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Section S1. East–west variations in major ion concentrations

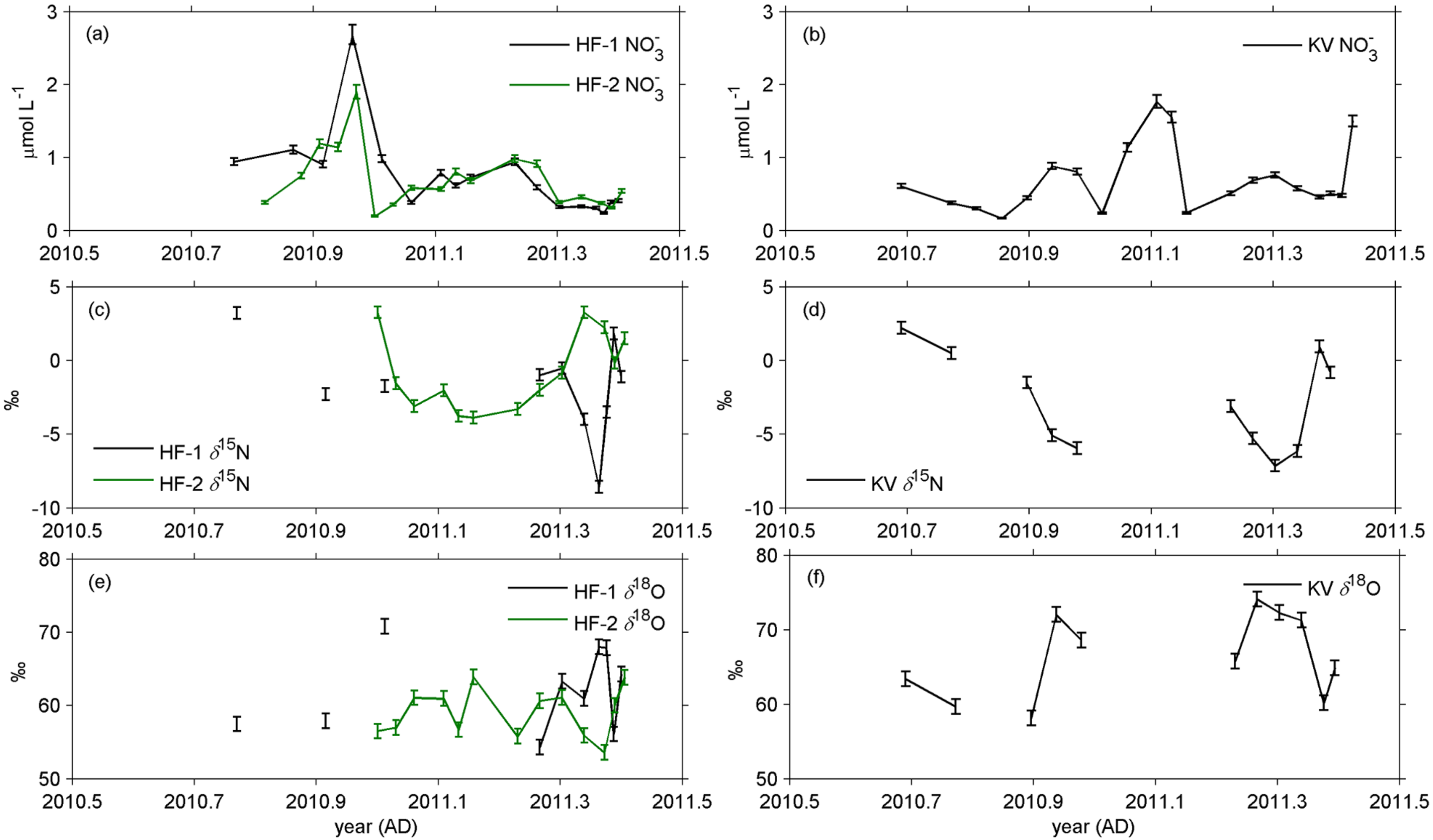
A decrease in [$nssSO_4^-$] and [$nssMg^{2+}$] from east to west can be observed in Fig. 2, with the mean concentrations of these ions showing a significant linear correlation (*R*> 0.93) with the longitudinal position (in degrees) of the sampling sites. No such significant linear correlation was found for [$nssCl^-$], [$nssK^+$] and [$nssCa^{2+}$] (Supplementary Table S2).

[NO_3^-] also shows a linear correlation with longitude (R = 0.93, p = 0.02, α = 0.05), with higher concentrations for AF-11snow than for the other sites (Supplementary Table S3). Note that due to the lack of isotopic data for the AF-11snow snow pit, we used the average isotope deltas of the AF-11 ice core for the linear correlation calculations. The linear correlation coefficients between longitude and isotope composition for the 2010-11 period were R = -0.77 (p = 0.13) and R = 0.59 (p = 0.3) for δ^{15}N and δ^{18}O, respectively (Supplementary Table S3).

Section S2. Main air transport patterns to Summit, Greenland

NO_3^- stable isotopes have been analysed in the atmosphere, snow and ice for a few High Arctic sites, mainly at Summit, Greenland (Freyer et al. 1996; Hastings et al. 2004; Hastings et al. 2009); Barrow, Alaska (Morin et al. 2012), Alert, Canada (Morin et al. 2007; Morin et al. 2008); Ny-Ålesund, Svalbard (Heaton et al. 2004; Amoroso et al. 2010; Morin et al. 2012; Björkman 2013); and the sites reported here.

It has been reported that 90 % of deposited NO_3^- is preserved at Summit (Burkhart et al. 2004). Although, photolysis has an impact on NO_3^- stable isotopes (Blunier et al. 2005; Frey et al. 2009), it has been recently found that this is not significantly affecting snow NO_3^- concentration or isotopes at Summit (Fibiger et al. 2013). Source regions contributing to the chemical load at Summit (72° 34’ N, 38° 27’ W, 3200 m a.s.l.) have been identified by Kahl et al. (1997) using a 40-year analysis of atmospheric trajectories. Daily 10-day back-trajectories arriving at a pressure of 500 hPa above Summit were calculated between 1946 and 1989. The results show that westerly flow to Summit is predominant. During winter, air transport is dominated by trajectories originating from East Asia (58 %) and North America (27 %). During spring, most of the trajectories originated from East Asia (34 %), the North Pacific (31 %) and North America (26 %). In summer, the North American component is dominant, with 46 % of the trajectories, while the North Pacific and East Asia contribute 23 % and 20 %, respectively. During autumn, trajectories are dominated by the North Pacific component (42 %), followed by East Asia (32 %) and North America (16 %) (Kahl et al. 1997).



Supplementary Fig. S1. [NO_3^-], $\delta^{15}N_{NO_3^-}$ and $\delta^{18}O_{NO_3^-}$ records measured in the (a, c, e) HF-1 and 2 and (b, d, f) KV snow pits at Holtedahlfonna and Kongsvegen, respectively. Error bars denote the analytical error of the measurements, i.e., 5 % maximum error for NO_3^- measurements, 0.4 ‰ and 1 ‰ for $\delta^{15}N_{NO_3^-}$ and $\delta^{18}O_{NO_3^-}$, respectively.

Supplementary Table S1. Values of $k = [X]/[Na^+]$ used in the calculation of non-sea salt ion concentrations. The values were obtained from the standard mean chemical composition of seawater with a practical salinity of 35.

Charge-equivalent concentration ratio k	
Cl^-/Na^+	1.16
Na^+/Na^+	1.00
$\frac{1}{2}SO_4^{2-}/Na^+$	0.12
K^+/Na^+	0.02
$\frac{1}{2}Ca^{2+}/Na^+$	0.04
$\frac{1}{2}Mg^{2+}/Na^+$	0.23

Supplementary Table S2. Input data used to obtain the linear correlations, *R* and *p* values described in section S1.

Site	Longitude (° E)	Latitude (° N)	Elevation (m a.s.l.)	Accumulation rate, water equivalent (m a^{-1})	Average charge-equivalent concentration (μmol L^{-1})				
					[$nssCl^-$]	[$nssSO_4^{2-}$]	[$nssK^+$]	[$nssCa^{2+}$]	[$nssMg^{2+}$]
KV	13.33	78.75	673	0.75	1.6	1.4	0.02	2	0.5
HF-1	13.39	79.14	1127	0.66	7	2	0.1	1	1
HF-2	13.39	79.14	1130	0.61	2	1	0.02	2	0.5
LF-11snow	17.43	78.82	1202	0.40	3	2	0.1	4	1
AF-11snow	23.97	79.83	757	0.69	5	4	0.1	0.4	2

	[$nssCl^-$]		[$nssSO_4^{2-}$]		[$nssK^+$]		[$nssCa^{2+}$]		[$nssMg^{2+}$]	
	R	*p*	*R*	*p*	*R*	*p*	*R*	*p*	*R*	*p*
Longitude	0.26	0.68	0.94	0.002	0.58	0.31	-0.28	0.65	0.93	0.02
Latitude	0.50	0.39	0.82	0.09	0.41	0.49	-0.75	0.15	0.84	0.07
Elevation	0.19	0.75	-0.37	0.54	0.29	0.64	0.49	0.40	-0.24	0.69
Accumulation	0.09	0.87	0.12	0.85	-0.39	0.51	-0.80	0.10	0.03	0.96

Supplementary Table S3. Input data used to obtain the linear correlations, *R* and *p* values described in section S1.

Site	Longitude (° E)	Latitude (° N)	Elevation (m a.s.l.)	Accumulation rate, water equivalent (m a^{-1})	[NO_3^-] (µmol L^{-1})	$\delta^{15}N_{NO_3^-}$ (‰)	$\delta^{18}O_{NO_3^-}$ (‰)
KV	13.33	78.75	673	0.75	0.7	-3	66
HF-1	13.39	79.14	1127	0.66	0.7	-2	62
HF-2	13.39	79.14	1130	0.61	0.7	-1	59
LF-11snow	17.43	78.82	1202	0.40	0.7	-11	80.99
AF-11snow	23.97	79.83	757	0.69	2	-9	72

	NO_3^- concentration		$\delta^{15}N_{NO_3^-}$		$\delta^{18}O_{NO_3^-}$	
	R	*p*	*R*	*p*	*R*	*p*
Longitude	0.93	0.02	-0.77	0.13	0.59	0.30
Latitude	0.91	0.03	-0.21	0.73	-0.04	0.95
Elevation	-0.50	0.38	-0.004	0.99	0.04	0.94
Accumulation	0.28	0.64	0.57	0.31	-0.64	0.25

Supplementary Table S4. Average source emission fraction (f) of combustion sources (stationary + mobile) (f_{comb}) and soil emissions (f_{soil}) of the USA, the European countries belonging to the Organisation for Economic Co-operation and Development (OECD) and the Russian Federation for the period 1970-2008 (EC-JRC/PBL EDGAR version 4.2).

Emission fraction	USA	OECD Europe	Russian Federation
f_{comb}	0.9852	0.9748	0.9792
f_{soil}	0.0148	0.0252	0.0208

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