

Interactive comment on "Deployment of a sequential two-photon laser induced fluorescence sensor for the detection of gaseous elemental mercury at ambient levels: fast, specific, ultrasensitive detection with parts-per-quadrillion sensitivity" by D. Bauer et al.

Anonymous Referee #2

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The paper describes a new detection method for Hg(0) together with first results from an intercomparison with commercial instruments. Improvements to the first-generation set-up are also briefly presented. This technique seems to be a very promising approach, especially in view of obtaining a higher sampling rate compared to currently used instrument. This work is certainly worth being published in AMT, but could benefit from some improvements.

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Given that the first goal of this paper is the description of the experimental set-up, I am missing details such as laser energy, beam sizes (for example you say one beam has been expanded for better overlap with the second beam: is the second dye laser producing a larger beam?) and PM voltage (do you change the voltage applied to the PM regularly and why? This is not clear, especially not how this is taken into account in the calibration). A schema of the set-up would be helpful.

Being not an Hg-expert, I had trouble following the different excitation schemes (page 5655 and 5656): maybe a figure showing the different transitions would help to make it easier? At this point I would appreciate an explanation about why you use two different excitation schemes? I was guessing that generating the 2nd generation laser wavelengths is easier (and you say so at the end, page 5672), but it would be good to have an explanation here. Maybe there are other reasons?

Throughout the manuscript I was wondering why you use two PM, and little by little I understood that this is for better signal. Please explain this when you mention the 2 PM for the first time (Page 5658). You mention that the set-up could be improved by using even more PM: do you have plans to do so? How about using a lens to collect more photons? Same page, you use a cold trap (what temperature): is this just for condensing water? Are you sure no Hg can be trapped? Same page, you use 254nm instead of 253.7 and 408 instead of 407.8: please use same wavelength everywhere.

Page 5659: you introduce the RAMIX manifold: the short explanation on this manifold that you give page 5666, would be better placed here.

Page 5663/4: you discuss the signal in air compared to helium. The paragraph is not very clear to me. You say before that the quenching rate for the higher states is not known, so what values did you use to calculate the expected fluorescence efficiency of 0.5%?

Page 5667: please explain in more detail what was the criteria that were used to change the PM voltage and how did you correct for. Figure 6, please change the

y-axis so one can see the full spike at 10am.

Page 5670, line 19: I guess it should say: ... because the PMT is closer... and not the cell. It seems that in Figure 12 the y-axis is not well chosen, and the signal is cut at 2.8.

Abbreviations are sometimes not well defined: Please define RGM the first time you use it (line 10 in the abstract). Line 21 of the abstract you could already introduce the abbreviation TGM. What is a KCI denuder (page 5654, line 9)?

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