## SUPPLEMENTARY MATERIAL

### 1. Snow pit and shallow core sampling

The Hansbreen snow pit was dug at 450 m a.s.l. (77.13°N; 15.46°E) on the 6<sup>th</sup> of May 2019. The snow pit was 290 cm deep, with a bulk density of 380 kg m<sup>-3</sup> and a total water equivalent (w.e.) of 109.6 cm. The identified snow layers were compact, with several ice strata, suggesting multiple melt or rain-on-snow events during the accumulation season. Snowpack temperature ranged from -4.2°C at the surface to -2.5°C at the snow-firn interface. The snow pit was dug using a clean aluminum shovel. Before sampling for trace element analysis, the snowpit wall was cleaned by removing the outer 20 cm of snow face with a dedicated clean polyethylene shovel to prevent possible contamination originating from the previous excavation. The snow was sampled using pre-cleaned polyethylene tubes at 10 cm depth increments. Nylon suits and gloves were worn during sampling to minimize contamination.

A 2 m deep firn-core was drilled using a coring hand drill (9 cm diameter; Kovacs Mark II Ice Coring System, US) at the bottom of the snowpit. The core was transported to the chemical laboratory of the Polish Polar Station Hornsund where it was weighed, providing density values between 510 kgm<sup>-3</sup> (firn) up to 900 kgm<sup>-3</sup> (ice) for a total of 143 cm of w.e. (Table S1).

#### 2. Hansbreen snow pit and shallow core sample preparation and treatment

The firm-core was scraped off with a clean knife to limit potential contamination by the drill and only the inner part was preserved for chemical analysis (Spolaor et al., 2012). The samples were melted at room temperature and immediately filtered through pre-rinsed Millipore MCE sterilized filters (0.22  $\mu$ m, 47 mm white gridded filters) and transferred to HDPE bottles that were then transported frozen to ISP-CNR laboratories in Venice. The snow pit samples were sampled from the snow wall using pre-cleaned vials, then transported frozen to Italy. Ice core samples were melted and acidified to 2% v/v with HNO<sub>3</sub> (UPA grade, Romil) for 24 hours before analysis. Snow pit samples were treated the same as the ice core samples; they were melted under a clean bench, filtered with Millipore MCE sterilized filters (0.22  $\mu$ m, 47 mm white gridded filters), and acidified for 24 hours prior to analysis. This procedure allowed us to measure the water soluble (leachable) fraction of the trace elements, but not the total abundance. We focused on the soluble fraction of trace elements because it is this fraction that is mobilized and more subject to the effects of liquid water present during the melting season. The determination of trace elements was carried out with inductively coupled plasma-mass spectrometry (ICP-MS) using an iCAP RQ (Thermo Scientific) instrument.

#### 3. Instrumental set-up and chemical Analysis

The ICP-MS (iCAP RQ, Thermo Scientific, US) was equipped with an ASX-560 autosampler (Teledyne Cetac Technologies), PolyPro PFE nebulizer, PFE cyclonic spray chamber thermostatted at  $2.7^{\circ}$ C, sapphire injector, quartz torch and Ni cones. The acquisition was performed at 1550 W of plasma RF power in Kinetic Energy Discrimination (KED) – high matrix mode, using He as the collision gas (4.3 mL min<sup>-1</sup>). Instrument parameters were optimized for best sensitivity in the whole mass range, and minimum oxides (<1%) and

double charges (<3%). A total of 57 elements were measured in triplicate acquisitions. Quantification was obtained by external calibration with multi-elemental standards prepared in Milli-Q water (18 M $\Omega$  cm<sup>-1</sup> at 25°C) with 2% v/v ultrapure grade HNO<sub>3</sub> (Romil) from a combination of certified level multi-elemental solutions IMS-101, IMS-102 and IMS-104 from UltraScientific. Rhodium was continuously spiked to the sample flow through a y-junction and acquired as the internal standard to compensate for possible matrix effects and instrumental drifts during the analytical run. The internal standard spike solution was also prepared in Milli-Q water with ultrapure grade HNO<sub>3</sub> 2% v/v from a certified level monoelemental solution (UltraScientific). Analytical quality control was performed by memory test blank (repeated analysis of ultrapure grade HNO<sub>3</sub> 2% v/v blank solution) after each sample and calibration verification (repeated analysis of reference standards) every 11 samples. The internal standard recovery was always >85% throughout the entire sequence of analysis.

#### 4. Calculation of the sea spray component, TE flux and element enrichment factor (EF)

To determine the different sources of elements in our snow and ice samples, we estimated the contribution of sea spray to total deposition by calculating the sea spray contribution (ssC). Considering the coastal location of our sampling site, maritime emissions are relatively high and the removal of this possible effect becomes necessary for a robust evaluation of other sources. Sodium in polar environments is commonly used as a sea spray tracer (Mafezzoli et al 2019; Lia et al 2017), especially in coastal areas where the crustal contribution for these elements is negligible (less than 1%). Using the sodium concentration as a maritime emission tracer and the average sea water composition proposed by Millero et al. 2008, we can calculate the sea spray contribution (and the relative percentage -  $ssC_s(\%)$ ) for each element with the formula:

1)  $[ssC_x] = [Na]_s * ([X]_{sw} / [Na]_{sw})$ 

# 2) $ssC_x(\%) = ([ssC_x]/[X]_s)*100$

where  $[X]_s$  = measured concentration of the trace element in question,  $[Na]_s$  = concentration of sodium in the sample,  $[X]_{sw}$  = concentration of the trace element in question in seawater,  $[Na]_{sw}$  = concentration of sodium in seawater. The contribution of sea spray emissions for each element is reported in Table 1.

The water-soluble trace element flux is a useful parameter for comparing snow layers with different densities. Snow layers with similar trace element concentrations might have an overall different mass content if the density is not equal. The flux is expressed as mg m<sup>-2</sup> and indicates the deposition flux that characterizes each layer. The flux is calculated as the trace element concentration multiplied by the snow water equivalent (w.e.) of each discrete layer, where the w.e. is a product of the layer thickness and density.

The enrichment factor (EF) of an element is calculated from the upper continental crust (UCC) abundance proposed by Wedepohl K, 1995. EF is calculated using barium, since Ba has high solubility and is negligible in sea spray emissions. The EF is calculated as:

3)  $EF_X = ([X]_s / [Ba]_s) / ([X]_{ucc} / [Ba]_{ucc}),$ 

where "X" is the element considered, "s" indicate the concentration measured in the sample, and "ucc" the abundance in the upper continental crust (values in square brackets signify concentrations of the given

element). An example of EF calculation and its explanation can be found in Barbante et al. 2017 and McConnel et al. 2008.

### REFERENCES

- Barbante, C., Spolaor, A., Cairns, W. R. L. & Boutron, C. Man's footprint on the Arctic environment as revealed by analysis of ice and snow. *Earth-Sci Rev* 168, 218-231 (2017).
- Lai, A. M., Shafer, M. M., Dibb, J. E., Polashenski, C. M. & Schauer, J. J. Elements and inorganic ions as source tracers in recent Greenland snow. *Atmos Environ* **164**, 205-215.
- Maffezzoli, N. *et al.* A 120.000-year record of sea ice in the North Atlantic? *Clim. Past* **15**, 2031-2051, doi:10.5194/cp-15-2031-2019 (2019).
- McConnell, J. R. & Edwards, R. Coal burning leaves toxic heavy metal legacy in the Arctic. *PNAS* **105**, 12140-12144 (2008).
- Millero, F. J., Feistel, R., Wright, D. G., and McDougall, T. J.: The composition of Standard Seawater and the definition of the Reference-Composition Salinity Scale, Deep Sea Research Part I: Oceanographic Research Papers, 55, 50-72, 2008.
- Spolaor A., P. Vallelonga, J. Gabrieli, G. Cozzi, C. Boutron, C. Barbante, Determination of Fe2+ and Fe3+ species by FIA-CRC-ICP-MS in Antarctic ice samples. *J Anal Atom Spectrom* 27, 310-317 (2012).
- Wedepohl K, H.: The composition of the continental crust, Geochim Cosmochim Acta, 59, 1217-1232, 1995.

**Table S1.** Density measurements of the Hansbreen annual snow pack and the shallow firn core. The table reports the name of each sample (sample ID), the snow pack depth coverage (sample length), the cumulative snow depth, the density measured corresponding to each sample interval, the corresponding w.e for each layer and the cumulative depth expressed as water equivalent (w.e.). The column "Type" indicates if the sample is from the snow pit (SP) or from the shallow ice core (IC).

Sample ID	Туре	Sample thickness (cm)	Cumulative depth (cm)	Density (gcm <sup>3</sup> )	W.e. (cm)	W.e. cumulative (cm)
HSB1	SP	10	10	0.280	2.8	2.8
HSB2	SP	10	20	0.265	2.7	5.5
HSB3	SP	10	30	0.441	4.4	9.9
HSB4	SP	10	40	0.341	3.4	13.3
HSB5	SP	10	50	0.399	4.0	17.3
HSB6	SP	10	60	0.351	3.5	20.8
HSB7	SP	10	70	0.342	3.4	24.2
HSB8	SP	10	80	0.341	3.4	27.6
HSB9	SP	10	90	0.413	4.1	31.7
HSB10	SP	10	100	0.422	4.2	35.9
HSB11	SP	10	110	0.422	4.2	40.1
HSB13	SP	10	130	0.422	4.2	44.3
HSB14	SP	10	140	0.422	4.2	48.5
HSB15	SP	10	150	0.401	4.0	52.5
HSB16	SP	10	160	0.311	3.1	55.6
HSB17	SP	10	170	0.366	3.7	59.2
HSB18	SP	10	180	0.372	3.7	63.0
HSB19	SP	10	190	0.321	3.2	66.2
HSB20	SP	10	200	0.395	4.0	70.1
HSB21	SP	10	210	0.446	4.5	74.6
HSB22	SP	10	220	0.421	4.2	78.8
HSB23	SP	10	230	0.417	4.2	82.9
HSB24	SP	10	240	0.412	4.1	87.0
HSB25	SP	10	250	0.423	4.2	91.2
HSB26	SP	10	260	0.378	3.8	95.0
HSB27	SP	10	270	0.361	3.6	98.6
HSB28	SP	10	280	0.372	3.7	102.3
HSB29	SP	10	290	0.372	3.7	106.0
H91	IC	9.5	299.5	0.641	6.1	112.1
H93	IC	17.5	317	0.512	8.9	121.0
H93B	IC	22.4	339.4	0.572	12.8	133.8
H94	IC	23	362.4	0.541	12.5	146.3
H95	IC	21.5	383.9	0.573	12.3	158.6
H96	IC	37.5	421.4	0.63	23.7	182.3
H97	IC	28	449.4	0.78	21.8	204.0
H98	IC	19	468.4	0.90	17.1	221.1
H99	IC	19	487.4	0.90	17.1	238.2
H910	IC	14.5	501.9	0.74	10.8	249.0

**Figure S1**.Distribution of the REE flux in the snow (0 - 290 cm depth) and firn samples (300 - 500 cm depth) (a) and of UCC normalized values (b). The comparison of the two graphs highlights 1) a common source for REEs for all the samples analyzed and 2) the presence of layers characterized by a different concentration of REEs.

