May 5, 2016

Dear Editor,

We appreciate the reviewers' suggestions which have considerably improved the manuscript (**acp-2016-7**). Enclosed are point-by-point responses to the reviewers. We hope that with these changes the manuscript will be suitable for publication in "**Atmospheric Chemistry and Physics**"

Thank you very much.

Sincerely,

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Response to Referee 2:

Comment 1

Abstract is written in the way with number reporting, there is no significant conclusions and any new discovery.

Response 1

As suggested, we revised the Abstract as follows on **Line 34 to Line 51**.

"In this study, mercury *(Hg) in dry and wet deposition, throughfall and litterfall, and Hg volatilization from soil were measured from August 2008 to February 2010 to identify the factors influencing the amount of atmospheric Hg deposited to forested areas in a temperate deciduous forest in Korea. For this location there was no significant correlation between the estimated dry deposition flux (litterfall + throughfall – wet deposition) (6.7 $\mu g\ m^{-2}\ yr^{-1}$) and directly measured dry deposition (9.9 $\mu g\ m^{-2}\ yr^{-1}$) likely due primarily to Hg losses from the litterfall collector. Dry deposition fluxes in cold seasons (fall and winter) were lower than in warmer seasons (spring and summer). The volume-weighted mean (VWM) Hg concentrations in both precipitation and throughfall were highest in winter likely due to increased scavenging by snow events. Since Korea experiences abundant rainfall in summer, VWM Hg concentrations in summer were lower than in other seasons. Litterfall fluxes were highest in the late fall to early winter when leaves were dropped from the trees (September to November). The cumulative annual Hg emission flux from soil was 6.8 $\mu g\ m^{-2}\ yr^{-1}$. Based on these data, the yearly deposition fluxes of Hg calculated using two input approaches (throughfall + litterfall or wet deposition + dry deposition), were 6.8 and 3.9 $\mu g\ m^{-2}\ yr^{-1}$ respectively. This is the first reported study which measured the amount of atmospheric Hg deposited to forested areas in Korea and thus our results provide useful information related to Hg fate and transport in this part of the world."*

Comment 2.

Line 69-71, the original papers were not cited and PBM is particle-bound Hg, how can it be adsorbed on PM? You could say oxidized Hg or GOM.

Response 2

As suggested, we revised text and reference as follows on **Line 70 to Line 71**.

"PBM is created by GEM or GOM adsorbing to a particle (Lai et al., 2011)."

Comment 3.

Line 72-74, re-write.

Response 3

As suggested, we revised as follows on Line **71 to Line 74**.

"..... Atmospheric PBM transport is significantly affected by its particle size distribution and may contribute to both wet and dry deposition (Lynam and Keeler, 2002). Wet and dry deposition of atmospheric Hg is an important input to the aquatic and terrestrial ecosystems"

Comment 4.

Line 84-86, not clear, also update the reference here Line 88-89

Response 4

As suggested, we revised and updated this text as follows on **Line 80 to Line 105**.

"Dry deposition to leaves compromises a large proportion of litterfall (Grigal, 2002; St. Louis et al., 2001). Previous investigations (Fu et al., 2009) estimated dry deposition to forested areas as litterfall + throughfall – wet deposition. However, there are many variables that can adversely influence this technique including reemitted Hg from beneath the canopy and sampling artifacts. Directly measuring dry deposition with a surrogate surface is an alternative approach, although there is no universally accepted method on how to make these measurements.

Hg deposited onto plant surfaces can be revolatilized, incorporated into tissue or washed off by precipitation (which is deemed throughfall) which often results in throughfall having higher Hg concentrations than precipitation (Iverfeldt, 1991; Kolka et al., 1999; Munthe et al., 1995; Choi et al., 2008; Grigal et al., 2000; Schwesig and Matzner, 2000).

Litterfall is dead plant material such as leaves, bark, needles and twigs that has fallen to the ground. Litterfall carries new Hg inputs from the atmosphere to the forest floor and also Hg recycled from volatilization from soils and other surfaces. Throughfall and litterfall contribute to the biochemical recycling of atmospheric Hg in forest systems (St. Louis et al., 2001) and are important Hg inputs that result in Hg accumulation in forest systems (Blackwell and Driscoll, 2015).

The deposition of Hg in the forest ecosystem is complicated because of complex interactions between atmospheric Hg and the canopy, including oxidation of Hg on leaf surfaces (Blackwell and Driscoll, 2015; Iverfeldt, 1991), deposition of GOM and PBM on leaf surfaces (Blackwell et al., 2014; Blackwell and Driscoll, 2015; St. Louis et al., 2001), stomatal uptake of atmospheric GEM (Fu et al., 2010; Iverfeldt, 1991; Lindberg et al., 1991; St. Louis et al., 2001), root uptake of dissolved Hg in soil and soil water and stomatal uptake of GEM that was volatilized from soils (Bishop et al., 1998; Cocking et al., 1995; Ma et al., 2015; St. Louis et al., 2001). Recycled Hg would increase throughfall and litterfall concentrations (St Louis. et al., 2001).”

Comment 5.

Line 77-79, add Selin et al., 2007 and Lindberg et al., 2007

Response 5

As suggested, we added Selin et al., 2007 and Lindberg et al., 2007 as follows on **Line 77 to Line 79**.

“…. , resulting in adverse health and ecological effects (Ma et al., 2013; Lindberg et al., 2007; Rolfhus et al., 2003; Selin et al., 2007; Weiss-Penzias et al., 2016; Zhu et al., 2014).”

Comment 6.

how does uptake via roots impact Hg deposition. Also stomatal uptake of Hg^0 emitted from soils?

Response 6

As suggested, we added information as follows on **Line 102 to Line 105**.

" .... root uptake of dissolved Hg in soil and soil water and stomatal uptake of GEM that was volatilized from soils (Bishop et al., 1998; Cocking et al., 1995; Ma et al., 2015; St. Louis et al., 2001). Recycled Hg would increase throughfall and litterfall concentrations (St Louis. et al., 2001)."

Comment 7.

I don't understand this sentence. Line 133, please discuss problems from using KCl coated quartz surface. Lyman et al., 2010; Huang et al., 2013/2015, McClure et al., 2015, Lynam and Keeler 2006 Sampling method, what are the time periods?

Response 7

As suggested, We revised and discussed problems from using KCl coated quartz surface as follows on **Line 141 to Line 154**.

"Some studies have investigated using a surrogate surfaces to measure dry deposition (Lyman et al., 2007; Peterson and Gustin, 2008). Surrogate surfaces is better control over exposure times than those provided with natural vegetation (Lai et al., 2011). Surrogate surfaces with cation exchange membranes could be collected very small aerosol fraction by diffusion (Lyman et al., 2007; Huang and Gustin, 2015b). However, we collected direct dry deposition using a surrogate surfaces with quartz filters. Similar to previous studies (Lai et al., 2011; Yi et al., 1996), the dry deposition sampler was equipped with a knife-edge surrogate surface (KSS) sampler with the collection media facing up. Forty seven-mm quartz filters were used to measure PBM deposition and KCl-coated quartz filters were used to measure GOM + PBM deposition. The quartz filter and KCl-coated quartz filter (soaked in KCl solution for 12h and dried on clean bench) were pre-baked in a quartz container at 900 ºC for PBM and 525 ºC for GOM + PBM. Before weekly sampling, the filters were placed on a filter holder base and held in place with a retaining ring and then were placed on the KSS. Filters exposed to the atmosphere for approximately one week and two side-by-side samples were deployed during each dry day."

Comment 8.

Analytical method, did the author develop the thermal desorption method? If not please cite references.

Response 8

As suggested, we added references as follows on **Line 187 to Line 188**.

"..... zero air passed through until the Hg concentration was zero (Kim et al., 2009; Kim et al., 2012)."

Comment 9.

If I understand this correctly, KCl QFF was heated to 525C and QFF was heated to 900C to separate GOM and PBM. Two questions here. 1. Is dry deposition collected up facing or down facing? and how up/down facing impact measurement? 2. Is this possible for GOM attach on QFF and quantified as PBM, and PBM attach on KCl-QFF and quantified as GOM?

Response 9

As suggested, we added information about sampling method of dry deposition as follows on **Line 141 to Line 154 and Line 192 to Line 193**.

Question 1 :

Some studies have investigated the use of surrogate surfaces to directly measure Hg dry deposition (Lyman et al., 2007; Peterson and Gustin, 2008). Surrogate surfaces allow better control over exposure times than those provided with natural vegetation (Lai et al., 2011). Surrogate surfaces with cation exchange membranes have been useful for measuring GOM however they may collect a very small aerosol fraction by diffusion (Lyman et al., 2007; Huang and Gustin, 2015b). Similar to previous studies (Lai et al., 2011; Yi et al., 1996), in this project the dry deposition sampler was equipped with a knife-edge surrogate surface (KSS) sampler with the collection media facing up. Forty seven-mm quartz filters were used to measure PBM deposition and KCl-coated quartz filters were used to measure GOM + PBM deposition. The quartz filter and KCl-coated quartz filter (soaked in KCl solution for 12h and dried on clean bench) were pre-baked in a quartz container at 900 ºC for PBM and 525 ºC for GOM + PBM. Before weekly sampling, the filters were placed on a filter holder

base and held in place with a retaining ring and then were placed on the KSS. Filters exposed to the atmosphere from approximately one week and two side-by-side samples were deployed during each dry day.

Question 2 :

It was assumed that GOM deposition was equal to the flux measured by the KCl-coated quartz filter minus the flux measured by the quartz filter.

Comment 10.

What is the recovery for the thermal desorption system? Recovery for Tekran 2537 direct injection 87% is too low usually from 93-107%. How many sampling time periods? Only 4 field blanks? Why? Volatilization from soil, what are MDL or blanks?

Response 10

As suggested, we revised section 2.4.1 and 2.4.4 as follows on **Line 209 to Line 214 and Line 232 to Line 235**.

"Automated daily calibration of Tekran 2537A routinely was performed using an internal permeation source. Two-point calibrations (zero and span) were performed separately for each pure gold cartridge. A recovery of 102 ± 2.9% (r^2 > 0.9995) (n = 4) was measured by directly injecting knowing amounts of five Hg standards which was connected to zero air. The Method Detection Limit (MDL) determined by measuring the Hg concentration in zero air was 0.04 ng m^{-3}. Additional information is described in the SI.....Before flux chamber measurements automated calibration was performed using the internal permeation source connected to the Tekran 2537A and Tekran 1110 dual sampling unit. External calibration and MDLs for this instrument are described above."

Comment 11.

Section 3.1, if you only have a short time period during each season, how can you really see the seasonal pattern? Please add more detail information for sampling plan.

Response 11

As suggested, we added information about sampling plan as follows on **Line 241 to Line 242**.

"Weekly sample were collected using quartz (PBM) and KCl coated quartz filters (GOM)"

Comment 12.

What statistical test are you using, please add information for every place you mention significant difference.

Response 12

As suggested, we added information about significant difference as follows on **Line 247, Line 265, Line 272, Line 290, Line 327, Line 396.**

"Nonparametric Mann-Whitney U tests....."

Comment 13.

Line 281, what is mechanical weathering?

Response 13

As suggested, we revised as follows on **Line 294 to Line 296**.

"Other possible sources of Hg in throughfall are leaching and biogeochemical recycling of Hg from foliage (St. Louis et al., 2001)."

Comment 14.

Most references are also out of date.

Response 14

As suggested, we added recent references.