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# *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

**Anonymous Referee #1**

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Richardson and Friedland present an interesting Dataset on a comparison of mercury in forest stands and soils between coniferous and deciduous forests. The authors found that the organic soil mercury pool in coniferous soils was significantly larger than the organic pool in nearby deciduous forests. Based on a simple box model they concluded that the mean residence time of Hg in coniferous forest soils was significantly longer than in deciduous forests. Given that with climate warming and more precipitation the forests will change more towards deciduous forests the authors conclude that also the coniferous forest soils will develop towards deciduous forests and as a consequence of this loose approx. 30 % of the mercury stored.

The study was carefully conducted and the paper is well written. I have however some



concerns about the box model that was established by the authors.

The authors developed a simple two-box model to calculate the mean residence time of mercury in organic and mineral soils. To set up this box model several simplifications were made. The authors also discuss several simplifications, which have been made what is very appreciated. Despite the mentioning of the simplification this leaves the question on how robust the results, the difference of mean residence time of Hg in coniferous and deciduous forest soils, are. The conclusions drawn from the difference in residence time are far reaching and therefore it is in my opinion justified to question the robustness of the model.

I think it would be important to provide more justifications for how the equations of the box model were set up. For example I cannot understand why the organic horizon does not have a volatilization flux but the mineral horizon has a volatilization loss. Also I cannot understand the reason why the input flux in the mineral soil depends on the litterfall flux and precipitation flux, which go into the organic horizon, or there is no dependency of the input flux in the mineral horizon on the size of the overlying organic horizon.

One important simplification was that the authors set the precipitation input equal for the two forest types. However other studies (e.g. Demers et al. 2007 and Blackwell and Driscoll 2015) found significant differences in throughfall Hg deposition between coniferous and deciduous forests. Demers et al 2007 for example found a 3 to 4.5 times higher througfall deposition in coniferous forests than in deciduous forests. In my opinion the negligence of this difference in throughfall deposition in the model needs to be shown to have an insignificant effect on the calculated mean residence times.

The contribution of litterfall to the total atmospheric deposition used in the model was approx. 50% for deciduous forests and only 10% for coniferous forests (Table S2). In particular the percentage of litterfall in coniferous forests is relatively low compared to studies measuring input fluxes with mass balance approaches (e.g. Gridal et al. 2000

Biogeochem, Demers et al. 2007 Ecol Appl., St Louis et al. 2001 ES&T). More recent studies based on stable Hg isotopes suggested even higher contributions of litterfall for deciduous (Demers et al. 2013, Glob. Giogeochem. Cycl.) as well as coniferous (Jiskra et al. 2015, ES&T) forests. It would be appreciated if the authors could discuss this discrepancy of litterfall contribution in particular in the coniferous forests compared to literature.

Another simplification is that the Hg deposition flux set to be constant to today's fluxes. It is clear that with changed in anthropogenic emissions the deposition flux has been highly variable in the last decades (e.g. Yin et al. 2014).

I think that a better justification for the model as it is set up as well as a sensitivity analysis on the appropriateness of the simplifications made would greatly enhance the robustness of the results.

Line Comments :

111465-L12-14 : Is an increase of precipitation expected for the whole globe or in Particular for the studied region ? Please provide a reference to climate model in addition to the Hg deposition model reference (Smith-Downey et al. 2010)

11467: What do the authors think is the reason for some sites being populated by coniferous forests and other sites by deciduous forests. Are there some ecological factors or some differences in the soil properties that favor one or the other type of forest stands?

114474-L20-L26: Given the high abundance of reduced sulfur in organic soils (in the order of 1mg/g) I find it hard to imagine that the sorption capacity for Hg (order of few hundred ng/g) is reached under natural uncontaminated conditions. Concerning Hg complexation by S in soils Skyllberg et al., 2006, ES&T might provide important insights

11475-L18-20 : Based on your measurements you estimate a approx. 5 to 10 times



lower litterfall flux for coniferous stands compared to deciduous stands (Table S2), how does this go along with the statement that : "Organic horizon Hg concentrations and pools may be greater at coniferous stands than at deciduous stands due to litterfall inputs..."?

11475-L2-4: The vertical profile of organic horizons also represent different age of the organic carbon (the lower the horizon generally the older is the organic carbon). Given that the atmospheric Hg deposition was very variable in the last decades to centuries (e.g. yin et al, 2014) the vertical Hg profile should also be discussed with respect to soil/OC age rsp. Time of deposition

1177-L1-L5 : If I understand this statement right, you suggest that with the change from coniferous forests to deciduous forests, the underlying soils would convert to soils similar to deciduous forests and therefore the Hg pools would adapt also to deciduous forest pools. The organic pools of coniferous forests have been formed from coniferous organic matter are therefore less decomposable (eg. higher C/N ratios and more lignin fraction as you explain in the introduction) than deciduous soils. I find it hard to understand how these soil conditions and as a consequence of that the Hg pool should change in short to mid-term when the vegetation on the soils changes. I would suggest to provide more explanation/justification for the suggested loss of 30 % of Hg in the soil pool based on vegetation change.

11483-L11-12 : On page 11481-L12 you concluded that the mean residence time of Hg in mineral soils of coniferous forests was significantly longer than of deciduous stands, how does this geo along with the statement : Âń We conclude that vegetation type significantly influenced Hg accumulation and retention in the organic horizons but not in the mineral horizons, which were controlled by soil properties."?

11483-L17: Did you mean microbial reduction and volatilization? (Hg can not be decomposed, please change the terminology)

Interactive comment on Biogeosciences Discuss., 12, 11463, 2015.