

Interactive comment on “Iodine speciation in rain, snow and aerosols and possible transfer of organically bound iodine species from aerosol to droplet phases” by B. S. Gilfedder et al.

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Gilfedder et al. present measurements of iodine components and their speciation in aerosol and rain samples from a number of locations worldwide and samples with higher time plus size resolution from Mace Head. Their findings confirm that iodate is often only a very minor component of total particulate iodine. They find that organic iodine is the dominant component. A large fraction, up to 15%, of this could not be identified with their IC-ICP-MS technique. Their findings seem to be consistent with recent suggestions in the literature of the formation of organic iodine via the reaction of HOI with dissolved organic matter. This is a very important contribution to the field and

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should trigger new laboratory studies in order to provide the kinetic information that is needed to improve our numerical models and thereby the quantitative understanding of the atmospheric iodine cycle. Other than I think that the lifetime of iodine should be discussed in more detail (see details below) I have only minor comments and suggest the paper to be published after they have been considered.

Minor comments

Title: I'm not sure if the second part of the title is adequately discussed in the paper. I suggest to either to do this or to drop the second part of the title. As all droplets grow on aerosol particles, the title might in any case be shortened to "Iodine speciation in rain, snow and aerosol particles".

p. 7978, l. 22: Maybe add that this indicates either a long lifetime of these components or a very rapid cycling.

p. 7978, l. 24/25: This has already been suggested a few years ago (Baker, 2005).

p. 7979, l. 25-27: For iodine this mechanism is not important to deplete particles compared to seawater.

p. 7980, l. 2/3: This is a finding that has been reported many times before, so I suggest to add "e.g." before the references.

p. 7982, section 2.2: Please explain in more detail how the snow sampling was made. Was it surface snow or taken from a snow pit? What time of year (or better: what date) where the samples taken?

p. 7984, l. 7: You refer to "results" but it is not clear for the reader where, or rather if, they are discussed. Please add.

p. 7984, l. 26: Change "minima" to "minimum".

p. 7986, l. 28: Doesn't this suggest that there might be either additional, terrestrial sources for iodine (as other particulate marine tracers like Na^+ do show a strong de-

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crease with distance from the coast) or, possibly more likely, that there is a very active recycling of iodine species between the gas and aerosol phases that increases the lifetime of atmospheric iodine over the lifetime of aerosol particles? We have found this in our model studies for bromine and chlorine (e.g. Pechtl and von Glasow, 2007) and it would be perceivable that this is happening for iodine as well. In order to test this "multiphase iodine recycling" in a numerical model more kinetic information about organic iodine species is required. Possibly such a recycling - if indeed occurring - could involve photochemical reactions as you list on p. 7988. As you state yourself (top of p. 7991), your sampling might be inadequate to resolve if there is either a day-night difference in the gas-aerosol partitioning or if even faster (light-dependent?) recycling reactions is occurring. If there is indeed light-dependent recycling between the gas and aerosol phase going on, you might "freeze" the chemical composition during sampling when you store the sample dark. Maybe you could add some discussion on this point.

p. 7990, R6: This had already been suggested by Baker, 2005.

p. 7990, l. 23: Are you really referring to R4 here - how is R4 dependent on the organic substrate?

p. 7991, l. 7-9: I assume that this implies that your "total iodine", as reported in text and tables, is "total soluble iodine". If this is the case, please make this more explicit in the text and captions to the tables. Are there any indications for an appreciable amount of insoluble iodine? Maybe you can refer to studies by Duce et al that used neutron activation to measure the "real" total iodine to put your "total (soluble) iodine" numbers into perspective.

p. 7992, l. 6-8: The composition of nucleation clusters should not be expected to be the same as those of the larger particles that you sampled.

Throughout the paper (incl. tables, figures and captions): Spelling of Mt. Egmont. This mountain was named after John Perceval, 2nd Earl of Egmont, not after a mountain of eggs.... Alternatively you could use the Maori name: Taranaki.

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Acknowledgements:

Check spelling of McGregor.

Tables:

Table 3: Isn't it surprising that in the Mt Egmont sample only 10% organic iodine was found whereas in Summit snow - even at 1m snow depth - it was a lot higher? Could you speculate about possible differences? Is the age of the snow comparable, what about post-depositional effects incl. snow photochemistry or lack thereof if deposition in winter?

Figures:

Figure 2: Caption: Change "doted" to "dotted" or rather "dashed".

References:

You sometimes (e.g. p. 7982, l. 6) refer to one of the Gilfedder et al. (2007) papers as "2006", please correct.

You list Pechtl et al. (2007) twice, I assume that one references was meant to be Pechtl et al., 2006.

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