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Atmospheric
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Discussions

Interactive comment on "Atmospheric mercury over sea ice during the OASIS-2009 campaign" *by* A. Steffen et al.

Anonymous Referee #1

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General Comments

This article presents the results of measurements of atmospheric mercury species and compounds believed to be linked to the elemental mercury oxidation, within the Arctic Circle at the time of year when Hg depletion events occur. The results are interesting because the measurements were made simultaneously over tundra and sea-ice, and give an insight into the different behaviour of deposited oxidised Hg compounds in the snow over sea-ice and land. The link between the retention of deposited Hg^{II} and the salinity of the underlying snow is, as the authors point out, important for modelling studies, and also in the context of a changing Arctic climate. It is obviously a great shame that the authors had problems with the Hg speciation unit which was at the

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inland site, however the sea-ice measurements make this a very valuable data set worthy of publication. The aspect that lets this manuscript down is the rather tortuous, and at times long winded and confused discussion. This section really does need to be rewritten before publication, and I have made some suggestions below. There are also a few places in which the descriptions / language seem imprecise, I have tried to highlight the most obvious of these as well.

Introduction

The discussion of AMDEs, and the fact that O_3 depletion is observed at the same time, and that both are believed to be the result of boundary layer bromine chemistry is unclear. The use of 'conversion' rather than 'oxidation' doesn't help the reader make the link between Hg^{II} and RGM. (p5689 line 18 onwards)

The comment on line 22, *These reactions have been shown to be in direct relation to the depletion of ozone*, could be changed to be more precise. There is a correlation?

Still on p5689 line 27), it is not the Hg that is collected on particles, but rather the particles that are collected on filters and then the Hg associated with the particles that is determined.

The sentence that follows seems unnecessary.

p5690 *It is believed that the majority of the* O_3 *depletion events occur over the Arctic Ocean and are meteorologically modulated to coastal measurement sites*, meteorologically modulated?

On page 5690 line 17 *sailboat*? Really?

And line 22; it is speciation data that is collected I would have thought, rather than speciation itself. This section does not explain that RGM is assumed to be Hg^{II}, or that RGM is comprised of gas phase oxidised Hg species.

The rest of the paragraph uses both Hg^{II} and RGM, perhaps it would be more clear to the reader not familiar with atmospheric Hg chemistry to use one or the other.

p5691 line 6 *the level of mercury drops off* ?

line 10. Looking at the cited articles I find the following:

Our simulation includes an improved treatment for the fate of $\mathrm{Hg^{II}}$ *deposited to snow. Photo-reduction of deposited* $\mathrm{Hg^{II}}$ *followed by* $\mathrm{Hg^{0}}$ *re-emission is known to take place but not all deposited* $\mathrm{Hg^{II}}$ *is easily reduced. Observational estimates of the reducible component of* $\mathrm{Hg^{II}}$ *range from less than 10% to more than 90%. Here we assume that 60% of deposited* $\mathrm{Hg^{II}}$ *is reducible, but test the sensitivity to this assumption.* from Fisher et al. The text says roughly 50% for some reason.

Simulations by GRAHM and its new snowpack/meltwater model for mercury suggest that, on an annual basis, the average fraction of mercury deposited onto snowpacks that is revolatilized increases with latitude from 39% between 30 and 45° N, to 57% from 45 to 60° N, 67% from 60 to 66.5° N, and 75% polewards of 66.5° N. from Durnford et al., which does not correspond to the values in the manuscript.

Methods

In the section on Max-DOAS it is stated that a radiative transfer model was used to simulate aerosol vertical profiles. As the authors refer to the results from this model in a later section it would be useful if the model used was described or named and a reference given.

Results and Discussion

Section 3.1

There are a few sentences which are a little unclear here:

Frost flowers are intricate ice crystals formed from brine wicked through the sea ice that is subsequently coated with air vapour deposition (Simpson et al., 2007; Douglas et al., 2012a) and have an appearance of a flower on the surface.,

air vapour deposition?

Using Max-DOAS, the position of the particle load and the thickness of the aerosol layer at the surface layer of the study site can be modeled to confirm the presence of *aerosols.*

The comment on the particle sizes that the instrument collects is not very well worded.

This confirms that there is a measured concentration of particles present in the measurement area at the lower surface air.

page 5698, line 16; emerged?

This small crystal forms in the air on clear, cold days and has a relatively large surface area onto which reactions can occur. They have been reported to have a length between 1 and 1000 μm,

onto which reactions can occur? length?

The discussion regarding the possible contribution of Hg adsorbed on diamond dust is divided into two parts, one in section 3.1 (p5699) where it is described as 'likely minor'. Then again in section 3.2 (p5700) where the hypothesis that diamond dust causes a peak in PHg is presented. So it a minor peak?

Section 3.2

The fact that PHg 'lags behind by a few hours' (how many?) does not indicate that this fraction of Hg is not photochemically derived, only that it is perhaps not directly formed by photochemical processes.

The following sentences are not very clear:

Interestingly, Event 3, where %RH is above 75%, is the warmest event may also decrease the potential of RGM to partition onto particles.

This close refrozen surface can reflect a strong depletion of GEM and elevated PHg (potentially a source of fresh sea salts). On 25 March the measurement site was further from the refrozen sea ice and but a closer distance from the open water (12.5 km); however, the air mass is coming from a different direction at this time and thus the northern sea ice may not be the biggest driver in activities during this event.

Section 3.4

It was also observed that after the spike in GEM concentrations at both locations

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the GEM is again oxidized and depleted from the air. How can this be true if no measurements of oxidised Hg were made at the inland site? Can transport of already Hg depleted air be ruled out?

The measurements are suggestive of higher Hg retention in the snow over sea-ice than the snow over land, and this is an extremely important finding in terms of the Hg cycle in the Arctic. However I am unsure that it can be considered as a definite fact as the result of one 12 day experimental campaign in one are of the Arctic, especially as the supporting speciation measurements over land were not performed successfully.
The discussion of AMDEs, ODEs and the atmospheric chemistry of Bromine containing compounds during Springtime in the Arctic boundary layer is rather convoluted, repetitive and at times confusing. The interplay between RGM (Hg^{II}) and Hg associated with particulates is also tortuous at times. The manuscript as whole is rather difficult to follow, and this is a shame given how interesting the results are.
The manuscript requires significant revision (and shortening) in order to present clearly and concisely the results obtained and the conclusions reached.