

Interactive comment on “Understanding atmospheric mercury speciation and mercury in snow over time at Alert, Canada” by A. Steffen et al.

Anonymous Referee #2

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Overview

The manuscript sums up the atmospheric Hg species concentration and Hg in snow measured at Alert over the last ten and fourteen years respectively. Unfortunately the manuscript describes this impressive data set in language that is sometimes clumsy, and sometimes rather inappropriate for a scientific article.

The findings show that a combination of very cold weather and relatively atmospheric aerosol lead to higher concentrations of Hg associated with particulate matter, and that as the temperatures warm and atmospheric PM concentrations decrease the

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proportion of RGM increases. The Hg in snow tends to reflect the RGM concentrations. This is not altogether a surprise.

I wonder whether the results presented here might have been included in a few extra paragraphs in the article recently published in ACP by Cole et al., "Ten-year trends of atmospheric mercury in the high Arctic compared to Canadian sub-Arctic and mid-latitude site"? I am not convinced that the results presented here merit publication in their own right.

As the authors are investigating the relative proportions of PHg to RGM as meteorological variables and time of year change, it seems very odd that in the whole of the manuscript there is not a single wind rose, no mention of wind direction, and not a single back trajectory plot. There is also no mention of total BrO columns, which as an indicator of where Hg oxidation is actually taking place can be useful. It is difficult to see how the relative concentrations of PHg and RGM can be discussed with no mention of the provenance of the air masses being sampled.

This article should either be rewritten and a more in depth analysis of the results included, or withdrawn.

Specific Points

Abstract

This sentence, "*In May, RGM transitions to be significantly higher than PHg and continues into June whereas PHg sharply drops down.*" is not well written. It is the concentrations of the species that are changing not the species themselves. Maybe something along these lines "*In May the high PHg and low RGM concentration regime of early Spring, undergoes a transition to a regime with higher RGM and much lower PHg concentrations*".

"Firstly, the ratio of PHg to RGM is favoured by low temperatures . . .", the high PHg to

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RGM ratio? I don't think that ratios themselves are favoured by any particular temperature, although they may well be temperature dependent.

"partitioning of oxidized mercury to produce PHg . . .", the authors are suggesting that oxidized Hg in the gas phase condenses on to pre-existing PM, or that RGM produces PHg in a particle formation process?

"The highest deposition of mercury to the snow in the spring at Alert is during and after the transition of PHg to RGM in the atmosphere." Again this seems as if the authors are suggesting that PHg becomes RGM, so it desorbs from the PM? Or are the authors referring to the high PHg concentration and high RGM concentration regimes.

Introduction

I agree that Hg has caused quite a stir, although it probably did it in scientific/policy/local government circles rather than in the air itself.

p17023,
I7, drive?

I12, unfortunately coincidental has two meanings, perhaps contemporaneously would avoid any possible confusion.

p17024,
I27, *"Arctic Haze is due to air masses originating from anthropogenic emissions in Europe, North America and the former Soviet Union, that are transported . . ."*, the air masses don't originate from emissions.

p17027
I think this section could be expressed differently, and the last sentence is missing a

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verb. “The ambient aerosol is pulled into the laboratory through a 3 m long, 10 cm diameter stainless steel vertical manifold at a flow rate of about 1000 L min⁻¹. Particles are sampled out of the manifold from near the center of the flow stream, about 30 cm up from the bottom of the manifold. From there the particles are delivered to the sampling devices via stainless steel tubing. The mean total residence time of a particle from outside to its measurement point is approximately 3 s and, at this point, the particle at approximately room temperature and the relative humidity (RH) is <50%.”

p17028

Really? It sounds as if someone had a long walk! “*The coolers are filled with snow and hand carried from Alert to Toronto where they remain frozen until analysis.*”

p17030

It is not clear to me why the results and conclusions were not included in the article cited below. “*Few long term mercury speciation measurements have been reported around temperate regions and only one for the Arctic (Cole et al., 2013).*”

Results

p17031

“*This annually occurring higher level of RGM in July is variable in concentration and unexpected but is not considered to be a result from AMDE chemistry. This study focuses on the springtime chemistry from March to June inclusive. Further in depth investigations into the annual cycling of PHg and RGM at Alert must be undertaken to explain these patterns.*”

I think the article would have had more weight if some more in depth investigations

had been presented here. I am not sure that the manuscript focusses on chemistry, most of the discussion is about meteorological parameters, aerosol loading and physisorption/condensation.

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I do not know much about the meteorology of Alert, but am surprised that there is no mention of prevailing wind directions (or absence of wind) in the discussion regarding the high PHg and high RGM concentration periods. Nor of whether the RGM is produced locally or whether the AMDEs observed are due to transport of already depleted air. There is no discussion, or even mention, of the BrO columns available from satellite observations. I would have thought that this would have been useful as an indicator of when Bromine chemistry 'switched on' either locally or within a region from which air was being transported to Alert. As pointed out in the overview the total lack of any mention of wind direction, air mass origin or back trajectory plot seems incredible.

Still in section 3.1, p17031

line 12 "trails off"?

line 14 "*a transition of PHg to RGM*" is this really what the authors mean? Are they suggesting that RGM is desorbing from the particulate phase?

Page 17032

"In Fig. 3, both RH and AWC follow the same pattern with temperature throughout the year and a similar steep increase in both parameters is shown March to May. It can be conceivable that the transition of PHg to RGM from April to May could be related to water absorption by aerosols. However, we suggest that other factors affect the transition of PHg to RGM more effectively and are described below."

A steep increase is observed / seen?, rather than shown. Transitions again, the authors need to be precise about the nature of the transition, concentration regime, or

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a physical process, as I mentioned above.

Transition is used a number of other times in the rest of the manuscript.

p17033

Hg+2 is an unusual nomenclature, Hg(II) would be more appropriate if the authors are referring to oxidation state.

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p17034

The following sentence is not very clear, does it mean that PHg is a function of particle volume only at certain times of the year?

“These results show that PHg is associated with higher particle volume for March and April, May is a transition month to lower particle volume and June shows no association with PHg.”

p17034

“The higher particle volume concentrations during January to April are linked with Arctic Haze (Barrie, 1986) and we hypothesize that the presence of Arctic haze is a significant contributor to the increased levels of PHg during this period.”

I think most people would agree that the Hg compounds which make up RGM are probably inorganic and really quite involatile. The fact that when it's very cold and the available aerosol surface area is abundant, RGM will tend to condense onto PM is not a hypothesis, it's common sense.

The sentences below need rephrasing. What is 'The atmospheric transition of mercury'?

“Each year PHg at Alert begins to increase in March and then climbs to a maximum in April and are concurrent with 25 sea salts and Arctic haze particle increases. Further

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study is required to identify which types of particles dominate both the atmospheric transition and deposition of mercury in the spring."

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Concerning the conclusions, I think they have been covered in the preceding discussion.

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