Response to Frank Wania

The referee comments are in black, while the author comments are in **bold print** and blue.

A well written paper on an important subject, based on a large new dataset that is analyzed thoroughly and rigorously. I particularly like the practical conclusions on how foliar uptake of mercury could be practically implemented in mercury fate models. I find little to criticise.

Thank you for the positive feedback to this manuscript and your comments.

Line 49: Delete comma before "during" and add "the water" before "vapor".

revised as suggested

Line 51: Delete comma before "during".

revised as suggested

Line 52: Delete "corresponding".

revised as suggested

Line 54: This sentence appears somewhat unmotivated in the abstract. Finding a proxy for stomatal conductance during an entire growing season was probably not among the original objectives of this study and is too hypothetical at this stage to merit inclusion in the abstract. It distracts from the main message. It also only merits a single sentence in the entire main body of the paper (line 480).

We agree that our suggestion to use foliar Hg concentration as a proxy for stomatal conductance is not discussed enough in the manuscript to merit a mention in the abstract. We will therefore delete the sentence in the abstract.

Line 65: use "exposure" instead of "exposition".

revised as suggested

Line 110 and line 122: 3515 foliage samples versus 3569 foliage samples. What is the reason for that discrepancy? Does either of these numbers include the outliers identified in line 165?

Thank you for the careful reading. The number of Hg concentrations in the dataset, on which the analysis is based, is indeed 3,569. After revision of all numbers describing the dataset, we understood that there must have been a confusion when counting the total sample number brought together from the data sets of ICP Forests, non ICP Forests (but provided by ICP Forests countries, only few samples) and Austrian Bio-Indicator Grid, which were all used together in the analysis. The total number of values of this dataset after blank correction is 3,569 Hg concentrations measured in 2,129 samples provided by ICP Forests countries (from both ICP Forests and non ICP Forests plots;

published at https://doi.org/10.5281/zenodo.5495179) and in 1,440 samples from the Austrian Bio-Indicator Grid. We will consistently change the numbers and equally check the numbers of subsets of samples per tree species and needle age class.

Line 137 to 138: Brandenburg and Baden-Wuerttemberg are not countries.

We will change "ICP Forests countries" to "ICP Forests members".

Line 200: The text prior to here makes reference to multiple needle year classes (Line 117/141, line 127-130). The sentence "by normalizing foliar Hg concentrations of samples to their respective life period in days from the beginning of the growing season (leaf flushing) to date of harvest" raises the question of how was this done for needles older than 1 season. (Judging from page 18 and figure 7, it appears that older needles were not subjected to the normalisation procedure and were used in an entirely separate analysis.)

Initially, we calculated daily Hg uptake rates of needles by dividing Hg concentrations with the respective number of days during the current-season needle life period + 356 days * needle age in years (e.g. 0; 1 year; 2 years etc.). However, the problem with this approach is, that we expect needle Hg uptake rates to slow down in multiple year needles (Wohlgemuth et al., 2020), which makes an average needle Hg uptake rate over the whole life period of older needles non-representative for a single growing season. Furthermore, the needle Hg uptake rate in winter and early spring is still unclear. Due to these issues, and because we directly compare foliar Hg uptake rates of broadleaves and needles, we decided to use Hg data from older needles exclusively in a separate analysis presented in Sect. 3.7. To make this clear, we will amend the sentence: "We therefore calculated foliar Hg uptake rates of current-season samples by normalizing foliar Hg concentrations to their respective life period…"

Line 216: Make it clear that you refer to water vapour here. This is advisable as there could in principle also be a mercury vapor pressure deficit.

We will explicitly specify water vapor pressure.

Line 250: Use "example" instead of "exemplary"

revised as suggested

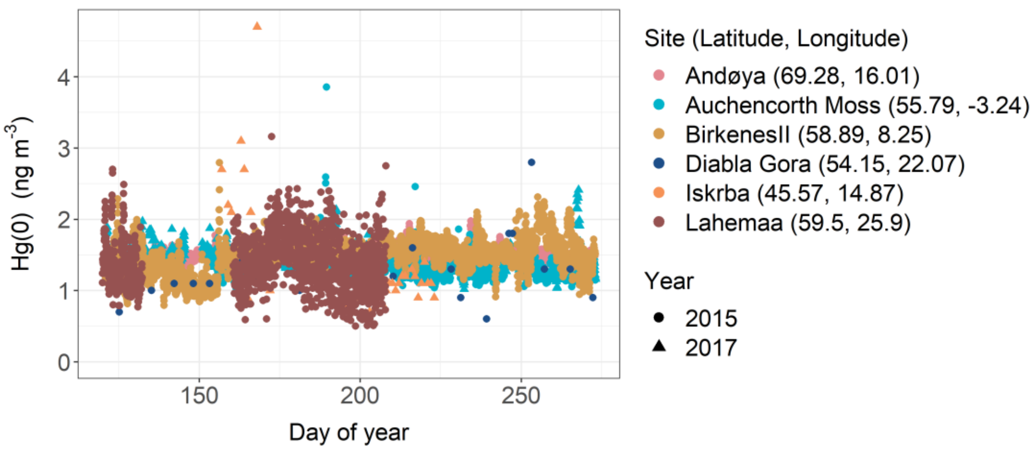
Figure S5: Maybe state that current-season needles are displayed.

The caption of Fig. S5 includes: "All samples represent current-season values."

Section 3.2 The comparison of foliar uptake rates across space suggests that variations in GEM concentrations in the atmosphere (in space and time) are deemed not to be important. That is likely correct, but should still be stated explicitly.

This is also important as some of the parameters explored later (soil dryness, VPD) could have a geographic component. You have to exclude the possibility that some of the observed relationships with these parameters are not artefact caused by a correlation of the parameters with atmospheric GEM concentrations (e.g. lower atmospheric GEM levels in the more water-stressed southern parts of Europe).

We agree that variations in GEM concentrations between sites have to be discussed, since it could indeed impact the results of the analysis. Unfortunately, we do not have GEM data available from any of the sampled forest sites to normalize foliar Hg uptake rates. In order to get a better understanding of GEM variation in Europe, we downloaded accessible EMEP Hg air data from terrestrial European stations between May 1st and Sept. 30th 2015 and 2017 from the EBAS database (http://ebas-data.nilu.no/Default.aspx). Like this, we obtained 11,900 Hg(0) values from 6 background sites (see figure below). We excluded available values from one site (Iskrba) in 2015 because air Hg values were abnormally low (0.41 ± 0.13 ng m^{-3}; mean ± sd).



Air Hg from EMEP measured at 6 different sites during May – Sept. 2015 and/or 2017.

We calculated the relative standard deviation of average air Hg values (rel.sd = 0.06) per site (including both years 2015 and 2017) to better represent the variation of Hg(0) among the sites. The relative standard deviation of daily Hg uptake rates of all tree species (Sect. 3.2) equals 0.64. We therefore will amend Section 3.2: "We were not able to normalize daily foliar Hg uptake rates with atmospheric Hg(0) concentrations at each respective sampling site and sample life period, as air Hg(0) measurements were unavailable for our sampling sites. The relative standard deviation of average air Hg(0) concentrations at 6 European measurement sites within the EMEP network between May and Sept. 2015 and 2017 (see Table S2 for details) was 0.06, which is lower than the relative standard deviation of the average daily Hg uptake rates between tree species and forest plots of 0.64 (Error! Reference source not found.**). We therefore argue, that the pronounced differences in median daily foliar Hg uptake rates between tree species cannot be exclusively explained by differences in atmospheric Hg(0) concentrations, but rather suggest a tree physiological cause. However, foliar Hg uptake rates should be normalized to ambient atmospheric Hg(0) concentrations, in particular when comparing foliar Hg observation between the northern and southern Hemisphere or over multi-decadal timescales."**

Additionally, we will add the following Section to the Supplement:

"In order to get a better understanding of the variation in atmospheric Hg(0) in Europe during the growing seasons 2015 and 2017, we obtained air Hg data from the European Monitoring and Evaluation Programme (EMEP). Air Hg measurements for 2015 and 2017 were available at 6 stations (Table S2). Selection of stations was based on availability of measurements in Europe at the relevant time intervals. Available measurements from one station (Iskrba) between May - Sept. 2015 were excluded from the dataset due to abnormally low air Hg values (0.41 ± 0.13 ng m^{-3}; mean ± sd). The temporal frequency of measurements (hourly to 6 days) and consequently the number of measurements varied between the different EMEP stations (Table S2)."

Table S 2. Details on air Hg measurements at 6 EMEP stations during the growing seasons 2015 and 2017.

Station name	EMEP code	coordinates (lat, lon)	altitude (m)	frequency	time coverage	air Hg (mean ± sd) (ng m^{-3})	n
Andøya	NO0090R	69.28, 16.01	380	hourly	May - Sept. 2015	1.50 ± 0.09	3371
Auchencorth Moss	GB0048R	55.79, -3.24	260	3hourly	May - Sept. 2015	1.33 ± 0.15	1384
				hourly	May - Sept. 2017	1.40 ± 0.12	2285
BirkenesII	NO0002R	58.39, 8.25	219	hourly	May - Sept. 2015	1.49 ± 0.24	3402
Diabla Gora	PL0005R	54.15, 22.07	157	6 days	May - Sept. 2015	1.26 ± 0.45	23
Iskrba	SI0008R	45.57, 14.87	520	daily	May - Sept. 2017	1.33 ± 0.80	39
Laheema	EE0009R	59.5, 25.9	32	hourly	May - July 2015	1.40 ± 0.38	1396

Concerning the possible impact of lower GEM in Southern Europe on the relationship of daily foliar Hg uptake rates with parameters explored in the manuscript, we believe, that this should not be a substantial issue for water VPD, since all VPD sites are located in Switzerland and Germany. We will amend the caption of Fig. 5: "All forest plots are located in Central Europe (latitude 46° - 54°), for which ambient air Hg(0) concentration is relatively constant (see Table S2 and Fig. S6)." With regard to the analysis of daily foliar Hg uptake rates and soil water, the range of latitude of evaluated forest plots is wider (41° - 55°) and potentially the impact of GEM on the presented relationship cannot be ignored. We will mention this caveat at the end of the respective section: "We also suggest determining the possible influence of additional parameters like gravel content and density of soils, tree root depth and atmospheric Hg(0), which could vary within the range of latitude (41° - 55°) of examined forest plots."

In Sect. 3.1 we will explicitly include the need to determine GEM when conducting experiments about the sensitivity of foliar Hg uptake to different parameters: "…the sensitivity of species-specific foliar Hg uptake normalized to air Hg(0) concentrations have to be determined in laboratory experiments with regards to elevated VPD, low soil water content or temperature."

Line 345: Figure 4 not 44

revised as suggested

Figure 4: Either use empty space at lower right for figure legend or vertically stack all three panels of the figure.

For a better overview, we will stack all three panels horizontally, with legend at the bottom.

Line 414: "In the future"

revised as suggested

Line 417: "gravel" is a soil parameter?

Thank you, correct is gravel content.

Line 424: When performing 54 linear regressions with a p value of 0.05, you would randomly expect 2 to 3 "significant" ones, even if there aren't any. Therefore, not too much should be made of the findings described on lines 423 to 427.

After repeating the analysis, we found, that there is no homoscedastic linear regression with a p value below the Bonferroni adjusted p value (here: 0.05/54 = 0.000925), i.e. the significant linear regression coefficients ($p < 0.05$) could indeed be false positive. We will state so in the manuscript: "None of the resulting 54 sets of linear regression coefficients were significant given a Bonferroni adjusted p-value = 0.000925." We will not further present the findings lines 423 - 427.

Line 463: "foliage takes up Hg(0) over the entire life time". Should this not be rephrased as "over the entire growing season" as the text earlier admits that mercury uptake during winter is poorly understood?

Yes, we will rephrase and amend the sentence: "We observed, that foliar Hg concentrations were highly correlated with foliage sampling date (Fig. 2), confirming the notion, that foliage takes up Hg(0) over the entire growing season and over multiple growing seasons in the case of coniferous needles (Fig. 7)."

Line 464: Again, I think it is necessary to state here that normalisation by the prevailing GEM concentration in the atmosphere is required when comparing foliar Hg uptake rates from different sites. That could be relevant when comparing between foliage from different hemispheres, and between foliage from areas with large differences in mercury source strength.

We will add the following sentence to the Conclusion: "For reasons of comparability, foliar Hg uptake rates should ideally be normalized to ambient air Hg(0) concentrations when large variation in atmospheric Hg(0) is expected (e.g. between northern and southern hemispheres, in polluted regions or over long timescales).