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Interactive comment on "The effects of meteorological parameters and diffusive barrier reuse on the sampling rate of a passive air sampler for gaseous mercury" *by* David S. McLagan et al.

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Reviewer: This paper reports on work towards development of a passive sampler for gaseous Hg. The paper reports on tests to determine the impact of meteorological parameters on the sampler. They found that RH did not affect the sampling rate and it increased slightly with wind speed. They also assess the utility of reusing Radiello sampler housings. In general I do not think this paper is of sufficient quality and novelty to be considered for publication in AMT.

Response: The reviewer provides no evidence to support an assessment of insufficient quality or novelty. In our manuscript we describe tests on the effects of wind speed, temperature and relative humidity on the sampling rate of a previously published passive air sampler (PAS) for gaseous Hg (McLagan et al. 2016a). Quantifying the sampling rate variability caused by meteorological conditions is a critical step in validating any PAS. This work had not been previously done for this sampler, and we feel we have approached this with utmost rigor. Given AMT's mandate of "validation of measurement instruments" and "advances in…measurement techniques for constituents…of the earth's atmosphere", we feel strongly that this contribution fits well within the scope of this journal.

Reviewer: I do not really understand the part of the sentence that starts with "with" just does not make sense so I would eliminate.

Response: We do not know what the reviewer is referring to in this comment. There are two sentences in the entire manuscript that start with the word "with" (pg. 9, line 199 and pg. 12, line 278) and both are grammatically correct. In case the reviewer is referring to line 278, we can rephrase the sentence to read: "When the protective shield is in place the SR was approximately 10% lower than without the protective shield for the yellow Radiello®."

Reviewer: Also is the resolution of this sampler sufficient for collecting meaningful data around the globe given the limited variability in concentrations outside of contaminated areas?

Response: The replicate precision of this sampler is 2±1 % relative standard deviation (McLagan et al. 2016a). Whether this is sufficient "resolution" for discerning the relatively small concentration differences of gaseous mercury "outside of contaminated areas" depends to a large degree on the extent to which the sampling rate is variable between different deployments. In other words, the work described in the submission is a necessary step towards establishing whether the sampler has the requisite level of accuracy and precision to monitor background levels of gaseous Hg. Our study shows that the effect of wind speed, temperature and relative humidity on the sampling rate is small and quantifiable. While this is necessary, it not yet sufficient to establish the smallest concentration difference the sampler is able to detect. This will require the determination and comparison of sampling rates in the field at sites that vary widely in terms of meteorology. Such work is currently ongoing.

Reviewer: Pg 6 Methods section It appears they had no simultaneous replication and the experiment seems quite crude there is no n and just looking at parallel wind speed is not sufficient.

Response: We measured the sampling rate of the PAS with the white Radiello at eight different wind speeds (wind still, 1, 1.5, 2, 3, 4, 5, 6 m/s) and those for the yellow Radiello at three wind speeds (windstill, 3, 6 m/s). Each of these measurements was triplicated, for a total of 33 individual sampling rate determinations. The high n in our study is apparent from Figure 1. We believe that this is a sampling design that compares very favorably with any that had been adopted in previous studies examining the relationship between wind speed and sampling rate in PASs. The following statement (in red) will be added to line 126 on pg 6 to increase clarity: "...the distance between 126 PASs and fan (see Fig. S1 and Fig. S2). For each wind speed triplicate PASs were deployed."

We do not report the average and standard deviation of the replicated measurements at different wind speeds, because it is impossible to exactly replicate exposure of PAS to wind in a sampling design (Zhang et al. 2013) that relies on individual electrical fans for each individual deployment (Lines 126-131 pg 6). Instead, we quantified the wind speeds at the front of the protective shield or diffusive housing of EACH individual sampler and used those individual data in the regressions displayed in Figure 1. The uncertainty of the sampling rate–wind speed relationships can be determined from the standard error of the slope of these relationships, which will be added to the updated manuscript.

Possibly the reviewer's reference to "parallel wind speed" not being sufficient, is meant to suggest that we should have also investigated the influence of the angle of wind incidence on the sampling rate. While this issue has not been investigated previously for any PAS for gaseous mercury or any PAS relying on the Radiello diffusive barrier, it has been studied in PAS for organic contaminants (May et al. 2011, Zhang et al. 2013, Gong et al. 2017). Because none of these PAS used a diffusive barrier, they are more susceptible to wind effects than our sampling design. Considering that our sampler design includes both a windshield and a diffusive barrier, we felt it not necessary to also include experiments with variable angles of wind incidence. However, that could be the subject of future experiments.

Reviewer: The temperature range tested is extremely low as is the relative humidity. Also, the temperature and relative humidity reported in the methods is not consistent with the abstract.

Response: The temperatures in our experiments ranged from -15 °C to +35 °C. This is not "extremely low" and covers the range of most average temperatures found on Earth. The relative humidity in our experiments ranged between 44 and 80 %. This was the maximum range achievable at the Biotron climate control chambers; increasing or decreasing relative humidity further while keeping temperature at 20 °C was not possible. While there are locations where relative humidity can be outside this range it does cover the mean range of relative humidity over land, 70-80% (Dai, 2006). We further note that the range of temperatures and relative humidity tested compares very favorably with those in other studies on the influence of meteorological conditions on the sampling rates of mercury passive samplers (Guo et al. 2014, Skov et al. 2007, Gustin et al. 2011, Brumbaugh et al. 2000).

The temperature and relative humidity ranges are not reported differently in the abstract, methods, or discussion, but to clarify the temperature range in the abstract (Line 18 pg 2) can be rewritten as: "-15 to +35 °C"

Also the caption of Table 1 can be updated to: "Table 1: Combinations of temperature and relative humidity during the eight experiments performed in climate-controlled chambers. The three relative humidity treatments were 44, 60, and 80% while the temperature was held constant at 20 °C. All treatments were used for the temperature experiments."

Reviewer: Pg 7 I do not really understand what they mean by replicated 5 times. Were 5 samplers deployed or was one sampler deployed in the center of the chamber or are there 5 chambers with the exact same conditions?

Response: The sentence on line 149 pg 7 will be updated to the following: "Each treatment included five replicates, all deployed in the same chamber over the same time period."

Reviewer: Pg 9 line 268 needs a reference and R2 should be r2 and there should be a p value and n

Response: The sentence does require a reference to the initial calibration study. It will be updated in the manuscript for clarification purposes as follows: "The addition of the windshield to the white Radiello® (configuration 1), which is the current method used for outdoor deployments with this PAS (McLagan et al., 2016a), reduced the effect of wind speed on the SR, particularly at higher wind speeds."

R2 will be changed to r2 and n = 44 will be added, and p-value for the exponentially transformed data (since we cannot run a significance test on untransformed log data; this p – value will be added to the Figure). r2, n and p-values will also be added to the other wind relationships (line 262, line 278)

Reviewer: P 14 line 337 self-citation in not appropriate here and in multiple other places in the paper. i.e. pg. 18 line 419

Response: We disagree and believe that citations to our earlier papers are appropriate, both in these two instances and elsewhere in the manuscript. On lines 337 and 419 we

cite our earlier review paper (McLagan et al. 2016b), in which we discuss (i) the two different ways in which temperature can affect the sampling rate of a PAS, and (ii) the two conflicting design criteria affecting the sampling rate of a PAS.

Reviewer: Pg 16 line 373 porosity of sampler housings has been demonstrated to be impacted by acid cleaning. How many times were the samplers subjected to these treatments?

Response: The following statement will be added to the manuscript at the end of line 436 pg 18: "Additionally, Gustin et al., (2011) suggested the porosity of high density poly-ethylene diffusive barriers can be affected by cleaning with HCl. While in this study we used HNO3 for cleaning purposes, the possibility of porosity changes caused by acid cleaning is further incentive to clean previously used Radiellos® with soap rather than acid or heat-acid."

The Radiellos were cleaned once for each method following the description in the methods (i.e. acid baths were for 6 hours). The following will be added to line 169 pg 7: "…water rinse and sonication and air drying). All Radiellos in each cleaning treatment were cleaned once according to the aforementioned methods."

Reviewer: Sampling rates should be compared to those in other papers

Response: The relationships and uncertainty of these relationships have been compared to other samplers throughout the paper (e.g. Lines 265-267, Lines 280-281, Lines 290-293, lines 352-358). The actual SRs determined in this study in the various experiments were frequently compared to the SR of the original calibration study by McLagan et al., (2016a). In that study the calibrated rates were compared to the SRs of other studies, especially that of Skov et al, (2007) which is physically the most similar sampler to that of our own.

We did observe one comparison that was not included in the manuscript and lines 329-330 will be updated as follows: "Relative humidity, tested at 44, 60, and 80% and a stable temperature of 20 °C, had no significant effect on SR ($p = 0.080$; see Fig. S5), which is similar to Guo et al., (2014) who also observed no effect from relative humidity on the SR of their PAS that uses the same sulphur-impregnated activated carbon sorbent.

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