List of relevant changes

- An introduction on Brenr as a potential proxy for past FYSI conditions has been added (Sect. 1).
- The whole experimental part together with the atmospheric modeling part are now presented in the Methods section (Sect. 2).
- Following the comment of Reviewer#1, the section on the Brenr calculation (Sect. 3) has been expanded and an additional analysis is performed. It now consists on 3 methods for the calculation of ssNa, based on chlorine, magnesium and calcium. Accordingly, we now include all the new measurements in the experimental description and plot all time series. On this topic, a section is added in the Appendix.
- The deglaciation section (Sect. 4.1 and Fig. 6) has been further analyzed; 2 additional marine records have been included.
- The transformation has now been removed. The general interpretation of the 120 kyr Brenr record is the same as in the previous version.

One major comment was addressed to the ice core age model, which is described in another paper. This paper has now been accepted for publication but at the moment of writing not yet published.

REVIEW – REFEREE#1

This paper presents a new 120,000 year record of Br measured in the Renland ice core. It then interprets this in terms of sea ice extent. The analytical record is an interesting and potentially important one, and the paper should eventually become publishable in Climate of the Past. It is written in a clear style. However there are a number of issues that need to be addressed before it can be published. These fall principally into three areas, the meaning of Br_enr, dating of the ice, and the transformation the authors adopt.

We thank the Reviewer for the time she/he took to review the manuscript. We agree with her/his comments and incorporated the suggested modifications, except for the ice core chronology, which is still not published. In particular, the introduction (Sect. 1.) is centered on the meaning/limitations of Brenr. The transformation has been removed. An extended section (Sect. 3.) has been added to deal with the calculation of ssNa (sea-salt sodium) concentrations, needed to calculate Brenr. Now three methods are used: chlorine, magnesium and calcium. The analytical records of this paper now include bromine, sodium, calcium, magnesium and chlorine. All time series are presented in the figures in the main text and the measurements are explained in the Methods section (Sect. 2.2). Two marine PIP records from the Norwegian Sea have been added.

1. The idea that some measure of Br enrichment may be associated with mainly first year sea ice (FYSI) is plausible and the authors have, in previous papers, made some kind of argument for it. However it is far from established and the paper is far too definite about that. It needs to spell out the caveats, and use the word "suggest", "might" and "may" a lot more. I would also argue to add a question mark to the title.

The idea that Br_enr is a FYSI proxy relies on a number of assumptions.

(a) The Br activation process leads to production of activated Br and to a depletion of Br in the salty surface on which it happens. As a result there can be both enhancements and depletions in what is deposited as snowfall, depending on the relative importance of depleted sea salt aerosol and gas-phase Br compounds that eventually get transported as (presumably) HBr. The implicit assumption used here is that the gas phase Br is transported to (in this case) Renland much more efficiently than the depleted sea salt aerosol. I agree with this, which is consistent with eg Simpson et al 2004. However the assumption needs to be stated, especially as this group has been inconsistent on that question, with Spolaor et al (2013), the first paper on this proxy, asserting exactly the opposite.

(b) There is also an assumption that HBr is produced only by the photochemical Br activation and not by a reaction between salt and acid analogous to that described on page 3, line 10 for Br. Again, the weak Cl fractionation suggests this may be true, but it needs stating.

(c) The paper assumes that Br activation takes place only on FYSI and not at all on MYSI. This is highly unlikely to be true, and the production ratio between these two forms of sea ice will be crucial to the interpretation. This is also the case for sea salt aerosol itself as shown in a modelling study by Rhodes et al (2018), where the use of sea salt as a sea ice proxy is crucially dependent on the extent to which FY and MYSI are involved in aerosol production. I don't expect a full modelling study here, but again it needs to be acknowledged that production on MYSI would affect the interpretation.

(d) Finally the authors essentially assume that their untransformed proxy equates to FYSI, taking no account of transport distance. It's actually quite plausible that their proxy decreases in very cold periods not because there is less FYSI but because it's much further from Renland. This may still argue for a

similar interpretation to the data, but is a more subtle mechanistic point that will eventually need to be tested in models.

I therefore ask the authors to state their assumptions and the caveats more fully, and I will suggest some places in the text where the interpretation should be more tentative. The introduction (Sect. 1.) now consists of a description of Brenr as a potential sea ice proxy and the limitations and uncertainties that still remain. We thus proceed with the interpretation of the Brenr record as a record of FYSI, but it is now clearly stated that a number of uncertainties still exist. A question mark to the title has been added. Please find the added text in italics.

1. Introduction: Br enr as a potential indicator for past sea ice conditions

The connection between Arctic sea ice and bromine was first identified through an anticorrelation between springtime ground level ozone (O_3) and filterable bromine air concentrations (Barrie et al., 1988). Large bromine oxide (BrO) column enhancements and simultaneous tropospheric ozone depletion were later found in Antarctica (Kreher et al., 1997). Satellite observations reveal aeographically-widespread 'bromine explosions', the sudden increase of atmospheric bromine concentrations during springtime occurring in both polar regions (Chance, 1998; Richter et al., 1998; Wagner and Platt, 1998). The mechanism proceeds via springtime photochemical heterogeneous reactions that lead to the activation of bromide, followed by the release and exponential increase of gas-phase bromine species in the polar troposphere (Vogt et al., 1996). Several saline substrates on fresh sea ice surfaces (hereafter referred as first-year sea ice, FYSI) were suggested as reservoirs of reactive sea salt aerosols (SSA) capable of sustaining bromine recycling (Abbatt et al., 2012; Saiz-Lopez and von Glasow, 2012). To date, both model studies (Yang et al., 2008, 2010) and experimental evidence (Pratt et al., 2013; Zhao et al., 2016) consider the deposited snow layer on FYSI (known as salty blowing snow) to be the most efficient substrate for SSA release and bromine activation. Atmospheric bromine and sodium thus originate from both oceanic and FYSI sea salt aerosol, where their concentration ratio is that of sea water mass: (Br/Na) m = 0.0062 (the subscript indicates 'marine', Millero et al. (2008)). Br enr values in ice core records, i.e. the bromine-to-sodium mass ratios beyond the sea water value (Eq. 1) were introduced by Spolaor et al. (2013b) as a potential proxy for past FYSI conditions within the ocean region influencing the ice core location. The basic assumption behind this idea is that bromine recycling on FYSI surfaces would increase the bromine-tosodium mass ratio beyond the sea water value in the atmosphere and at the ice core location, thus isolating effect of FYSI- induced bromine recycling from the sea salt aerosol contribution, the latter being interfered by oceanic emissions. Further studies have provided some evidence on the validity of Br enr as a FYSI indicator (Spolaor et al., 2013a, 2014, 2016a). At present, however, this interpretation is still challenged by several uncertainties related to the different variables which play a role in the involved chemical and physical processes. These can be grouped into three categorizes: bromine activation, transport and deposition/postdeposition. They are briefly discussed here. Pratt et al. (2013) showed that saline snow collected on Arctic tundra or first-year sea ice surfaces can serve as an efficient reservoir for bromine activation. They point out, however, that acidity of snow is a pre-requisite for bromine activation, as well as internal snow pack air chemistry (e.g. concentration of $\cdot OH$, NO⁻³/NO₂). A modification in these factors would alter the bromine production efficiency at constant FYSI extent. Aged sea ice (second-year sea ice or older) is possibly contributing to bromine activation due to a non-zero salt content, but the amount of this second order source effect is unknown. Bromine travels from the source region to the ice core location both in the aerosol and gas phases, while sodium is only found in the aerosol phase. To date, the partitioning of bromine species between the two phases isn't clear. Additionally, some studies have reported that bromine recycling takes place

within the plume even during the transport (Zhao et al., 2016), leading to possible bromine depletion from the aerosol in favor to the gas phase. Overall, the final observed Brenr values in ice samples depend on the relative importance of aerosol and gas-phase bromine. It appears that fine aerosol are enriched in bromine, while coarse particles are depleted (Legrand et al. 2016, Koenig pers. Comm.). Additionally, the two phases likely have different atmospheric residence time, thus the final bromineto-sodium ratio found in the snow might be function of the transport duration. On this topic, Simpson et al. (2005) experimentally showed that sodium is washed out from the atmosphere faster than bromine, suggesting the role of longer lasting gas-phase bromine. On the other hand, a model run by Spolaor et al. (2013b) showed opposite results. To which extent transport processes can interfere with a source effect is yet to be resolved. The last set of uncertainties relates to the variables associated to the deposition of bromine and sodium, such as the variability in accumulation rates over long time scales, which would impact the relative importance of the wet and dry contributions of the two species. Finally, photolytic reemission of bromine from the snowpack could be responsible for bromine loss and decreased Brenr values.

We acknowledge the above-mentioned open questions on the validity of Brenr as a FYSI proxy and clearly point out that further studies are needed to shade light to these uncertainties. We thus proceed with the interpretation of Brenr as a FYSI proxy bearing in mind that other interpretations could, at present, explain a Brenr record. Here, we consider the available evidence regarding past sea ice conditions in the Nordic Seas, and present a bromine enrichment record from the RECAP ice core, located in coastal East Greenland. Because of its location, RECAP should record the fingerprint of sea ice in the North Atlantic. We compare our the RECAP Brenr record with sea ice reconstructions from five marine sediment cores drilled within the Renland source area: the Fram Strait, the Norwegian Sea and the North Icelandic shelf.

2. This seems to be the first paper in which RECAP data have been presented covering the glacial, and is therefore the one in which the age model is effectively first presented. Dating Renland is far from simple, and it is completely unacceptable to rely on a reference to an in preparation paper (Simonsen) which the reader has no sight of. If this is the first paper published then this one needs to demonstrate that the dating is plausible – in the appendix or SI I would expect at minimum an age-depth profile, a table of tie points, and an estimate of age uncertainty. Without that, the reader has no idea how robust the comparisons to the NGRIP isotope record are (ideally we would see the Renland water isotope record in this paper but I appreciate that must be the subject of another paper and that this paper is out of sequence). In the absence of a substantial dating appendix/SI, this paper should not be published until a full description of the dating has been published in another paper.

For the ice core chronology we refer to the Supplementary of Simonsen et al. "East Greenland ice core dust record reveals timing of post-Eemian advance of Greenland ice sheet" (Nat. Comm. In press, 2019)

3. While it is a clever idea, I am not convinced by the value of the transformation. The paper claims that this "linearizes" the record, but how does it do that? The number that is achieved has not in any sense been shown to be linear with any climate variable, be it FYSI extent, total sea ice extent or anything else. What has been done is to give a spurious air of quantitativeness to a proxy that remains at present qualitative. In Fig 5 the panel where the data are colour coded red or blue is good, and I understand the temptation to try to turn it into a curve that can be compared with the IP25 record in Figure 6 but I feel it should be resisted. If the authors cannot resist producing something I suggest a different alternative:

(a) Have the y-axis go up to 7.4 (or whatever the change point is considered to be) and then decreases back to zero again, and plot the actual Br_enr value either above or below the line, maintaining its colour coding . (Mathematically this is achieved by making eq5 simply Br_enr=-Br_enr, plotting it with axis reversed and using the absolute value of Br_enr as the axis label). To indicate the uncertainty about the T threshold however, you should plot both the actual and the transformed Br_enr values for data within 2 sigma of the chosen temperature threshold so that it is obvious that there are two possible states at some depths. This procedure will produce a curve similar to the one in Figure 6 but without producing spurious new numbers.

The transformation has been removed, and we kept only the records with the two regimes color coded in red and blue.

More detailed comments

Page 1, abstract, needs to be more tentative: line 3 I suggest "and tentatively reconstruct"; line 6 "what we interpret as the transition from MYSO to FYSI started at 17.6 kyr"; line 8 "our proxy interpreted as FYSI reached its maximum"; line 10 "sea ice extent was probably greatest". We have now written the abstract clearly stating the uncertainties still exist in the use of Brenr.

Abstract. Although it has been demonstrated that the speed and magnitude of recent Arctic sea ice decline is unprecedented for the past 1,450 years, few records are available to provide a paleoclimate context for Arctic sea ice extent. Bromine enrichment in ice cores has been suggested to indicate the extent of newly formed sea ice areas. Despite the similarities among sea ice indicators and ice core bromine enrichment records, uncertainties still exist regarding the quantitative linkages between bromine reactive chemistry and the first year sea ice surface. Here we present a 120,000 year record of bromine enrichment from the RECAP ice core, coastal East Greenland, and interpret it as a record of first-year sea ice. We compare it to existing sea ice records from marine cores and tentatively reconstruct past sea ice conditions in the North Atlantic as far north as the Fram Strait (50-85 °N). We find that during the last deglaciation, the transition from multi-year sea ice to first-year sea ice started at \sim 17.5 kyr, synchronous with sea ice reductions observed in the eastern Nordic seas and with the increase of North Atlantic ocean temperature. First-year sea ice reached its maximum at 12.4-11.8 kyr, after which open-water conditions started to dominate, as supported by sea ice records from the eastern Nordic seas and the North Icelandic shelf. Our results show that over the last 120,000 years, multi-year sea ice extent was greatest during Marine Isotope Stage (MIS) 2 and possibly during MIS 4, with more extended first-year sea ice during MIS 3 and MIS 5. Sea ice extent during the Holocene (MIS 1) has been less than at any time in the last 120,000 years.

Page 2, line 1 "reservoirs" Thanks.

Page 3, equations 2-3 and surrounding text. I appreciate that correcting for crustal Na in Greenland is not straightforward, but you need to explain what your rationale is here better, and when you do, it doesn't completely make sense. You are implicitly assuming that when the salt/acid reaction has created excess Cl, there is no terrestrial Na (eq 3); and that if there is a Cl/Na ratio less than 1.8 that is entirely because of terrestrial Na, and not at all because of the salt/acid reaction leading to removal of Cl from aerosol. I am not sure why either of those assumptions should be correct. Normally one would use a crustal element such as Al or perhaps Ca to put limits on the terrestrial

correction. In the absence of that you need to revisit your method and at minimum state its limitations and suggest how much it may affect your results.

We greatly thank the referee for rising the issue. We also agree in the fact that both processes (terrestrial sodium inputs and dechlorination processes) could act simultaneously to modify the Na/Cl ratios. By looking at their 1994 paper, Hansson et al. (1994) wrote "The mean ratio between Cl- and Na+ never exceeds the sea water ratio for any climatic stage, i.e. never indicating an excess of Cl-, which is a further indication that the deviation from the sea water ratio is due to an additional (crustal) source of Na+ and not a fractionation process in the atmosphere between the sea salt elements". Our measurements, show in fact that the Cl/Na do exceed 1.8.

Thus, sea salt sodium (ssNa) has been calculated in two additional ways: using calcium and magnesium, both measured by ICP-SFMS (discontinuously in the Holocene, continuously in the glacial section). The analysis is described in Section 3. We also dedicate a section in the Appendix in which we infer from present day studies on desert storms the ratio of Ca/Mg and Ca/Na in 'asian dust'. These numbers are used to calculate ssNa. As far as calcium is concerned, Renland calcium concentrations showed unexpectedly high values during the glacial. This has been previously shown also in the GRIP core, probably due to the appearance of gypsum/carbonated dust. We thus account for the high calcium levels to provide a calculation of ssNa based on calcium, which turns out very similar to ssNa calculated using magnesium. We therefore consider two ssNa curves and two Brenr curves: Br_{enr, Mg} and Br_{enr, Cl}, calculated respectively from chlorine and magnesium concentrations. We will not attach here this whole section since it consists of a number of pages, plots and tables. It will be provided in the modified version of the MS.

Page 3, line 15-20. Thank you for using the word "suggesting" here. However I am not sure what point you are making in line 19-20. Why would we expect Holocene-like ice extent at 120 kyr when we are already well into the glacial inception?

We agree. The sentence has been written more clearly.

Page 3, line 26-28. The modelling study of Rhodes et al (Rhodes, R. H. et al., GRL, 45(11), 5572-5580, doi:10.1029/2018gl077403) doesn't really support your interpretation, as in present conditions, it says that both sites are overwhelmingly seeing OW conditions (Rhodes was looking at the influence of sea salt aerosol but I imagine modelling Br would produce a similar result). Thanks for rising this point. Rhodes et al. (2018) modeled an annual SISS/OOSS (sea ice sea salt / open ocean sea salt) of 0.21 at NEEM (fSISS annually integrated, Table S2 and Fig. 1), while at Renland this ratio drops to 0.04, meaning that at NEEM the sea ice influence would be 5 times greater than that at Renland. The ratio of the bromine enrichment values at the two locations are roughly 3-4 (if we consider Holocene averages, Fig. 3 in our manuscript), pointing to a similar conclusion, at least if a relative sea ice vs open ocean contribution are considered. On an absolute scale, it appears from Rhodes et al. (2018) that NEEM is indeed more influenced by open ocean rather than sea ice, at least just considering sodium. We therefore modify the text to better indicate that the different Brenr values at the two locations might reflect the RELATIVE influence of sea ice, being 3 to 5 times higher at NEEM than Renland.

The difference between Holocene values for the two sites could also be partly related to the fact that the Br_enr measure is not really ideal because it does not measure the amount of Br reaching the site but rather the ratio of Br/sea salt aerosol. NGRIP of course receives much less sea salt than Renland so just a small amount of gas phase Br can induce a Br_enr >>1. This may not fully explain the difference between the sites but it is a factor. It's too late now because this measure is embedded in

the literature, but the use of Br_excess ([Br-Br_seawater] instead of [Br/Br_seawater]) would have avoided this problem.

We probably agree, but we point out that a core1-core2 comparison between Br_excess curves could be interfered by the difference in snow accumulation.

Page 4, line 8 "a number of" Thanks.

Page 6, last line "Our reconstruction suggests.." Done.

Figure 2. I appreciate that Kindler only goes to 10 kyr but it is unhelpful that we don't see a climate record from 10-0 kyr. Can you infill with an 18O record to at least indicate to the reader that Holocene temperatures remain warm.

We now used NGRIP δ^{18} O in Fig. 4 and specified that in the caption of Fig. 6.

Fig 3 caption, Fig 6 caption: Tzedakis et al is not an appropriate reference for orbital parameters. Please cite original papers by either Berger or Laskar. Thanks.

Appendix B. I am wondering why you call this "sea salt aerosol source area" when what you are interested in is the source of the gas phase Br. The back trajectories are for air masses so certainly not specific to aerosol.

Both gas-phase (Br) and aerosols (Br,Na) contribute to Br_enr, so both need to be considered, so this paragraph (now Sect. 2.3) is now named "*Atmospheric reanalysis: the source region of bromine and sodium for the RECAP ice core*".

Fig B1 caption line 2 "constraint" (but can you explain what you mean by this constraint). Only the trajectories that crossed the 900 hPa isosurface were selected and considered for the analysis. The term constraint has been removed.

Fig B1 caption. Sorry for my ignorance but I never heard of a nabla before, can't you call it an inverted triangle? We now use diamond symbols.

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REVIEW - REFEREE#2

The paper by Maffezzoli presents the first data on sea ice coverage of the North Atlantic and the Greenland, Iceland, Norwegian (GIN) Seas based on an ice core recently drilled on a coastal ice cap from East Greenland (Renland ice core). Following the previous approach by Spolaor et al. (2016) they use the Br enrichment above sea salt concentrations (Br_enr) linked to halogen explosions occurring on seasonal sea ice surfaces. In principle, the data and argumentation of the paper are convincing although at some points not explained in detail enough. The paper represents an important contribution to the field that will be of interest for ice core specialists, marine geologists and modelers alike. In its current state, however, the paper still suffers from some language issues (see annotated pdf file attached), some structural deficits and some issues with the argumentations that I outline in my general comments below. I am convinced that these changes can be accommodated and recommend to accept the paper after major revisions.

We really thank Reviewer#2 for the time she/he took in making a constructive review. We now modified the paper structure by incorporating all the analyses in the whole Methods section of the main text (except for a study on 'asian' dust chemical composition in the Appendix). The deglaciation section has been further analyzed and includes two additional PIP records. The Brenr curves are now two (Br_{enr,Mg} and Br_{enr,Cl}), depending on how ssNa is calculated (we refer to the answers to Reviewer#1 on this topic). The Brenr-transformation has now been removed. We thank Reviewer#2 for finding the paper interesting and for spending time to produce the annotated pdf.

General comments

- Page 2 Line 17-23, "We compare . . . Belt and Muller, 2013)": This text should only come at the end of section 2

Done.

- Section 2, 2nd paragraph: In this paragraph it is stated that the origin of Renland sea salt is the North Atlantic and the text refers to Appendix B. This is a crucial piece of information and should not be hidden in an Appendix but included in the main text.

This analysis is now part of the Section 2.3 (main text): "*Atmospheric reanalysis: the source region of bromine and sodium for the RECAP ice core*".

Moreover some more information on this statement would be helpful for the reader:

a) sea salt aerosol has a pronounced seasonal cycle with a broad maximum in the winter half-year. This also holds for eastern coastal sites (Oyabu et al., Polar Science, 2016). Thus, the Renland sea salt record is mainly representative for the winter half-year. The back trajectory analyses should be done both for the winter and summer half-year separately and shown in two panels.

The atmospheric reanalysis has been done on a seasonal basis; the results are shown in the 4 panels (seasons) of Figure 1. We note, however, that although sea salts inputs have winter maxima, bromine explosions occur during springtime and so do Br_enr values, as it has been (broadly) observed in shallow core records (Spolaor et al., 2014, Maffezzoli et al., 2017, Vallelonga et al., 2017).

Note also that due to the limited lifetime of sea salt aerosol, short trajectories are likely to bring more sea-salt aerosol to Renland compared to longer trajectories.

Thanks for the comment. A line has been added: "...although the ocean regions closer to Renland are expected to be more significant as per the observed chemical signature".

b) transport pathway (trajectories) is one side of the coin, the sea ice source is the other. An additional figure/panel showing the multiyear and first year sea ice distribution in spring (at its maximum) would be helpful to support the claim that the source regions identified in the trajectory study are covered by FYSI or OW. The National Snow and Ice Data Center provides this information (https://nsidc.org/data/nsidc-0611).

A sea ice age plot has been added (Figure. 2), showing a representative winter maximum (Feb 1987) and summer minimum (Sept 2012) during the satellite era.

- A method section is missing. Again, this is hidden in an Appendix A but should be part of the main text.

The Method section is now fully described in the main text (Sect. 2) and includes three subsections: - 2.1: The 2015 RECAP ice core

- 2.2: Experimental: determination of Br, Na, Cl, Ca and Mg by mass spectroscopy
- 2.3: Atmospheric reanalysis: the source region of bromine and sodium for the RECAP ice core

- Page 3 line 15-20, "The Br_enr . . . Holocene value.": This text should be the beginning of section 3 These lines have been now moved to the "Results and Discussion" section (Sect. 4).

- Page 3 line 22-29. Here the NEEM Br_enr record is mentioned and compared to Renland. In order to allow the reader to make this comparison, the NEEM data should be shown in one of the figures! Done (Fig. 4).

- Page 4 line 1: Here the expression "tipping point" is used. There is no clear definition of what a tipping point is, but in climate science it is usually used for a rapid regime shift (see IPCC). The change from MYSI to FYSI to OW, however, is likely a gradual process. Accordingly, I would suggest to avoid the expression tipping point in the manuscript. Along this line, I think section 3.2 (linearization of the Br_enr record) and its application on the 120 kyr record in section 3.3 does not provide added value and in fact is misleading due to the gradual nature of the MYSI/FYSI/OW transition. Instead of trying to force this mathematically to a monotonous function, I would recommend to just use color bars in the figures underlying the records to indicate where MYSI, FYSI or OW dominate the Br_enr record. Moreover, mean Greenland temperature may not be the only parameter determining the amount of FYSI present (see next comment).

We agree, the term tipping point has now been removed. We also removed the Brenr-transformation.

- The high resolution data presented in Figure 3 clearly show that the YD is the time period of largest Br_enr (clear maximum) during termination I, thus the largest FYSI presence. In particular Br_enr is clearly higher in the YD than in the OD period. Note that there is no similar Br_enr maximum during the OD/BA transition as seen in the YD at the point when temperature during the OD/BA transition is crossing the same temperature as found in the YD. Either this point is just missed in the record (unlikely), or NGRIP temperature alone is not able to fully explain the observations. Here additional information could be used to elaborate on this issue. First of all, FYSI is strongly dependent on the seasonal temperature variation, this should be mentioned somewhere. Models suggest (Buizert et al., Science, 2014) that temperature seasonality during the YD and OD was much higher than during the BA. This could explain why the YD has higher FYSI than the BA. The difference between YD and OD sea ice conditions (Br_enr levels) may potentially be explained by the overall much lower temperatures encountered during the OD than in the YD (Buizert et al., 2014), which may push the OD sea ice

regime toward more MYSI. This difference may be linked to the generally higher AMOC strength in the YD compared to the OD (McManus et al., Nature 2004).

- Page 4 line 29-33: Here the paper by Rasmussen is referred to, but I am not sure based on the text provided - whether it is referred to correctly and whether the statement made in the manuscript is correct. Rasmussen et al. (2016) claim that in the North Atlantic south of Iceland SST warming already starts during stadial conditions, while in the GIN seas the warming starts only with the Greenland DO onset, i.e., when sea ice rapidly declines. Rasmussen et al., do not explicitly discuss the YD/BA/OD transitions and in fact their record does not show a clear early warming during the reduced AMOC conditions of the YD and OD. Accordingly, to make this statement would require high resolution Br_enr data for selected DO events from the Renland record, which are not available yet. I would suggest to remove this statement and also the reference to mean ocean temperatures, Antarctica and CO2.

Thanks for these comment. We will provide a single comment. This section (4.1) has now been analyzed in greater detail. We agree with the comments on the differences between YD/BA and OD, adding also that the availability freshwater from melting ice sheets during the deglaciation could have facilitated the formation of fresh sea ice surfaces. We agree on the comparison with Rasmussen, which has now been removed. We left however the sentence on the mean ocean temperatures, Antarctica and CO2 since we believe it could be a meaningful point for a broader perspective. Two additional PIP records from the Norwegian Sea has been added the Figure 6. The section of the deglaciation now reads:

4.1 The last deglaciation and the dual Br enr regimes

We now consider in further detail the last deglaciation, when a number of ocean temperature, salinity, circulation and sea ice changes are observed in the Nordic seas (Fig. 6). Marine-derived local sea ice records from both the Svalbard margin and the Norwegian Sea indicate (Fig. 6e,f) that near-perennial sea ice (PIP 25 \approx 0.5-1) was present during MIS 2 until ~17 kyr (17.6 kyr recorded in the Svalbard margin), the onset of a major breakup of extensive sea ice cover, during Heinrich Event 1 (18 to 15 kyr). Synchronous to within a few centuries, several modifications relevant to the North Atlantic ocean are observed (Fig. 6), including sea water surface freshening and warming in the polar and subpolar North Atlantic (the 67 °N Dokken and Jansen 1999 record is shown as an example in Fig. 6c) and a near total cessation of the Atlantic Meridional Overturning Circulation (AMOC, Fig. 6d). Generally low to intermediate PIP 25 values (PIP 25 \approx 0-0.5) are reported in the Svalbard Margin and in the Norwegian Sea in the \sim 17-12 kyr period (Fig. 6e,f,g), with a slight increasing trend throughout the Bølling-Allerød (BA) and a broad maximum reached during the Younger Dryas (YD), suggesting that seasonal sea ice conditions were dominating this period. Other studies from marine records in the Nordic Seas records also suggest milder sea ice conditions during the BA and increased sea ice during the YD (Belt et al., 2015; Cabedo-Sanz et al., 2016). In contrast, a record from the northern Icelandic Shelf (Fig. 6e) shows that here the sea ice conditions remained near-perennial from 14.7 to 11.7 kyr (PIP 25 \approx 0.5-1). The authors (Xiao et al., 2017) suggest that this pattern of more severe sea ice conditions in the north of Iceland is, at least during the BA and the YD, linked to the flow of warmer waters from the North Atlantic Current, influencing sea ice melting in the eastern Nordic Seas, whereas the Icelandic shelf is influenced by colder polar waters from the East Greenland Current and the East Icelandic Current.

The RECAP ice core was resampled at sub-centennial resolution to better constrain the timing of sea ice changes through the deglaciation in the 50-85 °N North Atlantic (Fig. 6b, squares). The $Br_{enr,Mg}$ serie ($Br_{enr,Cl}$ would lead to the same results) would indicate that FYSI started to increase in the North Atlantic, concurrent to a reduction of MYSI, at ~17.5 kyr, synchronous with local PIP 25 decrease in

the Svalbard margin and eastern Nordic Seas and in response to sea surface temperature warming in the North Atlantic. This findings would also suggest that North Atlantic sea ice changes occurred in concert with temperature and circulation changes of the underlying surface waters. We note that this time period also coincides with the initiation of dealacial changes in mean ocean temperature, Antarctic temperatures and atmospheric CO 2 concentrations toward interglacial values (Bereiter et al., 2018). North Atlantic FYSI continued to increase throughout the BA (except for one point at 12.7-12.4 kyr at the onset of the YD) until a maximum at 12.4-11.8 kyr during the YD, when a clear Br_{enr.Ma} maximum is observed (Fig. 6). From the comparison between the marine and ice core results, we infer that, during the 17-12 kyr period, the 50-85 °N-integrated North Atlantic sea ice changed from MYSI to FYSI. Local sea ice was also melting at \sim 17 kyr in the eastern Nordic Seas, likely influenced by the North Atlantic Current, while, at least from 14.7 to 11.7 kyr, sea ice was still near-perennial at the *North Icelandic shelf, possibly due to the influence of cold waters carried by the East Greenland* Current. Following its maximum value at 12.4-11.8 kyr, Br enr (i.e. FYSI) started to decrease (Fig. 6b). We suggest that from this point-in-time, the Br enr indicator now shifts to the FYSI/OW regime (Fig. 5), and the North Atlantic basin became largely ice free. A retreating FYSI scenario is also recorded in all 5 marine cores (decreasing PIP 25 to \approx 0-0.4 during the Early Holocene, Fig. 6e,f,g), suggesting that open water conditions progressively developed in the whole North Atlantic basin, sustained by increasing heat transport from the North Atlantic Current and a strengthened AMOC since \sim 11.7 kyr (McManus et al., 2004; Ritz et al., 2013).

Since Br enr is assumed to be an increasing function of FYSI, its decrease would point to either OW or MYSI conditions, following either the FYSI/OW or the FYSI/MYSI regimes (Fig. 5). At any point in time, only one regime is considered to be in place, and we suggest a simple model in which a temperature threshold could be the discriminating variable setting the regime type. Since a change of regime is observed during the deglaciation, with maximum Br enr values (i.e. FYSI) at 11.8-12.4 kyr, we set the threshold to be the mean NGRIP temperature reconstructed for that period: T NGRIP =- 4.6±0.9 (20) °C (the two lines in Fig. 6a). In every ice sample of the 120,000 year record the regime type (FYSI/MYSI or FYSI/OW, see Fig. 5) can thus be determined according to its integrated temperature value with respect to the temperature threshold: FYSI/MYSI for a lower temperature value and FYSI/OW for a higher temperature value.

According to this simple model the deglaciation is characterized by the FYSI/MYSI regime until the onset of the Bølling-Allerød (except few points at which the regime type depends on the chosen threshold value, Sect. 4.2), while the FYSI/OW regime operated from that point forward. We note that there is no similar Br enr maximum at the onset of the Bølling-Allerød as seen in the YD at the point when NGRIP temperature is crossing the same temperature as found in the YD. The NGRIP temperature alone appears therefore not to be able to fully explain the observations. The possible explanation of higher Br enr values during the Younger Dryas compared to the Bølling-Allerød may reside in the higher seasonal temperature variations (Buizert et al., 2014) and freshwater inputs from melting ice sheets in the former period, both promoting the formation of seasonal sea ice. Conversely, the lower Br enr values (hence to greater MYSI in the FYSI/MYSI regime) during the Older Dryas compared to the Bølling-Allerød and the Younger Dryas may be linked to the overall much lower temperatures during this period (Buizert et al., 2014), higher surface water salinity due to less freshwater inputs from melting ice sheets and a generally weaker AMOC.

- Page 6, line 26. Here you refer to the GI numbers. These should be included in the figures Done.

- Include Fig. 1 in Fig. 2 Fig. 4 now shows all the measured records from RECAP.

- Add the NEEM record in Fig. 2 or provide a separate figure for the NEEM/Renland comparison. Done (Fig. 4).

- Add color bars for sea ice conditions underlying figure 2 As it is the first plot, no error bars are colored in Figure 4. The colors appear in Figure 7, where the two regimes are discussed.

- add color bars for sea ice conditions underlying figure 3 The two regimes are just indicated with two colored bands (Fig. 6).

- remove figure 5 Done.

- remove the transformed BR_enr in figure 6, add color bars Done (Fig. 7).

- move Appendix A to a Method section Done, now Sections 2.1 and 2.2 (Methods).

- move Appendix B to section 2 and add more information as outlined above Done, now Section 2.3 (Methods).

Specific Comments - see annotated pdf We really thank the Reviewer for the annotated pdf.

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REVIEW - REFEREE#3

As the first two reviewers already raised the main concerns of the manuscript for which I totally agree, I will only focus on a general comment. Throughout of the manuscript the authors take granted that the enrichment of Br above sea water has only one explanation, namely the first-year sea ice extension. The discussion that follows never question this hypothesis and only interprets enriched Br as the first-year sea ice extension. Such concordance is so strong in the paper that enriched Br and first-year sea ice extension are used indifferently, like synonyms.

The use in ice core/snow of Br-excess as a proxy of sea ice extent was initiated in 2013 by the present author's team. Since then, this team has published around 10 papers with most of them using the same adequacy Br-excess = FYSI to reconstruct sea ice in Russian & Canada Arctic, East Antarctica, in East Greenland.

However, quoting their own conclusion in their 2013 original paper: "Although further investigation is required to characterize potential depositional and post-depositional processes, these preliminary findings suggest that I and Br can be linked to variability in the spring maximum sea ice extension and seasonal sea ice surface area (Spolaor et al., The Cryosphere, 2013)". Requirement and suggesting are the two most important terms in their conclusions. With time "suggest" has become a certitude and "require" has turned into still awaiting. I could not find in ISI database any papers treating this hypothesis at the process level as originally suggested the authors.

In J Abbath's Nat Geos comment of Pratt et al. 2013 paper (which results are at the heart of their hypothesis), I quote "Whether the rising fraction of young sea ice will enhance snowpack bromine production and release, and concomitant changes in atmospheric chemistry, remains to be seen (J. Abbath, Nat Geo, 2013)". Interestingly, Pratt's paper argues that acidity of snow is a prerequisite for Br activation as well as internal snow pack air chemistry, e.g OH snow pack production, nitrate/nitrite concentration etc. As the relative scavenging efficiency between ssNa and gaseous Br seems to be the main driver of the Br-excess, change in precipitation regime can potentially greatly influence the Br/Na ratio. A transect study from coast the inland along a strong accumulation gradient will be very informative in this view. The speciation of Bry species, air snow transfer are also important parameters to consider. In summary, the current literature gives little concrete elements to definitely link Br-excess and FYSI extension. Increase production efficiency at constant extension is for instance never considering, neither change in scavenging precipitation, etc. A correct scientific approach should be: 1-Establish the hypothesis 2- Test it against observations, determine the sensitivity to parameters 3- And use it within its limits It seems to me that the step 2 is currently missing for Br-excess. There is a long list of acclaimed proxies in the ice core community that after a throughout investigation turned out to have been interpreted too simply (just few of them MSA, MSA/sulfate ratio, water isotopes, levoglucosan, nitrate concentration).

In conclusion, the authors should clearly state that their hypothesis is not yet fully demonstrated and their conclusions are only hypothetical. Currently, other interpretations are possibles.

We thank Reviewer#3 for her/his comment. We agree on the general point that the uncertainties related to the Brenr have to be clearly stated. The manuscript introduction now focuses on Brenr as a potential indicator of past sea ice conditions (1. Introduction: Brenr as a potential indicator for past sea ice conditions). The uncertainties and the key aspects that require future studies and investigations are

discussed. This paragraph includes both aspects that Reviewer#3 and Reviewer#1 pointed out, as well as other few aspects. This section is structured into three general aspects related to Brenr: activation, transport and deposition/postdeposition. The reader can find this section attached in the answer file to Reviewer#1 (pp2,3).

120,000 year record of sea ice in the North Atlantic ?

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Abstract. Although it has been demonstrated that the speed and magnitude of recent Arctic sea ice decline is unprecedented for the past 1,450 years, few records are available to provide a paleoclimate context for Arctic sea ice extent. Bromine enrichment in ice cores has been suggested to indicate the extent of newly formed sea ice areas. Despite the similarities among sea ice indicators and ice core bromine enrichment records, uncertainties still exist regarding the quantitative linkages between

- 5 bromine reactive chemistry and the first year sea ice surface. Here we present a 120kyr,000 year record of bromine enrichment from the RECAP ice core, coastal East Greenland, and interpret it as a record of first-year sea ice. We compare it to existing sea ice records from marine cores and tentatively reconstruct past sea ice conditions in the North Atlantic as far north as the entrance of the Aretic Ocean-Fram Strait (50-85 °N). Bromine enrichment has been previously employed to reconstruct first-year sea ice (FYSI) in the Canadian Arctic over the last glacial cycle. We find that during the last deglaciation, the transition
- 10 from multi-year sea ice (MYSI) to FYSI to first-year sea ice started at ~17.6-17.5 kyr, synchronous with sea ice reductions observed in the eastern Nordic seas (Müller and Stein, 2014; Hoff et al., 2016) and with the increase of North Atlantic ocean temperature(Dokken and Jansen, 1999). FYSI First-year sea ice reached its maximum extent at 12.4-11.8 kyr during the Younger Dryas, after which open-water conditions started to dominate, as supported by sea ice records from the eastern Nordic seas and the North Icelandic shelf. Our results show that over the last 120,000 years, multi-year sea ice extent was greatest
- 15 during Marine Isotope Stage (MIS) 2 and MIS4, with decreased levels during MIS3 and the onset of the last glacial period (late-MIS5), possibly during MIS 4, with more extended first-year sea ice during MIS 3 and MIS 5. Sea ice extent during the last 10 kyr (Holocene /MIS1Holocene (MIS 1) has been less than at any time in the last 120kyr,000 years.

1 Introduction: Br_{enr} as a potential indicator for past sea ice conditions

The connection between Arctic sea ice and bromine was first identified through an anticorrelation between springtime ground
 level ozone (O₃) and filterable bromine <u>air</u> concentrations (Barrie et al., 1988). Large bromine oxide (BrO) column enhancements and simultaneous tropospheric ozone depletion were later found in Antarctica (Kreher et al., 1997). Satellite observations

reveal geographically-widespread 'bromine explosions', the sudden increase of atmospheric bromine concentrations during springtime occurring in both polar regions (Chance, 1998; Richter et al., 1998; Wagner and Platt, 1998). The primary sources of reactive bromine in the polar atmosphere remained unclear until bromine recycling over fresh sea ice surfaces was suggested to occur at polar sunrise. The mechanism proceeds via springtime photochemical heterogeneous reactions over fresh sea ice

- 5 surfaces that lead to the activation of bromide, followed by the release and exponential increase of gas-phase bromine species in the polar troposphere (Vogt et al., 1996). Several saline substrates on fresh sea ice surfaces (hereafter referred as first-year sea ice, FYSI) were suggested as reservoir reservoirs of reactive sea salt aerosol aerosols (SSA) capable of sustaining bromine recycling (Abbatt et al., 2012; Saiz-Lopez and von Glasow, 2012). To date, both model studies (Yang et al., 2008, 2010) and experimental evidence (Pratt et al., 2013; Zhao et al., 2016) (Pratt et al., 2013; Zhao et al., 2013; Zhao et al., 2013; Zhao et al., 2013; Zhao et al., 2014; Frey et al., 2019) consider the de-
- 10 posited snow layer on FYSI (known as salty blowing snow) to be the most efficient substrate for SSA release and bromine activation.

Atmospheric bromine and sodium thus originate from both oceanic and FYSI sea salt aerosol, where they are found in mass ratios identical to that of sea water . As a consequence of their concentration ratio is that of sea water mass: $(Br/Na)_m = 0.0062$ (the subscript indicates 'marine', Millero et al. (2008)). Br_{enr} values in ice core records, i.e. the bromine-to-sodium mass ratios

- 15 beyond the sea water value (Eq. 1) were introduced by Spolaor et al. (2013b) as a potential proxy for past FYSI conditions within the ocean region influencing the ice core location. The basic assumption behind this idea is that bromine recycling on FYSI surfaces , the atmospheric would increase the bromine-to-sodium mass ratio is increased with respect to that of sea water, leading to a bromine enrichment (), observed in polar snow. Measurements of beyond the sea water value in the atmosphere and at the ice core location, thus isolating effect of FYSI-induced bromine recycling from the sea salt aerosol contribution.
- 20 the latter being interfered by oceanic emissions. Further studies have provided some evidence on the validity of Br_{enr} in ice cores can thus be used to reconstruct FYSI conditions (Spolaor et al., 2014). The first Arctic glacial-interglacial investigation of as a FYSI indicator (Spolaor et al., 2013a, 2014, 2016a). At present, however, this interpretation is still challenged by several uncertainties related to the different variables which play a role in the involved chemical and physical processes (Abbatt, 2013). These can be grouped into three categorizes: bromine activation, transport and deposition/postdeposition. They are briefly
- 25 discussed here.

Pratt et al. (2013) showed that saline snow collected on Arctic tundra or first-year sea ice surfaces can serve as an efficient reservoir for bromine activation. They point out, however, that acidity of snow is a pre-requisite for bromine activation, as well as internal snow pack air chemistry (e.g. concentration of OH, NO_3^-/NO_2^-). A modification in these factors would alter the bromine production efficiency at constant FYSI extent. Aged sea ice (second-year sea ice or older) is possibly contributing to

30 bromine activation due to a non-zero salt content, but the amount of this second order source effect is unknown. Bromine travels from the source region to the ice core location both in the aerosol and gas phases, while sodium is only found in the aerosol phase. To date, the partitioning of bromine species between the two phases isn't clear. Additionally, some studies have reported that bromine recycling takes place within the plume even during the transport (Zhao et al., 2016), leading to possible bromine depletion from the aerosol in favor to the gas phase. Overall, the final observed Br_{enr} was performed on the

35 NEEM ice core, located in Northwest Greenland and representing sea ice aerosol from the Canadian archipelago, Baffin Bay

and Hudson Bay (Spolaor et al., 2016b). The NEEM 120 kyr sea ice reconstruction found greater values in ice samples depend on the relative importance of aerosol and gas-phase bromine. It appears that fine aerosol are enriched in bromine, while coarse particles are depleted (Legrand et al. 2016, Koenig pers. comm.). Additionally, the two phases likely have different atmospheric residence time, thus the final bromine-to-sodium ratio found in the snow might be function of the transport duration. On

5 this topic, Simpson et al. (2005) experimentally showed that sodium is washed out from the atmosphere faster than bromine, suggesting the role of longer lasting gas-phase bromine. On the other hand, a model run by Spolaor et al. (2013b) showed opposite results. To which extent transport processes can interfere with a source effect is yet to be resolved.

The last set of uncertainties relates to the variables associated to the deposition of bromine and sodium, such as the variability in accumulation rates over long time scales, which would impact the relative importance of the wet and dry contributions of the

10 two species. Finally, photolytic reemission of bromine from the snowpack could be responsible for bromine loss and decreased Br_{enr} valuesduring the Holocene and interstadials, pointing to greater FYSI conditions during such periods. Lower values during colder climate phases indicated more extended multi-year sea ice (MYSI) coverage.

2 The RECAP ice core and the record

We acknowledge the above-mentioned open questions on the validity of Br_{enr} as a FYSI proxy and clearly point out that
further studies are needed to shade light to these uncertainties. We thus proceed with the interpretation of Br_{enr} as a FYSI proxy bearing in mind that other variables could, at present, explain a Br_{enr} record.

Here, we investigate consider the available evidence regarding past sea ice conditions in the North AtlanticOcean, based on the record extracted, and present a bromine enrichment record from the RECAP ice core, located in coastal East Greenland. Because of its location, the record is sensitive to ocean processes and sea ice dynamics (Cuevas et al., 2018)RECAP should

20 record the fingerprint of sea ice in the North Atlantic. We compare our sea ice record with results from three the RECAP Br_{enr} record with sea ice reconstructions from five marine sediment cores drilled within the Renland source area: the Fram Strait, the Norwegian Sea and the North Icelandic shelf (Fig. 1).

2 Methods

2.1 The 2015 RECAP ice core

- 25 The RECAP (REnland ice CAP) ice core was retrieved from the Renland ice cap (71° 18' 18" N; 26° 43' 24" W; 2315 m a.s.l.) from May 13th to June 12th, 2015. The ice cap is located on the Renland peninsula and is independent of the main Greenland ice sheet, with fjords to the north and south. A 98 mm diameter ice core was recovered to 584 m (bedrock). The drilling occurred in a dry borehole to 130 m depth and estisol 140 drilling fluid was used for the remaining depth. The record covers the last 120,800 years. The core age model is described in full details in the Supplementary of Simonsen et al., Nat.
- 30 Comm., 2019 (accepted). The index is asemi-quantitative indicator of the local sea ice condition at the marine core location. It

is calculated by coupling the sediment concentration of , a biomarker produced by diatoms living in seasonal sea ice , with an open waterphytoplankton biomarker (brassicasterol or dinosterol, hence or)

The ice core samples dedicated to mass spectroscopy measurements (n=1205) were collected and automatically decontaminated from a continuous ice core melting system as part of the RECAP Continuous Flow Analysis (CFA) campaign conducted at

- 5 the University of Copenhagen in Autumn 2015. The ice core was melted at a speed of approx. 3 cm min⁻¹ on a gold coated plate copper melter head (Bigler et al., 2011; Kaufmann et al., 2008). Meltwater was collected continuously from the melter head into pre-cleaned polyethylene vials (cleaned with ultrapure water, > 18.2 M Ω cm⁻¹) at two different depth resolutions. From the ice cap surface to a depth of 535.15 m, samples incorporated ice meltwater spanning depths of 55 cm. From a depth of 535.15 m to the ice cap bedrock the samples integrated 18.3 cm. The time resolution is annual to multicentennial in the
- 10 Holocene and from centennial to millennial in the glacial section. After collection, the samples were immediately refrozen at -30 °C and kept in the dark until mass spectroscopy analyses to reduce bromine photolysis reactions.

2.2 Experimental: determination of Br, Na, Cl, Ca and Mg by mass spectroscopy

The samples were shipped to Ca' Foscari University of Venice (Italy, n=770) and Curtin University of Technology (Perth, Australia, n=435) for determination of bromine (Br), sodium (Na), chlorine (Cl), calcium (Ca) and magnesium (Mg) by

- 15 Inductively Coupled Plasma Mass Spectroscopy. Only bromine and sodium were measured in the samples analyzed at the Italian lab, while all elements were quantified with the Australian setup. The depth and age ranges associated with the Italian samples are: surface-150 m (2015 AD-328 yr b2k); 165-219 m (383-636 yr BP); 234-413 m (727-2857 yr b2k); 441-495 m (3522-5749 yr b2k). 435 samples were measured in Australia. The depth and age ranges associated with the Australian samples are: 150-165 m (328-383 yr b2k); 219-234 m (636-727 yr b2k); 413-441 m (2857-3522 yr b2k); 495-562 m (5749-120788 yr
- 20 <u>b2k).</u>

University Ca' Foscari of Venice, Italy

Bromine and sodium (⁷⁹Br and ²³Na) were determined by Collision Reaction Cell-Inductively Coupled Plasma-Mass Spectrometry (CRC-ICP-MS, Agilent 7500cx, Agilent, California, USA). The introduction system consisted of an ASX-520 autosampler (CETAC Technologies, Omaha, USA) and Scott spray chamber fitted with a MicroFlow PFA nebulizer. The sample flow was

25 kept at 100 μ Lmin⁻¹. All reagents and standard solutions were prepared with ultrapure water (UPW, 18.2 M Ω cm⁻¹). Nitric acid (65% v/v, trace metal grade, Romil, Cambridge, UK) and UPW washes (2 minutes each respectively) were used for background recovery after every sample analysis. The nitric acid washing concentration was lowered to 2%. The experimental routine (standards and calibrations), as well as the overall instrument performance (detection limits and reproducibility) are the same as in Spolaor et al. (2016b). Briefly, the index is a dimentionless number varying from 0 to 1: \approx

30 Curtin University of Technology, Perth, Australia

The analyses were performed by Inductively Coupled Plasma Sector Field Mass Spectroscopy in reverse Nier-Johnson geometry (ICP-SFMS, Element XR, Thermo Fisher, Germany) inside a Class 100 clean room environment at Curtin University TRACE facility (Trace Research Advanced Clean Environment Facility). The ICP-SFMS introduction system consisted of an Elemental Scientific Inc. (ESI, Omaha, USA) syringe-pumped autosampler (Seafast II) with a 1 indicates perennial sea ice cover; ≈ 0

indicates open water conditions, while intermediate values reflect seasonal sea ice (Müller et al., 2011; Belt and Müller, 2013). mL PFA capillary injection loop and using an ultrapure water carrier. A 1 ppb indium internal standard in 5% v/v nitric acid (HNO₃, double PFA distilled) was mixed inline at a flow rate of 25 μ Lmin⁻¹ using a T-split (final flow rate of 400 μ Lmin⁻¹, take-up time 1.5 min). Nebulization occurred in a peltier cooled (2 °C) quartz cyclonic spray chamber (PC3, ESI), fitted with a

- 5 PFA micro-concentric nebulizer (PFA-ST, ESI). Bromine, sodium and magnesium, chlorine and calcium isotopes (⁷⁹Br, ²³Na, ²⁴Mg, ³⁵Cl, ⁴⁴Ca) were detected in medium resolution (10% valley resolution of 4000 amu) and normalized to ¹¹⁵In. Memory effects were reduced by rinsing the system between samples with high purity HNO₃ (3%) and UPW. One procedural blank and one quality controlled standard (QC) were analyzed every 5 samples to monitor the system stability. The detection limits, calculated as 3σ of the blank values (n=80) were: 0.18 ppb (Br); 1.1 (Na); 0.2 ppb (Mg); 1.6 ppb (Ca) and 4.6 ppb (Cl). The
- 10 majority of the sample concentrations were above the detection limits for all elements (>97%). The relative standard deviations of the control standard samples (n=82) concentrations were monitored over >100 hours and were 9% (Br); 4% (Na); 3% (Mg); 4% (Ca) and 13% (Cl). Calibration standards were prepared by sequential dilution (7 standards) of NIST traceable commercial standards (High-Purity Standards, (Charleston, USA)). All materials used for the analytical preparations were systematically cleaned with UPW (18.2>MΩcm⁻¹) and double PFA distilled ultrapure HNO₃ (3%, prepared from IQ grade HNO₃, Choice
- 15 <u>Analytical Pty Ltd, Australia) throughout.</u>

The RECAP ice core was drilled in 2015 in the Renland peninsula and covers the last 120, 000 years (Appendix ??). A back trajectory analysis spanning the 2000-2016 period indicates that Renland is associated with a different SSA source area compared to the NEEM ice core (Appendix 2.3). The source of Renland SSA has been constrained to the ocean basin A laboratory intercomparison between sodium and bromine measurements was performed on a common set of samples

20 (n=140) to investigate differences between the analytical techniques and laboratories, as described in Vallelonga et al. 2017. The correlations and the gradients between the measured concentrations in the two setups are $\rho(Na_{IT}-Na_{AUS})=0.99$ (n=140; p<0.01), $m_{Na}=1.08\pm0.01$ (1 σ) for sodium and $\rho(Br_{IT}-Br_{AUS})=0.93$ (n=140; p<0.01), $m_{Br}=1.08\pm0.02$ (1 σ) for bromine.

2.3 Atmospheric reanalysis: the source region of bromine and sodium for the RECAP ice core

To estimate the source of bromine and sodium deposited at Renland, daily back trajectories were calculated from 2000 AD

- 25 to 2016 AD with HYSPLIT4 (Stein et al., 2015; Draxler et al., 1999; Draxler and Hess, 1998, 1997), using publicly available NCEP/NCAR Global Reanalysis meteorological data (1948-present), with a 2.5° resolution in both latitude and longitude (Kalnay et al., 1996). The back trajectories were started daily on an hourly basis at 500 m above the Renland elevation (71.305 °N, 26.723 °W, 2315 m a.s.l.) for the 17 year time span. The trajectory time was set to be 72 hours, representing the average atmospheric lifetime of sea salt aerosol (Lewis and Schwartz, 2004) and likely a lower limit for inorganic gas-phase bromine
- 30 compounds. To access the potential marine sources of bromine and sodium, only a subset of the all trajectories is considered. Such selection limits the ensemble to only such trajectories that crossed the marine boundary layer (MBL), defined here as the 900 hPa isosurface (corresponding to approximately 1000 m a.s.l.), for at least 10 hours. This pressure value was chosen according to Lewis and Schwartz (2004) and references within. One way to display a map of MBL layer crossings is the Residence Time Analysis, a spatial distribution of trajectory endpoints (Ashbaugh et al., 1985). This map indicates that

75% of the signal originates from the North Atlantic Ocean, extending in latitude from 50° N to 85° N (corresponding to the North Atlantic up to the Fram Strait) and in longitude from the western coasts of Norway and the UK to East Greenland (Fig. ??). Thus Renland receives a negligible SSA contribution from West Greenland waters. The source 1), although the ocean regions closer to Renland are expected to be more significant as per the observed chemical signature. A minor contribution is

5 expected from aerosols and gas-phase bromine originated from coastal waters off West Greenland. The consistency of sea ice reconstructions from the RECAP core and the Nordic Sea sediment cores (Sect. 4.2) suggests that the source area could extend to these regions throughout the last 90 kyr. For the overall interpretation of the RECAP record, the source area is therefore assumed to be the 75% contour region of the Residence Time Analysis endpoint distribution (Fig. 1). Such region is nowadays mostly dominated by open water (OW) and FYSI conditions, with only minor contributions of MYSI, FYSI grown in situ and

10 MYSI transported south from the Arctic Ocean alongside the East Greenland coastline via the Transpolar Drift -(Fig. 2).

3 Calculation of bromine enrichment (Brenr) and Brenr time series

Sodium and bromine concentrations were determined by Inductively Coupled Plasma Mass Spectroscopy (ICP-MS, Appendix 2.2). The bromine enrichment values , , were calculated according to the following equations in the ice samples are calculated from the departure from the sea water abundance, the latter inferred from sodium (hereafter sea-salt sodium, ssNa):

$$Br_{enr} = \frac{Br/ssNa}{(Br/Na)_m}$$
(1)

$$\underline{ssNa} = \longleftrightarrow < 1.8 \tag{2}$$

 $\underline{ssNa} Na = \underline{Na} \longleftrightarrow \geq 1.8$ (3)

where Brand ssNa ssNa + nssNa

15

where Br, ssNa and nssNa are the bromineand, sea-salt and non-sea-salt sodium concentrations in the ice samples $\frac{1.8}{1.8}$ 20 and respectively, and $(Br/Na)_m = 0.0062$ are respectively the chlorine/sodium and bromine/sodium sea water mass ratios (Millero et al., 2008).

Since sodium has a potential crustal contribution is the bromine-to-sodium mass ratio in sea water (Millero et al., 2008), assumed constant in time and space. Since sodium concentrations in ice cores can be interfered by terrestrial inputs (nssNa, from sodium oxide Na₂O), which can be significant especially during generally contribute up to $\approx 10-50\%$ during glacial

- 25 arid periods (i.e. stadials), it was necessary to extract its marine contribution. For this purpose, we used chlorine, which was measured along with bromine and sodium by ICP-SFMS (nssNa and ssNa have to be evaluated to calculate Br_{enr} (Eq. 1-3). We estimate the ssNa concentrations by using three methods. They make use of chlorine, magnesium and calcium concentrations. The decrease of the Cl/Na record in Fig. ??) . Chlorine/sodium mass ratios greater than that of seawater are observed in polar ice cores during warm climate periods due to mass ratio from the sea water reference (1.8 from Millero et al. 2008)
- 30 can be related to extra sodium inputs of terrestrial origin. Thus, the ssNa can be calculated from chlorine if the measured

chlorine-to-sodium ratio becomes less than 1.8, while zero nssNa is assumed otherwise (Eqs. 4-5):

$$ssNa = \frac{Cl}{1.8} \longleftrightarrow \frac{Cl}{Na} < 1.8$$

$$ssNa = Na \longleftrightarrow \frac{Cl}{Na} \ge 1.8$$
(4)
(5)

 $\underbrace{\text{Unfortunately, sea salt aerosols are affected by}_{\text{acid-scavenged}} (HNO_3 \text{ and } H_2SO_4) \text{ dechlorination } \underbrace{\text{of sea salt aerosols processes}}_{\text{processes}} \underbrace{\text{Constraint}}_{\text{processes}} \underbrace{\text{Constraint}}_$

- 5 (e.g. 2NaCl + H₂SO₄ → 2HCl + Na₂SO₄). The removal of , in which chlorine is removed as gas-phase HClis believed to be more efficient during warmer elimatic conditions, resulting in . These processes would increase the Cl/Na ratios in ice beyond the sea water reference (Legrand and Delmas, 1988), especially in warmer climatic conditions, since HCl has is believed to have a longer atmospheric residence time than SSA. No correction to sodium is therefore applied when In fact, RECAP Holocene Cl/Na ratios (≈ 3) suggest that dechlorination processes do occur and chlorine is also deposited as HCl.
- 10 Additionally, this result shows that HCl loss from the snowpack after deposition is limited, opposite to what is observed in Antarctica (Dome C) (Röthlisberger et al., 2003). The huge difference between Dome C and Renland accumulation rates likely explain such differences. In glacial ice, and especially during MIS2 and MIS4, the effect of dust neutralization of nitric and sulfuric acids would reduce chlorine loss both after deposition and during transport, therefore making the above correction more trustworthy (Wolff et al., 1995). However, the relative contributions of dechlorination processes and dust loading as per
- 15 the observed Cl/Na ≥ 1.8. The sodium crustal contribution was found to be up to 29% during 60-70 kyr and ratios in the ice are difficult to quantify. A correction based on Cl/Na ratios was used by Hansson (1994) to calculate the ssNa and nssNa concentrations in the 1988-drilled Renland core. In particular, they found that nssNa accounts for respectively 17% and 24% of the total sodium during MIS2 and MIS4. If the same correction is applied to our measurements, we find values of 15% at ~23 kyr, consistent with the resultsfrom the Renland 1988 ice core (Hansson, 1994), and 20-30% during MIS2 and MIS4, close to
- 20 their findings.

The time series (Fig. 4) displays values greater than A better alternative to calculate ssNa and nssNa would be to use an element of terrestrial origin. Magnesium contains both a marine and a terrestrial signature. By using reference mass ratios of Na/Mg in sea water and in the Earth upper continental crust, we are able to calculate the marine sodium contribution (Eq. 8). Often in ice core studies, the sea-salt and non-sea-salt contributions are calculated using global-mean reference values of

- 25 chemical composition of terrestrial elements (see for example Rudnick and Gao 2003, Table 1throughout the record, suggesting a persistent FYSI influence. Values are on average low during the Holocene (). However, the spatial variability of the dust mineralogy can heavily impact the results. In our case, the terrestrial value (Na/Mg)t to be used in the calculation (the subscript 't' refers to 'terrestrial') would require knowledge of the geochemical composition of the dust deposited at Renland. In remote Greenlandic ice cores, the provenance of aeolian mineral dust is believed to be the deserts in North East Asia, as
- 30 inferred from Sr, Nd and Pb isotopic ratios (Biscaye et al., 1997). We therefore carried out a compilation of present day Na/Mg mass ratios of dust from North East Asian deserts, assuming that such an Asian dust source remained the dominant one over the last 120 kyr. For such analysis, the reader is referred to the Appendix (??), that considers n=3.8, RMS(root mean square)6 studies of dust composition (Na, Mg and Ca) from the Gobi and desert regions of Mongolian and northern China deserts. This

analysis (Appendix ??) shows that, on average, dust from these regions exhibits the following ratios:

$$(Na/Mg)_t = 1.23 \tag{6}$$

(7)

 $(Na/Ca)_t = 0.38$

5

It is worth noting that these values differ substantially from global average values (Rudnick and Gao, 2003), especially for Na/Ca.

By using the sodium-to-magnesium ratios in asian dust $(Na/Mg)_t = 1.4$, MIS1) and during the coldest glacialphases, with 1.23 (Eq. 6) and in sea water $(Mg/Na)_m = 0.12$ (Millero et al., 2008) (the subscript 'm' refers to 'marine'), we are able to calculate ssNa and ssMg (Eqs 8-9):

$$ssNa = \frac{Na - (Na/Mg)_{t} \cdot Mg}{1 - (Mg/Na)_{m} \cdot (Na/Mg)_{t}}$$
(8)

10
$$\operatorname{ssMg} = \frac{(\operatorname{Mg/Na})_{\mathrm{m}} \cdot Na - (\operatorname{Mg/Na})_{\mathrm{m}} \cdot (\operatorname{Na/Mg})_{\mathrm{t}} \cdot Mg}{1 - (\operatorname{Mg/Na})_{\mathrm{m}} \cdot (\operatorname{Na/Mg})_{\mathrm{t}}}$$
(9)

In the Holocene, on average ssNa \approx 0.99Na, while ssMg \approx 0.50-0.90Mg. During MIS2, ssNa \approx 0.6–0.7Na and ssMg \approx 0.20–0.30Mg, while during MIS4 ssNa \approx 0.50–0.60Na and ssMg \approx 0.10–0.20Mg. In those Holocene samples where magnesium was not measured, we assume Na = ssNa, making on average a 1% error.

- The same procedure using calcium has been used to calculate ssNa from Antarctic ice cores (Röthlisberger et al., 2002),
 by solving the equations similar to Eq. 8 and Eq. 9 with Mg replaced by Ca. In the RECAP record, such correction (with asian dust (Na/Ca)_t=0.38 and (Ca/Na)_m = 0.038 (Millero et al., 2008) unrealistically predicts ssNa ≈ 0 throughout the glacial period. Small changes in the (Na/Ca)_t value do not significantly change this result. It appears that higher-than expected calcium is deposited at Renland, and therefore a lower (Na/Ca)_t is to be used. Excess of calcium during the glacial period, has been reported in Greenlandic ice cores, in the form of calcite (CaCO₃), dolomite (CaMg(CO₃)₂) and
- 20 gypsum (CaSO₄ · 2H₂O) (Mayewski et al., 1994; Maggi, 1997; De Angelis et al., 1997). In their investigation of the GRIP core Legrand and Mayewski 1997 find larger increase in sufate (SO_4^{2-}) in Greenland compared to Antarctica during glacial times. Such large enhancements of the sulfate level are well correlated with calcium increases, but not with MSA, suggesting that sulfate levels are related to nonbiogenic sulfur sources (gypsum emissions from deserts, for instance) (Legrand and Mayewski, 1997). Different calcium sources were likely active during the glacial as compared to warmer periods, but it is unclear whether these
- 25 were new active continental sources or continental shelfs which became exposed due to a lowered sea level. Similarly to what has been observed in other Greenland ice cores, extra calcium is also found in the RECAP core during the glacialminima at ~, as illustrated from the scatter plot with magnesium (Fig. 3). From the calcium-magnesium relation, we estimate that during the coldest and most arid glacial times (Ca/Mg)_t = 23kyr (±1 (blue fit in Fig. 3), while in other periods we assume that the composition remained the same as the modern Asian dust composition (Appendix A):

$$30 \quad \left(\frac{Ca}{Mg}\right)_{t,modern} = 3.7 \tag{10}$$

This hypothesis is substantiated by the value found for the lower envelope for the Holocene, interstadial late MIS5 samples (green points in Fig. 3) as well as for composition of the bottom 22 meters of the core (562–584 m, red points in Fig. 3): $(Ca/Mg)_t = 3.23 \pm 0.2(1 \text{ (red fit)})$. We note that these bottom measurements do not appear in any time series since the core chronology ends at 120 kyr (562 m), although we here suggest that the RECAP bottom 22 meters may date back to the

5 previous interglacial, the Eemian.

Sea-salt sodium concentrations (ssNa) can therefore be calculated using calcium, by replacing in Eq. 8 Mg \rightarrow Ca and by using (Ca/Na)_m = 0.038 and (Na/Ca)_t calculated by:

$$\left(\frac{Na}{Ca}\right)_{t} = \frac{(Na/Mg)_{t,modern}}{(Ca/Mg)_{t,glacialfit}} = \frac{1.23}{23} = 0.054$$
(11)

This latter value is not too far to the value (0.036) empirically found by De Angelis et al. 1997, who calculated ssNa in the
GRIP core based on a mixed chlorine-calcium method. The calculation of RECAP nssCa reveals that calcium contains almost a purely crustal signature: nssCa ≥ 0.95Ca throughout the record.

The ssNa curves calculated with magnesium and calcium (ssNa_{Mg}, ssNa_{Ca}) differ by less than 1σ), while appreciable differences between these two curves and the chlorine one (ssNa_{Cl}) are only found during MIS2) and at \sim 72 kyr (MIS4 (Fig. 4b). Since ssNa_{Mg} \simeq ssNa_{Ca}, for the following discussion we will only consider the two Br_{enr} = 2.6±0.4(series based on

- 15 the chlorine and magnesium correction: $Br_{enr,Cl}$ and $Br_{enr,Mg}$ (Fig. 4g). Standard error propagation is carried out to yield the final Br_{enr} uncertainties. This analysis along with past studies demonstrate that the sea-salt and non-sea-salt calculations of elemental concentrations depend on the chemical composition of the dust that is deposited at the ice core site. The dust composition has a spatial variability which should be considered, while the use of tabulated reference values should be discouraged.
- From the calculated RECAP Br_{enr} curves, we now investigate past sea ice conditions in the 50-85 °N North Atlantic Ocean, based on the aforementioned hypotheses on the Br_{enr} use as an indicator of first-year sea ice conditions. Because of the RECAP location, the record is sensitive to ocean processes and sea ice dynamics (Cuevas et al., 2018). We compare our sea ice record with PIP₂₅ results from three marine sediment cores drilled within the Renland source area: the Fram Strait, the Norwegian Sea and the North Icelandic shelf. The PIP₂₅ index is a semi-quantitative indicator of the local sea ice condition
- 25 at the marine core location. It is calculated by coupling the sediment concentration of IP_{25} , a biomarker produced by diatoms living in seasonal sea ice, with an open water phytoplankton biomarker (brassicasterol or dinosterol, hence P_BIP_{25} or P_DIP_{25}). Briefly, the PIP₂₅ index is a dimensionless number varying from 0 to 1), MIS4). At the Eemian termination: PIP₂₅ ≈ 1 indicates perennial sea ice cover; PIP₂₅ ≈ 0 indicates open water conditions, while intermediate PIP₂₅ values reflect seasonal sea ice (Müller et al., 2011; Belt and Müller, 2013).

30 4 Results and discussion

The two Br_{enr} time series ($Br_{enr,Cl}$ and $Br_{enr,Mg}$), based on either chlorine or magnesium for the for sea-salt sodium calculation, display values greater than 1 throughout the record, suggesting some FYSI signature throughout the last 120 kyr in the North

Atlantic (Fig. 4g). The very low values found in the Holocene (on average $Br_{enr,Hol} = 3.9$, RMS(root mean square) = 1.5) suggest minimum FYSI during the current interglacial. Although the record does not extend to the Eemian period, it shows that at the inception of the last glacial period, ~120kyr,000 years ago, Br_{enr} was slightly higher with respect to what is observed during values were higher than the Holocene (on average $Br_{enr,120kyr} = 4.95.4$, RMS = 1.1, MIS5n=38), suggesting that more

5 FYSI was present in this period with respect to at that time. Since 120 kyr ago the Br_{enr} levels increased until the Greenland Interstadial 21 (GI-21), ~85-80 kyr ago ($Br_{enr,85-80 kyr} \simeq 8$). Hereafter, they decreased throughout late MIS 5. From late-MIS 5 to MIS 4, rising Br_{enr} values are observed, but a statistically significant difference between the two Br_{enr} curves result from the two ssNa corrections, with the mean Holocenevalue. $Br_{enr,Mg}$ series showing higher values than $Br_{enr,Cl}$. The two series converge again at the beginning of MIS 3, with values around 7±1. From ~ 50-40 kyr both series start to decrease. The

10 decrease is more significant for $Br_{enr,Cl}$. Minimum Br_{enr} values are reached at the Last Glacial Maximum during MIS 2. The deglaciation reveals a Br_{enr} increase up to ca. 12,000 years ago, followed by a steady decrease towards the Holocene.

5 Results and discussion

The first Arctic glacial-interglacial investigation of Br_{enr} was performed on the NEEM ice core, located in Northwest Greenland. At NEEM, the main source regions for sea ice-related processes are believed to the Canadian archipelago, the Baffin Bay and

- 15 the Hudson Bay (Spolaor et al., 2016b; Schüpbach et al., 2018). The 120,000 year NEEM record (Spolaor et al., 2016b) showed higher Br_{enr} values during the Holocene and interstatials, compared to the glacial statials (Fig. 4f). This was interpreted with greater FYSI conditions existing during such warmer periods, while the lower Br_{enr} values during colder climate phases would indicate more extended multi-year sea ice (MYSI) coverage in those ocean regions. In accordance with their respective $\frac{SSA}{Source}$ regions, the Canadian Arctic sea ice reconstruction Br_{enr} record from the NEEM ice core (Spolaor et al., 2016b) is
- 20 markedly different from the North Atlantic sea ice record reconstructed here<u>RECAP</u> one (Fig. 4). In the NEEM core, Br_{enr} was positively correlated to δ^{18} O throughout the entire climatic record(Spolaor et al., 2016b). In contrast, at Renland, such a consistent correlation is not present, and Br_{enr} is at times lower during warmer climate periods (e.g. during the Holocene). We suggest that this difference originates from the fact that during warm periods, while the NEEM site is influenced by FYSI in the Canadian Aretic, Baffin Bay and Hudson Bay, Renlandfaces mostly the relative FYSI-to-OW influence is greater at NEEM
- 25 than at Renland, the latter being mostly dominated by OW conditions in the North Atlantic. Since a lower value indicates a reduced bromine activation. This is supported, at least for present conditions, by a model study that investigated the Arctic spatial variability of the ratio of sea ice to open ocean sodium loadings (Rhodes et al., 2018), where it was found that such ratio at NEEM is ~5 times higher than Renland.

Since the increase in atmospheric bromine is believed to reflect the strength of bromine recycling from FYSI surfaces,

30 low values of Br_{enr} can could indicate either OW or MYSI conditions. Hence, at RenlandThus, we suggest that the former occurs during the Holocene, while the latter occurred during the coldest parts of the glacial. Br_{enr} is therefore a signature, at Renland, of contrasting sea ice states: FYSI/MYSI and FYSI/OW. We also suggest that the former (FYSI/MYSI) occurs during generally colder periods while the latter (FYSI/OW) occurs during generally warmer periods. In the framework of this dual

regime behavior, if the ice core time resolution is high enough, an increase of Br_{enr} values, followed by a maximum and a negative trend should be a 'tipping point' is to be expected whenever there is a shift from one regime to the other signature of an ocean facing gradual changes from open ocean to multi-year sea ice conditions, or viceversa, in case of monotonically warming or cooling ocean temperatures (Fig. 5). One such shift occurs One such regime shift is observed in the Br_{enr} record during

- 5 the deglaciation (Fig. 4g). At the Last Glacial Maximum (LGM, ~23 kyr), the low enrichment value (values ($Br_{enr,Mg,LGM} = 3.3\pm0.3$; $Br_{enr,Cl,LGM} = 3.2\pm0.2$) suggests suggest reduced FYSI recycling and therefore increased MYSI (increased extensive MYSI conditions (blue arrow in Fig. 5; OW conditions are considered very unlikelyin this cold climatic phase). Moving unlikely). Transitioning from the Last Glacial Maximum into the Holocene, Br_{enr} increases until ~12 kyr ($Br_{enr,Mg,12kyr} = 8.7\pm0.9$; $Br_{enr,Cl,12kyr} = 7.5\pm0.5$), indicating maximum FYSI at this time (Fig. 4 and Fig. 6). Hereafter, a decrease is observed,
- as Br_{enr} continues to drop during the Early Holocene (Fig. 4and the proxy operates in the FYSI/OW regime (red arrow in Fig. 5).

4.1 The last deglaciation and the dual Brenr regimes

We now consider in further detail the last deglaciation, when a number of ocean temperature, salinityand circulation modifications eoincide with changes in Nordic seas and North Atlantic sea ice as reconstructed here., circulation and sea ice changes are

- 15 <u>observed in the Nordic seas (Fig. 6). Marine-derived local sea ice</u> records from both the Svalbard margin (eastern Fram Strait, from Müller and Stein 2014 and the Norwegian Sea , from Hoff et al. 2016) indicate (Fig. 6e,f) that near-perennial sea ice (PIP₂₅ \approx 0.5-1) was present during MIS 2 until ~17 kyr (17.6 kyr recorded in the Svalbard margin), the onset of a major breakup of extensive sea ice cover, during Heinrich Event 1 (18 to 15 kyr). Synchronous to within a few centuries, several modifications relevant to the North Atlantic ocean are observed (Fig. 6), including sea water surface freshening and warming in
- the polar and subpolar North Atlantic (a record from the 67 °N , Dokken and Jansen 1999 , Dokken and Jansen 1999 record is shown as an example in Fig. 6c) and a near total cessation of the Atlantic Meridional Overturning Circulation (McManus et al., 2004).
 (AMOC, Fig. 6d). Generally low to intermediate PIP₂₅ values (PIP₂₅ ≈ 0-0.5) are reported at the Fram Strait from this point until in the Svalbard Margin and in the Norwegian Sea in the ~12 kyr 17-12 kyr period (Fig. 6e,f,g), with a slight increasing trend throughout the Bølling-Allerød (BA) and a broad maximum reached during the Younger Dryas (YD), suggesting that
- 25 seasonal sea ice conditions were present in the ~17-12 kyr period. Marine dominating this period. Other studies from marine records in the eastern Nordic Seas also indicate Nordic Seas records also suggest milder sea ice conditions during the BA and increased sea ice during the YD (Belt et al., 2015; Cabedo-Sanz et al., 2016). In contrast, a record from the northern Icelandic Shelf (Xiao et al. 2017, Fig. 6e) shows that here the sea ice conditions remained near-perennial from 14.7 to 11.7 kyr (PIP₂₅ ≈ 0.5-1). Xiao et al., The authors (Xiao et al., 2017) suggest that this pattern of more severe sea ice conditions in the north of
- 30 Iceland is, at least during the BA and the YD, linked to the flow of warmer waters from the North Atlantic Current, influencing sea ice melting in the eastern Nordic Seas, whereas the Icelandic shelf is influenced by colder polar waters from the East Greenland Current and the East Icelandic Current.

The RECAP ice core was resampled at sub-centennial resolution to better constrain the timing of sea ice changes through the deglaciation in the 50-85 °N North Atlantic sampling region. These data (Fig. 6, stars) b, squares). The Br_{enr,Mg} serie (Br_{enr,Cl}

would lead to the same results) would indicate that FYSI (i.e.) started to increase in the North Atlantic, concurrent to a reduction of MYSI, at ~17.5 kyr, synchronous with local PIP_{25} decrease in the Fram Strait Svalbard margin and eastern Nordic Seas and in response to sea surface temperature warming in the North Atlantic. This suggests finding would also suggest that North Atlantic sea ice changes occurred in concert with temperature and circulation changes of the underlying surface

- 5 waters. We note that this time period also coincides with the initiation of deglacial changes in mean ocean temperature, Antarctic temperatures and atmospheric CO₂ concentrations toward interglacial values (Bereiter et al., 2018). The synchronicity between the warmings in the Nordie Seas and in Antarctica, already reported at the onset of Dansgaard-Oeschger events (Rasmussen et al., 2016), can be thus extended to the sea ice in the North Atlantic. North Atlantic FYSI increases continued to increase throughout the BA (except for one point at 12.7-12.4 kyr at the onset of the YD) until a maximum at 12.4-11.8
- 10 kyr during the YD, when a clear Br_{enr,Mg} maximum is observed (Fig. 6b). From the comparison between the marine and ice core results, we infer that, during the 17-12 kyr period, the 50-85 °N-integrated North Atlantic sea ice changed from MYSI to FYSI. Local sea ice was also melting at ~17 kyr, in the eastern Nordic Seas, likely influenced by the North Atlantic Current, while, at least from 14.7 to 11.7 kyr, sea ice was still near-perennial over the at the North Icelandic shelf, as influenced by polar waters carried on possibly due to the influence of cold waters carried by the East Greenland Current.
- Following its maximum value at 12.4-11.8 kyr, Br enr Br_{enr} (i.e. FYSI) started to decrease (Fig. 6b). We suggest that from this point-in-time, the Br_{enr} indicator now shifts to the FYSI/OW regime (Fig. 5), and therefore lower values indicate more extensive OW the North Atlantic basin became largely ice free. A retreating FYSI scenario is also recorded in all three 5 marine cores (decreasing PIP₂₅ to ≈ 0-0.4 during the Early Holocene, Fig. 6e,f,g), suggesting that OW open water conditions progressively developed in the whole North Atlantic basin, sustained by increasing heat transport from the North Atlantic 20 Current and a strengthened AMOC since ~11.7 kyr (McManus et al., 2004; Ritz et al., 2013).

4.2 **Dual regime and transformation**

25

Since Br_{enr} is assumed to be an increasing function of FYSI, its decrease points would point to either OW or MYSI conditions(Fig. 5), following either the FYSI/OW or the FYSI/MYSI regimes (Fig. 5). At any point in time, only one regime is considered to be in place, and we suggest that a simple model in which a temperature threshold is could be the discriminating variable setting the regime type. Since a change of regime is observed during the deglaciation, with maximum Br_{enr} values (i.e. FYSI) at 11.8-12.4 kyr, we set the threshold to be the mean NGRIP temperature reconstructed for that period: T_{NGRIP} =-44.6±0.9 (2 σ) °C . The (the two lines in Fig. 6a). In every ice sample of the 120,000 year record the regime type (FYSI/MYSI or FYSI/OW, see Fig. 5) is can thus be determined according to such a temperature threshold. The regime type in each ice sample is represented by the color of the error bars in Fig. ??: blue for the its integrated temperature value with respect to the

30 temperature threshold: FYSI/MYSI for a lower temperature value and FYSI/OW for a higher temperature value. According to this simple model the deglaciation is characterized by the FYSI/MYSI regime and red for until the onset of the Bølling-Allerød (except few points at which the regime type depends on the chosen threshold value, Sect. 4.2), while the FYSI/OW regime - We suggest a procedure to linearize the operated from that point forward. We note that there is no similar Br_{enr} indicator with respect to the two climate regimes. The method is again based on the observation that during the deglaciation, maximum at the onset of the Bølling-Allerød as seen in the YD at the point when NGRIP temperature is crossing the same temperature as found in the YD. The NGRIP temperature alone appears therefore not to be able to fully explain the observations. The possible

- 5 explanation of higher Br_{enr} reaches its maximum value (i.e. maximum FYSI) at 11.8-12.4 kyr: = 7.4±0.5 (1) (Fig. 4 and Fig. 6). We now introduce the transformed-values during the Younger Dryas compared to the Bølling-Allerød may reside in the higher seasonal temperature variations (Buizert et al., 2014) and freshwater inputs from melting ice sheets in the former period, both promoting the formation of seasonal sea ice. Conversely, the lower Br_{enr} indicator, which compensates for the decrease from this maximum value due to increasing MYSI conditions (only-values (hence to greater MYSI in the FYSI/MYSI regime)
- 10 with an increase of the same magnitude (Eq. ??). No transformation affects the ice samples in the FYSI/OW regime (Eq. ??).

 $\begin{array}{l} \mathrm{Br}_{\mathrm{enr}} \longrightarrow \mathrm{Br}_{\mathrm{enr}} \\ T > \mathrm{T}_{\mathrm{NGRIP}}(12.4 - 11.8 \mathrm{kyr}) = -44.6 \pm 0.9 (2\sigma)^{\circ} \mathrm{C} \end{array}$

$$\begin{split} \mathbf{Br}_{\mathrm{enr}} &\longrightarrow \overline{\mathbf{Br}_{\mathrm{enr}}} + \left(\overline{\mathbf{Br}_{\mathrm{enr}}} - \mathbf{Br}_{\mathrm{enr}}\right) \\ T &< \mathbf{T}_{\mathrm{NGRIP}}(12.4 - 11.8 \mathrm{kyr}) = -44.6 \pm 0.9 (2\sigma)^{\circ} \mathrm{C} \end{split}$$

during the Older Dryas compared to the Bølling-Allerød and the Younger Dryas may be linked to the overall much lower temperatures during this period (Buizert et al., 2014), higher surface water salinity due to less freshwater inputs from melting

15 ice sheets and a generally weaker AMOC. Standard error propagation is carried out when transforming in Eq. ??.

The transformed-variable

4.2 The 120,000 year Br_{enr} record

We now apply the previously mentioned temperature-based discrimination of the two sea ice regimes to the 120,000 year
 Br_{enr,Mg} record (Fig. ??). The same analysis performed on the Br_{enr,Cl} curve can be found in the Appendix (Fig. ??, lower panels) is now to be interpreted as being linearly variable between OW and MYSI conditions, therefore providing an absolute index of sea ice extent within the Renland source area.

??). The regime type in each ice sample is represented by the color of the error bars: blue for the FYSI/MYSI regime and red for the FYSI/OW regime. In order to test the sensitivity of the regime output on the threshold value, 4 scenarios are

computed, using a $\pm 1\sigma$ and a $\pm 2\sigma$ value around the temperature threshold mean value $\div \overline{T} = -44.6$ °C (Fig. ??. Left: -2:top; +??). Except 2 :bottom. Right: -1:top; +(1) samples at 20 kyr showing a different regime type during the deglaciation (in the \overline{T} -2(1) σ :bottom). We note that for the simple 2-state model applied here, discontinuities could occur for adjacent samples integrating temperature values close to the temperature threshold. Only at a few time periods, however, is scenario- top panels in Fig. ??), the regime discrimination significantly affected by the chosen threshold value (see them discussed mentioned in the following section). MIS1 and MIS5 are characterized by the 'warm' discussion is invariant with respect to the 4 scenarios. MIS 5 is characterized by increasing Br_{enr.Mg} values in the FYSI/OW regime , while the 'cold' from 120 to 80 kyr ago

5 (Greenland Interstadial, GI-21). We interpret this trend with increasing FYSI extent in the North Atlantic. From the end of GI-21 the Brenr regime changes to FYSI/MYSI regime occurs during MIS2 and MIS4. MIS3 shows a mixture between the two regimes.

4.3 The 120,000 year sea ice record

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The two North Atlantic sea ice time series, calculated by applying the as the NGRIP temperature drops during late-MIS 5 and MIS 4. As compared to the levels reached during GI-21 (8±2transformation (i.e. allowing for the biggest discrepancies), are

shown in Fig. ??. It is worth noting that since the sample integrates 1500 years, these two records have been downscaled to 1500 years.

Some discrepancies between the two reconstructed sea ice records are found. The first one is at the 23.3-17.3 kyr (Last Glacial Maximum), with reduced sea ice predicted for the 'lower' temperature threshold (see also Fig. ??). The second one,

- 15 at 35.3-33.8 kyr (integrating Greenland Stadial GS-7, and Greenland Interstadial GI-7), results in opposite sea ice scenarios. Finally, at 72.8-71.3 kyr (integrating GI-19.2-1), lower Br_{enr,Mg} values are found during GS-21, GS-20 and GS-20), both scenarios indicate a decrease in sea ice but of slightly different magnitude. The output of the transformation is sensitive to the chosen temperature threshold, and therefore could result in anomalous sea ice states during those climate periods when the temperature was close to the threshold. If the GS-19 (5±2) and on average during MIS 4 (7±1transformations are considered
- 20). We interpret the combined effect of decreased Br_{enr,Mg} values and a FYSI/MYSI regime with an increase of MYSI extent in the North Atlantic from late-MIS 5 to MIS 4. The Br_{enr,Cl} curve shows even lower values during MIS 4, predicting more extended MYSI than the Br_{enr,Mg} record (Fig. ??). Overall, from 120 kyr ago to MIS 4, the change of Br_{enr} regime (from FYSI/OW to FYSI/MYSI) is opposite to what is observed during the deglaciation (from FYSI/MYSI to FYSI/OW): from a relatively warm climate 120 kyr ago, the cooling trend is characterized by an increasing FYSI extent until the point (GI-21)
- 25 in which the FYSI starts to be replaced by MYSI. This maximum Br_{enr,Mg} value at the time of the regime change (GI-21) is similar to the peak Br_{enr,Mg} value reached during the deglaciation: Br_{enr,Mg}(12 kyr) ≃ Br_{enr,Mg}(GI-21) ≃ 8. The same result holds if one considers the Br_{enr,Cl} serie (Fig. ??, right panels), the two predicted sea ice scenarios would still display a slight discrepancy during the LGM, and during ??). Unlike the longer lasting GI-21, the Greenland Interstatials GI-20 and GI-19.2 and GS-20, while a sea ice reduction would be predicted for both scenarios during 35.3-33.8 kyr, likely associated
- 30 with a sea ice decrease that occurred during GI-7. display very low Br_{enr} values ($\simeq 3-5$). A possible explanation for these low values could be found in a fast replacement (at least not captured by the time resolution of the Br_{enr} record) of MYSI by OW conditions. Higher time resolved measurements would be needed to test this hypothesis.

With the exception of the aforementioned discrepant time periods, both reconstructions show that, over the last 120 kyr, sea ice was greatest during MIS2 and MIS4, the two climate periods characterized by the coldest temperatures and reduced

insolation Moving from MIS 4 to MIS 3, the $Br_{enr,Mg}$ values remain within 1 σ of each other. This suggests similar FYSI extent in the two periods, although the FYSI/MYSI regime operating during MIS 4 could suggest extra MYSI extent during this period, as compared to MIS 3, the latter being generally characterized by a mixture of both Br_{enr} regimes. This hypothesis is supported by the $Br_{enr,Cl}$ series, which shows lower values during MIS 4 than MIS 3 (Fig. ??). Sea ice during MIS3 ??).

- 5 A sea ice record in the Norwegian Sea (Hoff et al., 2016) also indicates more perennial sea ice conditions during MIS 4 than during MIS 3. The time resolution does not allow a deep investigation on DO events during MIS 3, although a low Br_{enr} value in the FYSI/OW regime during the GI-12 (Br_{enr,Mg}(46.0-46.6 kyr)=5.2±0.7) suggests a shift to open water conditions during the interstadial, similarly to GI-19.2 and MIS5 was less extensive than during MIS2GI-20. Possibly, similar sea ice dynamics were in play during these DO events. Increased time resolution is however needed to better characterize the DO events.
- 10 MIS 2 is characterized by a FYSI/MIS4. In particular, sea ice at the onset of the last glacial period (~120 kyr)was slightly more extended than during the late Holocene (MIS1), characterized by the lowest sea ice levels of the entire 120 kyr record. These results agree with local sea ice conditions reported from the Norwegian Sea for the last 90 kyr (Hoff et al. 2016, Fig. ??). Dansgaard-Oeschger (DO)millennial-scale oscillations cannot be fully resolved since our procedure has been downscaled to 1500 year resolution, although the model does indicate a sea ice decrease during GI (glacial interstadial)7, 19 MYSI regime
- 15 and progressively decreasing values of both Br_{enr,Mg} and 20, while a sea ice increase is found during GS-25. The RECAP ice core was subject to considerable glaciological thinning in the section containing glacial ice, therefore the sampling resolution used here is insufficient to resolve all DO transitions. Nonetheless, sea ice variability on DO-timescales has been reported for the Canadian Arctic, and comparable resolution is offered by the forthcoming EastGRIP ice core, located in Northeast GreenlandBr_{enr,Cl} series (Fig. ?? and Fig. ??), reaching their respective minima at ~23 kyr ago, during the Last Glacial
- 20 Maximum: $Br_{enr,Mg}(LGM)=3.3\pm0.3$; $Br_{enr,Cl}(LGM)=3.2\pm0.2$. We interpret this negative trend with progressively increasing MYSI conditions in the whole North Atlantic, that reached a maximum during the LGM. Maximum PIP_{25} values are also reported in the Norwegian Sea at this time, indicating perennial sea ice conditions at this time.

The deglaciation is characterized by increasing FYSI conditions until the mid-Younger Dryas (12.4-11.8 kyr ago), followed by a return to the FYSI/OW regime and decreasing Br_{enr} values as open waters progressively replaced sea ice in the Holocene.

25 A clear sea ice decline is also reported in the Norwegian Sea following the LGM towards the current interglacial (Fig. 6 and Fig. ??).

5 Conclusions and outlook

We present a method to reconstruct past sea ice conditions 120,000 year record of bromine enrichment (Br_{enr}) from the RECAP ice core, and interpret it in terms of first-year sea ice (FYSI) variability in the 50-85 °N-integrated North Atlantic ocean for the

30 last 120,000 years from sodium and bromine measurements in the RECAP ice core. Our reconstruction shows. The record suggests that, during the last deglaciation, sea ice started its transformation to transform from multi-year sea ice to first-year sea ice ~17.6 kyr 17.5 kyr ago, probably triggered by increasing surface ocean water temperatures. Increasing first-year sea ice conditions are observed throughout the Older Dryas, the Bølling-Allerød and the Younger Dryas. During these periods,

availability of freshwater from melting ice sheets, seasonal temperature variations and AMOC strength likely played a role in driving the sea ice changes. The maximum first-year sea ice signature is found at 12.4-11.8 kyr ago, during the Younger Dryas, whereupon open ocean was the dominant condition during the Holocene (MIS+MIS_1). Although the RECAP sea ice record does not extend back to the warmest period of MIS5MIS 5, it does show that sea ice extent during the Holocene is lower than

- 5 at any time in the past 120kyr. Sea iceextent was maximum during MIS2 and MIS4. Intermediate conditions, if compared to MIS2/MIS4, were present during MIS3 and late-MIS5.,000 years. Minimum first-year sea ice, likely associated with maximum multi-year sea ice conditions existed during MIS 2 and possibly during MIS 4. Compared to MIS 2, greater first-year sea ice extent existed during MIS 3 and MIS 5. Increased time resolution is needed to fully resolve Dansgaard-Oeschger oscillations. Our sea ice reconstruction provides the first ice core-based observation of sea ice in the 50-85° N-integrated North Atlantic,
- 10 an area where oceandynamics has profound global climatic implications. This record will provide critical constrains on past North Atlantic sea iceconditions to climate models, advancing the knowledge of Arctic climate science, its feedbacks and implications for the whole climate systemHowever, the data indicate that during GI-12, GI-19.2 and GI-20, large extent of first-year sea ice could have been replaced by open ocean, while large first-year sea ice areas existed during GI-21.

These analyses and conclusions rely on a number of hypotheses and assumptions mainly concerning the validity of Brenr

15 as a proxy for first-year sea ice. Efforts are still needed in the direction of validating these assumption and investigating the limitations and processes that affect the Br_{enr} signature in present deposition and in old ice core records.

Data availability. The RECAP ice core data will be made available on NOAA paleoclimate and PANGAEA online data archives.

Author contributions. P.V. and A.S. conceived the experiment. N.M., R.E., P.V., A.S, H.A.K and C.T. collected the samples and ran the experimental analyses. N.M. analyzed and interpreted the results. N.M. wrote the manuscript with inputs from all authors.

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Figure 1. Cl/Na mass ratio time series used for the calculation Modern (2000-2016) source region of ssNaatmospheric sodium and bromine deposited at the Renland site estimated from 3-day back trajectory calculations. NGRIP On the z-axis is represented the number of Marine Boundary Layer (central GreenlandMBL) temperature reconstruction (blackcrossings per unit area, Kindler et al. 2014/derived by selecting only those trajectories which crossed the 900 hPa isosurface for at least 10 hours (see Sect. 2.3). The red line contours 75% of the distribution integral and it is considered to be the bromine and sodium source region. It covers the North Atlantic Ocean from ~50° N to 85° N in latitude. A minor contribution is expected from West Greenland waters. Note that areas outside the 75% line with counts <0.02 crossings km⁻² are not coloured. The analysis is performed on a seasonal basis. The Renland Clice cap is marked with a star; the NEEM and NGRIP core sites are marked with a circle. The diamonds indicate the marine cores discussed in the text: Svalbard margin (MSM5/Na mass ratio time series-5-712-2, Müller and Stein 2014); North Icelandic shelf (blueMD99-2272, Xiao et al. 2017) and the Norwegian Sea (JM11-FI-19PC, Hoff et al. 2016; MD99-2272, Muschitiello et al. 2019; JM99-1200, Cabedo-Sanz et al. 2013).The-



Figure 2. Map of modern Arctic sea water reference value ice age shown in representative extreme conditions: winter maximum (1.8February 1987, left) and summer minimum (September 2012, right) is indicated by the dotted line. A sodium crustal correction was applied only for ice samples. The marine cores discussed in the text are indicated with Cl/Na < 1.8colored diamonds. See Fig. ??-1 for the RECAP and NGRIP core locationsnames and references. The sea ice age data are from NSIDC/NASA (v.3) (Tschudi et al., 2016).'yr b2k' = years before AD 2000.



Figure 3. 120 kyr record of sodium Calcium and bromine-magnesium concentrations in the RECAP iee-core. Samples with high (a>100 ppb) Concentrations calcium concentrations are drawn in blue. The red samples of bromine the bottom 22 meters (Br, grey562–584 m depth) are not dated but may consist of Eemian ice. All others samples are drawn in green. The Ca/Mg ratio varies between 3 ± 1 (Asian dust) and sea-salt sodium 23±1 (ssNa, blackgypsum and/or excess of carbonate). The last 8 kyr sea water ratio (Ca/Mg)_m=0.32 cannot be identified since both elements are 100 year averages dominated by terrestrial contributions. The experimental uncertainties



Figure 4. 120 kyr time series of analyte concentrations in the RECAP ice core. The last 8 kyr are 100 year averages. (a) bromine; (b) total sodium and sea-salt sodium (ssNa) series calculated using respectively chlorine (green), magnesium (blue) and calcium (red); (c) chlorine; (d) calcium (black) and magnesium (blue); (e) NGRIP δ^{18} O from North Greenland Ice Core Project members (2004); (f) NEEM Br_{enr} from Spolaor et al. (2016b); (g) RECAP Br_{enr} series (Br_{enr,Cl}, green and Br_{enr,Mg}, black) calculated using chlorine and magnesium respectively.



Figure 5. Schematic illustration of marine and (Isuggested) are indicatedice core-based sea ice indicators as a function of different sea ice conditions. Br_{enr} (bice core) NGRIP temperature reconstruction (Kindler et al., 2014) and IP₂₅ (sediment core) increase as a function of first-year sea ice (FYSI) area. Low values can thus indicate either open water (eOW) Bromine enrichmentor multi-year sea ice (MYSI) conditions, depending on the climate state. The PIP₂₅ index is a marine-derived semi-quantitative indicator of local sea ice conditions at the marine core location. PIP₂₅ \sim 0 (ealeulated with EqPIP₂₅ \sim 1) indicates open water (perennial sea ice, i. 1-??-??e. MYSI) conditions, while intermediate PIP₂₅ values reflect seasonal sea ice (i. The error bars are propagated from e. FYSI). Br_{enr} values in the experimental bromine RECAP core are believed to indicate a FYSI/OW regime in a 'warm' climate state and sodium uncertainties FYSI/MYSI in a 'cold' climate state.

Schematic illustration of marine and ice core-based sea ice indicators as a function of different sea ice conditions. (ice core) and (sediment core) increase as a function of FYSI area. Low values can thus indicate either open water (OW) or multi-year sea ice (MYSI) conditions, depending on the climate state. The index is a marine-derived semi-quantitative indicator of local sea ice conditions at the marine core location. ~ 0 (~ 1) indicates open water (perennial sea ice, i.e. MYSI) conditions, while

5 intermediate values reflect seasonal sea ice (i.e. FYSI). The transformed-reflects an integrated sea ice extent within the RECAP core area of influence (North Atlantic, 50-85 °N). The indicated numbers (2 and 12) are associated with the range of oscillation of the transformed-variable as a result of the transformation (see also Fig. ??).



Figure 6. Climate records during the last deglaciation. (a) The black line is the NGRIP air temperature reconstruction (Kindler et al., 2014) . The light grey band corresponds to single temperature uncertainties ($\pm 1\sigma = 2.5$ °C). The two horizontal lines correspond to a $\pm 2\sigma$ deviation from the mean temperature (-44.6 °C) found during the 12.4-11.8 kyr period of maximum Brenr, Mg (dark-grey vertical band). The red T_{NGRIP} record does not extend before 10 kyr but the temperature is not expected to cross the threshold anytime in the Holocene. The black line is the 21 June daily mean insolation at 65 °N (Tzedakis et al., 2017)(Laskar et al., 2004). (b) RECAP Brenr, Mg (thick black line) and repeat sampling conducted at higher resolution (starsquares). Greater Since for the high-resolved measurements magnesium was not measured, for the calculation of the sea-salt sodium concentrations and the Brenr, Mg values indicate more extensive FYSI(Eq. 1-8) we used the magnesium values of the 4 low-resolution samples. Over the time period covered by the repeat measurements, nssNa $\simeq 10-20\%$. The average Br_{enr,Mg} value measured during the Holocene is indicated by a diamond. (c) Planktonic δ^{18} O record (expressed in permil) from sediment core MD95-2010 (66° 41.05' N; 04° 33.97' E, from Dokken and Jansen 1999). Lower δ^{18} O values indicate warmer and fresher ocean waters. (d) records from Record of ${}^{231}Pa/{}^{230}Th$ ratios within a sediment core retrieved in the Norwegian Sea deep western subtropical Atlantic (blue 33° 42' N 57° 35' W). Sedimentary ²³¹Pa/²³⁰Th are believed to reflect Atlantic meridional overturning circulation (AMOC) strength (McManus et al., 2004). (e, core JM11-FI-19PC, Hoff et al. 2016g), PIP₂₅ records from the Svalbard margin (green, core MSM5/5-712-2, Müller and Stein 2014)and from, the North Icelandic shelf (red, core MD99-2272, Xiao et al. 2017), and the Norwegian Sea (core JM11-FI-19PC from Hoff et al. 2016), core JM99-1200 from Cabedo-Sanz et al. 2013 and core MD99-2284 from Muschitiello et al. 2019). The PIP₂₅ scale varies from perennial sea ice (PIP₂₅ \approx 1) to open water (PIP₂₅ \approx 0) conditions (see Fig. 5). All time series are plotted on their original chronologies. See Fig. 22 for the PIP₂₅ core locations.



Br_{enr,Mg} regimes and transformationaccording to the NGRIP temperature threshold (see Fig. Upper panels: ?? for the equivalent analysis using Br_{enr,Cl}), a) reconstructed NGRIP temperature (grey, Kindler et al. 2014). The temperature threshold black line corresponds to value is the mean temperature (ŦT±2σ: left; T±1σ: right) during the 11.8-12.4 kyr period, when the change of Br_{enr,Mg} regime was observed (see Fig. 6). The black time series is the temperature profile downscaled to the measured Br_{enr} resolution. Middle panels: discrimination of the regimes computed according to the integrated temperature value with respect to the threshold. The FYSI/OW (FYSI/MYSI) regime is indicated with red error bars (blue error bars). The =7.4±0.5 value is indicated with a dotted line. Lower panels: transformation of according to Eq. ?? and Eq. ??. The transformed-is calculated by reflecting the measured values about the =7.4±0.5 axis, only in the FYSI/MYSI regime (blue error bars). Some discrepancies between numbers indicate the four scenarios are found at 17.3-23.3 kyr (LGM), 33.8-35.3 kyr (GS-7 and GI-7) and 71.3-72.8 kyr Greenland Interstadials (GI-19.2 and GS-20GI) :

120 kyr record of sea ice discussed in the North Atlantic. (a) NGRIP temperature reconstruction, from Kindler et al. 2014text. (b) 21 June daily mean insolation at 65 °N, from Tzedakis et al. 2017(Laskar 291., 2004). (c) 90 kyr $P_D IP_{25}$ record from the Norwegian Sea (grey, from Hoff et al. 2016) and 5-point running mean (black). (d) RECAP ice core 1500 year resolution record discrimination of transformed-the Br_{enr,Mg} regimes computed according to the T+2(black) and T-2(green) integrated temperature threshold (see Section ??, Fig. ??). The

660 Appendix A: The RECAP ice core samples Modern Na/Mg and Na/Ca ratios in dust from Northern Asian deserts

The RECAP (REnland ice CAP) ice core was retrieved from the Renland ice cap (71° 18' 18" N; 26° 43' 24" W; 2315 m a.s.l.) from May 13th to June 12th, 2015. The ice cap is located on the Renland peninsula and is independent of the main Greenland ice sheet, with fjords to the north and south. A 98 diameter ice core was recovered to 584 (bedrock). The drilling occurred in a dry borehole to 130 m depth and estisol 140 drilling fluid was used for the remaining depth. The record covers

- 665 the last 120 kyr. The age-depth model is based on annual layer counting for the interval 0-4 kyr, using the StratiCounter algorithm (Winstrup et al., 2012), tied to fit 25 GICC05-based age markers (Vinther et al., 2006). From 4 to 11.7 kyr, it is created using a shape-preserving piecewise cubic interpolation between 16 GICC05-based markers while from 11.7 to 120.8 kyr the chronology is created by linear interpolation between 74 age markers, the last 3 being estimated from gas-correlation between NGRIP and RECAP and air measurements (Simonsen et al., in preparation). The ice core samples (aim of this
- 670 section is to obtain a reference value for the present day mass ratios of Na/Mg and Na/Ca in Asian dust, since the Gobi and desert regions of Mongolian and northern China (hereafter referred to as Asian dust) is believed to be the main source region of dust found in Greenlandic ice cores during the glacial period, as inferred from Sr. Nd and Pb isotope composition (Biscaye et al., 1997).

We consider n=1205) were collected and automatically decontaminated from a continuous ice core melting system as part of

- the RECAP Continuous Flow Analysis (CFA) campaign conducted at the University of Copenhagen in Autumn 2015. The ice core was melted at a speed of approx. 3 on a gold coated plate copper melter head (Bigler et al., 2011; Kaufmann et al., 2008). Meltwater was collected continuously from the melter head into pre-cleaned polyethylene vials (cleaned with ultrapure water, > 18.2) at two different depth resolutions. From the ice cap surface to a depth of 535.15, samples incorporated ice meltwater spanning depths of 55. From a depth of 535.15 to the ice cap bedrock the samples integrated 18.3. The time resolution is
- 680 annual to multicentennial in the Holocene and from centennial to millennial in the glacial section. After collection, the samples were immediately refrozen at -30 °and kept in the dark to reduce bromine photolysis reactions. The samples were shipped to Cà Foscari University of Venice (Italy, n6 published studies in which the chemical composition of dust sampled during "Desert Events" (DS) was analyzed in China. Often these studies are carried out in the framework of pollution-related research. We also include composition data from dust sampled in-situ in desert areas. For details on the single studies, we refer the reader
- to the references in Table A1. We note that our compilation only considers dust samples originated (or sampled directly) from deserts. Data from trajectories that traveled over other areas (e.g. marine or others) are not considered.
 By considering all the above-mentioned studies, the mean Na/Mg mass ratio is 1.23 (median = 770) and Curtin University of Technology (Perth, Australia, n1.26). The mean Na/Ca mass ratio is 0.38 (median = 435) for determination of bromine () and sodium () by Inductively Coupled Plasma Mass Spectroscopy (CRC-ICP-MS and ICP- SFMS, respectively, see Sect. 2.2).

690 A1 Analytical determination of bromine and sodium

A total of 1205 meltwater samples, collected during the Continuous Flow Analysis campaign, were measured in two laboratories. 770 samples were measured in Italy. The depth and age ranges associated with the Italian samples are: surface-150 m (2015

AD-328 vr b2k); 165-219 m (383-636 vr BP); 234-413 m (727-2857 vr b2k) ; 441-495 m (3522-5749 vr b2k).435 samples were measured in Australia. The depth and age ranges associated with the Australian samples are: 150-165 m (328-383 vr b2k); 219-234 m (636-727 vr b2k); 413-441 m (2857-3522 vr b2k); 495-562 m (5749-120788 vr b2k) 0.33).

University Ca' Foscari of Venice, ItalyBromine and sodium (and) were determined by Collision Reaction Cell-Inductively Coupled Plasma-Mass Spectrometry (CRC-ICP-MS, Agilent 7500ex, Agilent, California, USA). The introduction system consisted of a ASX-520 autosampler (CETAC Technologies, Omaha, USA) and Scott spray chamber fitted with a MicroFlow PFA nebulizer. The sample flow was kept at 100. All reagents and standard solutions were prepared with ultrapure water

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(UPW, 18.2). Nitric acid (5% vThe mean Ca/v, trace metal grade, Romil, Cambridge, UK) and UPW washes (2 minutes each respectively)were used for background recovery after every sample analysis. The experimental routine (standards and ealibrations), as well as the overall instrument performance (detection limits and reproducibility) are the same as in Spolaor et al. 2016bMg mass ratio is $3.66 \pmod{3.55}$.

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The analyses were performed by Sector Field Inductively Coupled Plasma Sector Field Mass Spectroscopy in reverse Nier-Johnson geometry (ICP-SFMS, Element XR;

Appendix B: Discrimination of the two Brenr, Cl regimes according to the NGRIP temperature threshold

Sea salt aerosol (SSA) source area for the Renland ice core. Spatial distribution of Marine Boundary Layer (MBL) crossings per unit area, derived by applying a 900 constrain for at least 10 hours (see text). The Renland ice cap is marked with a star; the NEEM and NGRIP core sites are marked with a circle. The nablas indicate the marine cores discussed in the text (green:

- 710 Svalbard margin, Müller and Stein 2014; azure: Norwegian Sea, Hoff et al. 2016; red: North Icelandic shelf, Xiao et al. 2017). The red lines indicate the 75%, 50% and 25% boundaries of the integrated distribution. The 75% contour line is considered to be the SSA source region for the Renland ice core. The source area covers the North Atlantic from ~50° N to 85° N in latitude. A small SSA contribution is expected from West Greenland waters. Note that areas outside the 75% line with counts <0.05 crossings are not coloured. To infer the sea salt aerosol (SSA) source area for the Renland ice core, daily back trajectories were
- 715 ealculated from 2000 AD to 2016 AD with Hysplit4 (Stein et al., 2015; Draxler et al., 1999; Draxler and Hess, 1998, 1997), using publicly available Reanalysis meteorological data, with a 2.5° resolution in both latitude and longitude (Kalnay et al., 1996). The back trajectories were started daily on an hourly basis at 500 above the Renland elevation (71.305 °N, 26.723 °W, 2315 m a.s.l.) for the 17 year time span. The trajectory time was set to be 72 hours, representing the average atmospheric lifetime of SSA (Lewis and Schwartz, 2004). To access the potential marine sources of SSA, a selection on the trajectories was implemented.
- 720 by keeping only those that crossed the marine boundary layer (MBL), defined here as the 900 isosurface (corresponding to approximately 1000 a.s.l.), for at least 10 hours. This pressure cutoff was chosen according to Lewis and Schwartz 2004 and references within. The spatial map of the trajectories projected onto the MBL field indicates that 75% of the signal originates from the area spanning from the North Atlantic to the Arctic Ocean (50-85 °N, Fig. ??). A minor contribution is expected from aerosols originated from coastal West Greenland. The consistency of sea ice reconstructions from the Renland ice core and
- 725 the Nordie Sea sediment cores (see main text) suggests that the Renland source area extends to these regions and this is valid

throughout the last 90 kyr. For the overall interpretation of the Renland record the source area is therefore assumed to be the



75% contour region.

Figure B1. Discrimination of the two $Br_{enr,Cl}$ regimes according to the NGRIP temperature threshold. This figure is equivalent to with $Br_{enr,Mg}$ replaced by $Br_{enr,Cl}$.