

Interactive comment on “Iodine speciation and size distribution in ambient aerosols at a coastal new particle formation hotspot of China” by Huan Yu et al.

Anonymous Referee #2

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The manuscript provides useful insights into chemical composition of newly formed particles and indicates iodine role in coastal nucleation events. Moreover, it shows a presence of organic iodine species in the nucleation mode particles and gives their semi-quantitative estimate. The manuscript gives valuable input into understanding of marine new particle formation and growth and, given the potential importance of new particle formation for clouds and climate, I would support the publication after the comments listed below are addressed.

General remarks

It is nice to see the evidence for organic iodine being present in the nucleation mode

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particles, however, it is still not clear whether these contribute to the formation of the new particles or the growth by iodine reactions with organics. The latter would actually be consistent with previous studies (Sipilä et al., 2016) that point to iodine oxoacids and iodine oxide vapours being the main drivers of particle formation and initial growth. Similarly, in your figure 9, I-organic adducts are present in the larger particle sizes than inorganic I (IO_x and I+I₃). Moreover, as shown in Figure 8, the difference between I-NPF and non-NPF events is mainly in relative contribution from inorganic I compounds (IO and I+I₃), which would support them being the drivers of NPF's. I would, therefore, recommend stating this point clearly in the abstract and conclusions or provide more extensive discussion.

Likewise, it doesn't seem that any iodine compounds were important for continental-NPF's, but this point is not clearly covered in the paper. Add more discussion on the drivers of continental-NPF's. Be clear what other than I- compounds are driving the continental-NPF as I- is not important here (Figures 8 and 9, also line 343). Also, lines 374-375: the hypothesis state that 'that iodine species in the accumulation mode during the continental NPF days were from the aging process of small iodine-containing particles' – where do these iodine particles come from in the continental air masses, are there iodine sources over the continent, elaborate? Lines 437-438 point to importance of aromatic iodine, but how relevant it is to NPF if existing in accumulation mode?

Abstract and conclusions state that farmed microalgae, as well as wild algae, could be an important source of new particle formation in the coastal areas of China, but statistics on the nucleation events is not provided, the statement that 'unless it was cloudy or rainy, strong NPF events were observed almost every day in April and May' is vague, provide frequency distributions or other quantitative information to evaluate that account. Move the text in lines 103-109 from experimental to result section and add more quantitative information on the occurrence of the NPF events.

Add figures for back trajectories to supplementary and references to them (e.g. line

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167, 360 and elsewhere);

I would suggest moving section 3.2 to methods rather than results as it describes the measurement method development.

I commend authors for being open with advantages and limitations of the technique, but more information is still needed. Material on lines 211-226 is very important, but difficult to follow for non-specialists, please provide more details on RT and its relevance to conclusions, elaborate, why longer RT points to bonds with organic. For example, the link between in-source fragmentation and weakly bond iodine-organic adducts is not so obvious, give more details and explanations. Similarly, provide explanation why 'all m/z 126.9039 peaks after RT=1 min in a sample by low energy MS scan can be deemed iodide-organic adducts and their total peak area should be proportional to the total concentration of the adducts.' (lines 224-226). It might be obvious for ion chromatography person, but not everyone. E.g. why RT of 1 min is important? (line 312 as well);

Provide more information on deductions from Figure 5, e.g. elaborate on the statement 'Therefore, each red circle without a black dot in -0.3~0 mass defect regime in Figure 5 should stand for an organic iodine compound.' (Lines 245-246);

The statement in the conclusions 'Our study provided important information of iodine speciation, concentration and its role in NPF' does not have any specific info, spell out that 'important information' here;

Specific comments

Lines 99-100: be consistent in dimensions; use either cm or m when describing inlet lengths;

Line 193: clarify what you mean by 'the contribution of ions were negligible';

Line: 233: what aerosol extract – high I or low?

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Lines 244-246: add '-' for mass defect of 0.0961 to be consistent with the ones below.

Line 248: Elaborate what are the 'typical' aerosol samples;

Lines 262-263: What MOUDI stages were analysed to produce the results in Table1, all? Provide info here;

Lines 321-322: provide info on the significance of the blank concentrations, where they comparable to the sample or significantly lower?

Lines 328-329: Provide info on what samples were used for regression analysis;

Figure 8: typo in 'continental';

Figure 9: Is it possible to provide overall mass size distribution calculated from SMPS here for reference by say using a constant density. It would give a feeling on how much mass was reconstructed. Otherwise, provide this information in the caption or description of the figure. Reference to Fig.2 is not very useful as it is very much time consuming to compare different figures (one is distribution over time, the other is averaged distribution);

Table 2: Explain what the percentage in the table is referring to (22.8 % of what?) as adding all lines gives higher percentage than 100%.

Sipilä, M., Sarnela, N., Jokinen, T., Henschel, H., Junninen, H., Kontkanen, J., Richters, S., Kangasluoma, J., Franchin, A., Peräkylä, O., Rissanen, M. P., Ehn, M., Vehkamäki, H., Kurten, T., Berndt, T., Petäjä, T., Worsnop, D., Ceburnis, D., Kerminen, V.-M., Kulmala, M., and O'Dowd, C.: Molecular-scale evidence of aerosol particle formation via sequential addition of HIO₃, *Nature*, advance online publication, 10.1038/nature19314 <http://www.nature.com/nature/journal/vaop/ncurrent/abs/nature19314.html#supplementary-information>, 2016.

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