Holocene atmospheric iodine evolution over the North Atlantic

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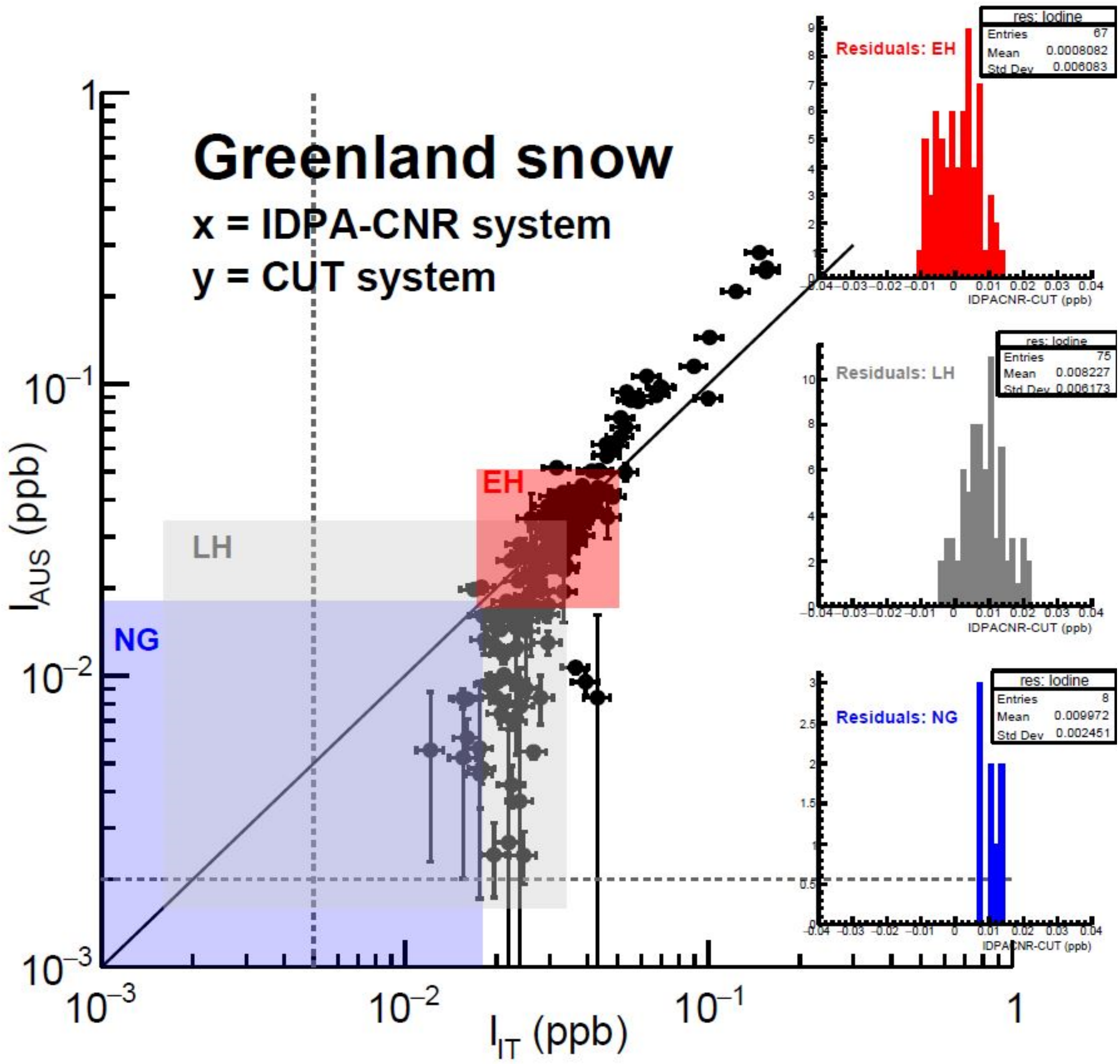

Figure S1: Iodine intercalibration between the IDPA-CNR and the CUT systems performed on Greenland surface snow iodine measurements. The colored areas reflect the average (± 2σ) iodine concentrations detected in the RECAP ice core (EH: Early Holocene; NG: Neoglacial Period; LH: Late Holocene)

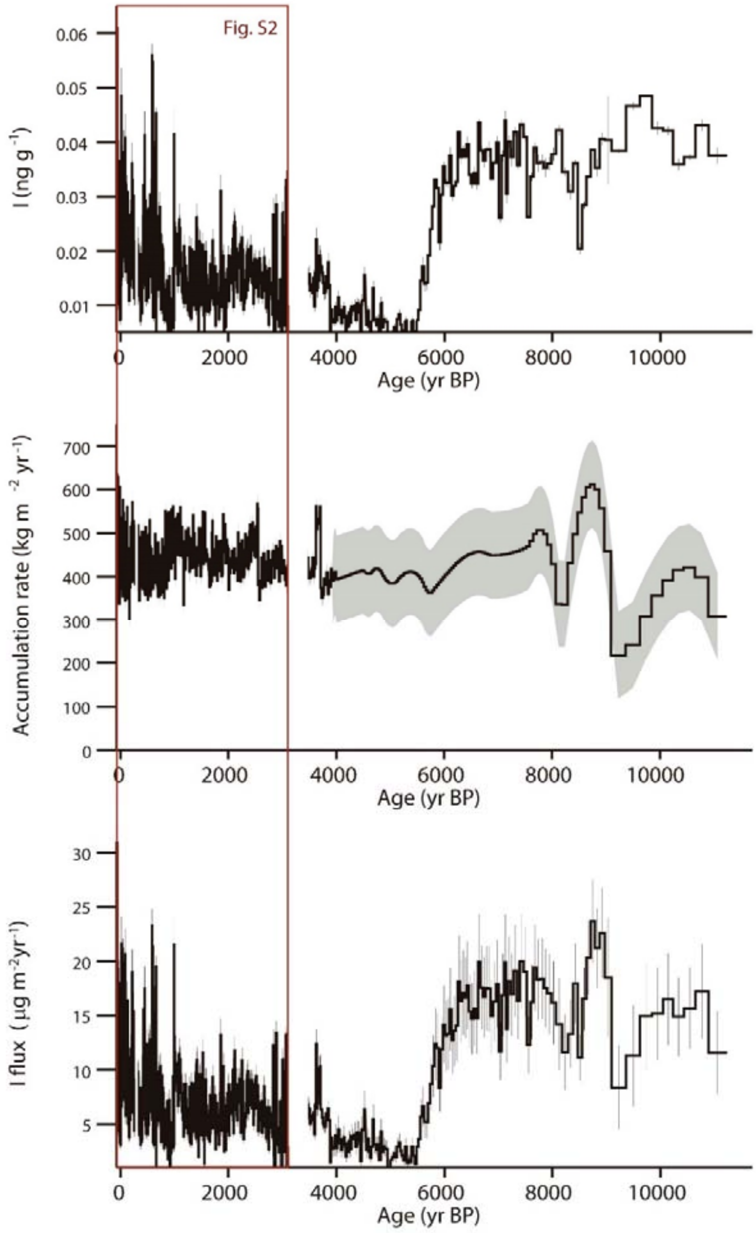

Figure S2. ReCAP ice core time series during the Holocene: Top: iodine concentration (1σ, experimental uncertainties). Middle: accumulation rates and associated uncertainties (1σ band). Bottom: iodine fluxes (1σ, propagated from the concentration and accumulation rate uncertainties). Iodine measurements are missing for the time intervals 275-320 yr BP and 3107-3476 yr BP due to instrumental errors during the analyses. The brown inset area is shown in Fig S2.

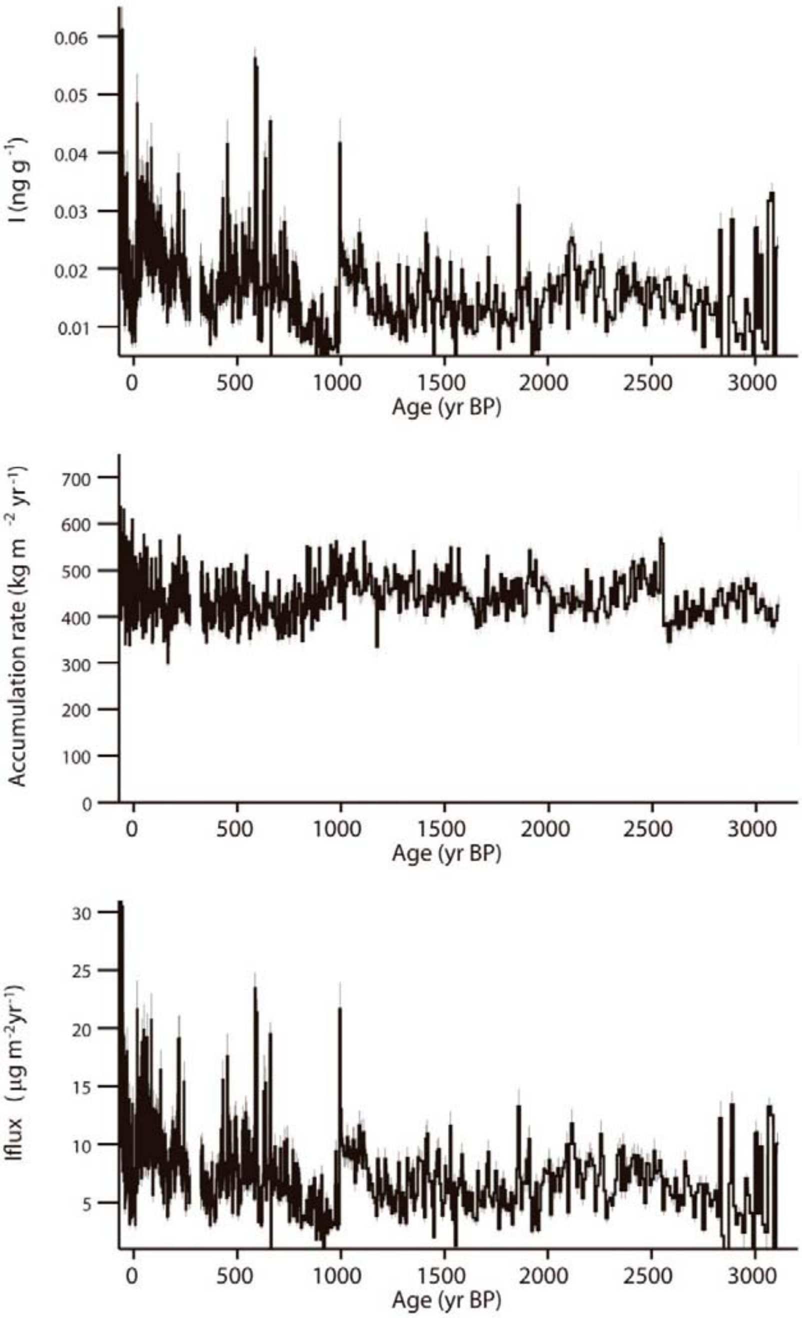

Fig. S3: **ReCAP ice core time series during the Late Holocene**: Top: iodine concentration (1σ, experimental uncertainties). Middle: accumulation rates (and 1σ uncertainties). Bottom: iodine fluxes (1σ, propagated from the concentration and accumulation rate uncertainties).

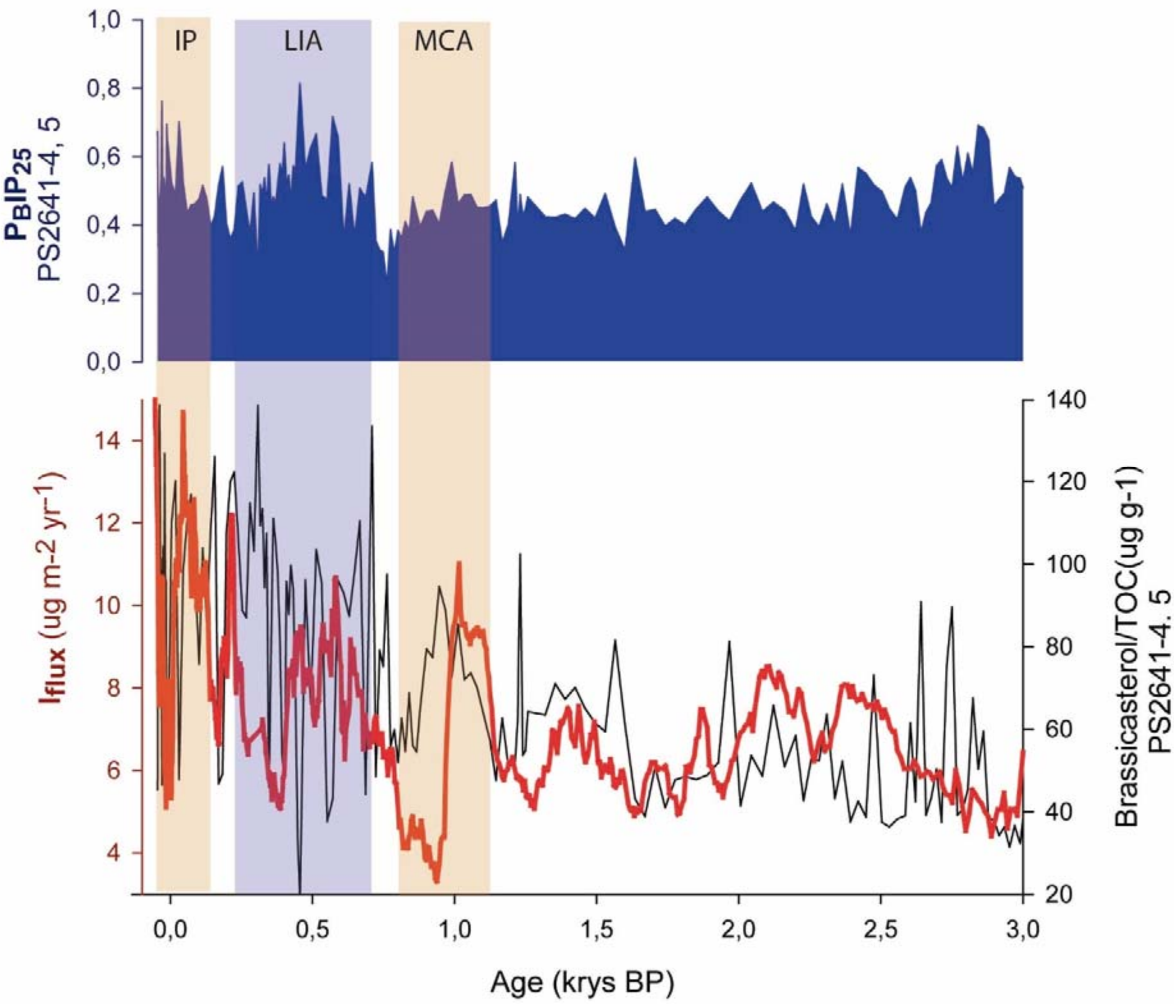

Fig. S4: **ReCAP ice core fluxes, primary productivity and sea-ice conditions in Eastern Greenland during the Late Holocene**: Bottom: I_{flux} 10-samples running average in ReCAP ice core (red) and *Brassicasterol* in eastern Greenland coastal shelf (core PS2641-4,5) (black) (*Kolling et al.*, 2017). Top: eastern Greenland coastal shelf sea-ice extent proxy P_BIP_{25} (*Kolling et al.*, 2017). Color bars indicate the last millennium main climatic phases (LIA: Little Ice Age; MCA; Medieval Climate Anomaly; IP: Industrial Period).

	[I ng/g] ReCAP	SST/°C	[iodide] seawater †	[O3] ppb	Wind speed (ms-1) ‡	Flux_HOI (nmol $m^{-2} d^{-1}$) †	Flux_I2 (nmol $m^{-2} d^{-1}$) †	Total Iodine Flux (nmol $m^{-2} d^{-1}$)	Inorganic emission sources (%)	Organic emission sources (%)
HTM	0,036	8,85	1,251E-08	10§	7	10,49	0,25	10,74	25	75
Neoglacial	0,008	8,57	1,212E-08	10§	7	9,87	0,24	10,10	100	0
Late Holocene	0,017	8,30	1,175E-08	10§	7	9,25	0,23	9,48	46,7	53,3
Present-day	0,038	9,10‡	1,288E-08	30‡	7	33,22	0,78	34,00	75*	25*

* Percentages obtained from Prados-Roman et al., (2015).
† Values obtained based on MacDonald et al. (2014) parametrization.
‡ Values obtained from Merra reanalyses dataset.
§ Values obtained from Volz and Kely (1988).

Table S1. THAMO modelled iodine emission fluxes during the Holocene main climatic phases.

	Holocene Thermal Maximum	Neoglacial	Late Holocene	HTM/ Neoglacial transition
[I] (ng/g)	**0,034**	**0,009**	**0,018**	**277%**
std	*0,01*	*0,00*	*0,01*	
I_{flux} (ug/m^2 yr)	**14,75**	**3,97**	**8,02**	**271%**
std	*4,38*	*2,16*	*4,09*	
Planktic foraminifera (ind/cm^2 kyr)	**6270**	**3653**	**5041**	**71.6%**
std	*4066*	*1365*	*2173*	
***Brassicasterol* (ug/g OC)**	**36,43**	**31,85**	**36,74**	**14.4%**
std	*7,69*	*4,13*	*5,30*	
Dinosterol (%)	**4,69**	**2,98**	**4,86**	**57.4%**
std	*2,40*	*0,67*	*1,58*	
T. Quinqueloba (%)	**32,87**	**19,35**	**21,83**	**70%**
std	*13,87*	*6,44*	*9,64*	
Arctic SST (ºC)	**10,40**	**9,81**	**9,88**	**6%**
std	*0,77*	*0,39*	*0,30*	
P_DIP_{25}	**0,09**	**0,19**	**0,30**	**-52.6%**
std	*0,05*	*0,02*	*0,08*	
IP_{25}	**0,14**	**0,84**	**1,52**	**-83.3%**
std	*0,17*	*0,31*	*0,69*	
71ºN July solar irradiance (W/m^2)	**519,17**	**493,69**	**487,57**	**5.2%**
std	*3,28*	*3,93*	*5,24*	

Table S2: Mean values and standard deviations (std) of iodine levels (iodine concentration and fluxes) in ReCAP ice core and mean values of environmental proxies reconstructed in the Arctic during the main climatic periods discussed in the text (i.e. Holocene Thermal Maximum, Neoglacial period and Late Holocene). From bottom to top: Iodine concentrations [I]; Iodine fluxes (I_{flux}); Planktic foraminifera [*M Telesiński et al.*, 2015; *Werner et al.*, 2013]; *Brassicasterol* and *dinosterol* (*Kolling et al.*, 2017; *Müller et al.*, 2012; *Werner et al.*, 2016); *T. quinqueloba* (Werner et al., 2013; Telesinski et al., 2015*)*; Sea surface temperature (SST) (*Bendle and Rosell-Melé*, 2004; *Justwan and Koç*, 2008; *Justwan et al.*, 2008); Sea-ice cover (P_DIP_{25} and IP_{25}) (*Cabedo-Sanz et al.*, 2016; *Werner et al.*, 2016; *Xiao et al.*, 2017); **i)** 71ºN July solar irradiance (*Laskar et al.*, 2004).

	A) Great Acceleration *(1950-Present-day)*	
	[Na]	[Ca]
[I]	0,252*	
Sig	*0,026*	
N	*78*	
	B) Late Holocene *(last 3.4 kyrs BP)*	
	[Na]	[Ca]
[I]	0,095*	*0,215**
Sig	*0.011*	*0,030*
N	*717*	*102*
	C) Neoglacial *(5.5-3.4 kyrs b2k)*	
[I]	**-0,42****	
Sig	0,000	
N	75	
	D) HTM *(11.7-5.5 kyrs BP)*	
[I]	-0,228	-0,009
Sig	*0,062*	*0,943*
N	*68*	*68*

(ρ=Pearson correlation coefficient, Sig=significance (*=significance<0.05, **=significance<0.01 highlighted in bold font)

Table S3. Great Acceleration, Late Holocene, Neoglacial and Holocene Thermal Maximum Pearson (ρ) correlation coefficients between iodine concentrations [I] in the Renland ice-core and sodium [Na] and [Ca] concentrations. [Ca] data are not available for the Great Acceleration and the Neoglacial Period.