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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "4D dispersion of total gaseous mercury derived from a mining source: identification of criteria to assess risks related with high concentrations of atmospheric mercury" *by* José M. Esbrí et al.

Anonymous Referee #3

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This manuscript presents an experimental design using atmospheric Hg monitoring instruments to improve the characterization of a Hg point source over time and space. There is indeed some interesting data and discussion in the manuscript. Nonetheless, I do not recommend the manuscript for publication for several reasons:

1. I feel it is not ideally suited to ACP. It is written as a methods paper based on its experimental design to improve the source characterization across four dimensions. Thus, I would recommend it's submission for Atmospheric Monitoring Techniques, or another similar journal. I do not feel it has the necessary impact or scope for ACP.

2. Given the direction of the paper (such that it is delivered as a methods style paper). I also see some short-comings here. The authors are validly critiquing the need for more time representative studies rather than short "snap-shots" in time that are typically made when taking mercury measurements (especially mobile ones) at source sites using active monitoring instruments. Yet their own work does exactly this. 4 snap-shots spread across the 4 seasons (I assume there is only one profile in each season). Are the days they did their horizontal transects truly representative of the whole season? Why is this approach any better than taking a single snap shot and describing the meteorological conditions present during said snap shot? This is particularly so because the sampling along the profiles was by changing location for each new sample, thus time can play a role in the observed concentration differences and not only spatial variation. Indeed, the authors even mention and discuss this, but it means changes in the measured concentrations can be related to both space and time. This exact point was raised in a study by McLagan et al., (2018). This study used passive samplers concurrently deployed in high numbers across the source area and the time integrated samples (over week long or seasonal deployments) give much more relevant data to assess chronic exposure risk and longer-term trends. The concurrent deployments mean concentration variability is limited to spatial differences. This study is highly relevant to this manuscript and should be discussed in detail (not referenced at all).

3. There is a lot of discussion of mixing layer or boundary layer characteristics based on only the TGM data measured at 3 different heights in the vertical profiles to a maximum of 3 m. Can these large scale phenomena (generally hundreds of metres be described with any certainty based of TGM measurements at three heights extending to only 3 m? I am highly skeptical of this. This applies to this whole section 3.1.

4. The methods section is lacking details. There is nothing describing when the horizontal profiles where made (time of day, date) and there is also nothing on the number of profiles made in each season. Thus, I have to assume each profile was only driven once per season? Thus, 4 "snap-shots-in-time". Details of the sampling instrumenta-

tion are also severely lacking. We need more details on the specific setup of the Tekran 2537B and the Lumex RA-915M to define the exact species being sampled. Heated lines, filters, sampling duration? At least reference another paper whose setup was followed. Were there any external injections to test the quality of the internal calibration source?

5. Some of the writing is also very heavy and needs to be made more concise. Whole paragraphs are used at times to make a point that could be summarised in a sentence and many sentences are very long and convoluted.

Specific comments: Abstract: Abstract is far too long. 680 words. It is heavy reading, where it should be a clear and consise summary.

Lines 47-49: This sentence really sums up one of the problems with this article. The writing is at times very convoluted and could be improved by making sentences more consise. Here stating "PBM & RGM are deposited on local or regional scales" or "PBM & RGM are deposited nearer to source" is enough.

Lines 49-50: “Once Hg is being deposited” should be “Once Hg HAS BEEN deposited”

Line 54-58: long, convoluted and repetitive sentence.

Line 64: “Metric scale” metric is the system, using this word to describe metre scales is very confusing. State "on the scale of metres"

Lines 72-74: Break this into two sentences.

Lines 79-80: There have been more recent studies on this very topic using passive samplers see McLagan et al. (2018). This study is highly relevant to this manuscript. And here seasonal differences are compared and the longer-term nature of the sampling method is ideal for chronic exposure assessment. Although it cannot make any diurnal assessment. This study should be discussed in detail in this manuscript.

Line 92: “Secular” wrong use of this word. It describes not being associated with

religion. i do not know of another definition such as that being the intention of the authors.

Lines 93-95: This is not ok. This does not need a Wiki quote. People know what the four dimensions are. Just like them without the Wiki reference.

Line 192: “...TGM concentrations close to zero...” Please change this to simply "lower". At no point do these concentrations get close to zero. especially considering typical background concentrations are less than 2ng/m3.

Lines 198-199: It seems difficult to state with much confidence that higher concentrations at ground level mean greater deposition. These are not flux measurements as there is a lot of influence of wind. It might be possible to also expect the higher elevation sample to be higher in mercury. Enrichment at the surface, especially in low wind conditions could suggests a source at the ground with decreasing concentration with elevation being caused by dilution with the less enriched air above. It makes sense there is little difference between the sampling heights in the day because the winds mix the system and little difference can be observed.

Figure 3: This is a poor figure. Simply categorizing the data as high medium or low removes any quantitative assessment of the data. This could be vastly improved by taking the mean of the three height measurements for teach hourly time period and then plotting the residuals of each sampling height against time. Thus describing the magnitude of differences.

Figure 4: Instead of presenting typical days with these weather patterns, why not present the mean data (and the number of days described by this weather) for each meteorological condition. The goes to the very heart of the purpose of the manuscript – to eliminate “snap-shots-in-time” and give better time integrated data.

Figure 5: why is the data so much more noisy in spring and autime than winter and summer in profile 3? This could be an analytical issue.

Lines 265-268: Couldn't this easily be confirmed with river water and sediment samples at each river crossing site?

Lines 279-281: Are they though? there looks to be little if any differences in overall concentrations of these profiles particularly for background concentrations based on Figure 5.

Lines 290-292: This may well be the case, but the sampling methods chosen do not relay any information as to whether this is a random and very short term spike in concentration or a longer-term trend. The measurement is merely a "snap-shot-in-time", making it exceedingly difficult to produce any assessment of chronic exposures.

Lines 298-299: again this is a short-coming of the method and an example of a time-related change in concentration rather than simply a spatial related change.

Lines 299-301: of course it is because wind increases dilution - it blows concentrations away and the mix with surrounding air depleted in TGM more rapidly.

Line 301: Why are we now talking about GEM and not TGM? This simply switched. Consistency of terminology please.

Line 307: But Profile 3 certainly does have emissions sources. You only have to look at the large spikes in TGM concentrations. The authors really needed to have a control profile, without any sources (rivers or mines) to make such a statement.

Conclusions: This point form conclusions is a little strange.

REFERENCES: McLagan, D. S., Monaci, F., Huang, H., Lei, Y. D., Mitchell, C. P., & Wania, F. (2019). Characterization and quantification of atmospheric mercury sources using passive air samplers. Journal of Geophysical Research: Atmospheres, 124(4), 2351-2362.

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