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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

Interactive comment on "Speciation of ^{127}I and ^{129}I in atmospheric aerosols at Risø, Denmark: insight into sources of iodine isotopes and their species transformations" *by* L. Y. Zhang et al.

Anonymous Referee #1

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This manuscript reports on the determination of chemical speciation of 127I and 129I in aerosol samples collected in Denmark in 2011 (shortly after the Fukushima nuclear accident) and 2014. The speciation work is very detailed and the study is very worthwhile, particularly since having this combination of measurements for both isotopes is rather rare. The results therefore provide a useful insight into the complex atmospheric chemistry of iodine. The data reported appear to be of good quality, the manuscript is well written and the conclusions drawn are for the most part sound. I have no hesitation in recommending that it is suitable for publication in ACP and congratulate the authors on producing an excellent study. However, I feel that in some cases the authors' inter-

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Discussion Paper

pretation of their data is a little speculative, and some editing of the text is required to better reflect the confidence that can be placed in some of this interpretation. I set out my specific and technical concerns below:

Specific comments

P25142, l 11. "Larger particles remained in the troposphere for about 20 days. . ." It would be helpful if the actual size range of the particles in question could be stated explicitly. Large (e.g. 20 um) particles would be expected to have lifetimes shorter than 20 days.

P25142, l 15. The expression "marine boundary layer" is very commonly used to describe the lowest levels of the troposphere in contact with, and directly influenced by, the ocean. From the context of the sentence I am not sure whether this is what the authors intend here. Do they actually mean the "sea surface"?

P25143, l 22. I find the statement "diluted by a factor of 1-20" to be ambiguous. It implies that not all the samples were diluted by the same factor, which I don't think was the case. Probably "diluted by a ratio of 1:20" would be more accurate.

P25144, l 17 (and several other instances throughout the manuscript). Here the authors report average 129I concentration and the standard deviation of that average to 4 significant figures. In my view this level of precision is not justified. Two significant figures would be quite enough in this case.

P25146, l 26. "Except for the Norwegian Sea.." the authors are implicitly excluding the North Sea (which has relatively high 129I seawater concentrations) from being part of the Atlantic Ocean. Perhaps it would be better to say "Except for the North Sea and Norwegian Sea..".

P25148, l 4-7. Here the authors cite the relatively low terrestrial emissions of iodine as the cause of the relatively low 129I concentrations in the aerosol concentrations observed in samples AE11-6 and AE11-7. If low terrestrial emission was the only

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factor involved then one would also expect these samples to have similarly low 127I concentrations, but this does not appear to be the case. Probably more important is the low 129I:127I ratio of the terrestrial emissions, due to their distance from the western (marine) 129I sources.

P28148, l 15. "dominant air masses during the sampling periods were westerly". Please specify exactly which time periods are referred to here, as several different time periods are discussed in the preceding sentences.

P28148, l 16. I do not understand what the authors intend by the word "secondarily" in this sentence.

P25148, l 18-21. I am confident that the authors' statement regarding the relative strengths of direct atmospheric and secondary marine emissions of 129I is correct, but I don't see how the data presented in this manuscript can allow one to draw that conclusion. I suggest that the words "It can therefore be concluded" be removed and the authors cite one or more of the studies that have compared atmospheric 129I concentrations with known emissions from Le Hague and Sellafield, as these do demonstrate this.

P25148, l 29. "Hence, iodine in marine aerosols directly participates in aerosol formation." I think there is something incorrect here. If the iodine is already in marine aerosols how can it participate in aerosol formation? Is this intended to mean that iodine nano-particles nucleate particle growth?

P25149, l, 1-8. Previous studies have shown that the speciation of iodine in rainfall and aerosol samples are rather different, even when the samples were collected at the same location over the same time periods (Gilfedder et al., ACP, 8, 6069-6084, 2008). One other factor contributing to this difference may be that the iodine present in the rainwater samples can originate from iodine present in the source cloud, as well as from material acquired during droplet descent, whereas the aerosol iodine is only representative of the material at ground level. I do not understand the point the authors

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wish to make regarding back trajectory analysis in the final sentence here.

P25150, l 4. “alternative primary pathways”. Alternative to what?

P25150, l 11-12. Gaseous SO2 can be formed by the oxidation of DMS, but in north-west Europe direct emission of SO2 from anthropogenic sources is more significant.

P25150, l 21. “We note that relatively low WSI 129I and 127I was measured in marine-sourced aerosols from the North Sea”. I agree that this appears to be the case for 127I, but I am not so convinced for 129I (from looking at the concentrations listed in Table 2).

P25151, l 29 – p25152, l4. While there is no question that the environmental lifetime of NRP 129I is far longer than that of Fukushima 129I, their atmospheric lifetimes are short and probably rather similar. Might it therefore be possible that RII is formed over longer time periods in some other compartment (the sea surface?) and enters the atmosphere through primary emission?

P25152, l 5-19. I find this paragraph to be contradictory and poorly argued. Initially the authors appear to discount soil as being a significant source of RII on the basis that little (<10%) 129I remains after NaOH leaching. They later state that the RII fraction might be associated with “metal oxides that originated by suspension of fine inorganic particles”. What might these fine inorganic particles be, if they are not of soil origin? They then state that a “relatively large fraction of iodine in soil and sediment has been observed in metal oxides associated form”. Does this not contradict the earlier statement, or does it indicate that the latter fraction is not RII? There is no evidence presented at all to support the final statement regarding the association of gaseous iodine with inorganic particles.

P25152, l 20 – p25153, l 3. I struggle to understand the authors’ interpretation of the relationships they found between iodine species concentrations and 7Be. Although the iodide/iodate ratios of 127I and 129I in the North Sea are different (Hou et al., EST, 41, 5993-5999, 2007), it is difficult to see how this could cause the speciation of emissions

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of the two isotopes from the sea surface to be significantly different. If emissions from the sea surface dominate behaviour of 129I, then this must also be the case for 127I.

P25155, l 6. Why should the calculated dry deposition estimate from this work be consistent with the wet deposition? These are different deposition mechanisms.

P25155, l 16. I recommend changing this to read ". . . of the water-soluble iodine in the aerosols measured in this study", since other studies has found different results.

P25155, l 22-24. This statement appears to be worded a little too strongly. The difference in 127I concentrations referred to here is only a factor of 2.

Technical comments

P25141, l 11. Please replace "to speculate" in this sentence. Its meaning is not clear.

P25141, l 23. The meaning of " might hardly be converted " is also not clear.

P25142, l 20. "with the only one being our previous study"

P25146, l 4. "A range of . . ."

P25146, l 15. " was observed in aerosols "

P25148, l 23. " result was not observed "

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