctuating soil water status and evaporative demand: modelling isohydric and anisohydric behaviours, *Journal of Experimental Botany*, 49, 419–432, doi:10.1093/jxb/49.Special_Issue.419, 1998.
- 715 Terrier, A., Girardin, M. P., Périé, C., Legendre, P., and Bergeron, Y.: Potential changes in forest composition could reduce impacts of climate change on boreal wildfires, *Ecological applications a publication of the Ecological Society of America*, 23, 21–35, doi:10.1890/12-0425.1, 2013.
- 720 Wang, X., Yuan, W., Lin, C.-J., Wu, F., and Feng, X.: Stable mercury isotopes stored in Masson *Pinus* tree rings as atmospheric mercury archives, *Journal of hazardous materials*, 415, 125678, doi:10.1016/j.jhazmat.2021.125678, 2021.
- 725 Wang, X., Yuan, W., Lin, C.-J., Zhang, L., Zhang, H., and Feng, X.: Climate and Vegetation As Primary Drivers for Global Mercury Storage in Surface Soil, *Environmental science & technology*, 53, 10665–10675, doi:10.1021/acs.est.9b02386, 2019.

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Yang, H., Ciais, P., Frappart, F., Li, X., Brandt, M., Fensholt, R., Fan, L., Saatchi, S., Besnard, S., Deng, Z., Bowring, S., and Wigneron, J.-P.: Global increase in biomass carbon stock dominated by growth of northern young forests over past decade, *Nat. Geosci.*, 16, 886–892, doi:10.1038/s41561-023-01274-4, 2023.

730 Zhang, H., Holmes, C. D., and Wu, S.: Impacts of changes in climate, land use and land cover on atmospheric mercury, *Atmospheric Environment*, 141, 230–244, doi:10.1016/j.atmosenv.2016.06.056, 2016.

735 Zhang, L., Qian, J.-L., and Planas, D.: Mercury concentration in tree rings of black spruce (*Picea mariana* Mill. B.S.P.) in boreal Quebec, Canada, *Water Air Soil Pollut*, 81, 163–173, doi:10.1007/BF00477263, 1995.

Zhou, J., Obrist, D., Dastoor, A., Jiskra, M., and Ryjkov, A.: Vegetation uptake of mercury and impacts on global cycling, *Nat Rev Earth Environ*, 2, 269–284, doi:10.1038/s43017-021-00146-y, 2021.

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Data availability

Tree-ring mercury concentration data of this study (sessile oak, Douglas fir) can be received from the following repository: Doi is added after acceptance.

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Author contributions

AL sampled the trees, dated and cut the tree rings, performed statistical analysis, interpreted the data, discussed the results and wrote the manuscript. AN sampled the trees, dated and cut the tree rings, discussed the results. JF interpreted the data, discussed the results and wrote the manuscript. PS performed the tree-ring mercury measurements. HB performed the tree-ring mercury measurements, interpreted the data, discussed the results and wrote the manuscript.

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Competing interests

The authors declare no competing interests.