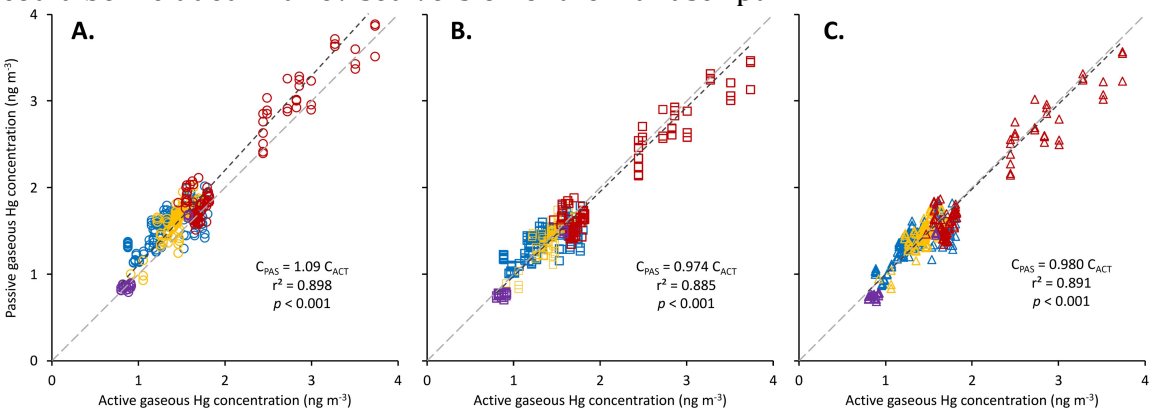
Response to Anonymous Referee #1

REVIEWER COMMENT: This paper provides data from around the world collected using a GEM passive sampler. Based on previous work with GOM passive samplers Huang and Gustin 2015 I doubt that this system collects GOM. I appreciate the effort to get so many people to collaborate.

RESPONSE: We appreciate the positive feedback and agree with the reviewer that the passive sampler is unlikely to take up GOM due to the diffusive barrier. We hope to confirm this with follow-up experiments in the near future.

REVIEWER COMMENT: The paper is well written, and the figures and tables relevant. I think it would be really useful if the authors would make the sites as they have separated them out in the text as urban, rural, and high elevation with different symbols on Figure 3.

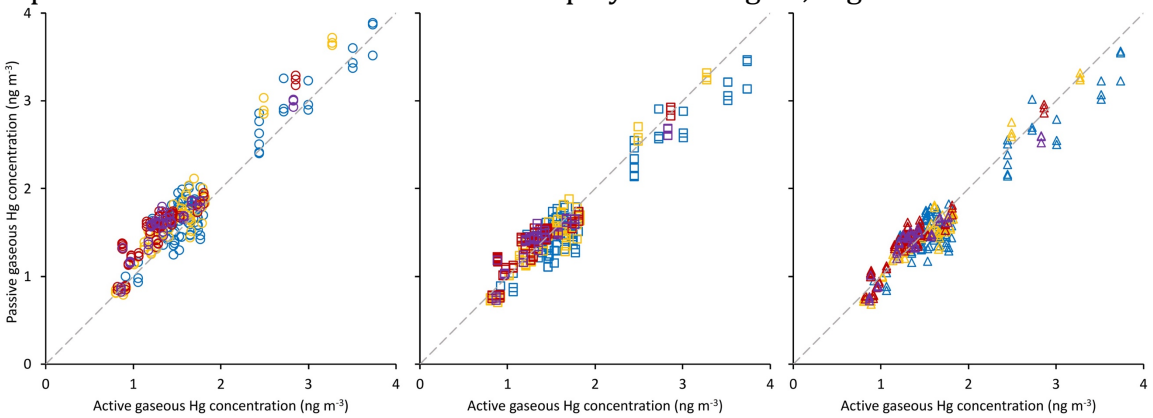
RESPONSE: A version of Figure 3 that takes up the reviewer's suggestion is given here and could be included in a revised version of the manuscript.



In this figure the data are divided by color according to site type: red – urban sites; blue – rural sites; purple – high altitude sites; and yellow – northern/Arctic sites. The fitted relationships are for all data combined.

REVIEWER COMMENT: I wonder also if it would be useful to make a graph that shows correlations of sampler uptake concentrations with Tekran concentrations that plot data based on the time resolution instead of lumping all into one figure?

RESPONSE: Another version of Figure 3, which uses differently colored markers to represent data obtained over different deployment lengths, is given here:



In this figure blue, yellow, red and purple markers indicate 1, 3, 6 and 12 month deployments, respectively. (There were only two 9 month deployments.)

No significant effect of deployment length on the MND of samples for either the recalibrated SR ($p = 0.082$) or the adjusted SR ($p = 0.298$) was observed. Thus, neither the recalibrated nor adjusted SRs nor the uncertainty of the sampler is dependent upon the length of deployment at these background concentrations over deployments from 1 to 12 months in length. This figure could be added to the supplementary information of a revised manuscript.

REVIEWER COMMENT: The limitation of this method is the long time resolution and lack of collection of GOM that is really the atmospheric form of most concern. The authors need to be honest about this. If there are short higher periods of exposure of GEM would the sampler resolve this in anyway given the very long sampling time?

RESPONSE: We agree that a passive air sampler cannot achieve the same temporal resolution as an active instrument. It is, however, noteworthy to point out that while the shortest deployment in the current study was one month, the sampler can take up amounts of Hg that are sufficient for reliable quantification in much shorter time periods. We have previously estimated that at atmospheric background levels (1.5 ng/m^3), a temporal resolution as short as 5 days is achievable (McLagan et al. 2016). At higher concentrations much shorter deployment periods are possible. For example, at concentrations of 10 ng/m^3, 100 ng/m^3 and 1000 ng/m^3, the shortest PAS deployment times to yield amounts above the MQL (0.86 ng of Hg) are estimated to be ~1 day, ~2 hours and ~20 minutes. As accuracy and precision may deteriorate close to the MQL, it is advisable to sample somewhat longer than those minimums.

We do not agree that the "lack of collection of GOM" is a limitation of the sampler. In fact, the sampler was specifically developed to monitor gaseous elemental mercury (GEM). The Minimata Convention (UNEP 2013) and recent papers on the state of atmospheric mercury science have stated the need for improved monitoring of GEM/total gaseous Hg (TGM) due to the limited and biased spatial coverage of current monitoring sites (e.g. Pirrone et al. 2013). Atmospheric models predict spatially highly variable GEM/TGM concentrations in some of the areas with the poorest GEM/TGM measurement coverage (Travnikov et al. 2017). Dry deposition of GEM appears be a much more important pathway for atmospheric Hg deposition than previously thought (Obrist et al. 2017). Accordingly, we do not necessarily share the opinion, that GOM is "the atmospheric form of most concern". GEM is by far the most prevalent atmospheric mercury species and contributes most to its global dispersion.

We take exception to the insinuation that we are somehow dishonest about the limitations of the PAS. The limited temporal resolution of the PAS has been spelled out on lines 286-288 ("The time-averaged nature of the concentrations measured by the PASs conceals much of the variability that generally occurs at shorter time resolution.") We are equally forthright about our current inability to establish with certainty whether GOM is being taken up by the PAS or not. On lines 526-529 we wrote: "we cannot yet conclude with certainty that GEM is indeed the sole analyte sorbed by the PASs. Furthermore, in all cases, the proportion of GOM (TGM minus GEM) in TGM measurements was close to the level of PAS uncertainty, which further reduces the strength of the conclusions that can be drawn."

REVIEWER COMMENT: I also wonder about the activated carbon material. Is the uptake only surficial or can Hg penetrate into the interior given the design of the sampler? Some discussion of the past use of this sampling system for other gases should be mentioned (O3, nitrogen compounds) as well as any limitations.

RESPONSE: The mercury taken up by the carbon sorbent is quantified by combusting all of the carbon in a total mercury analyzer and not by desorbing it thermally. Therefore, while it may be of academic interest to explore the nature of the uptake of Hg in the sorbent, it has no bearing on the performance of the sampler. It also means that a comparison with other vapors that are analyzed differently is unlikely to provide useful insights.

REVIEWER COMMENT: Linear regression is not R2 it is r2 and these should be associated with a p-value.

RESPONSE: Both r^2 an R^2 are in common usage to designate the coefficient of determination. The revised version of Figure 3 given above includes the p-values associated with the regressions.

REVIEWER COMMENT: I also am not sure of the utility of this method overall.

RESPONSE: We hope that we eventually will be able to convince this reviewer of the tremendous potential that the passive air sampler for mercury holds. We have already applied the sampler in a number of studies (e.g. characterization and quantification of mercury from area sources, identification of unknown mercury sources), which will be published in the near future. The potential applications are varied and numerous.

REVIEWER COMMENT: I would personally not promote this as a device that could be used for personal exposure sampling especially since GEM concentrations measured are so below the human health exposure limit. This certainly needs to be tested before being promoted.

RESPONSE: While the maximum gaseous mercury concentrations measured in the current study are below 4 ng/m^3, we have already applied the sampler to record concentrations that are more than four orders of magnitude higher. While we think that the sampler has the potential to be used for monitoring mercury in workplace atmospheres and for personal exposure sampling, we agree with the reviewer that more testing is required, before the PAS can be confidently applied for this purpose.

REVIEWER COMMENT: Figure 1. I think it is misleading to present lines for 2 points and even 3 when the intercept starts with the blank. The blank has been subtracted from the sample so I am not sure if it is a relevant point. I think it might be more appropriate to just present data from each location with the same time resolution. This may better illustrate the different slopes. Just a thought.

RESPONSE: We assume the reviewer refers to Figure 2. We do not agree with the reviewer's suggestions. Yes, the blank has been subtracted from the samples, hence each uptake period starts at the origin and not at the blank level. The slope of each curve, even those with only two or three data points, corresponds to an uptake rate (ng/day) over the deployment period and conveys valuable information on whether the sampler remains in the linear uptake phase. The passive data from each time period are given in the current figure format and the different slopes are also apparent, as the scales of each graph are the same.

REFERENCES

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