Ozone catalytic destruction by halogen species and new particle formation and cloud condensation nuclei formation from iodine species are two important processes with potential climate implications. Most previous field studies focus on group-level measurements, while the vertical distributions of reactive halogens are much sparser. Many global chemical transport models have illustrated the potential role of bromine and iodine species in atmospheric chemistry and suggested that their impacts are ubiquitous. However, aircraft measurements have often been lacking to support these theses. The non-sea-salt aerosols (nSSA) bromine and iodine data presented in this paper, as part of the ATom campaign, are unique and severely needed for global model evaluations of the atmospheric chemistry impact of bromide and iodine species. Furthermore, several recent studies have highlighted the impact of iodine oxoacids in atmospheric new particle formation processes, which potentially contribute to cloud condensation nuclei formation and influence the aerosol indirect effect by affecting cloud properties. The data provided in this study will certainly be used by global models to constrain aerosol formation from iodine species.

This paper presents a comprehensive study showing the ubiquitous presence of nSSA containing trace amounts of bromine and iodine. One strength of the PALMS used in this study is that it separates aerosol types thereby can easily and directly associates bromine and iodine mass concentrations to different aerosol source types. The study finds that bromine and iodine are present in a significant fraction of nSSA, with biomass burning identified as a primary source. Additionally, it suggests that pervasive secondary sources of bromine and iodine are needed to explain their concentrations in nSSA. The finding of the correlation between biomass burning and elevated nSSA iodine and bromine is novel and represents a missing source of bromine and iodine in global models, as suggested by the comparison to GEOS-Chem results.

Overall, the paper is well-written, and the data presentation is clear and clean with no cluttered plots. The choice to place many similar plots in the appendix is encouraged to prevent overcrowding the main text. Therefore, I recommend this paper for publication in ACP after minor revisions.

Major:

One major problem with this paper is that the narrative emphasises two aspects: 1) bromine and iodine are ubiquitous in the nSSA throughout the atmosphere, and 2) bromine and iodine mass concentrations are negligible compared to organics and sulfate. I believe the discussion should not stop here. A critical difference between particulate iodine/bromine and sulfate/organics is that iodine and bromine are actively involved in heterogeneous processes, thereby are efficiently recycled, whereas sulfate and organics primarily remain in the particles once condensed. In other words, mass concentration is not a sufficient metric for characterising the importance of halogen species. It would be a pity for this study to stop at this point. The study integrates the GEOS-Chem simulation, and it should be relatively straightforward for the authors to delve further, such as by correlating iodine mass concentrations with its impact (or

the potential impact, by any reasonable metric) on odd oxygen. This would help readers understand the significance of low mass concentrations in practical terms. Is it important or not? As the authors pointed out, many factors influence the impact of iodine on odd oxygen, which has been previously explored. However, providing such a calculation, correlating mass concentration to atmospheric impact along the tracks of ATom, is critical and has the potential to be widely cited (I would for sure cite this in upcoming studies).

It would be great if the authors could present the I+IO data in the appendix. The CH3I measurement from the GEOS-Chem simulation captures well what ATom provides. Including I+IO as a metric for future evaluation of iodine chemistry and aerosol formation would be highly valuable.

After obtaining these data in the authors' preferred formats, please correlate the bromine/iodine mass concentrations with their atmospheric implications throughout the manuscript, especially in the abstract and conclusions. This will undoubtedly enhance the manuscript's significance and "citability."

Another major problem is the insufficient emphasis on the importance of heterogeneous processes, i.e., the recycling of halogen species between the gas and particulate phases. As detailed below, several raised hypotheses could at least be partially explained by these heterogeneous processes. While determining the exact reasons for the under/over-estimated bromine/iodine levels of GEOS/Chem, and gas-to-particle partitioning processes in the measurements, are outside the scope of this study, mentioning this possibility would provide a more complete discussion.

Minor:

Suggested title: Widespread Trace Bromine and Iodine in Remote Tropospheric Non-Sea-Salt Aerosols. I believe this will highlight the key message of this study. However, I will leave this to the authors to decide whether to adopt the suggestion or not.

Lines 6-7: it should also note that low concentrations of halogens can already introduce significant impact.

Line 20: Sherwen et al. 2016b should be quoted first as Sherwen et al. 2016a.

Lines 42-44: the lines are a bit confusing. The iodine contribution to secondary aerosols is direct but the bromide contribution is indirect through its a bit limited capability (compared with chlorine radicals) to react with organic species. It should be clarified.

Lines 45-47: it should be noted that the prevailing evidence currently shows iodine oxoacids play key roles in iodine secondary aerosol formation processes, while iodine oxides play relatively smaller roles. However, since I believe the reviewer 1 might have a differing opinion, it is the best for this study to give more generic expression such as "iodine species" to pass the review process.

Lines 49-51: Finkenzeller et al., 2023 does not show iodic acid can nucleate on its own. Please rephrase the reference to their results.

Line 58: please convert ng m−3 to pmol mol −1 which is frequently used in this study. Also line 58: please provide the measured range of the Koenig et al. (2020).

Lines 133-134: I do not understand how masses 95 and 97 can affect the bromide measurement? Is this instrument specific problem? Please specify.

From the section 2.2, it seems the systematic errors of the reported bromine and iodine concentrations are more likely the lower limit than higher limit. It would be great if such systematic error can be reflected in the abstract and conclusions where such numbers are quoted.

Lines: 254-255: This is interesting. However, the explanation is not convincing enough since gaseous and particulate bromine and iodine are constantly exchanging through heterogenous processes. Therefore, this cannot be a single way of accumulation of mass. It is also likely that the dry air prohibits such exchange, therefore resulting in net accumulation of bromide and iodine in aerosols.

Lines 330-336: Is there a correlation between days since boundary layer influence and relative humidity? Could the authors confirm this? Additionally, could the authors discuss whether such findings are due to bromine/iodine accumulation or shifts in gas/particle partitioning caused by changes in temperature and relative humidity?

Lines 337-344: following the same as the last comment, could the change in RH also a possibility?

Lines 351-353: going into the same line - difficult for me to imagine how RH is going to prevent condensable iodine species to condense onto particles, as laboratory experiments have pointed to a kinetic condensation of species such as HIO_3, which is the dominant mass contributor in at least the MBL[1]. However, the reverse may be more true: high RH may promote the release of iodine from particles, thereby reducing the iodine particle mass concentration.

Lines365-366: another line of evidence supports potential RH effect.

Lines 367-383: meteoric aerosols presumably contain a lot of iron which promotes the conversion of iodide to molecular iodine[2]. This may be another reason for the strikingly lower aerosol iodine in meteoric aerosols than the sulfate-organic-nitrate / biomass burning aerosols.

Lines 384-393: along the same line, is it possible to check the correlation between metals such as iron vs. iodine content?

Lines 412-413: inorganic emissions of what? Presumably halogens?

Lines 459-460: The statement that water reduces iodine particle formation in the MBL is unfortunately incorrect. Experiments conducted under atmospheric conditions generally indicate a positive enhancement of particle nucleation rates with increasing water content. Specifically for iodine aerosol nucleation, recent evidence suggests that relative humidity above 2% has negligible impact on iodine aerosol nucleation in the marine boundary layer[3]. Please remove this statement. Furthermore, the high levels of iodine in nSSA are also likely attributable to missing heterogeneous chemistry processes.

Lines 469-472: could the authors please specify what are the major iodine components in nSSA in GEOS-Chem for a comparison with the ATom measumements? Ideally it should be quantitative.

Lines 469-477 are difficult to follow, and more context is needed for general readers regarding what is consistent and what is not consistent between the conclusions of this study and Koenig et al. 2020[4]. While reformulating the discussion, it is worth mentioning that the higher iodate

in the LS would already indicates lower regeneration of iodine to the gas phase, thereby may lead to lower gas phase reactive iodine, assuming similar total nSSA iodine in the LS and UT.

References:

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(2) Fudge, A. J.; Sykes, K. W. 25. The Reaction between Ferric and Iodide Ions. Part I. Kinetics and Mechanism. *J. Chem. Soc. Resumed* **1952**, 119. https://doi.org/10.1039/jr9520000119.

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