

## A point-to-point response and relevant changes made in the revised manuscript

### Anonymous Referee #3

*The paper by Yu et al. is an important study in quantifying iodine species in atmospheric aerosols. Given their low concentrations, but dominant role in new particle formation, studies like this offer significant advancement in understanding processes where direct measurement of iodine species is always very challenging or not yet developed. The study is well written and should be accepted after addressing mostly minor (but often important) comments.*

We thank the referee for his important comments and careful examination of the manuscript. Point-to-point responses are given below. All relevant changes made in the revised manuscript are highlighted in yellow color.

#### **Comments**

*Line 17. quadrupole (several cases thereafter).*

Re: corrected.

*Line 25. The sentence is rather pointless in the abstract as it should summarize significant findings, not just "shedding light".*

Re: we thank the referee for pointing out this. Now the sentence is changed to be more specific about what work we have done in this study and moved to line 18-20:

“Our study provided important information of iodine speciation, size distribution and its role in NPF in the context of heavy air pollution in China’s coastal areas.”

The significant findings are subsequently summarized in the following lines 20-27 of the abstract.

*Line 30. O<sub>3</sub> is largely depleted due to mainstream NO<sub>x</sub> chemistry, especially taking into account miniscule amount of iodine compounds.*

Re: here we change to

“The atmospheric impact of iodine includes ozone (O<sub>3</sub>) depletion, altering HO<sub>x</sub> and NO<sub>x</sub> chemistry, mercury oxidation and aerosol formation.”

*Line 57. knowledge gap instead of unknown, because we already know quite a bit.*

Re: here we change “unknown” to “knowledge gap”

*Line 71. techniques, not experiments.*

Re: we change “experiments” to “techniques”

*Line 116. What fraction of 72h sampling period were actual NPF events? Could be easily estimated from concurrent SMPS/NAIS measurements. It is an important estimate considering different iodine sources in coastal/inland environment.*

Re:

For I-NPF, it can be easily estimated from SMPS/NAIS that nucleation mode particle occurred in about 30 hours out of the 72 h sampling period. This means the average iodine concentration reported in Figure 8 and 9 may underestimate the concentrations during the I-NPF period by 40%. On the other hand, for the continental NPF events, iodine resided in accumulation mode particles from 0.3 to 1  $\mu\text{m}$ , which received contributions from both NPF and non-NPF sources. It becomes difficult to tell which period is dominated by NPF and which period is not. Therefore, it is impossible to estimate the percentage of underestimation for the continental NPF samples, as well as the samples collected at the inland urban site.

To be consistent, we reported 72 h average concentrations in the manuscript. This is also a common practice for all off-line measurement studies, which cannot provide time-dependent information. In line 353-356 we have pointed out this issue:

“It should be noted that, identical to previous aerosol iodine speciation studies, the concentration reported here ( $\text{pmol m}^{-3}$ ) is an average over the entire period of 3 sampling days. Thus, iodine concentrations during the intense NPF periods should be higher than the values reported in this work.”

In the captions of Figure 8 and 9, we clearly state that we report the average concentration during the NPF days, not the NPF periods.

*Line 177. There is a question to what extent those intense I-NPF events were driven by ordinary coastal algae induced emissions as compared to dried farmed algae. Without an estimate it is little reason to compare with natural environment like Mace Head which is not even located at a hot spot.*

Re:

With only one fixed observation site, this study cannot provide further information about the contributions from wild or farmed algae sources. Now we delete the words “Similar to the NPF events observed at Mace Head...” in line 182.

*Line 189. It is important to consider not only nucleation relevant gaseous iodine species, but also condensation sink which limits the production of new particles.*

Re: we thank the referee for pointing out the possible reasons why new particle concentrations differed so much in the I-NPF and continental NPF events. However, we were discussing ion/particle ratios and the possible

importance of ions in the NPF in this paragraph. So it is not appropriate to add irrelevant discussions about gaseous nucleating precursors and CS.

*Line 209. Quadrupole*

Re: corrected

*Line 238. "from" typed twice.*

Re: corrected

*Line 299. mode not model.*

Re: corrected

*Line 303. PM1 or PM0.5 would be adequate substitutes too were size-segregated samples not available.*

Re:

Thanks for pointing out this. Now the sentence in lines 313-315 is changed to

“To avoid artificial formation of iodide-metal complexes during the sample extraction process, our result highlights the importance of collecting PM<sub>0.5</sub>, PM<sub>1</sub> or size-segregated samples instead of PM<sub>2.5</sub> or PM<sub>10</sub>.”

*Line 323. limitations not weaknesses.*

Re: corrected

*Line 324. ...there still a possibility...*

Re: corrected to “it is still possible that”

*Line 327. the most serious. Also wasn't sulphate and nitrate high in this study as considered?*

Re: corrected. Sulfate and nitrate may be higher in accumulation mode than other size ranges. The sentence is changed to

“The underestimation may be the most serious if there are high concentrations of co-eluting sulfate, nitrate and ammonium in the aerosol samples of accumulation mode.”

*Line 330. two significant digits are sufficient, unless better than 1% precision which is impossible in regression analysis.*

Re: the R<sup>2</sup> is corrected to 0.94.

*Line 336. over the entire period of 3 sampling days.*

Re: corrected

*Line 337. iodine concentrations were certainly higher and it can be estimated by the duration of NPF events and iodine concentrations outside of NPF events.*

Re:

As we explained above, the durations of NPF and non-NPF are not straightforward to differentiate for continental NPF events, during which iodine resided in 0.3-1  $\mu\text{m}$  particles. To be consistent, we reported 72 h average concentrations in the manuscript. This is also a common issue for all off-line measurement studies, which cannot provide time-dependent information.

*Line 341. uncertainties of semi-quantification are partially related to bulk mass matrix interferences, so distributions are not highly accurate.*

Re: we change to

“Although not highly accurate, relative distribution in different sizes is less affected by the uncertainties of semi-quantification.”

*Line 349. high iodide concentrations*

Re: corrected.

*Line 351. most likely originating from the partitioning*

Re: corrected.

*Line 355. The role of sulfuric acid in new particle growth has long been established.*

Re: The word “interesting” is deleted. The sentence in line 372 is changed to

“The finding of  $\text{HSO}_4^-$  in nucleation mode (Figure 6 and 9a) indicates that  $\text{H}_2\text{SO}_4$  also contributed to new particle growth during the I-NPF days.”

*Line 389. : : :the rest of the data*

Re: corrected.

*Line 393. please note the numbers claimed for comparison.*

Re: the numbers are added.

*Line 413. counted towards organic iodine...*

Re: corrected.

*Line 420. coastal locations of western Europe...*

Re: corrected.

*Line 423. This claim is overblown out of proportions, because the impact of algal farms would be limited by a small physical footprint of the entire coastline area.*

Re:

With only one fixed observation site, this manuscript cannot provide further quantitative information about the contributions from different algae sources. We didn't claim any proportion information in these lines. Now the sentence in line 443 is changed to

“Therefore, wild algae, as well as farmed algae, could be an important source of new particle formation in the coastal areas of China.”

*Figure 7.  $R^2$  is not a correlation coefficient, but variance which is interpreted as the percentage of variance between two variable to the total variance.*

Re:

Thank you for pointing out this mistake. Now we simply use  $R^2$  in the manuscript.

*Figure 8. “continental” misspell.*

Re: corrected.