Reviewer #2: Review of the manuscript entitled "Mercury in coniferous and deciduous upland forests in Northern New England, USA: Implications from climate change" reference BG-2015-255

General Comments:

In my opinion, the contribution of this manuscript is under the scope of the journal and it addresses a relevant question regarding Hg biogeochemistry in forests ecosystems as it is the potential effect of climate change on tree vegetation substitution. To this, the authors have made a considerable effort in terms of sampling and analysing different components involved in Hg dynamics in forest ecosystems such as the own vegetation (both foliar a wood samples) and the soils, especially focused in the diversity of organic horizons, but also take into account the mineral soil. Although the paper does not present novel concepts or ideas for Hg behaviour in forest ecosystems, the existing ones were correctly applied as tools for the estimation of Hg residence in different soil layers which, in addition to valuable new data of Hg in soils and plant samples, are the true significant contribution of this paper to the wide body of scientific literature on Hg biogeochemistry in terrestrial ecosystems.

The title of the manuscript reflects in a suitable way the contents of the manuscript, being the abstract a good source of information for those readers who only can access to this part of the publication. The keywords reflect adequately most of the issues assessed along the manuscript and the introduction is very informative, considering several aspects around Hg dynamics in forest ecosystems, which is the central aspect of the paper. Moreover, the proposed objectives are clearly established and related to the subject under investigation, being all of them finally achieved.

Material and methods section is sufficiently informative for stands selection, soil and plant sampling and Hg analysis. Thus the description of the whole experiment done is complete and precise. However, although the authors made several assumptions to carry out calculations of mean residence time of Hg (MRT) in soil layers and most of them are valid in terms of their use for a rough approximation of Hg cycling in forest soils, some of them could be under discussion as they can create uncertainties in the final results. In this sense, it must be said that the authors already considered explicitly in the text the doubts that could be arise from some of these assumptions.

The results and discussion section is clearly presented as mathematical formulae, symbols, abbreviations, and units were correctly used, whereas the results exposed and their interpretations are strongly consistent. The conclusions outlined by the authors are mostly substantial and supported by the results obtained and their consequent interpretations. However, I have some discrepancies about some of the conclusions.

In general, the authors present in a suitable way its own study, particularly those aspects which could be considered as new contributions but, at the same time, take into account results from similar studies.

The manuscript is properly organized in terms of presentation, structure and length. The use of the language is precise, and all the tables and figures included in the manuscript, as well as the information included in the supplementary material, are useful. Finally, the number of references is adequate and they are consistent with the most recent studies published in the issue under the scope of the present manuscript.

As final consideration, I consider that the data set showed by the authors in this paper and their interpretations may contribute to extent the borders of the studies on Hg biogeochemistry in forest ecosystems.

Thus, in my opinion, the paper has sufficient originality and interest and it is suitable and deserve be published in Biogeosciences after **minor considerations**. The following points, which are only personal suggestions or opinions, could help to the authors to make some slight improvement in the manuscript content.

Specific Comments:

The pages and line numbers of the following comments come from the pdf version of the manuscript published in Biogeosciences Discussions.

<u>Material and Methods</u>:

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

Page 7 line 22-23: This is only a doubt. The mass of O subhorizons 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?

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.

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?

<u>Results and Discussion</u>:

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?

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?

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.

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.

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.

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?

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.

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.

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?

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?

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.

Page 18, lines 25-27: Part of this sentence is repeated, as it was already considered in a previous phrase. Please check it.

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?

<u>Conclusions</u>:

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.

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?

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.