Author comment on

"Bromine, iodine and sodium in surface snow along the 2013 Talos Dome – GV7 traverse (Northern Victoria Land, East Antarctica)" by Maffezzoli et al.

Anonymous Referee #2

We would like to thank Referee 2 for the extensive review and the time taken to read the paper. The introduction has been extensively revised, especially for the analytical and dating parts. We acknowledge that an error was found in calculating the sea ice area, this has now been corrected.

Major concerns

Analytical information is lacking. The authors present very few analytical details, instead refering readers to the Spolaor et al, 2013a paper. However, some information will be specific to the Maffezzoli paper and should be included, and some additional information would be useful.
We agree with the reviewer comment and have added the requested information. The citation of the method was wrong (2013a). The correct reference was the 2014 paper. The analytical part has been corrected and expanded including the following details (see below).
For example, what was the residual standard deviation for Br, I, and Na for the current work?
"The reproducibility of the measurements was carried out by repeated measurements of standard samples within the calibration range. The residual standard deviations were respectively 5 % (bromine), 3 % (sodium) and 2 % (iodine). "
How were the standards prepared – gravimetric or volumetric methods?
The standards were prepared by gravimetric method.
Standard concentrations ranged between 10 and 4000 ppt, but for for which species? Spoloar 2013 refers to iodine and bromine being calibrated in this way – but what standards were used for sodium? Presumably halogen standards were separate from sodium standards – did this introduce any uncertainties into the analyses?
We thank to referee for the comment.
The standard solutions were prepared from separate stock 1000 ppm standard solutions of the three analytes. The primary solution was then diluted for the calibration purposes into 6 bromine and iodine standards (0.01, 0.05, 0.1, 0.5, 1 and 4 ppb) and 6 sodium standards (0.5, 1, 5, 10, 50 and 100 ppb). Previous tests have suggested that mixing of standards didn't change the response signal compared to separate standard solution.
I am not so familiar with this technique – does it analyse Na, I, and Br in a single run? If not, what uncertainties does this introduce in terms of instrument drift etc? Does the method use columns to separate out the elements of interest? If so, what columns were used?
Yes, the analytes are detected almost simultaneously (instrument detects each analyte 5 times and averages the response). No column separation was used: the sample was introduced with a cyclonic Peltier-cooled spray chamber (ESI, Omaha, USA) directly injected into the ICP-SFMS at a flow rate of 0.4 mL min^{-1}.
Was any reference material used to really pin down the analytical technique, given the extremely low concentrations being measured?
There is no halogen certified reference material that we know of.
Exposure of samples to direct light was minimised, but how long were they actually exposed for?
"The samples were exposed to laboratory light for a maximum time of one hour."
How long were the samples melted for before they were analysed?
"The samples were melted maximum one hour before measurements."
Were there any repeat analyses of samples carried out?
Every data point is the average of 5 almost simultaneous instrumental detections. Precautions were taken to assure that each of the 5 detections were consistent (less than 2% variations).
How much liquid water was needed for each analysis?
"The sample flow was 0.4 mL min^{-1} for a total sample volume of approximately 5.0 mL".

Presumably the samples were re-frozen before being shipped to Copenhagen for isotopic analysis. Is this likely to have introduced any problems/uncertainties into the analyses?

Sub sample aliquots for isotopic measurements were taken during the melting phases. The aliquots were immediately refrozen and shipped frozen to Copenhagen. We don't think that this procedure introduced any major uncertainty since the time span in which the samples were melted was on the order of 10 minutes and precautions were taken to minimize evaporation (septum-sealed glass vials were used to these aliquots).

Finally, the manuscript states that during sampling, every tool was repeatedly cleaned with ultrapure water – how many times is "repeatedly". What effort was made to ensure that the tools were clean?

Rephrased as: "Every tool was cleaned each time a piece of sample was decontaminated into three serial baths of ultrapure water, which was changed every 10 washes."

The analytical section has been reviewed:

2.2 Analytical measurements

Total sodium (Na), bromine (Br) and iodine (I) concentrations were determined by Inductively Coupled Plasma - Sector Field Mass Spectrometry (ICP-SFMS Element2, ThermoFischer, Bremen, Germany) at Cà Foscari University of Venice, following the methodology described in Spolaor et al., 2014.

The samples were melted one hour before measurements. During this time exposure from direct light was reduced by covering them with aluminum foils, minimizing bromine and iodine photolysis reactions.

The introduction system consisted of a cyclonic Peltier-cooled spray chamber (ESI, Omaha, USA). The operational flow rate was kept at 0.4 mL min^{-1}, for an overall sample volume of 5.0 mL. Each sample determination consisted of 5 instrumental detections (less than 2% variations between them). The 5 values were then averaged to provide the final quantification.

Each analytical run (10 samples) ended with a HNO_3 (5%) and UPW cleaning session of 3 min to ensure a stable background level throughout the analysis.

The external standards that were used to calibrate the analytes were prepared by gravimetric method by diluting separate stock 1000 ppm IC solution (TraceCERT® purity grade, Sigma-Aldrich, MO, USA) of the three analytes into a primary solution, which was further diluted for into 6 bromine and iodine standards (0.01, 0.05, 0.1, 0.5, 1 and 4 ppb) and 6 sodium standards (0.5, 1, 5, 10, 50 and 100 ppb).

The calibration regression lines showed correlation coefficients $R^2>0.99$ (N=6, p=0.05). The detection limits, calculated as three times the standard deviation of the blanks, were 50 and 5 ppt for bromine and iodine respectively and 0.8 ppb for sodium. The reproducibility of the measurements was carried out by repeated measurements of standard samples within the calibration range. The residual standard deviations (RSD) were respectively 5 % (bromine), 3 % (sodium) and 2 % (iodine).

Procedural UPW blanks were analyzed periodically to test the cleanliness of the instrument lines.

Stable isotopes of water (^{18}O and D) measurements were conducted on sub sample aliquots, which were immediately refrozen and shipped to the Center for Ice and Climate (Copenhagen, Denmark). Analyses were carried out using a Cavity Ring-Down Spectrometer (Picarro, Santa Clara, USA) using the method described by Gkinis et al. (2010). Septum-sealed glass vials were used for these measurements to prevent any sample evaporation during the experimental phases.

Dating: The conclusions drawn in the paper rely heavily on correct dating of the cores, i.e. correctly allocating samples to specific seasons. This is done using stable water isotopes, and the manuscript states (line 140) that "isotope ratio minima (representing mid-winter) can be easily identified". But the authors must give the criteria used for such identification, if other than just by eye. As the authors state, there is clearly a question over assignment of the 2013 mid-winter – how did the authors select the one chosen?
I also wonder what happened to the top layer of core 6..? Was it damaged during sampling? The accumulation rate of core 10 is less than core 9, so one would expect the snow at 2m depth to be older in core 10 than in core 9; however, this is not the case as presented in the paper.
It makes me wonder whether mid-winter assignments in core 10 are out by a year? Certainly, more information on the criteria used to allocate winter minima must be given for cases where it is not completely clear.
If the Referee refers to assignment of year 2013 in core 10, it is clear that the surface of core 10 is loaded with a layer of snow with mixed isotopic value. The effect of this layer of disturbed snow is to push the ordered stratigraphy downwards, resulting in 'younger' snow at the bottom compared to core 9. Therefore, winter 2013 in core 10 is suggested, but summer values (beginning of 2013) are clearly visible (and that is a tie point in Fig. 2).
Year 2013 in core 10 has been removed from the calculation of % annual bromine enrichment (now Fig. 7b) and in the calculation of the fluxes (now Fig. 9) because of the these reasons.
The top part of core 6 has probably been lost due to wind erosion. Though, we concluded from the isotopic values that the surface is the start of a 'cold' period.
The methodology to assign winters has been written more clearly. Lines 139-154 have been rephrased:

"The cores were dated based on the seasonal variations identified in the stable water isotopes (both $\delta^{18}O$ and δD). Midwinters were associated to the relative minima of the isotopic curves (Fig. 2). In case a winter isotopic plateau was found, the center of the plateau was associated to midwinter depth (2011 in core GV7; 2012 and 2011 in core 8; 2010 in core 6). Almost all the cores cover the period between 2010 and late 2013, providing four years of snow deposition. The only exception is represented by core 6, whose upper layer is missing.
The annual deposition signal looks less clear in the two cores that were drilled at the sites with the highest elevation and the closest to the Ross Sea, cores TD and 10, and especially for 2013 in core 10. The two sites are probably the most affected by surface remobilization and isotopic diffusion due to low accumulation. Indeed, non-uniformities in the shallow snow layers such as sastrugi, dunes, wind crusts and other features have been identified as an important aspect of the surface morphology around the Talos Dome area (Frezzotti et al., 2004; 2007).
The annual accumulation rates were calculated by selecting the depth intervals included within consecutive maximum or minimum $\delta^{18}O$ values (Table 2). Each snow layer within this interval (i.e. sampling resolution, 5 cm) was multiplied by the density of the snow at that depth, the density curves having the same resolution. The contributions were summed over the annual thickness. Table 2 also includes accumulation rates in Victoria Land reported from previous studies. The GV5 site is located between sites 10 and 9 (Fig. 1)."

Please explain fully how accumulation rates were calculated, rather than just saying (line 153) that measured density profiles were accounted for – explain the method.
The methodology to calculate the accumulation rate has been written more clearly, see comment above.

Was only 1 core taken in each site? How do you account for variability between cores drilled at the same site? If this is not considered important, then please say so and explain why.
The main goal of the traverse was to compare halogen seasonality of deposition in a wide area, since no transect before this one has ever been performed for this purpose. It would have been surely interesting to investigate the spatial variability in each location but this was not possible due to logistic constrains. The traverse between Mario Zucchelli and Concordia stations (planned for 2017/18) will provide a more complete picture especially to elucideate gas-phase vs aerosol fractionation and deposition.

The authors must provide justification on why they chose to use the stake farm data for Talos Dome over their own method. It is not enough to reject your method, which is used on all other sample sites, just because it does not agree with stake farm data at TD. This selection raises concerns over the validity of the method used for all the other sites.

Talos Dome, is known to be a difficult location due isotopic diffusion and non-uniformities (Frezzotti et al., 2004; 2007). Therefore, and because the value in Talos Dome is the only one that is inconsistent with literature values, we decided to disregard the core from the % annual bromine enrichment (now Fig. 7b) and the flux calculations (now Fig. 9) and show its chemistry variability in the supplementary on a depth scale. The other values are consistent with published data, therefore we used them.

Below are more minor comments:

- Line 16: halogen chemistry does not only occur through release of sea salt rich aerosols; various saline condensed phases have been suggested;

Rephrased: " .. sea salt aerosols and other saline condensed phases ..".

- line 18: the statement "halogen species in polar snow samples are shown to be closely related to sea ice extent" is too strong – there is clearly a link to sea ice, but this is far from quantified, so how close it is related is not yet known.

'closely' has been removed.

-line 25: the transect revealed homogeneous fluxes" – what type of fluxes? Air-to-snow..? Snow-to-air..?

Modified to 'air-to-snow'.

-line 27: "flux measurements are consistent with the uniform values of BrO and IO"... uniform in time or space..?

Uniform in space.

Rephrased: " .. BrO and IO concentrations detected from satellites over the traverse area."

Line 36 – there is no tropospheric ozone layer; remove the word "layer"

The word ''layer'' has been removed.

Line 36 – there are many other papers that should be referenced here other than Barrie et al. There are also some excellent reviews, e.g. Simpson et al. 2007, Abbatt et al. 2012, that should also be included here.

The references have been added.

Line 38 – explain why young sea ice surfaces have high salinity, and how they are thus a source of bromine compounds (presumably you mean to the atmosphere). Also, what are "bromine halides"..??

This sentence has been reviewed:

"Although the ocean is the main reservoir of sea salts, various condensed phases of high salinity are found on young sea ice surfaces. During seawater freezing, brine is separated from the frozen water matrix and expulsion processes lead to both upward and downward movement, as temperature decreases (Abbatt et al., 2012). Therefore, high salinity brine, frost flowers and salty blowing snow make newly formed sea ice surfaces a highly efficient substrate for inorganic halogen halides and for their activation and release in the atmosphere (Saiz-Lopez et al., 2012b). "

Line 43 – include the reactions and improve the wording here

The sentence has been reviewed:

"Reactive halogen species are involved in cyclic reactions between halogen radicals, their oxides and ozone. Reactions R1-3 show the main reactions for bromine. Atomic bromine radicals result from photolysis of molecular bromine, leading to formation of bromine oxide, BrO, through the uptake of ozone:

$$Br_2 \rightarrow 2Br \qquad (1)$$

$$Br + O_3 \rightarrow BrO + O_2 \quad (2)$$

$$BrO + BrO \rightarrow Br + Br + O_2 \quad (3)$$

Self reaction of BrO may form 2 bromine atoms (85%) or a Br_2 molecule (15%) which is readily photolyzed. The mechanism has a catalytic behavior that destroys ozone."

Line 45- MAX-DOAS generally refers to a ground-based instrument technique; SCIAMACHY is a satellite-borne uv-vis-nir spectrometer that quantifies BrO and IO columns using the DOAS technique. Please re-word. Also state which satellite instrument you refer to.
The sentence has been reviewed:
"High concentrations of tropospheric vertical columns of BrO and IO have been confirmed by SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmo- spheric CartograpHY) satellite observations over Antarctic sea ice (Schönhardt et al., 2012)."

Line 47 – "from halogen-rich condensed phases (e.g. sea salt aerosol)" – there are likely to be others as well.
Rephrased as: "Bromine can then be recycled and re-emitted from halogen-rich condensed phases (such as sea salt aerosol or other saline solutions)"

Line 49: Such reactions ... lead to enhancement of bromine in the deposition in the surface snowpack - give a reference to support this statement.
We agree with the Referee, as this is a central point. The original hypothesis of bromine enrichment has been recently supported by a chemistry model in the 2016b paper. The reference is added and the sentence made clearer. The topic is further addressed later on in this paper, answering to Referee1 comment in Line 189.
"Such reactions, known as bromine explosions, lead to enhanced bromine in the atmosphere. A recent 1D chemistry model simulation predicted an increase of bromine deposition on surface snowpack after 24/48 hours of recycling over first year sea ice (Spolaor et al., 2016b)."

Line 51 – is the Spolaor reference 2016 a or b?
It's reference 2016 a and b, updated.

Line 52 – the Vogt paper is a modelling study, so does not include primary information on iodine sources
The Vogt reference has been replaced by the Saiz-Lopez et al., 2012a review.

Line 66 – DMS is produced from DMSP, not directly from phytoplankton
Rephrased as: "...dimethylsulfide (DMS), which is produced by phytoplankton synthesis of DMSP".

Line 70- I believe Mulvaney and Pasteur were the first to report post-depositional movement of MSA in ice cores.
The references have been added and "mostly in Greenland" has been removed.

Line 78 – "Back trajectory calculations show that favourable events of air mass advection from the sea ice surface to TD are rare but likely to occur" – what calculations are these? Which model? Did the authors run them, or are they in a published paper? If the latter, then give references. If the former, give more information. An example plot would be useful with some sort of analysis of air mass origin.
This is a central point in the choice of sea ice sector and in the interpretation of the fluxes, so we appreciate the importance of communicating this clearly.
An extensive study in this area on the provenance of air masses is presented in the paper by Scarchilli et al. (2010) which is mentioned in section 3.3. This paper shows that (analyzing daily back trajectories from 1980 to 2001) air masses arriving in Talos Dome are clustered into two main sources: the Ross sea and the Indian

ocean sector (Fig. 4 in the Scarchilli paper). Moving from Talos Dome to the Indian ocean (where cores GV7, 8, 7 and 6 were drilled) we expect this sector to be more and more important. Hence our choice of the sea ice sector.
Another paper, Sala et al. (2008) has studied provenance of air masses from back trajectory calculations from 1990 to 2002, pointing out sea ice influence at Talos Dome.
Both references are now mentioned here.

Actually the point being made in paragraph of line 72 to line 82 is not clear.
The paragraph has been rewritten to state more clearly that this area is influenced by marine and sea ice air advection. This has been demonstrated by measurements of chemical species in the snow (traverse studies and longer cores) and back trajectory calculations. The 'dust' part and its refences have been removed.

"Victoria Land has been intensively studied for the past two decades. The Taylor Dome (Grootes et al., 2001) and Talos Dome (Stenni et al., 2011) deep ice cores respectively provide 150 kyr and 300 kyr climatic records directly influenced by marine airmasses. Sala et al. (2008) pointed out the presence of marine compounds (ikaite) at Talos Dome, typically formed at the early stages of sea ice formation. Their back trajectory calculations also showed that favourable events for air mass advection from the sea ice surface to Talos Dome are rare but likely to occur. An extensive study by Scarchilli et al. (2011) on provenance of air masses has shown that Talos Dome receives 50% of its total precipitation from the west (Indian Ocean), 30% from the east (Ross Sea and Pacific Ocean) and approximately 15% from the interior. Within the framework of the ITASE program (International Trans-Antarctic Scientific Expedition, Mayewski et al., 2005), several traverses were carried out to evaluate the spatial patterns of isotopic values and chemical species linked to marine influence (Magand et al., 2004; Proposito et al., 2002; Becagli et al., 2004, 2005; Benassai et al., 2005)."

Line 90 and 93 – "Indian **ocean** sector"
Corrected.

Line 92 – Friess et al did not report Br in their snow pit.
Corrected.

Line 104 – it would help the reader to give the distance between the cores.
Table 1 has been updated with un extra column showing core-to-core distances in km.

Line 184 – The description of bromide release needs improving. e.g."bromide … is recycled over halogen-rich sea ice surfaces" – these are the sources; recycling is different. Please clarify what you mean.
Rephrased: "Despite bromine being a sea salt marker like sodium, it is activated when gas phase HOBr oxidizes bromide over halogen rich sea ice surfaces (i.e. first year sea ice, FYSI) and suspended sea salt aerosol, and exponentially released as Br_2".

Line 187 – give the phases of the reactants and products in reaction 1
The phases have been written. The following reaction, $Br(g) + HO_2(aq) \rightarrow HBr(g) + O_2$(g), has also been added in response to Referee1 to motivate the possibility of enrichment in the following sentence.

Line 191 – add reference to Simpson et al. GRL (32), 2005
Added.

Lines 198 to 200 should be used as the figure captions in Figs 4 to 7 – what is currently used is inadequate.
Captions have been rewritten accordingly.

Line 205 typo: "maximum values in during late"
Corrected into 'maximum values in late..'.

Line 209 and Figure 9a – how was sea ice area calculated; did it rely on the threshold of >15% sea ice in a pixel? More details are needed. But further, and importantly, there is no justification given of why the 130 to 190° sector is chosen. This is why proper assessment of air mass origin and history (as detailed above) becomes important.

The sea ice area was calculated by multiplying the sea ice concentration value in each pixel by the pixel area (25X25 km^2). A mistake in the sea ice computation was found: the new figure (Fig. 7) shows the correct values (the trend doesn't show any difference). No threshold was applied to the sea ice concentration values.

As in the Referee comment at line 78, we referred to Scarchilli et al. (2011) for the choice of the sea ice sector. The sector boundaries have been rewritten as 130°E-170°W throughout the text.

The sentence has been reviewed:

"Sea ice area in the 130°E-170°W sector was calculated for the 2010-2013 period using publicly available NSIDC passive microwave sea ice concentration data (Meier et al., 2013), by multiplying the sea concentration value in each grid pixel by the area of the pixel (25 x 25 km^2) and integrating over the domain. The longitude sector was decided on the basis of Scarchilli et al. (2011), who concluded that air masses arriving in this area originate from the Ross sea and from the Indian ocean sector, by analyzing 5 day back trajectories from 1980 to 2001."

Line 215 – explain why you used the data from Syowa and why they are the right data to use (presumably because they are similar latitude, but you need to say that).

In sight of a similar comment from Reviewer 1, we calculated the solar irradiance at 71°S, 158°E with Tropospheric Ultraviolet and Visible (TUV) model. The figure has been updated with such values (no visible difference appears in the normalized SI*radiation magenta band). The caption has also been revised.

Line 216 – Figure 9b relies entirely on correctly dating the cores, and correctly attributing the months to the measurements. This goes back to my concern about dating, above. There needs to be complete assurance that this has been done correctly.

Year 2013 of core 10 and the Talos Dome core have been removed this figure, in accordance to the previous comments and findings on dating and accumulation values. Each contribution of each core to the overall deposition trend has been added on top. The blue band shows 1sigma.

Line 237 and Fig 10 – the figure shown originates in a paper by Anja Schoenhardt – the original reference needs to be provided, not the secondary one (Spoloar et al). Also, these data are not tropospheric measurements – they are vertical columns from space that have not been adjusted for any stratospheric component.

The picture has been produced for the Spolaor et al. (2014) paper; the reference therefore has been left. "Tropospheric" has been removed.

Line 244 – the polar night does not "start" in winter – please re-phrase

Rephrased as: "... in winter, when absence of sunlight inhibits photoactivation."

Section 4 – the Conclusions need to be re-visited in light of the above comments.

The conclusions have been reviewed:

"The 2013/14 Talos Dome – GV7 traverse provided an opportunity to expand the existing sodium dataset in Victoria Land and investigate important features of bromine and iodine temporal and spatial variabilities, so far only available in Antarctica at Law Dome and Neumayer station.

The accumulation rates agree with previous studies, with increasing values from the Ross Sea to the Southern Ocean. Accumulation rates calculated for Talos Dome are higher than previously reported, likely caused by isotopic diffusion and remobilization at this site. Further studies are required at this site in order to access the reproducibility of the climate signal. The locations near the Southern Ocean exhibit high variability due to the higher accumulation.

Sodium and bromine concentrations in the snow samples result in a positive bromine enrichment to seawater, confirming the sea ice influence in the area for the extra bromine deposition. While sodium does not capture clear sub-annual variations associated with sea ice, bromine enrichment shows consistent seasonal variabilities with late spring maxima. It is possible to relate such seasonality to the combined effect of sea ice growth and sunlight, which trigger photochemistry above fresh sea ice. The timing of deposition is coherent among Victoria Land, Law Dome (Indian sector) and Neumayer (Atlantic sector). Iodine shows an average value of 0.04 ppb, similar to background values observed in the Antarctic coastal locations of Law Dome and Neumayer. Unlike those locations, low iodine annual variability and no consistent seasonality of the signal are observed in the traverse samples.

The spatial variability study reveals homogeneous fluxes of Na, Br, and I over the transect length, with an increase in absolute values and variability at the sites close to the Indian Ocean, due to high accumulation and proximity to the coasts. Uniform satellite values of BrO and IO over Victoria Land are consistent with the snow measurements. A fractionation due to distance of these potential proxies is not found probably due to the combined double input of air masses from the Ross Sea and the Indian Ocean.
A transect covering larger distances and directed towards the interior of the plateau would give an insight on this feature, especially clarifying the spatial pattern of bromine enrichment with respect to differences in gas-phase and aerosol depositions. "

In particular, the "Uniform satellite values of BrO and IO over Victoria Land confirm the snow measurements" is far too strong. They might "be consistent" with the snow measurements, but they do not confirm them.
Rephrased accordingly.
Finally, line 291, the halogens are not yet "proxies" as they are not rigorously demonstrated – they are potential-proxies, but not yet proven.
"Potential" has been added.

Table 3 – include also the median, given the statement in the text about high episodes, the median then becomes important.
The table was removed as suggested by Referee #1.

Fig 1 – it would be useful to have latitude/longitudes on the lower maps or the inset maps to clarify how the zoomed-in map relates to the continent-wide ones.
The lower map has been modified as suggested by Referee #1 (and Figure 8 has been removed). It now shows the max/min sea ice concentrations and the sea ice sector which is considered. Some lat/long values in both top and bottom panels have been added for clarification.

Added references

(1) Abbatt, J. P. D., Thomas, J. L., Abrahamsson, K., Boxe, C., Granfors, A., Jones, A. E., King, M. D., Saiz-Lopez, A., Shepson, P. B., Sodeau, J., Toohey, D. W., Toubin, C., Von Glasow, R., Wren, S. N. and Yang, X.: Halogen activation via interactions with environmental ice and snow in the polar lower troposphere and other regions, Atmos. Chem. Phys., 12(14), 6237–6271, doi:10.5194/acp-12-6237-2012, 2012.
(2) Simpson, W. R., Von Glasow, R., Riedel, K., Anderson, P., Ariya, P., Bottenheim, J., Burrows, J. and Carpenter, L. J.: Halogens and their role in polar boundary-layer ozone depletion, Atmos. Chem. Phys. Atmos. Chem. Phys., 7, 4375–4418, doi:10.5194/acpd-7-4285-2007, 2007.

(3) Saiz-Lopez, A., Plane, J. M. C., Baker, A. R., Carpenter, L. J., von Glasow, R., Gómez Martín, J. C., McFiggans, G. and Saunders, R. W.: Atmospheric Chemistry of Iodine, Chem. Rev., 112(3), 1773–1804, doi:10.1021/cr200029u, 2012a.

(4) Mulvaney, R., Pasteur, E. C., Peel, D. A., Saltzman, E. S. and Whung, P.-Y.: The ratio of MSA to non-sea-salt sulphate in Antarctic Peninsula ice cores, Tellus B, 44(4), 295–303, doi:10.1034/j.1600-0889.1992.t01-2-00007.x, 1992.

(5) Pasteur, E. C. and Mulvaney, R.: Migration of methane sulphonate in Antarctic firn and ice, J. Geophys. Res. Atmos., 105(D9), 11525–11534, doi:10.1029/2000JD900006, 2000.

(6) Simpson, W. R., Alvarez-Aviles, L., Douglas, T. A., Sturm, M. and Domine, F.: Halogens in the coastal snow pack near Barrow, Alaska: Evidence for active bromine air-snow chemistry during springtime, Geophys. Res. Lett., 32(4), 1–4, doi:10.1029/2004GL021748, 2005.

Below the new figures:

Figure 1. (a) Schematic map of the traverse area and coring sites, marked with stars. The cores were drilled between Nov 20th 2013 and Jan 8th 2014 (early austral summer). (b) Maximum (left, August 2011) and minimum (right, January 2010) sea ice concentrations in the 130°E-170°W sector for the 2010-2013 time interval covered by the core records (NSIDC data from Meier et al., 2013). The traverse location is marked with an ellipse.

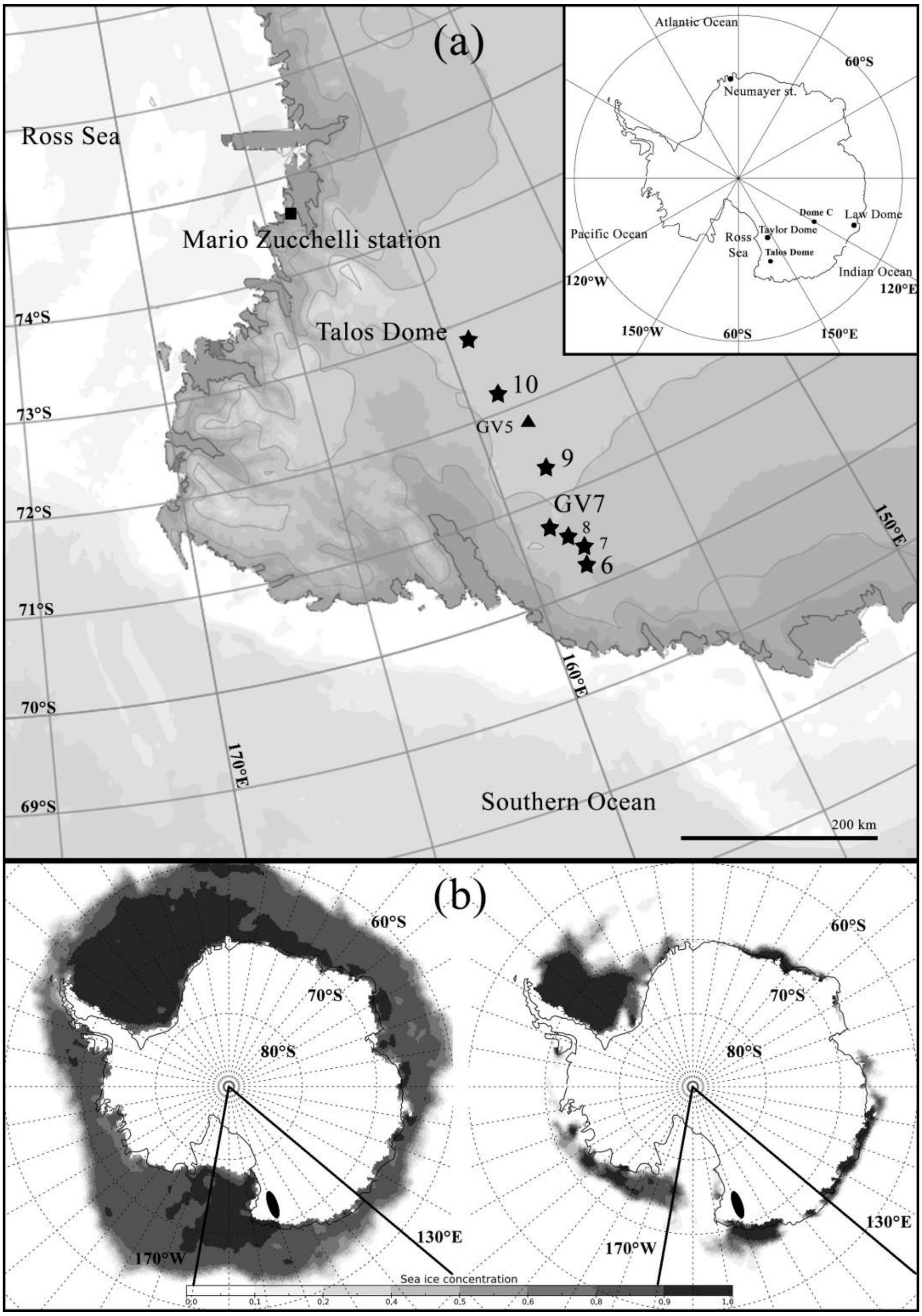


Figure 3. Distribution of bromine enrichment values within cores TD, 10, 9 (blue) and GV7, 8, 7, 6 (red). The dashed line indicates the seawater value (Br$_{enr}$ = 1).

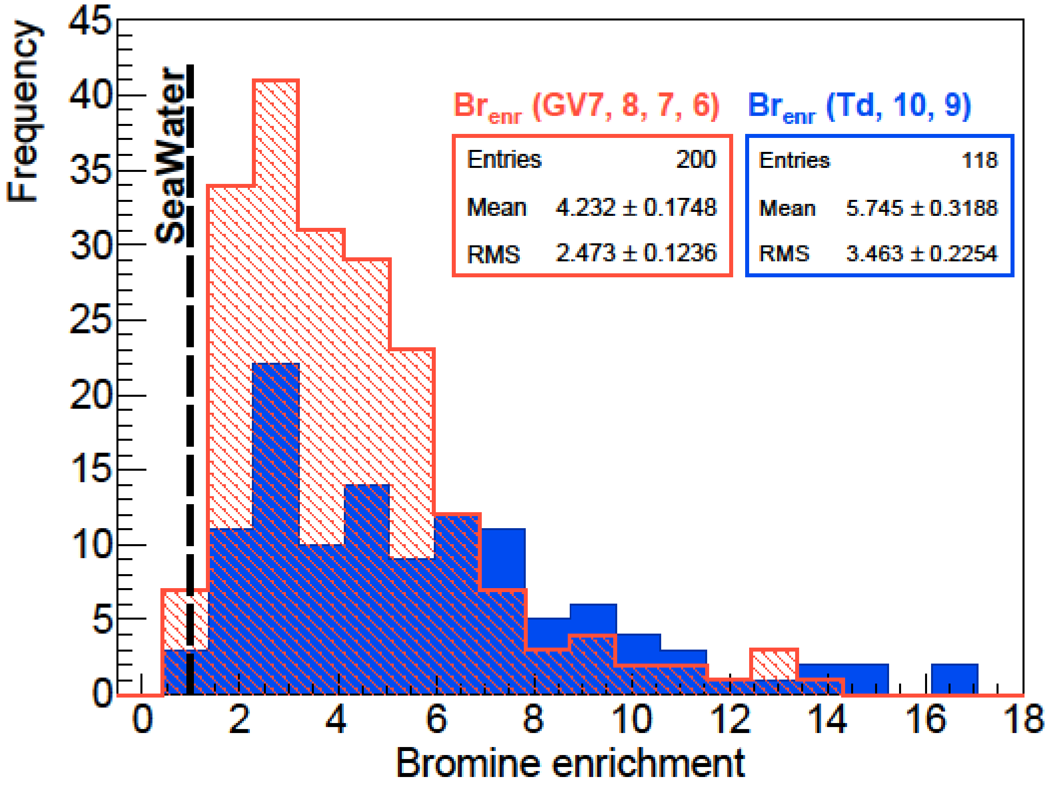


Figure 4. Variability of δ^{18}O (upper panel), Na (middle top panel, left axis), Br (middle top panel, right axis), Brenr (middle bottom panel, left axis), nssBr (middle bottom panel, right axis), and I (bottom panel) in cores 10 (left) and 9 (right). Thick lines represent 3-month running means of the raw data (circles).

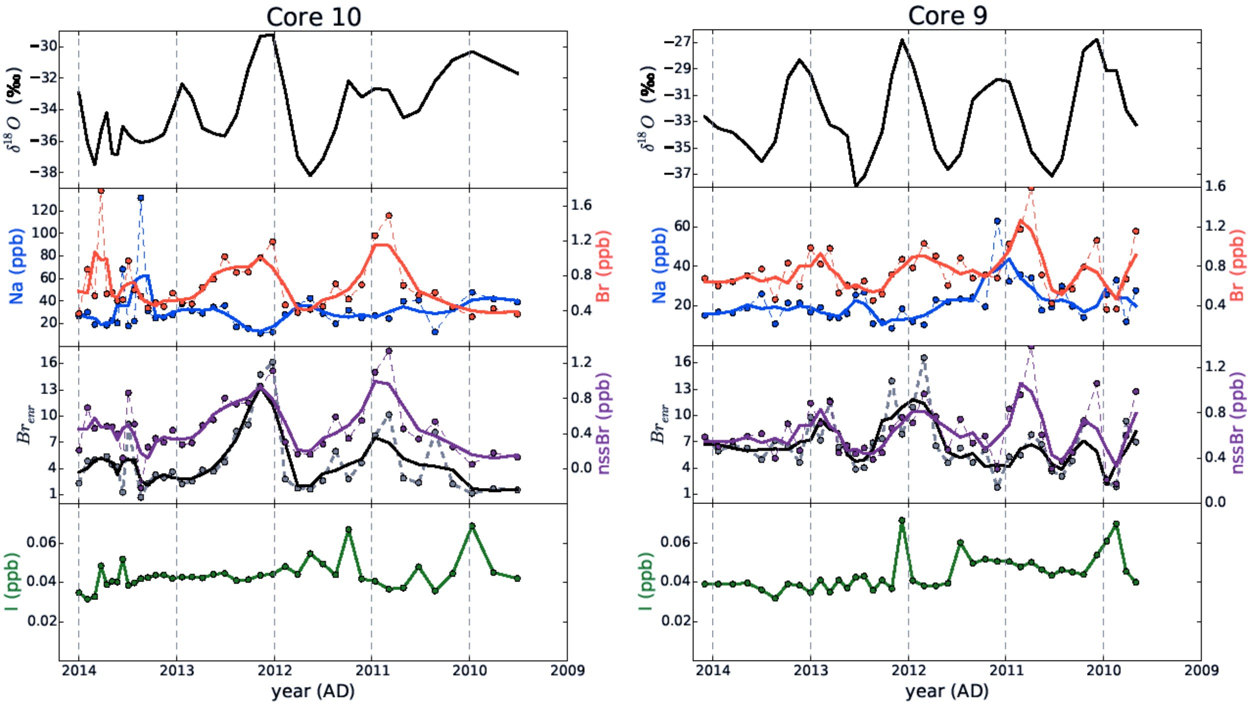


Figure 5. Variability of $\delta^{18}O$ (upper panel), Na (middle top panel, left axis), Br (middle top panel, right axis), Brenr (middle bottom panel, left axis), nssBr (middle bottom panel, right axis), and I (bottom panel) in cores GV7 (left) and 8 (right). Thick lines represent 3-month running means of the raw data (circles).

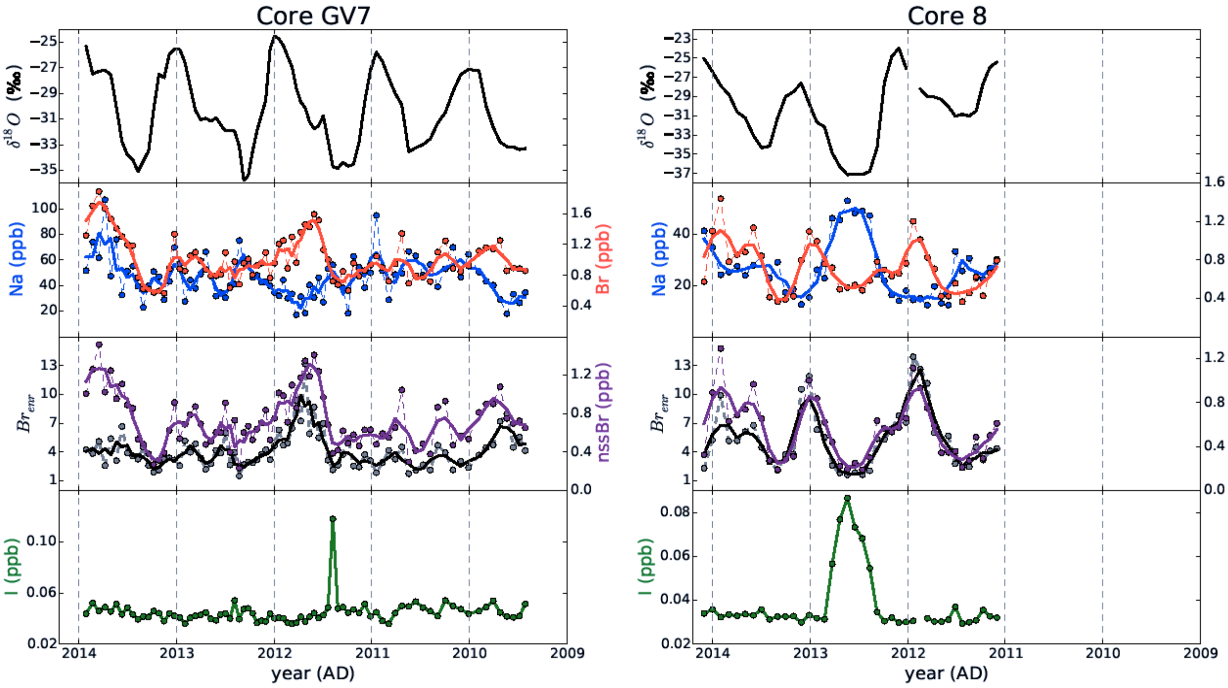


Figure 6. Variability of δ^{18}O (upper panel), Na (middle top panel, left axis), Br (middle top panel, right axis), Brenr (middle bottom panel, left axis), nssBr (middle bottom panel, right axis), and I (bottom panel) in cores 7 (left) and 6 (right). Thick lines represent 3-month running means of the raw data (circles).

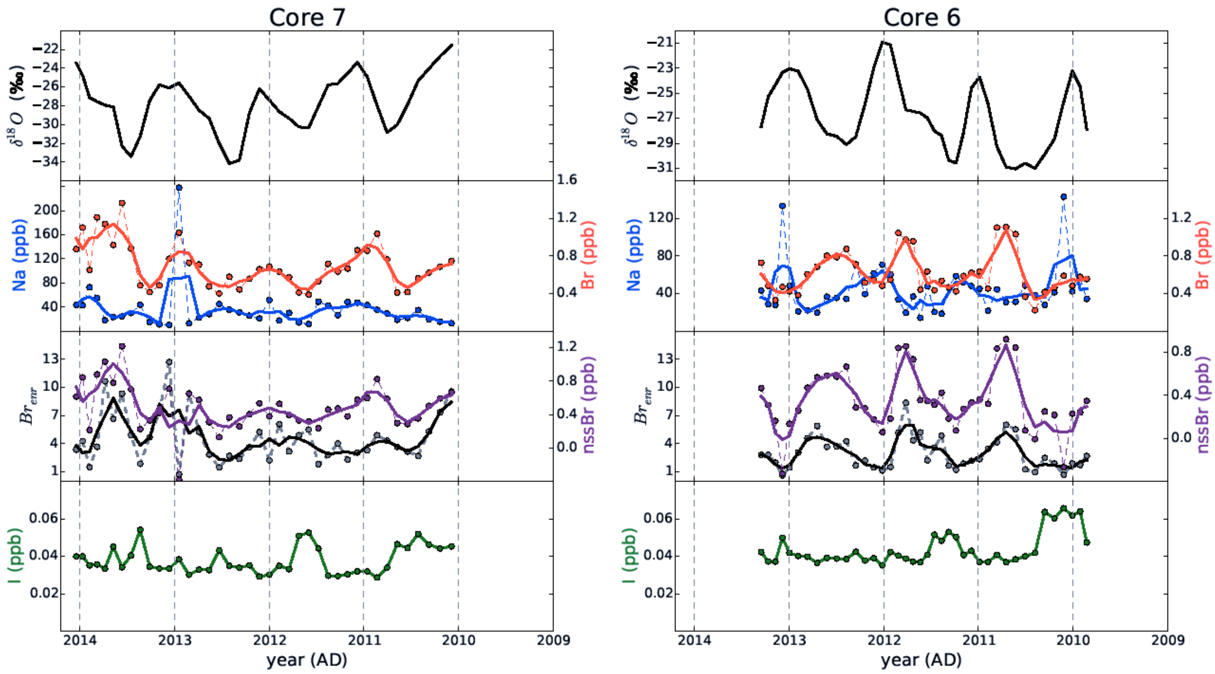


Figure 7. (a) Monthly values of sea ice area (blue) within the 130°E-170°W sector from 2010 to 2013 (±1σ, month variability) and daily average (24 hours) total downwelling spectral irradiance (red), calculated using the TUV model at 71° S, 158° E. Each irradiance calculation was set the 15th day of each month, in 2012. **(b)** Seasonality of annual bromine enrichment along the traverse: the monthly trend shows a seasonal feature with maximum in Spring. Each line refers to a core of the transect (±1σ, shaded blue area). The month averages are displayed in black. The systematic uncertainties associated to the dating are shown as verticals error bars. The magenta band represents the product distribution of normalized sea ice area and insolation, expressed in annual percentage. **(c)** Monthly sea ice area values (blue) from 2010 to 2013, with annual values of FYSI (red) and averaged bromine enrichment (black).

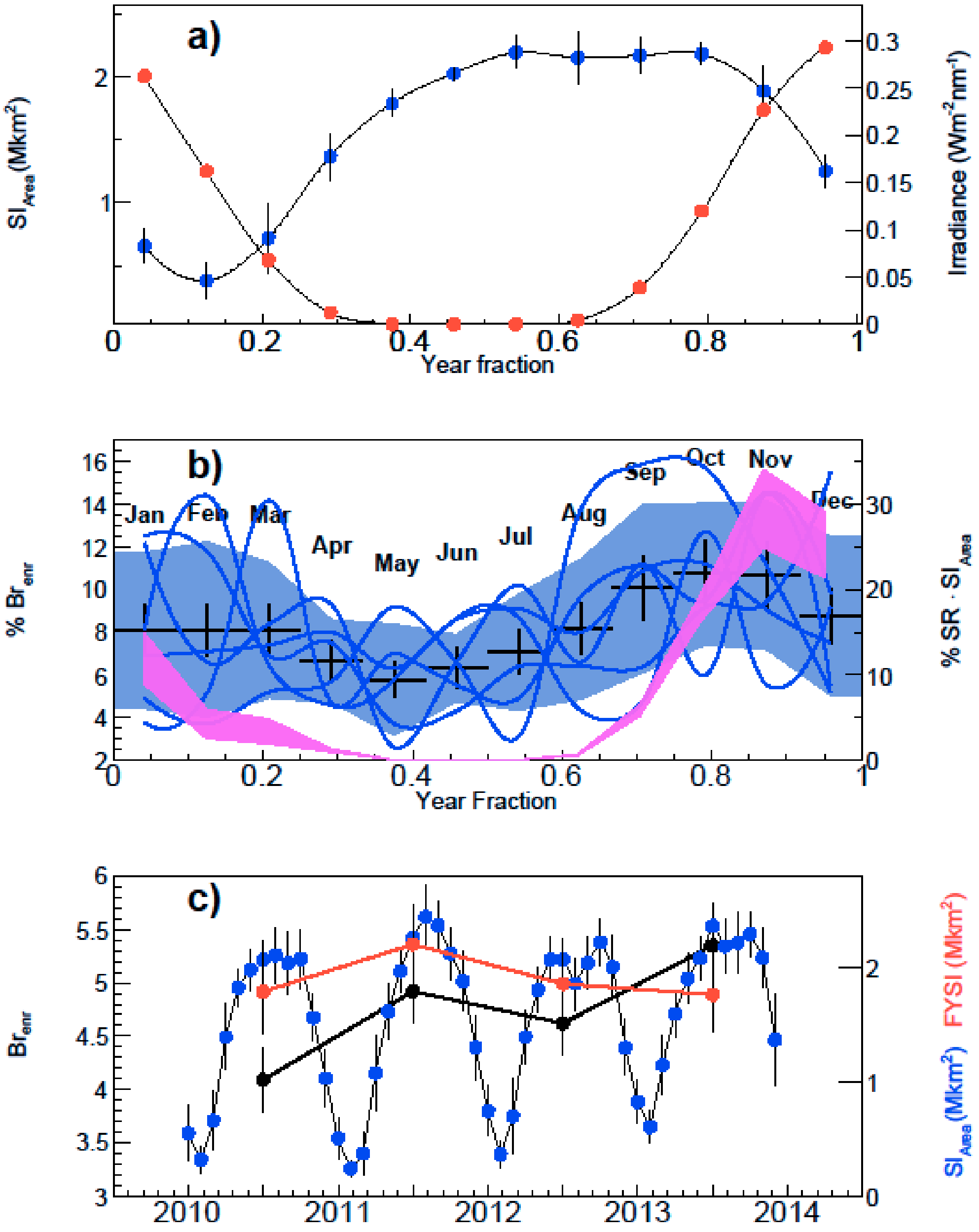
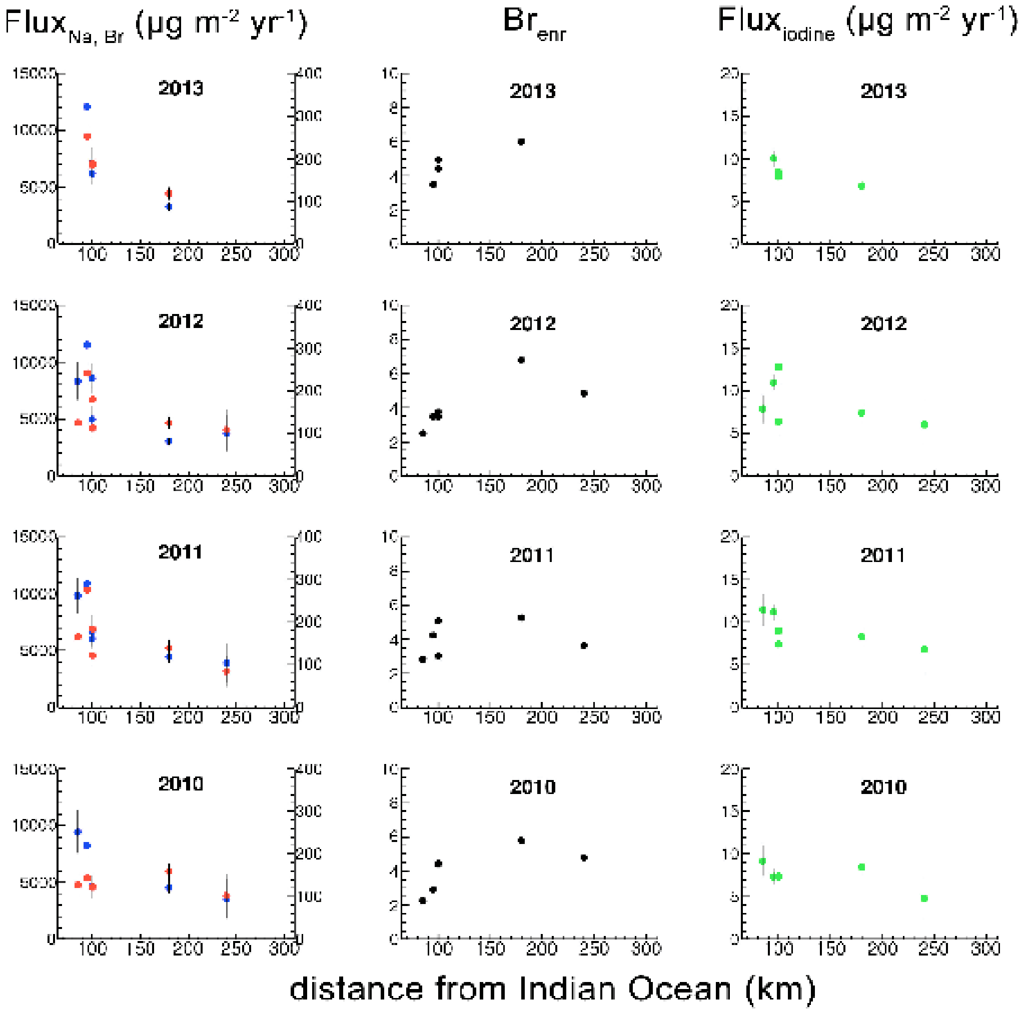


Figure 9. Mean annual fluxes of sodium (blue, left axes), bromine (red, right axes), iodine (green) and bromine enrichment values (black), as a function of distance from the Indian Ocean. Each dot represents a location along the traverse.



Supplementary material

The following figure shows the measurements related to the Talos Dome core.

Figure. Variability of δ^{18}O (upper panel), Na (middle top panel, left axis), Br (middle top panel, right axis), Brenr (middle bottom panel, left axis), nssBr (middle bottom panel, right axis), and I (bottom panel) in the Talos Dome core on a depth scale. Thick lines represent 15 cm running means of the raw data (circles).