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## ***Interactive comment on “Fate of mercury in tree litter during decomposition” by A. K. Pokharel and D. Obrist***

**A. K. Pokharel and D. Obrist**

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Fate of mercury in tree litter during decomposition

A.K. Pokharel and D. Obrist Biogeosciences Discuss., 8, 2593–2627, 2011

Dear Dr. Wang,

Please find below our detailed response to the two anonymous referees in regards to our manuscript. We greatly appreciate the thorough and excellent feedbacks and constructive critiques of both reviewers. Please find below a point-by-point response to the reviewer's comments.

Attached to this file, please find a revised Figure 3, and edited Tables 1 and 2. Please

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Discussion Paper



also find a supplemental file of the original BGD manuscript where all text are highlighted to reflect changes in the text that were performed to address the reviewer's concerns.

With best regards Daniel Obrist and A.K. Pokharel

#### Response to Reviewer Comments:

Reviewer 1: (i) In the materials and methods you mention that woody litter component were removed. Why do you choose to do so? Do you think that Hg associated with these types of debris could represent an important part of Hg dynamic in litter? The large majority of our samples was composed of foliage litter, and sticks and woody components were minor contributions to surface litter at all sites. We removed these woody components to facilitate comparison across sites, so that we were able to clearly compare the patterns of Hg in foliage litter across all four sites. We clarified this in the Methods Section.

(ii) I also have concern about the fact that litter samples were homogenized. I'm not sure of what does it mean (mixing/crushing the litter?). I wonder if the homogenization process will not enhance the degradation process of organic matter and emission of gaseous mercury. We simply well mixed leaves and needles for sample homogeneity but did not crush litter, so we don't expect this to affect observed patterns. We clarified this in the manuscript.

(iii) Finally, you used ultrapurified Millipore water in your controlled laboratory study. Is this water representative of rainwater? Do you think that rainwater composition (without Hg) could change something about the results obtained (I'm thinking about pH, chemical composition...). We discussed that we used (Hg-free) Millipore water to avoid adding new Hg to decomposing litter samples. We now clarify in the discussion section that the use Millipore water may have affected decomposition in the laboratory compared to field conditions (that receive regular rainwater), but that this was the only way to evaluate the behavior of Hg in litter without adding additional Hg inputs.

**BGD**

8, C1461–C1470, 2011

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(iv) I think that you should find a better way to present results of the field litter decomposition study. As you didn't present the results in table 1 (although you are referring to this table in section 3.3) and mix these results with the laboratory study in fig.3, it is very difficult for the reader to have a comprehensive vision of these data. You should add a figure or a table with initial and t=12 month values of all measured parameters during field study. The section 3.3 appeared quite confuse for me and I didn't really understand the way you calculate the Hg enrichment of field sample (lines 16 – 20 page 2605) and what is the difference with enrichment resented just above (lines 10-11 page 2605). This is a very good point. We added new Figure panels A and B in Figure 3 that now clearly show Hg concentrations and Hg/C ratios in field litter samples prior to (i.e., starting concentration) and after (i.e., after 12 months) of field exposure. We also clarified the degree of Hg enrichments in the text.

(v) Under the controlled laboratory study, you worked under dark conditions. In the discussion section, you didn't talk about the role of solar radiation on both Hg and SOM degradation/mineralization. These processes can explain differences between the field and the controlled conditions of the study. What can be the role of photodegradation of soil organic matter on Hg cycling in soils? This parameter may also explain differences of litter degradation observed with other studies (page 2607) (see selected paper below). This is a very good point that we now clearly discuss. We now mention and discuss that artificial conditions in the laboratory may account for differences to field results as well. Particularly, we mention that darkness in the laboratory may have caused less photoreduction (e.g., Graydon et al., 2008), and we mention effects of light exposure on litter decomposition (i.e., through reduction in litter decomposition; Austin and Vivanco, 2006).

(vi) The C/N ratio section (page 2607) will gain of being enhanced with further readings (selected paper below). The paragraph on Hg solubility is quite interesting and some aspect of the discussion may be related to studies that assessed the fate of mercury between terrestrial and aquatic ecosystems and more particularly the role of

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Interactive  
Comment

organic matter quality. Some authors have suggested that Hg sorption increased with increasing terrestrial organic matter degradation state (see selected paper below). We expanded our discussion in regards to organic matter degradation and links to Hg accumulation and sorption, the role of C/N ratio, and added suggested references (Grondin et al., 1995; Teisserenc et al., 2011). We also refer to the studies mentioned by the reviewer (Finzi et al., 1998; Cote et al., 2000; Smolander et al., 2005) to clarify that in addition to C/N ratios and/or degradation states, other biogeochemical processes likely differ between litter types (such as mineralization rates, microbial biomass, dissolved C and N dynamics).

Specific comments. (vii) From line 27 page 2596, to line 7 page 2597. This section should not be included within the introduction and should be removed. This information is already available in the Materials and Methods section. The introduction should end with objectives and hypotheses of research. We shortened this section and we understand the reviewer concerns about method descriptions in the introduction. However, we would like to keep a short overview of the methods here because it helps to clarify the detailed hypothesis that follow. The introduction does currently end with clear hypotheses.

(viii) Line 3, page 2597. Please define “N”. I wonder why you didn’t mention nitrogen or C/N ratio in the abstract? We feel that the link to C/N ratios is interesting, but that it is unclear to what degree C/N ratios directly versus decomposition rates or other biogeochemical processes contribute to these patterns (see above). We hence decided not to make this a major point in the abstract.

(ix) Sampling site description: A better way to present collection site may include a table with site description, location, elevation and precipitation. We feel that the current description is clear and accurate. We don’t think addition of a new table is necessary.

(x) Lines 3-5 and 12-14 page 2599, repetition. Please simplify. Done

(xi) Lines 3-4, page 2600 : Concentrations are measured, ratios are calculated. Please

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remove ratio from sentence. Done

(xii) Table 1 is quite difficult to read and huge amount of data is presented. Maybe you should present first data on C, N and C/N data and then Hg data. There is also a repetition between table 1 and Fig 1, 2 and 3. In the table you also present Dry mass/C concentration/C mass but in fig 1 you present Dry mass/C mass/C concentration. If you keep the table (maybe in annex) please be consistent. We changed that table to be consistent with Figure 1. We would like keep Table 1 as it shows the details of all results that other authors may find useful.

(xiii) Technical corrections. Line 21 page 2604 : Bonferroni - Line 16 page 2606 : Bonferroni - Line 9 page 2615 : Driscoll Done

Anonymous Referee #2 (xiv) In general it would be nice to have a few more details about how well the laboratory experiments mimicked the in situ decomposition process (for example the three week wetting period and its implications for evasion). Associated with this first point, what kinds of bounds need to be placed on the flux estimates provided in the experiments when trying to extrapolate to field conditions? Can this even be done with any confidence in the authors' opinions and if not what needs to be done to generate comparable field numbers? Given the simplification of the wetting and drying cycles in soils in the laboratory experiments (and the differing composition of rainwater) – what do the authors feel the experimentally mobilized fractions represent? This is an excellent point, and was also raised by reviewer #1. We clarified that differences in the field results may also be due to different experimental settings (e.g., solar radiation and temperature, Millipore water, and wetting/drying cycle. We also clarify in the summary section that it is unclear if and to what degree 1laboratory findings directly relate to in situ field conditions, and that the laboratory samples experienced artificial conditions where litter samples were kept in an artificial environment, were removed from soils, kept in darkness, and watered regularly with Millipore water."

(xv) A lot of the experimental results seem to be driven by the large temporal change

**BGD**

8, C1461–C1470, 2011

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[Interactive Discussion](#)

[Discussion Paper](#)



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in Hg concentrations Hg:C ratios etc in the aspen litter and contrasted by the pine litter example where little change was observed. Could the authors include in their discussion some explanation about the composition of the aspen litter and the types of carbon pools in these forests compared to the pine forest litter? For example, the lignins in the pine forests are known to be highly resistant to degradation and this helps to explain the observed results. Linking back the results presented in both the field and experimental analysis to fundamental understanding about the composition and types of different carbon pools would be very helpful for increasing the impact of this work. We added some discussion point in regards to differences between aspen and pine litter. However, we'd like to be cautious and not generalize too much (e.g., between coniferous versus deciduous litter types) based on results of only four litter types. We now clarify differences between litter types, refer to differences in C/N ratios and decomposition rates, and mention additional studies that discuss biogeochemical differences between litter types (Grondin et al., 1995; Teisserenc et al., 2011; Finzi et al., 1998; Cote et al., 2000; Smolander et al., 2005). We now also mention how our results compare to a large, continental-scale investigation across 14 forest sites that show that Hg accumulation patterns are mainly driven by south-to-north gradients, and less by litter species/type.

(xvi) I find the use of the term “sorbed” Hg throughout the manuscript confusing in that it is used to describe the combined processes of external inputs of Hg that are bound (sorbed) to the soil matrix. For clarity, can the authors be as specific as possible (e.g., inputs of atmospheric mercury to soils etc.). We clarified this point, we now specify what Hg we refer to when using the term “sorb” or “sorption”.

(xvii) Specific Comments: Page 2597, line 16, “un-decomposed” is redundant when referring to “fresh surface litter”. Corrected (xviii) Page 2598, line 21, I have not heard of these types of glass bottles for this type o experimental design. Is there a reason these jars were chosen? There is no specific reason other than soda-lime glass with PTFE-lined lids can be acid-cleaned well and should be very inert for Hg.

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Interactive  
Comment

(xix) Page 2600, lines 1-5: How much field variability was there between C:N:Hg ratios prior to decomposition study for a specific forest-litter type? If no estimates are available here is there some way of bounding this from previous work? Would there also be expected variability in decomposition rates across samples randomly collected at the sampling site? Is there any way to quantify this? We already present variability of both laboratory and field samples prior to exposure (we present means and standard deviations of replicate samples in tables and figures). In our experience, variability in Hg concentrations (or Hg/C ratios) are unlikely to be specific to forest types, but may possibly depend on the number of species present (Obrist et al., 2011, ES&T).

(xx) Page 2608, lines 15 onward – There is much discussion in the manuscript about “confirmation of hypotheses” that makes me a little uncomfortable since the questions themselves are not set up as testable hypotheses with a null that is rejected or accepted according to the scientific method. I don’t think this is a major problem but it would be nice if the authors could be more careful in how they imply they came to specific conclusions. Generally the experimental results point to another observation or support a premise that was not specifically “tested” here. That is a valid point and we agree. We reworded these parts in the discussion to reflect this concern.

(xxi) Page 2609, line 10-onward – The references cited throughout this manuscript are in general comprehensive. There is one recent paper by Smith-Downey et al, 2010 that we think would add to the discussion and interpretation of these results that is not cited. We added this paper in our discussion of Hg re-emissions upon decomposition (Smith-Downey et al., 2010, estimated a re-emission fraction of 16% upon decomposition of organic matter in soils).

(xxii) Page 2611, line 10: Why not look at the ranges of total deposition (wet+dry) from combined modeling and field observations for each of the different forest sites? We think that this would exceed the scope of this work, particularly given the large uncertainty such modeling would contain given that we don’t have any measurements of atmospheric Hg or speciation at any of these sites.

[Full Screen / Esc](#)[Printer-friendly Version](#)[Interactive Discussion](#)[Discussion Paper](#)

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**Interactive  
Comment**

(xxiii) Figures and tables - In general these are difficult to read at this time due to the size of font used. Please adjust for final publication. In addition, Table 2 repeats text and I would recommend avoiding such duplication. There are also examples in the text where the length could be shortened by referring to the Tables/Figures rather than repeating the results. We will work with the editorial office to adjust font size in Tables. We removed duplications in Table 2.

Please also note the supplement to this comment:

<http://www.biogeosciences-discuss.net/8/C1461/2011/bgd-8-C1461-2011-supplement.pdf>

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Interactive comment on Biogeosciences Discuss., 8, 2593, 2011.

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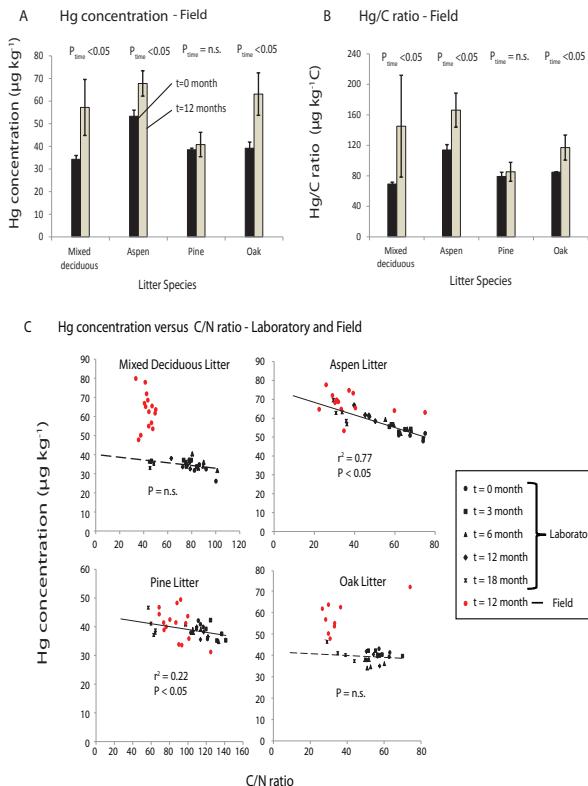
Interactive Discussion

Discussion Paper



Interactive  
Comment

Figure 3



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Discussion Paper

**Fig. 1.** Figure 3 - Revised

Table 1 : Dry mass, C mass, C concentration, Hg concentration, Hg mass, Hg/C ratio, N mass, N concentration, C/N ratio, and Hg/N ratio of laboratory samples.

Time (months)	Dry mass (g)				C mass (g)				C concentration (%)			
	Mixed deciduous	Aspen	Pine	Oak	Mixed deciduous	Aspen	Pine	Oak	Mixed deciduous	Aspen	Pine	Oak
0	24.1±0.4	23.9±0.3	21.3±0.6	11.1±1.2	11.70±0.30	11.29±0.99	10.80±0.29	5.33±0.58	48.56±0.67	47.28±0.53	50.80±0.48	47.90±0.14
3	22.2±0.5	22.5±0.9	20.9±0.4	10.6±1.1	11.01±0.13	10.24±0.40	10.59±0.28	5.11±0.55	49.72±0.50	45.54±0.81	50.65±0.43	48.16±0.55
6	21.8±0.5	21.3±0.4	20.6±0.4	10.4±1.2	10.94±0.24	10.15±0.20	9.25±0.20	4.41±0.54	50.26±0.40	47.66±0.23	44.98±0.52	42.50±0.58
12	19.9±0.5	16.8±1.0	19.9±0.5	9.5±0.6	9.54±0.20	7.70±0.52	9.70±0.24	4.38±0.29	47.74±0.40	45.70±0.31	48.88±0.30	46.22±0.42
18	19.4±0.3	16.4±0.9	19.7±0.6	8.6±0.7	9.64±0.20	7.52±0.47	10.08±0.25	4.09±0.34	49.58±0.43	45.87±0.61	51.27±0.48	47.40±0.79
Time (months)	Hg concentration (µg kg⁻¹)				Hg mass (ng)				Hg/C ratio (µg kg⁻¹C)			
	Mixed deciduous	Aspen	Pine	Oak	Mixed deciduous	Aspen	Pine	Oak	Mixed deciduous	Aspen	Pine	Oak
0	32.43±4.17	50.16±1.60	39.28±2.52	40.67±1.53	782±97	1198±30	834±57	452±48	66.8±8.9	106.1±3.3	77.4±5.6	84.9±3.3
3	35.52±1.67	54.60±1.51	37.85±2.97	40.81±1.21	788±53	1228±52	792±61	430±37	71.5±4.0	119.9±5.0	74.8±6.3	84.8±2.6
6	35.00±3.39	55.68±2.89	38.36±2.27	36.08±1.84	764±71	1174±44	790±46	378±55	69.7±7.3	115.6±6.3	85.3±5.0	84.9±3.9
12	34.94±2.14	61.82±3.08	37.87±1.83	38.99±2.67	700±62	1036±52	754±42	372±24	73.2±5.0	135.3±7.4	77.5±3.7	84.4±5.2
18	35.53±1.55	62.13±4.95	40.30±3.84	40.25±3.82	690±26	1016±31	792±84	348±36	71.7±3.5	135.6±12.2	78.6±7.5	84.9±9.0
Time (months)	N mass (g)				N concentration (%)				C/N ratio			
	Mixed deciduous	Aspen	Pine	Oak	Mixed deciduous	Aspen	Pine	Oak	Mixed deciduous	Aspen	Pine	Oak
0	0.134±0.011	0.161±0.013	0.096±0.007	0.090±0.012	0.56±0.05	0.67±0.06	0.45±0.03	0.81±0.05	87.76±8.68	70.55±5.66	113.25±6.72	59.21±3.43
3	0.142±0.008	0.166±0.013	0.081±0.006	0.088±0.010	0.64±0.03	0.74±0.04	0.39±0.03	0.83±0.09	77.61±4.38	62.09±4.50	130.81±8.76	58.69±6.93
6	0.123±0.010	0.164±0.012	0.084±0.009	0.083±0.010	0.56±0.04	0.77±0.07	0.41±0.05	0.80±0.05	89.40±7.71	62.37±5.18	111.22±13.75	53.03±4.14
12	0.130±0.015	0.167±0.012	0.085±0.004	0.081±0.005	0.65±0.06	1.00±0.09	0.43±0.02	0.86±0.06	73.82±6.54	45.94±3.89	114.36±5.31	54.05±3.48
18	0.210±0.007	0.227±0.011	0.164±0.011	0.116±0.020	1.07±0.03	1.40±0.12	0.83±0.05	1.35±0.19	46.23±1.40	33.20±3.00	61.77±3.28	35.86±5.81
Time (months)	Hg/N ratio (µg kg⁻¹N)											
	Mixed deciduous	Aspen	Pine	Oak								
0	5808.8±3784.7	7480.8±5919.1	8753.8±700.6	5027.0±334.7								
3	5541.9±340.2	7434.3±364.1	9748.9±582.3	4963.0±463.7								
6	6195.4±383.3	7187.9±167.8	9433.8±632.4	4497.7±348.1								
12	5385.5±336.6	6192.7±214.9	8849.5±374.6	4552.1±307.1								
18	3313.4±185.1	4474.9±187.2	4836.1±219.5	3018.7±334.2								

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