

Interactive comment on “Mercury in coniferous and deciduous upland forests in Northern New England, USA: implications from climate change” by J. B. Richardson and A. J. Friedland

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We are grateful for the reviewers for insightful comments to improve our study and corrections found. Below, we outline in a point-by-point response how we addressed concerns through additional analyses and expanded our discussion where suggested.

Review Comment #1 Page 6, lines 16-18: In figure 4, the species is *Picea rubens* instead *Picea rubrus* as it in line 16. Please, check it.

Author response #1: The species has been corrected from *Picea rubrus* to *Picea rubens*.

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Review Comment #2 Page 7 line 22-23: This is only a doubt. The mass of O sub-horizons estimated from the samples separated in the laboratory was that for samples collected using the 3 15*15 cm sections or was that derived from the material removed when you used the 50*50 cm wooden frame?

Author response #2: The organic horizons were collected as three 15x15 cm blocks and separated in the laboratory. This has been clarified to “In the laboratory, the 15iCf 15 cm blocks of organic horizon were separated into Oi (litter layer), Oe (fermentation layer), and Oa (humified layer) horizons. Roots > 5 mm in diameter were removed and organic horizon layers were air-dried at 25 °C to a constant mass.”.

Review Comment #3 Page 8, lines 5-8: Only as consideration. Attending to soil pH values (which are strongly acid in most of the soils analyzed), the calculation of SOM by loss of ignition would not be necessary as SOM could be estimated directly using total C and the Van Bemmelen factor.

Author response #3: The Van Bemmelen factor utilizes makes an equally uncertain assumption but generally agrees to our SOM values in the mineral soil with 2 % of our LOI estimations.

Review Comment #4 Page 8, line 9: There is no information about the procedure for determination of particle size distribution in mineral soil horizons. Please, could you include a brief description or a reference of the method used?

Author response #4: We utilized the particle size analysis from Methods of Soil Analysis and have added the citation and reference for Gee and Bauder, 1986.

Review Comment #5 Page 11, lines 21-22: Did the authors consider any explanation for this difference in clay content among mineral soil horizons? Could be this related to podzolization?

Author response #5: We have added a brief explanation to the discussion about the difference in clay fraction within the soil profile, and we attribute the variation in clay

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to Al and Fe oxy-hydroxides from podsolization and cite the reference do Valle et al., 2005, *Chemosphere* 58, 779-792.

Review Comment #6 Page 12, lines 3-4: No asterisks were shown in figure 1 for E and Bs horizons. So, were the differences in soil pH also significant for these horizons between both types of forest?

Author response #6: The soil pH was not significantly different for the E and Bhs horizons and this has been corrected throughout the manuscript.

Review Comment #7 Page 14, lines 12-13: Regarding sesquioxides, there are several studies that revealed the role of these soil compounds in Hg retention and storage, especially in podzolic soils such as those assessed in the present study. Perhaps the authors could add some comments regarding this issue.

Author response #7: We have added the following sentences to address this point: "Sesquioxides, Al and Fe oxy-hydroxides, are important inorganic surfaces that may sorb Hg directly, or provide surfaces for organo-mineral complexation (Garbriel and Williamson, 2004; do Valle et al., 2005). These are particularly of interest for the Bhs horizons, which have accumulations of both sesquioxides and SOM."

Review Comment #8 Page 14, lines 24-25: This is only a personal opinion to put under author's consideration. Perhaps the reduction (24%) in Hg soil pool due to potential changes in vegetation could be overestimated. It should be take into account that part of the Hg storage in the soil, especially for mineral soil horizons, is consequence of long-term soil processes which extent in time more than a few decades, which could be probably the time spam for the change in vegetation from coniferous to deciduous forest. The idea behind this comment is that, Hg pool in mineral horizons is expected to be less dynamic than that from organic horizons as in them, Hg is probably bound (strongly bound) to well humified organic matter or Fe and Al oxyhydroxides. Thus, the response of the Hg pool in mineral horizons could be not significantly modified due to a vegetation change.

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Author response #8: We completely agree that changes to the mineral soil horizons Hg pool is controlled by many other processes than vegetation and on a different time scale as well. However, the main conclusion from Page 14 is that there is a 24% reduction in the total soil pool, primarily due to differences in the organic horizons. The organic horizons are expected to be dynamic and directly influenced by the vegetation.

Review Comment #9 Page 15, lines 13-14: It seems, from figure 3, that the greater differences in EHg occur in the Oa subhorizons and in the E horizons. Please, check the sentence you wrote in lines 13-14 of this page.

Author response #9: All EHg concentrations were similar between vegetation type and with depth. This has been corrected throughout the manuscript.

Review Comment #10: Page 15, lines 18-19: In regard to this comment, the change in vegetation suggested by the authors could lead to a minor acidification in organic horizons which, at the same time, could result in a minor production of dissolved organic matter which is considered one of the main Hg transporters in podzols. Did you consider the Hg mobility in these terms and their potential consequences in a scenario of vegetation change?

Author response #10: We observed a more alkaline pH at deciduous stands, and thus a shift in vegetation would be expected to raise the pH. We now briefly mention this potential effect at the end of subsection 3.2.2.

Review Comment #11 Page 16, lines 3-14: Regarding this paragraph, it is missed some information about the distribution of Fe and Al oxyhydroxides in the mineral horizons. Results from these soil compounds could help to a better interpretation of the greater exchangeability of Hg in mineral horizons. A comment related to this will be welcomed.

Author response #11: We have added the following sentence to the paragraph: "Sesquioxides, such as hematite, goethite, and gibbsite, and SOM in the Bhs horizon

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can increase Hg complexation (Gabriel and Williamson, 2004).”.

Review Comment #12 Page 16, lines 22-23: I consider that could be better to maintain the use of the scientific names of the trees throughout the text, instead of the local/common names.

Author response #12: This has been adopted, now only scientific names are used consistently through the text.

Review Comment #13 Page 17, line 6: Is there any reason to use capital letters for common tree names when in the previous page you used lower case letters?

Author response #13: We now only use the scientific names, where the genus is capitalized and the species is lower case.

Review Comment #14 Page 17, lines 22-23: This comment is, in general, related to this subsection (3.3.1). It is worried that, although the authors compared and discussed widely with other studies, there was no references to your own data of foliar Hg concentrations. Please, could you do somewhat about this?

Author response #14: We previously made two references to our own foliar Hg concentration data in subsection 3.3.1. However, we have added to the discussion and have added the following sentence: “For example, the Hg concentration for the foliage of *Picea rubens*, one of the three coniferous species, was not significantly different than the foliage of *Acer* spp., and *Betula* spp., common deciduous genera (Fig. 4).”.

Review Comment #15 Page 18, lines 20-21: In figure 5, data of Hg pools in organic and mineral horizons from deciduous-dominated stands are not in bold as the other values of Hg pools and fluxes. Please, check it.

Author response #15: The deciduous-dominated stands pools values have been emboldened.

Review Comment #16 Page 18, lines 25-27: Part of this sentence is repeated, as it

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was already considered in a previous phrase. Please check it.

Author response #16: This has been corrected and now reads: “In spite of two-thirds less woody biomass at coniferous stands than deciduous stands, the aboveground THg pools at coniferous stands were greater than deciduous stands.”.

Review Comment #17 Page 20, lines 3-12: This comment is regarding uncertainties in Hg MRT estimation treated in these lines. Could be the potential mobilization of Hg from organic horizons to mineral horizons, and its consequent accumulation in illuvial horizons of podzols, another source of uncertainty in MRT calculation in the studied soils?

Author response #17: This is certainly true and we have added the following sentences to the discussion in subsection 3.4: “Fourth, the mode of transport of Hg is not taken into account in the box models. Mercury transport via dissolved SOM leaching and illuviation in the mineral soil by organo-mineral aggregate transport are more complex than investigated.”.

Review Comment #18: Page 21, lines 20-21: I am somewhat disagree with the latter part of this conclusion. The authors consider that physico-chemical soil properties are scarcely involved in Hg mobility. I think that this perspective is due to the approach used by the authors to asses the fraction of Hg mobility. Possibly, as in podzols is widely recognized that Hg could be mobilized by DOC, other soil extractions more closely related with organic matter dynamics (such as Na-pyrophosphate) could be a better approach to determine a true mobile Hg fraction in soil. So, I suggest that the authors should consider that the approach used to estimate Hg mobility was not the more suitable one.

Author response #18: We do not discuss the physicochemical soil properties with respect to mobility, but rather their influence on Hg accumulation in soil. We agree that the soil properties are important for Hg mobility but since we did not discuss DOC or other organic matter focused extractions, we limited our discussion on such topics. We

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have added the following sentence to the methods “We chose to investigate exchangeable Hg to consider dissolved Hg transport, rather than transport of Hg by dissolved organic compounds.”. Also we added the following sentence to our results and discussion section: “However, our method is limited in quantifying the pool of Hg that may be mobilized via particulate or DOC leaching. For this reason, increased Hg mobility may have been better examined by quantifying Hg bound to mobile forms of organic matter rather than EHg that may become mobilized after dissolution.”.

Review Comment #19: Page 21, lines 25-26: Could be this estimation wrong? 12 g ha⁻¹ * 7000 ha = 84000 g Hg, i.e. 84 kg Hg (instead of 840 kg). Could you check this?

Author response #19: This calculation was incorrect. I contacted the authors from which the 7000 ha estimate was obtained and they provided the correct area of the current coniferous area in New England, which is 2.2 million hectares. 12 g Hg ha⁻¹ across 2.2 million hectares is 27 Mg of Hg.

Review Comment #20: Page 22, lines 1-2: I would also include the possibility of the accumulation of Hg, leached from uppermost mineral soil horizons, in illuvial Bs/Bhs horizons of podzols soils. It should be consider the role of these subsurface soil horizons in Hg storage in upland soils, as well as its environmental function as an additional barrier against Hg mobilization to groundwaters and surface waters.

Author response #20: We have added illuviation as a potential process of Hg loss from the organic horizons.

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