The authors report the results of quite detailed analyses of aerosol composition collected during a ship expedition on the Atlantic between England and the southern tip of South America. The main analyte is iodine, in particular the speciation of particle-bound iodine into four fractions (iodide, iodate, total iodine, soluble organic iodine). For the interpretation of the results a number of other components (e.g. trace metals) are also determined. The results of the sophisticated analysis are also evaluated with the use of back trajectories. Since different size fractions are always sampled, there is also particle size-relevant information, which in turn is used by the authors to interpret sources. One of the main conclusions is the influence of aerosol acidity on the chemistry and thus the occurrence of the different iodine species in atmospheric aerosol particles. The manuscript is well written, is based on an excellent data set, and the conclusions drawn are well justified. The topic is certainly relevant to ACP readers and therefore I recommend publication with only minor changes.

We thank the reviewer for their careful consideration of our manuscript and helpful comments.

Page 5, line 94: missing word !?

We have rewritten the text here. The amended text is:

"For the determination of soluble TMs, samples were extracted into ~ 1 M ammonium acetate solution followed by analysis using inductively coupled plasma – optical emission spectroscopy (ICP-OES). Full details of these methods have been reported in Baker and Jickells (2017)."

Page 5, line 103: an "as" is missing

This has been inserted.

The authors discuss in several passages the iodine chemistry, e.g. the reactions of IO3- to HOI, SOI and I- and quote from Baker 2005 and Pechtl et al. 2007. Since without any question the speciation is in the center of the work and also in the title the possible influence of the chemistry on the iodine species is pointed out, I would find it helpful for the reader to compile the central chemical reactions from Baker 2005 and Pechtl et al. 2007 for this paper. In doing so the understanding of the discussion would be simplified from my point of view.

Thank you for this suggestion. The most important reactions in this context are the acid-dependent reduction of iodate:

 $IO_3^- + I^- + 2H^+ \leftrightarrow HIO_2 + HOI$

And the formation of SOI and its subsequent decay to iodide:

 $\mathsf{HOI} + \mathsf{DOM} \iff \mathsf{SOI} \iff \mathsf{DOM} + \mathsf{I}^{-} \mathsf{H}^{+}$

The latter reaction was included as HOI + DOM \leftrightarrow DOM + I⁺ + H⁺ by Pechtl et al. because they did not include an organic iodine fraction (SOI) in their model. We have added these reactions at lines 56 - 58. We have also amended the description of the Pechtl et al. model on lines 51 - 53, as the original description of this was slightly inaccurate.