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Interactive comment on "Holocene atmospheric iodine evolution over the North Atlantic" *by* Juan Pablo Corella et al.

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Referee #2: In this study, the first ice core iodine record spanning the Holocene is presented. Iodine levels were high during the early Holocene and industrialization relative to the late Holocene. Chemical transport model results of inorganic iodine sources and transport are compared to ice core iodine levels. This comparison suggests that marine biogenic sources of iodine were higher during the early Holocene relative to the late Holocene. This increase in biogenic sources is reasonable in the early Holocene given that other reconstructions show that sea surface temperatures in the North Atlantic were warmer, salinity was higher, sea ice extent was lower, and primary productivity of subpolar planktonic species was higher. This study is well-written and well-organized. It should be published because it is the first record iodine covering the early Holocene

and provides strong links to climatic factors that could explain the unexpectedly high levels of early Holocene iodine. Hopefully, it will inspire future study of iodine species in ice cores.

Response: We appreciate the positive feedback from reviewer 2. Please, find below the responses to all the comments made by referee #2

Major comments

Referee #2: Section 1, lines 36-37: This sentence is confusing. What exactly is meant by "has a global contribution of up to 27% of the total rate of ozone loss?"

Response: We are referring to reactive iodine as an important ozone-depleting family in the troposphere. Specifically, we are saying that reactive iodine is responsible of up to 27% of the ozone loss in both the marine boundary layer and the upper troposphere. We have rephrased that statement to clarify the meaning.

Referee #2: Section 1, lines 58-68: Two ice cores are reported as having the only iodine records, but further in the paragraph, a third ice core record is discussed. Please clarify.

Response: We have modified the sentence to clarify the iodine reconstruction in polar regions available up to date

Referee #2: Section 2.1, line 91-96. Please discuss the depth-age scale in more detail because (it is not yet published?). Perhaps include a depth-age scale in the supplement. Which volcanic markers were used and how well-spaced are they throughout the record? What are the uncertainties?

Response: The ReCAP ice core record covers the last 120 kyr BP and is currently in press (Simonsen et al., 2019). The RECAP timescale down to 458.3 m (4048.1 y b2k) was produced using the StratiCounter automated layer counting software (https://github.com/maiwinstrup/StratiCounter) [Winstrup, 2016)]. The software was constrained to fit 28 volcanic marker horizons dated in other Greenland ice cores

(DYE-3, GRIP, GRIP and NEEMS1) formalized in the Greenland Ice Core Chronology 2005 (Rasmussen et al., 2013). Below 458.3 m, the timescale was derived automatically using a shape-preserving piecewise cubic interpolation (Vinther, et al., 2008) between 15 chronological tie points (volcanic markers or climate transition matches) used to constrain the timescale back to 11703 y b2k. As the RECAP timescale is synchronized to GICC05, it inherits the uncertainty budget associated with that timescale and presented in the aforementioned references.

Referee #2: Section 2.2, lines 99-100. Please include the sampling resolution at various depths. Clearly, several more samples were analysed in more recent years which makes sense as the fractions were collected via the CFA system. The easiest way to do this may be to include a mean sampling resolution for the time periods discussed in each section (results).

Response: The iodine and sodium measurement temporal resolution ranged from subannual in the upper metres during the Great Acceleration (average resolution of 1.31 samples/yr), subdecadal resolution during the Late Holocene (average resolution of 0.23 samples/yr), and sub-centennial resolution during the Neoglacial and the HTM (average resolution of 0.035 and 0.011 samples/yr respectively). This information has been added in the revised manuscript

Referee #2: Section 3: Please use the same units when possible. It is difficult for the reader to understand readily the comparisons between different periods of time and the model versus the observations when the units are changed.

Response: We have now changed the unit of the modelling results throughout the whole text from nmol m-2 d-1 to μg m-2 y-1 according to the observations units.

Referee #2: Section 3: It would be useful to discuss the relationship between concentration and flux here. Did the flux calculation confirm that remobilization processes were not consequential? Are there any important differences when flux is used?

Response: In ReCAP ice core, reconstructed accumulation rates shown in Figure S2 shows reasonably stable levels for the last 8000 years, suggesting relatively constant snowfall and surface conditions. Variability in the snow accumulation (derived from annual layer thickness calculations) occurring prior to 8000 years ago may be due to glaciological flow effects already identified in the previous Renland ice core drilled in 1988 (Hansson et al., 1994) just 2 km from the site of the 2015 RECAP ice core (this work).The absence of any significant correlation between the accumulation rates and the concentration of impurities such as iodine and ssa also indicate that the Holocene iodine variability in ReCAP ice core is neither related to the effects of changing snow deposition rates. Furthermore, both iodine concentration and fluxes shown similar trends. This agreement between concentration and fluxes reinforces the explanation that source effects (i.e. marine bioproductivity and sea ice dynamics) rather than relative changes in wet/dry deposition are driving the iodine levels variability throughout the Holocene. We have mentioned this in the revised version of the manuscript.

Referee #2: Line 141: Start a new paragraph here? How does the second part of this paragraph relate to the first part?

Response: Done, we have modified this section by adding a new sub-section regarding re-mobilization processes in snow and ice

Referee #2: 3.1, lines 182-183 the authors state that there is no correlation. Is it possible to also include a time series of calcium and sodium measurements in the supplement?

Response: Correlation coefficients of sodium and calcium are shown in Table S3 and in section. Sodium profiles are shown in Maffezzoli et al (2018) while calcium dataset is available upon request. We have stated this in the revised manuscript

Referee #2: Lines 191: Please use the same units throughout the manuscript when possible.

Response: Done

Referee #2: Lines 192-193: How would the ssa contribution change over time? Is the lack of a correlation during the early Holocene purely due to the biogenic contribution overwhelming the other signals? Are they still there?

Response: Sea salt aerosol (ssa) inputs to RECAP are a combination of sea salt directly lofted from open ocean (wave breaking and storms) as well as sea salt incorporated into blowing snow over sea ice. When iodine is primarily emitted through organic processes there should be less correlation between ssa and iodine. When iodine is primarily emitted through inorganic processes, there should be a greater correlation. The reviewer's description is therefore accurate, that the lack of correlation during the early Holocene supports the interpretation of a dominance of organic iodine emission sources. As described in Supplementary Table S1, the THAMO model results indicate organic iodine emissions were effectively zero during the Neoglacial, but were active before and after the Neoglacial and continue to the present. Ssa showed an abrupt increased at the beginning of the Holocene and fluctuating values throughout the Holocene. This variability is described in Maffezzoli et al. (2018). During the HTM the biogenic contribution would be much higher than the iodine related to atmospheric ssa deposition. This is well reflected in the I/Na ratio that have been added to Fig. S4 in the revised version of the manuscript. However, ssa, as well as dust, could also be minor contributors of the iodine levels during the HTM. This has been reflected in the revised manuscript.

Referee #2: Lines 205-208: Please include the ice core values here and clarify how this conclusion was made, given the values provided.

Response: We have added the mean iodine depositional fluxes from the ice core during this period. The modelling results during this period show that inorganic iodine emissions were significantly higher than the organic iodine emissions (Table 1) which is in agreement with the decreased in bioproductivity shown in Fig. 2 and described

in the text. We have now provided the values for organic iodine emissions in the new version of Table S1 in order to make this clearer

Referee #2: Lines 221-223: Replace "a higher frequency" with "higher frequency variability." I agree that this is pretty clearly due to increased sampling resolution. If anything, averaging or smoothing could be used to compare the late Holocene to the early Holocene. The late Holocene levels may only appear to show higher frequency variability because there are more samples.

Response: We agree with reviewer. Thus, we have rephrased the sentence as follows: "The iodine time series in this part of the record reveals a higher frequency variability mainly due to a reduced ice compression towards the surface and thus an increased temporal resolution of the samples"

Referee #2: The focus of this study is iodine and should remain iodine. Do other halogens, like bromine, that were measured in the ReCAP ice core (Maffezzoli et al., 2018, in re-view), also show the same high levels during the early Holocene? Are the processes governing the sources, transport, and deposition of other halogens so different that they should not be included in the supplement?

Response: Neither Br nor Cl show that abrupt difference between Early Holocene and the rest of the Holocene. Bromine enrichments found in ReCAP ice core are mostly influenced by the autocatalytic processes in Multi-Year Vs First-Year sea-ice that are fully described in Maffezzoli et al (2018). On the other hand, Cl is dependent on ssa variability and other complex recycling processes in the polar atmosphere that are beyond the scope of this paper. Therefore, in our opinion it would not make sense to add the other halogens in the supplementary material.

Referee #2: Consider adding a section about the modelling results. These results are used to explain the interpretation of the ice core record, but it would be useful to have an understanding of the results prior to the comparison.

Response: We have elaborated on the modelling section. We have added more information about THAMO and described with more detail the outcome of the modelling results in the revised version of the manuscript. On top of that we have added a new figure in the supplementary material (Figure S3) that displays the modelled atmospheric reactive iodine according to the different oceanic iodine emissions during the present day and the different Holocene periods discussed in the text.

Figures and tables

Referee #2: fig. 1 add mapping software reference and color bar if going to include bathymetry

Response: We have added the software reference (Ocean Data View, Schlitzer, R., 2015, http://odv.awi.de) and the colour bar in the new version of the figure

Referee #2: fig.2 add indication of detection limits to iodine concentrations

Response: We have added the information of the detection limits in the Figure caption

Referee #2: Table S1: Please consider moving table S1 to the main text.

Response: Done, Table S1 has been incorporated to the main text as table 1. The table now includes the inorganic and organic oceanic emission in the same units of the ice core fluxes (μg m-2 y-1).

Referee #2: Please add more to this caption. What are the sources (ice core versus model inputs/outputs) in each column?

Response: We have now including more information in the caption of the table, defining the different fluxes included in the organic and inorganic iodine columns. Minor Comments:

Referee #2: Abstract, line 22: change to: “in the record were found”

Response: Done

Referee #2: line 30: “allows for the detailed”

Response: Done

Referee #2: line 32: “key factor for understanding”

Response: Done

Referee #2: line 42: change to “involve”

Response: Done

Referee #2: line 59: “Ice core”

Response: Done

Referee #2: line 66: “allow for a”

Response: Done

Referee #2: Rename section 2.1 “Study site and dating” or “Study site and age scale”

Response: Done

Referee #2: Line 108: “with the stability of the instrumental”

Response: Done

Referee #2: Line 126: remove “The model”

Response: Done

Referee #2: Line 129: “all of the”

Response: Done

Referee #2: Line 138: add comma before “respectively”

Response: Done

Referee #2: Line 141: remove “thus” before “Holocene”

Response: Done

Referee #2: Line 146: remove “occurring” before “before”

Response: Done

Referee #2: Line 203: change to “algal”

Response: Done

Referee #2: Line 205: change to “both of these processes”

Response: Done

Referee #2: Line207, 220: “concentrations”

Response: Done

Referee #2: Line 221: commas before and after “respectively”

Response: Done

Referee #2: Line 241:comma before “resulting”

Response:

Referee #2: Line 251: “to accelerate considerably”

Response: Done

References Hansson, M. E., The Renland ice core. A Northern Hemisphere record of aerosol composition over 120,000 years. Tellus B: Chemical and Physical Meteorology, 46(5), 390-418, 1994.

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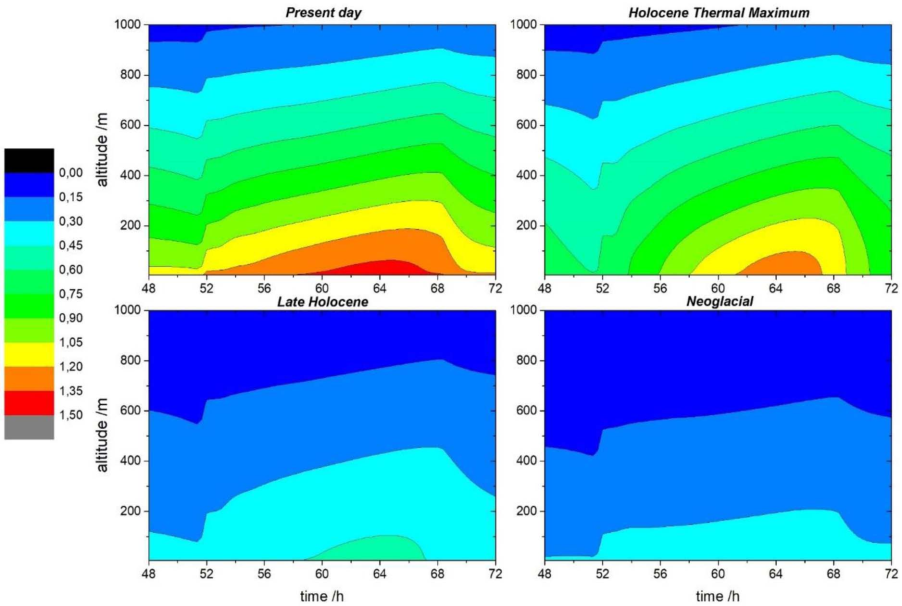

Fig. 1. Figure S3. Diurnal variation of reactive iodine modelled by THAMO in different scenarios of [O3] and VSLs e missions fluxes

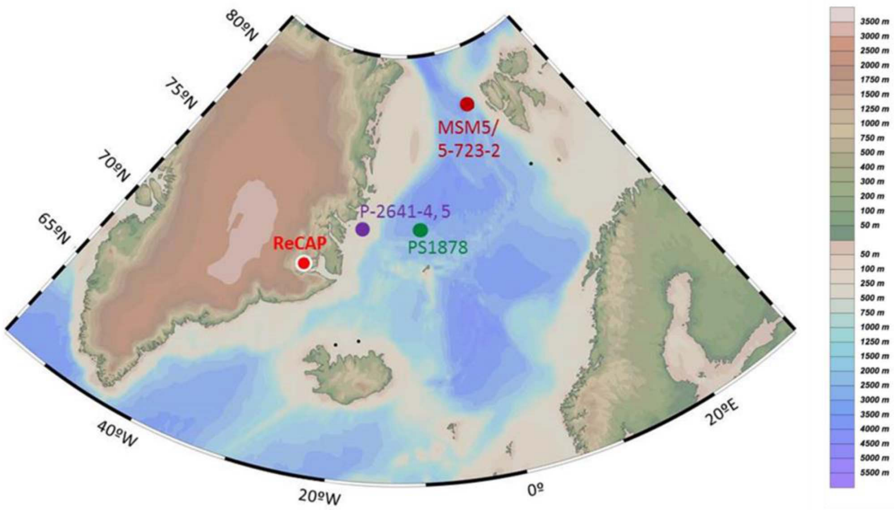

Fig. 2. Fig. 1: Location of the ReCAP ice core (red) and other marine paleoceanographic archives in the Nordic Seas discussed in the text.