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Supplement of

Canadian forest fires, Icelandic volcanoes and increased local dust observed in six shallow Greenland firn cores

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S1-The Copenhagen CFA set up for the analysis of the shallow firn cores

In Figure S1 a flow chart of the CFA setup used to analyse the firn cores is presented.

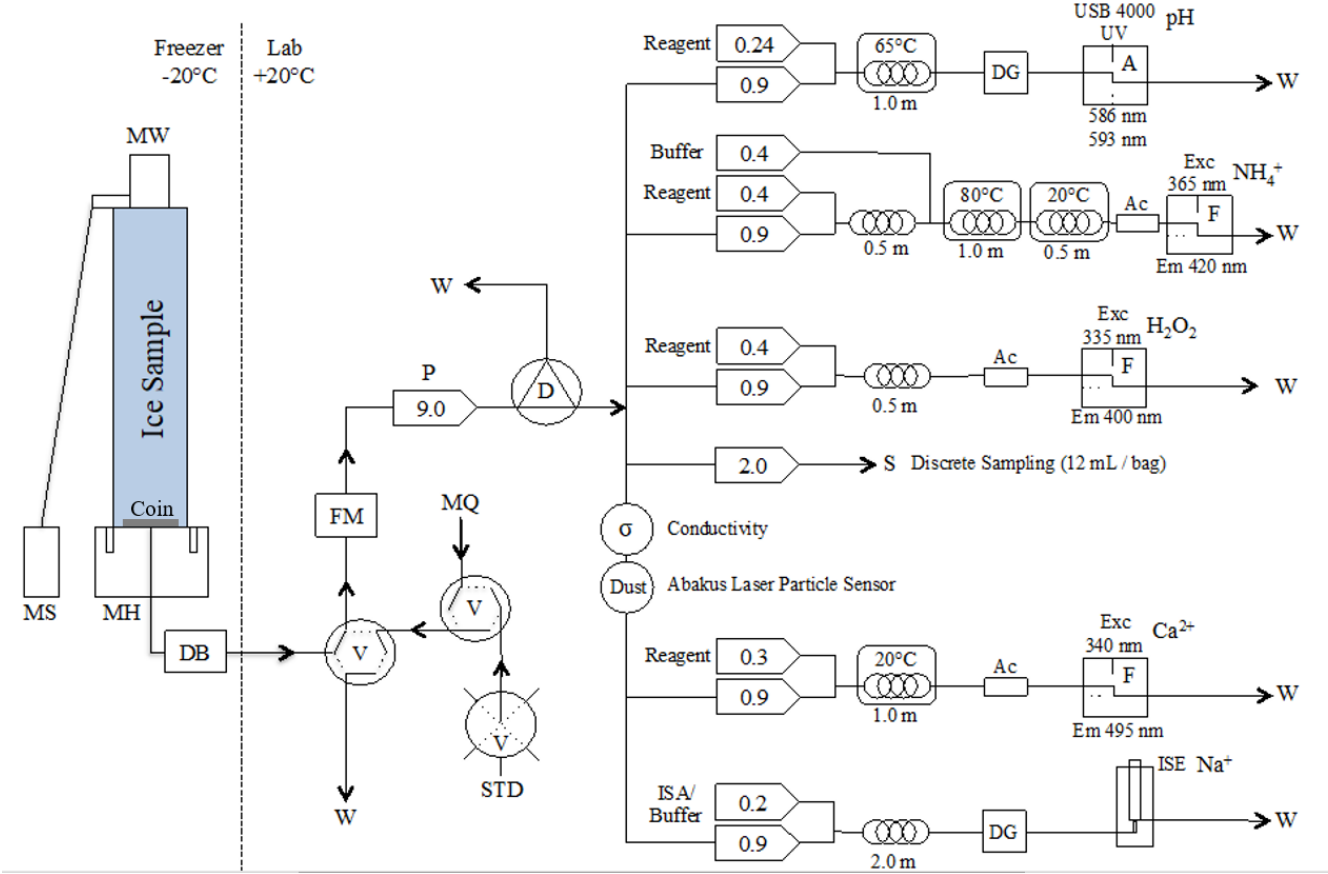

Figure S1: Copenhagen CFA (Bigler et al., 2011) set up used to measure the 6 firn cores. Flowchart of the Copenhagen continuous flow analysis system setup with a melt head (MH) melting the ice sample, melting speed encoder (MS), melting weight (MW), Coin added for firn analysis, debubbler (DB) selection and injection valves (V) for standard solutions (STD) and blanks (MQ), waste (W), flow meter (FM), peristaltic pumps (P), triangular debubbler (D), buffer and reagents, reaction coils, Accurel membrane debubblers (Ac), absorption (A), fluorescence (F), discrete sampling line (S), conductivity detector (σ), particle sensor (Dust) and a vacuum degassing unit. Arrows indicate flow directions. Flow rates within the pump system (mL min^{-1}), temperatures, reaction coil lengths and detector light wavelengths are shown in the flowchart.

Insoluble dust was determined using an Abakus laser particle detector with a LDS-23/25bs sensor type (Klotz, Simonsen et al., 2018) and the conductivity using a bench-top conductivity meter (3082 with micro flow cell 829, Amber Science). The measurements of ammonium, hydrogen peroxide and calcium is based upon fluorescence methods (Kaufmann et al., 2008; Bigler et al., 2011) using photomultiplier detectors (PMT-FL, FIAlab instruments) and spectrofluorimeters set to specific (Sigg et al., 1994a). Acid was determined as described in Kjær et al. 2016a using a dye-based method. In Table S1 and S2 a full list of reagents and instrumentation used is presented.

Table S1. Overview of measurement instruments of the Copenhagen CFA system (Bigler et al., 2011).

Parameter	Instrument
Conductivity	Amber Science 3082 Multi-Function Conductivity Meter
Dust	Abakus LDS 23/25bs sensor, Klotz; ASL-1600-20, Sensirion
Ca^{2+}, NH_4^+, H_2O_2	PMT-FL Fluorometer, FIAlab Instruments
H^+	Ocean Optics USB 4000 Spectrometer
Na^+	PerfectION^{TM} comb Na^+, S220 SevenCompact^{TM}, Mettler Toledo

Table S2. Reagent and buffer solutions for fluorescence methods (Röthlisberger et al., 2000; Sigg et al., 1994b; Bigler et al., 2011; Kjær et al., 2016)

NH_4^+ reagent	**Components**	Lifetime: 2 days
950 mL	Milli-Q	
1.43 g	Fluoraldehyde o-Phthaldialdehyde reagent solution (OPA)	
60 mL	Ethanol	
NH_4^+ buffer	**Components**	Lifetime: 2 days
1L	Milli Q	
35.8 g	Sodium phosphate dibasic dodecahydrate ($Na_2HPO_4 \cdot 12H_2O$)	
600 µg	Sodiumhydroxide (NaOH >32%)	
100 µg	Formaldehyde (HCHO >37%)	
0.8 g	Sodium sulfite (Na_2SO_3)	
H_2O_2 reagent	**Components**	Lifetime: 3 days
1 L	Milli-Q	
0.61 g	4-ethylphenol	
5 mg	Peroxidase type II	
6.18 g	Boric acid (H_3Bo_3)	
7.46 g	Potassium chloride (KCl)	
150 µg	Sodium hydroxide (NaOH)	
Ca^{2+} reagent	**Components**	Lifetime: 1 days
800 mL	Milli-Q	
20 mg	Quin-2 Potassium hydrate	

2.91 g	PIPES	
1 - 1.5 mL	Sodium hydroxide (NaOH) to buffer pH7	
pH reagent	**Components**	Lifetime: >1 week
900 mL	Milli-Q	
0.025 g	Bromophenol Blue	
0.025 g	Chlorophenol Red	
250 µg	Brij L23	

The NH_4^+ and Ca^{2+} was calibrated using 3 standards made from a multi-element stock solution, acid was calibrated with a two-point calibration made from a 0.1M HCl (Fluka), and the H_2O_2 was calibrated based on a two-point calibration using a Peroxide stock solution (30% H_2O_2). The standards concentration preparation scheme is presented in table S3.

Table S3. Concentrations of CFA standard solutions used to calibrate the data

Ca^{2+}, NH_4^+, Na^+	**Multi-element standard**	**Milli-Q**	**Concentration**
	µL	mL	ppb
Stock solution			10^5
Std 1	20	200	10
Std 2	50	200	25
Std 3	200	200	100
H_2O_2	**Peroxide standard**	**Milli-Q**	**Concentration**
	µL	mL	ppb
Stock solution			3×10^8
1st dilution	30	100	9×10^4
Std 1 (2nd dilution)	50	75	60
Std 2 (2nd dilution)	100	75	120
pH (H^+)	**Acidity standard HCL**	**Milli-Q**	**Concentration**
	µL	mL	µM
Stock solution			3.65×10^6
1st dilution	600	60	991
Std 1 (2nd dilution)	1200	120	9.8
Std 2 (2nd dilution)	2400	120	19.6

S2 Data on a depth scale

Below is shown from top the calibrated data of peroxide, Calcium, Insoluble Dust, Ammonium, Conductivity and Acidity on a depth scale together with in red vertical lines the annual layers chosen for the six individual firn cores.

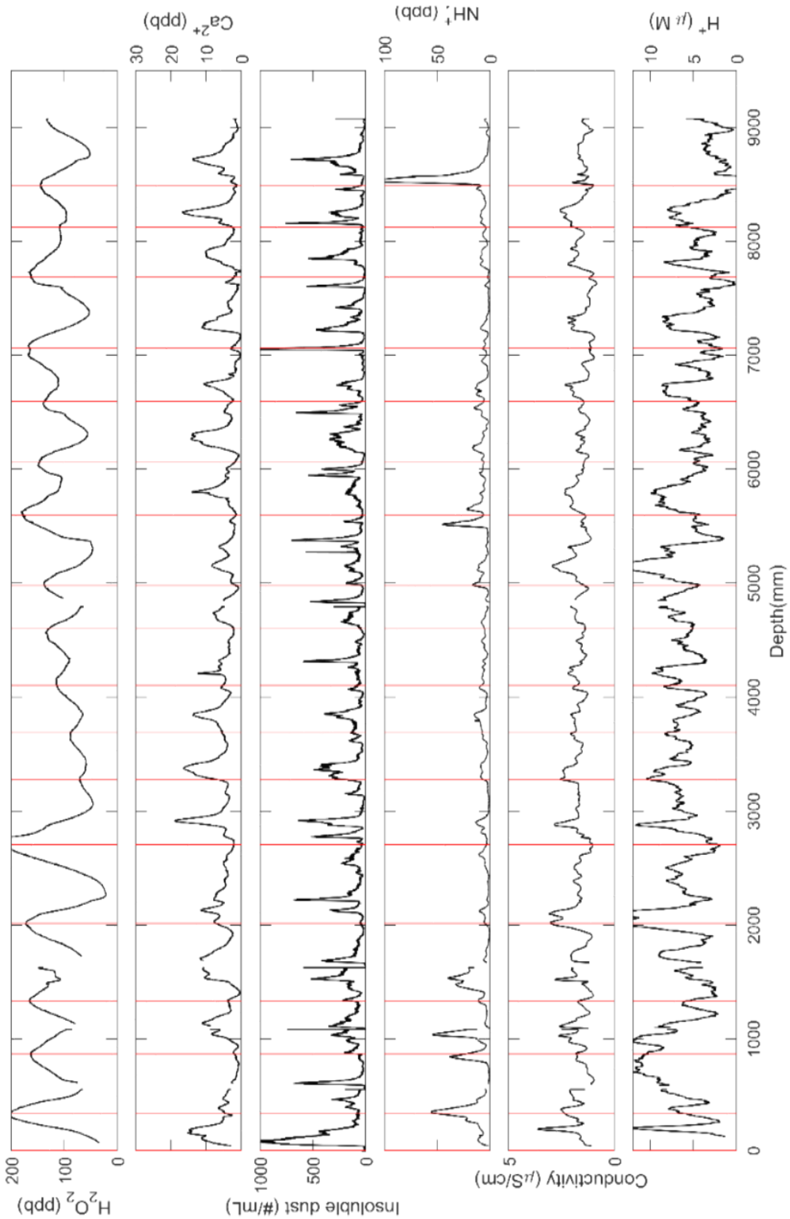

Figure S2: T2015-A1 data on a depth scale together with chosen annual layers (red vertical lines) used for generating the age scale.

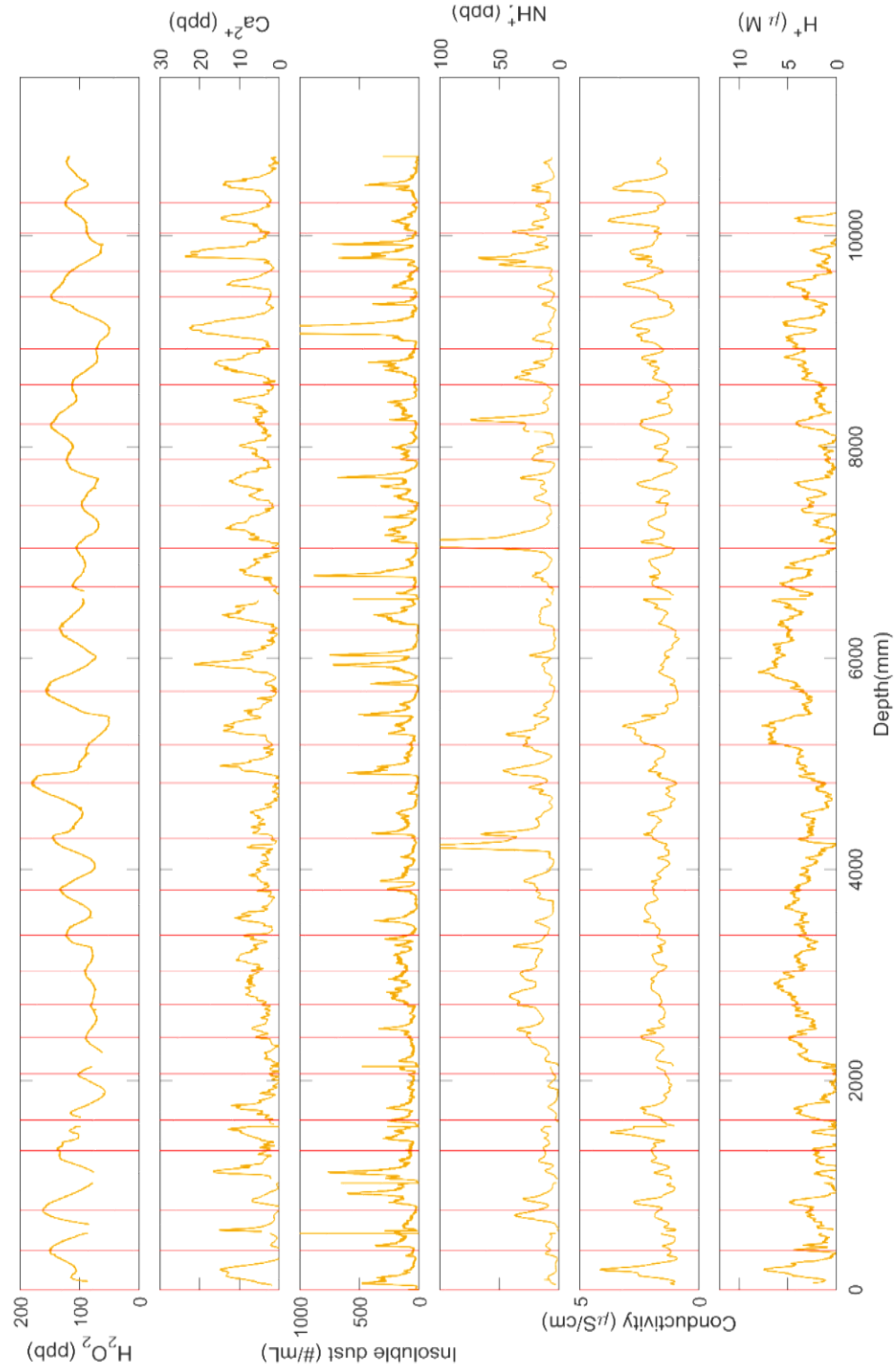

Figure S3: T2015-A2 data on a depth scale together with chosen annual layers (red vertical lines) used for generating the age scale.

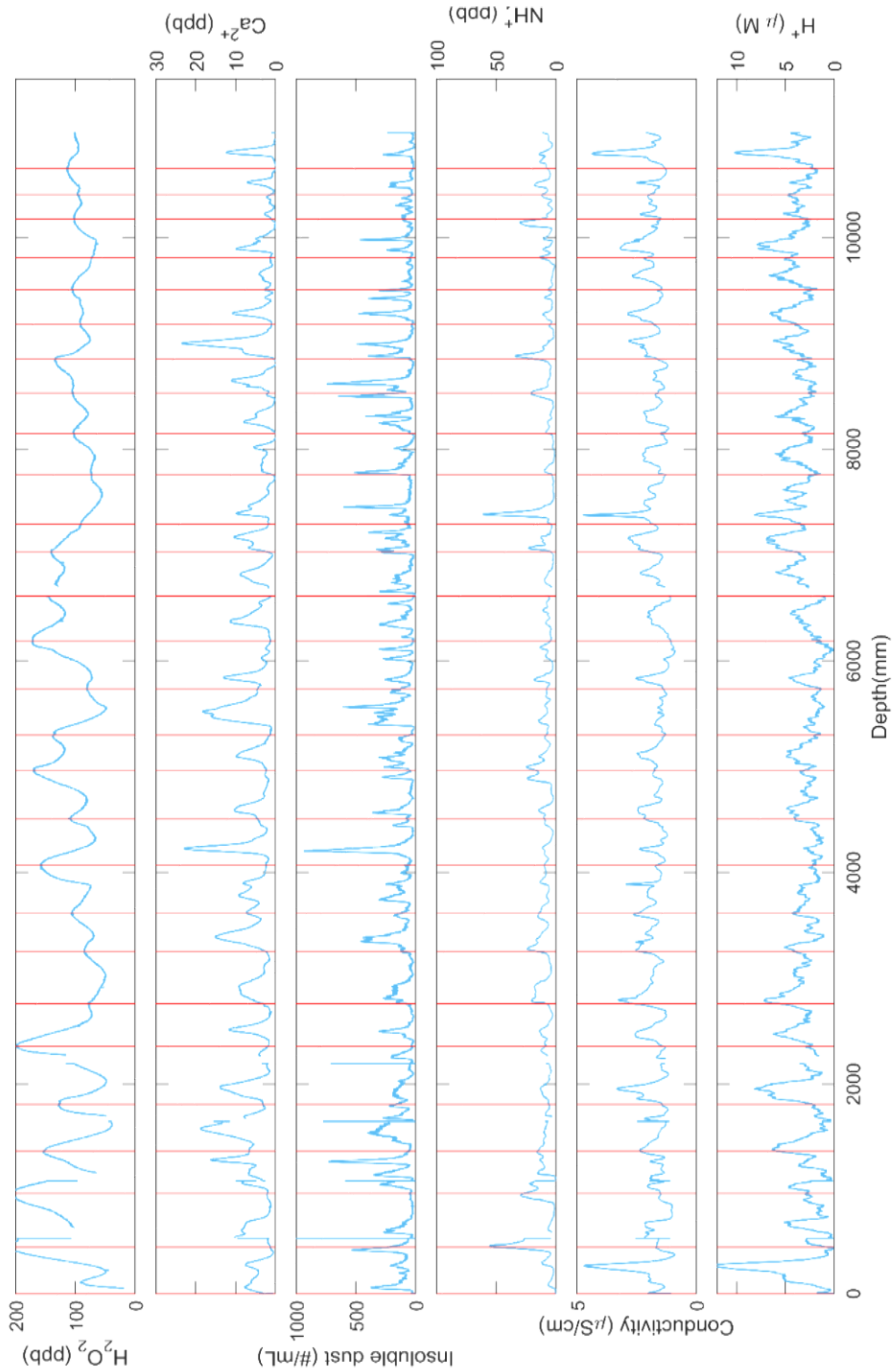

Figure S4: T2015-A3 data on a depth scale together with chosen annual layers (red vertical lines) used for generating the age scale.

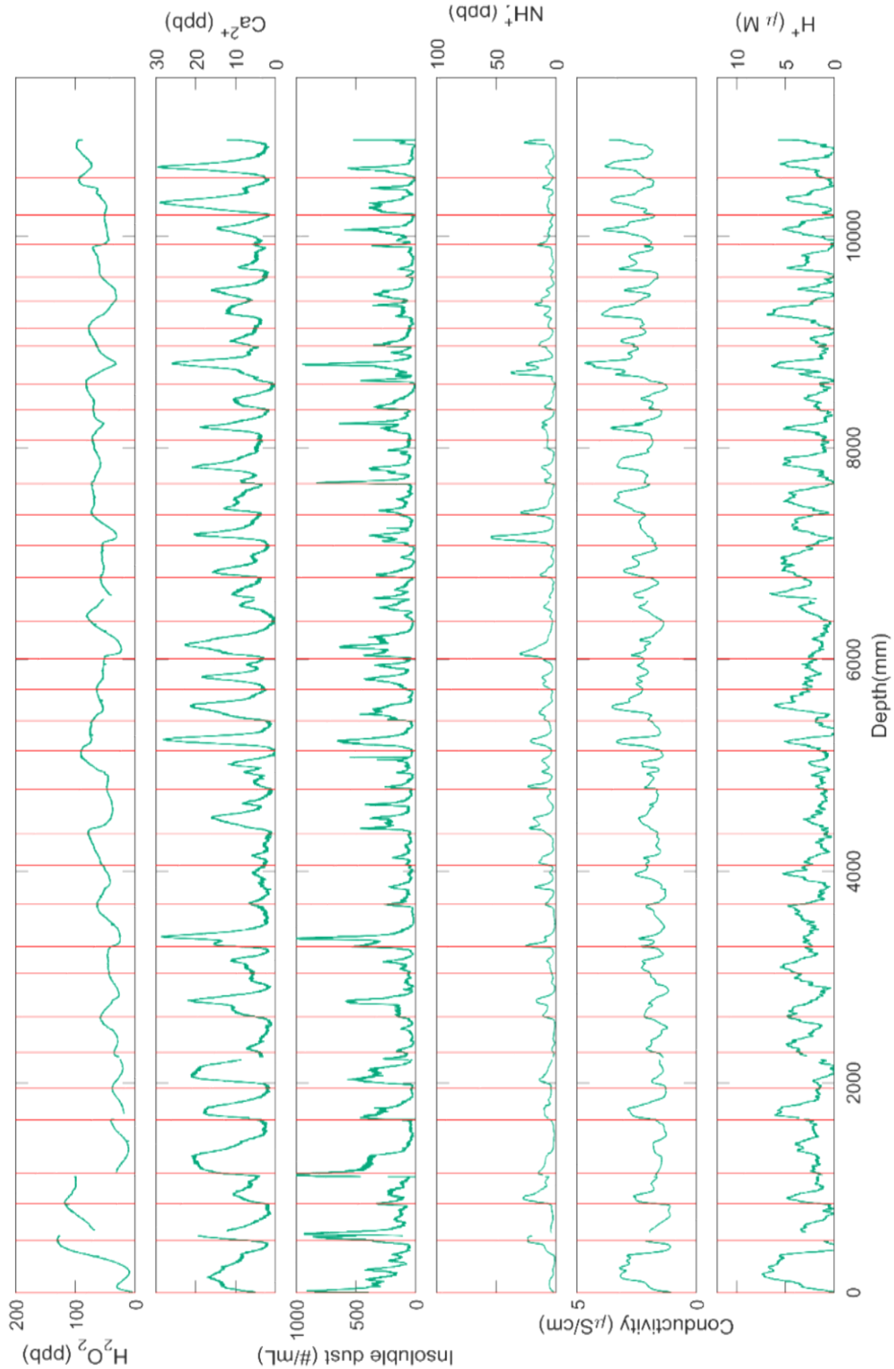

Figure S5: T2015-A4 data on a depth scale together with chosen annual layers (red vertical lines) used for generating the age scale.

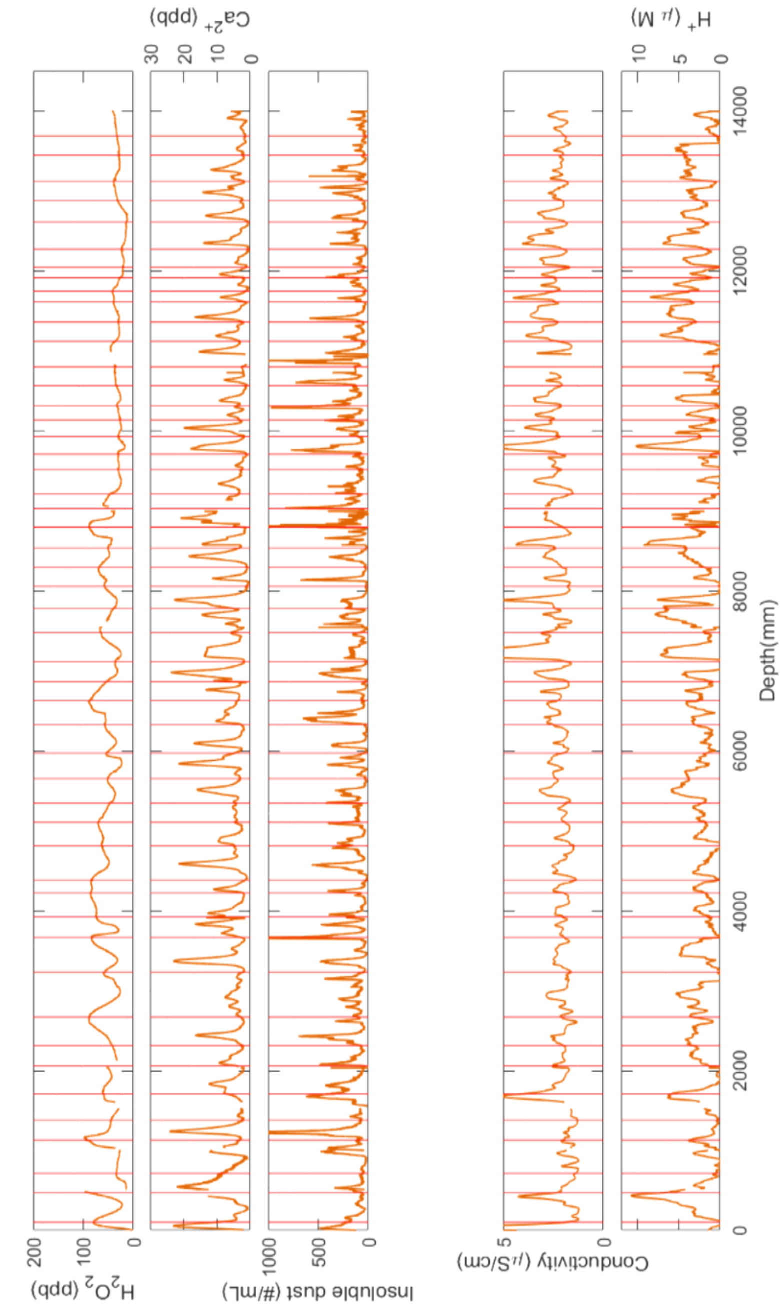

Figure S6: T2015-A5 data on a depth scale together with chosen annual layers (red vertical lines) used for generating the age scale. Note for this core ammonium was not determined.

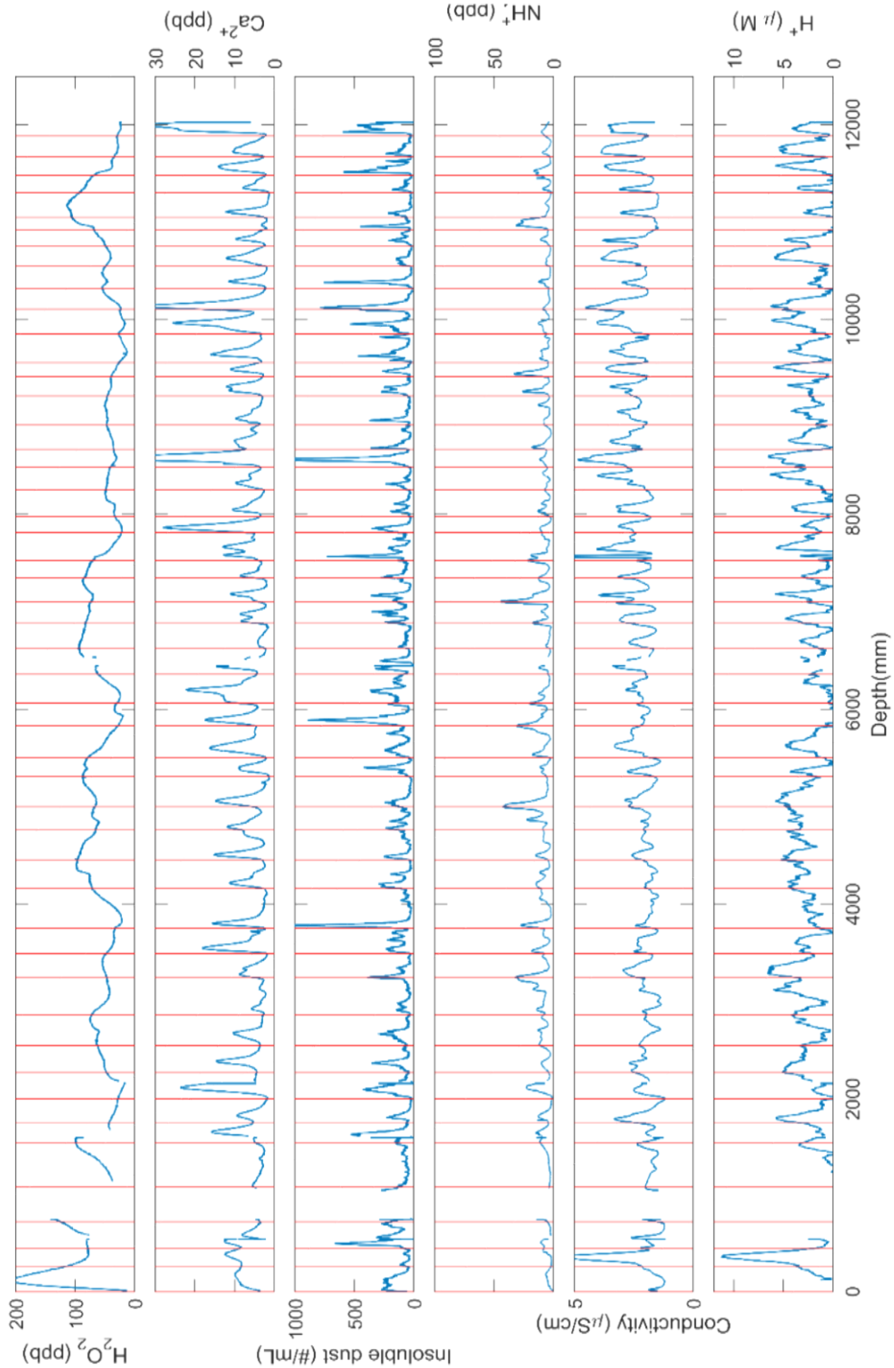

Figure S7: T2015-A6 data on a depth scale together with chosen annual layers (red vertical lines) used for generating the age scale.

	Full period						2000 onwards					
	T2015-A1	T2015-A2	T2015-A3	T2015-A4	T2015-A5	T2015-A6	T2015-A1	T2015-A2	T2015-A3	T2015-A4	T2015-A5	T2015-A6
Dust (Particles >1	*33.0*	*32.7*	*34.0*	*36.6*	*41.0*	*35.1*	*33.5*	*39.6*	*37.4*	*38.2*	*51.5*	*38.2*
μm /mL)	**89.9**	**86.4**	**79.2**	**99.4**	**95.9**	**78.2**	**95.6**	**91.5**	**97.5**	**103.8**	**99.3**	**79.2**
	224.3	*184.3*	*191.5*	*263.2*	*240.1*	*194.3*	*237.0*	*190.2*	*200.2*	*307.6*	*278.2*	*195.7*
Ca^{2+}	*2.2*	*1.9*	*1.3*	*2.7*	*2.2*	*2.9*	*2.3*	*1.3*	*2.0*	*2.3*	*2.5*	*2.9*
(ppb)	**4.3**	**4.3**	**3.4**	**6.0**	**4.2**	**5.6**	**4.4**	**3.6**	**4.6**	**5.3**	**4.8**	**5.3**
	8.7	*9.5*	*8.2*	*13.4*	*9.5*	*11.1*	*8.6*	*8.0*	*9.0*	*14.0*	*9.7*	*9.9*
H_2O_2	*67*	*76*	*72*	*34*	*27*	*29*	*66*	*77*	*69*	*27*	*32*	*35*
(ppb)	**108**	**100**	**96**	**56**	**38**	**49**	**112**	**100**	**103**	**46**	**56**	**55**
	148	*129*	*132*	*75*	*66*	*84*	*152*	*131*	*150*	*78*	*81*	*96*
NH_4^+	*2.2*	*5.6*	*3.3*	*2.5*		*3.2*	*2.2*	*4.7*	*4.3*	*2.3*		*2.8*
(ppb)	**5.8**	**11.6**	**7.0**	**5.1**		**6.0**	**5.5**	**11.8**	**8.0**	**5.2**		**6.3**
	10.8	*25.7*	*12.6*	*10.6*		*11.2*	*11.0*	*24.10*	*13.4*	*11.8*		*11.1*
H^+	*3.15*	*0.52*	*1.88*	*0.86*	*0.74*	*0.53*	*3.59*	*1.01*	*1.54*	*1.01*	*0.47*	*0.97*
(μM)	**5.67**	**2.78**	**3.24**	**2.10**	**2.40**	**2.34**	**5.99**	**3.21**	**2.66**	**2.08**	**1.96**	**2.73**
	8.32	*4.43*	*4.78*	*4.14*	*4.43*	*4.23*	*8.63*	*4.76*	*4.31*	*4.11*	*3.31*	*4.46*
Conductivity	*1.27*	*1.28*	*1.41*	*1.56*	*1.79*	*1.70*	*1.27*	*1.25*	*1.34*	*1.41*	*1.78*	*1.52*
(μS/cm)	**1.65**	**1.68**	**1.73**	**2.02**	**2.27**	**2.17**	**1.66**	**1.60**	**1.65**	**1.76**	**1.94**	**1.94**
	1.99	*2.18*	*2.22*	*2.75*	*2.91*	*3.05*	*1.99*	*2.01*	*2.10*	*2.22*	*2.35*	*2.35*

Table S4: Concentrations of impurities. 15%, median (50%) and 85% quantiles based on the monthly mean records for each of the compounds analysed is shown for each of the 6 traverse cores. 15% and 85% shown in italic. Left for the full temporal record and right for the period 2000 AD and onwards to make comparable estimates between sites. See also Figure 3 for a whiskers plot of the data.

S3 -Tables of correlations

Below tables of Pearson correlations between the records are presented. The correlations are made on both annual means and monthly mean records. The maximum available overlap between years have been used.

NH_4^+ (Month)	T2015-A2		T2015-A3		T2015-A4		T2015-A5		T2015-A6	
	R	p	R	p	R	p	R	p	R	p
T2015-A1	0.61	0.000	0.42	0.000	0.47	0.000			0.36	0.000
T2015-A2			0.38	0.000	0.24	0.000			0.50	0.000
T2015-A3					0.28	0.000			0.23	0.000
T2015-A4									0.23	0.000

NH_4^+ (Annual)	T2015-A2		T2015-A3		*T2015-A4*		T2015-A5		T2015-A6	
	R	p	R	*p*	*R*	p	R	p	R	p
T2015-A1	0.46	0.050	0.61	0.005	0.64	0.003			0.29	0.236
T2015-A2			0.42	0.027	0.29	0.132			0.68	0.000
T2015-A3					0.32	0.092			0.32	0.090
T2015-A4									0.49	0.003

H_2O_2 (Month)	T2015-A2		T2015-A3		T2015-A4		T2015-A5		T2015-A6	
	R	p	R	p	R	p	R	p	R	p
T2015-A1	0.63	0.000	0.75	0.000	0.48	0.000	NaN	NaN	0.26	0.000
T2015-A2			0.56	0.000	0.34	0.000	NaN	NaN	0.24	0.000
T2015-A3					0.51	0.000	NaN	NaN	0.21	0.000
T2015-A4							NaN	NaN	0.22	0.000
T2015-A5										

H_2O_2 (Annual)	T2015-A2		T2015-A3		*T2015-A4*		T2015-A5		T2015-A6	
	R	p	R	*p*	*R*	p	R	p	R	p
T2015-A1	0.44	0.058	0.80	0.000	0.62	0.005	NaN	NaN	0.19	0.427
T2015-A2			0.49	0.009	0.39	0.040	NaN	NaN	0.26	0.188
T2015-A3					0.54	0.003	NaN	NaN	0.21	0.273
T2015-A4							NaN	NaN	0.13	0.448
T2015-A5										

Ca_2^+ (Month)	T2015 -A2		T2015 -A3		T2015 -A4		T2015 -A5		T2015 -A6	
	R	p	R	p	R	p	R	p	R	p
T2015-A1	0.49	0.000	0.56	0.000	0.49	0.000	0.16	0.018	0.27	0.000
T2015-A2			0.35	0.000	0.37	0.000	0.32	0.000	0.16	0.004
T2015-A3					0.36	0.000	0.37	0.000	0.44	0.000
T2015-A4							0.24	0.000	0.34	0.000
T2015-A5									0.18	0.000

Ca_2^+ (Annual)	T2015 -A2		T2015 -A3		*T2015 -A4*		T2015 -A5		T2015 -A6	
	R	p	R	*p*	*R*	p	R	p	R	p
T2015-A1	0.23	0.351	0.38	0.104	0.50	0.030	-0.27	0.261	0.34	0.160
T2015-A2			0.06	0.763	0.28	0.145	-0.16	0.421	-0.12	0.534
T2015-A3					0.28	0.143	0.03	0.880	-0.09	0.625
T2015-A4							-0.19	0.288	0.39	0.024
T2015-A5									-0.05	0.742

Dust (Month)	T2015 -A2		T2015 -A3		T2015 -A4		T2015 -A5		T2015 -A6	
	R	p	R	p	R	p	R	p	R	p
T2015-A1	0.33	0.000	0.19	0.006	0.21	0.002	0.09	0.170	0.15	0.027
T2015-A2			0.18	0.001	0.12	0.030	0.12	0.032	0.02	0.783
T2015-A3					0.12	0.027	0.25	0.000	0.30	0.000
T2015-A4							0.10	0.049	0.14	0.007
T2015-A5									0.07	0.080

Dust (Annual)	T2015 -A2		T2015 -A3		*T2015 -A4*		T2015 -A5		T2015 -A6	
	R	p	R	*p*	*R*	p	R	p	R	p
T2015-A1	0.62	0.004	0.09	0.729	0.57	0.011	0.02	0.919	0.67	0.002
T2015-A2			-0.03	0.864	0.23	0.233	0.05	0.810	-0.03	0.899
T2015-A3					0.38	0.043	0.10	0.618	0.22	0.261
T2015-A4							0.17	0.322	0.22	0.205
T2015-A5									-0.10	0.511

H^{+}_{dye} **(Month)**	T2015-A2		T2015-A3		T2015-A4		T2015-A5		T2015-A6	
	R	p	R	p	R	p	R	p	R	p
T2015-A1	0.10	0.138	0.11	0.120	0.14	0.043	-0.22	0.001	-0.06	0.447
T2015-A2			0.12	0.035	0.23	0.000	-0.06	0.300	0.09	0.121
T2015-A3					0.35	0.000	0.24	0.000	0.02	0.665
T2015-A4							0.11	0.039	0.07	0.176
T2015-A5									0.07	0.118

H^{+}_{dye} **(Annual)**	T2015-A2		T2015-A3		*T2015-A4*		T2015-A5		T2015-A6	
	R	p	R	*p*	*R*	p	R	p	R	p
T2015-A1	0.05	0.823	-0.32	0.184	0.05	0.829	-0.36	0.133	-0.04	0.884
T2015-A2			-0.22	0.262	0.27	0.162	-0.41	0.029	0.19	0.352
T2015-A3					0.05	0.790	0.43	0.019	-0.05	0.785
T2015-A4							-0.14	0.435	-0.18	0.306
T2015-A5									0.00	0.993

Cond (Month)	T2015-A2		T2015-A3		T2015-A4		T2015-A5		T2015-A6	
	R	p	R	p	R	p	R	p	R	p
T2015-A1	0.30	0.000	0.51	0.000	0.20	0.003	0.05	0.444	-0.11	0.133
T2015-A2			0.41	0.000	0.34	0.000	0.16	0.004	0.02	0.785
T2015-A3					0.34	0.000	0.23	0.000	0.18	0.001
T2015-A4							0.26	0.000	0.27	0.000
T2015-A5									0.29	0.000

Cond (Annual)	T2015-A2		T2015-A3		*T2015-A4*		T2015-A5		T2015-A6	
	R	p	R	*p*	*R*	p	R	p	R	p
T2015-A1	0.28	0.244	0.50	0.028	-0.18	0.472	0.08	0.749	0.26	0.275
T2015-A2			0.39	0.040	0.15	0.432	-0.03	0.884	0.03	0.875
T2015-A3					0.16	0.406	0.41	0.026	0.23	0.228
T2015-A4							0.18	0.300	0.41	0.015
T2015-A5									0.35	0.014

S4-Additional analysis of the data

Conductivity in the 6 shallow cores are presented in Figure S8.

Table S5 presents the correlation of the excess ammonium between the 5 sites for which ammonium was analyzed.

In Figure S6 the particle dust fluxes are presented with time for small particles 1.25-2.9 µm, medium particles 2.9-8.13 µm and large particles 8.13-10.5 µm and Table S7 presents a similar analysis as that of Table 2, but for the longer period 1988-2015 covered by cores T2015A2 through T2015-A6).

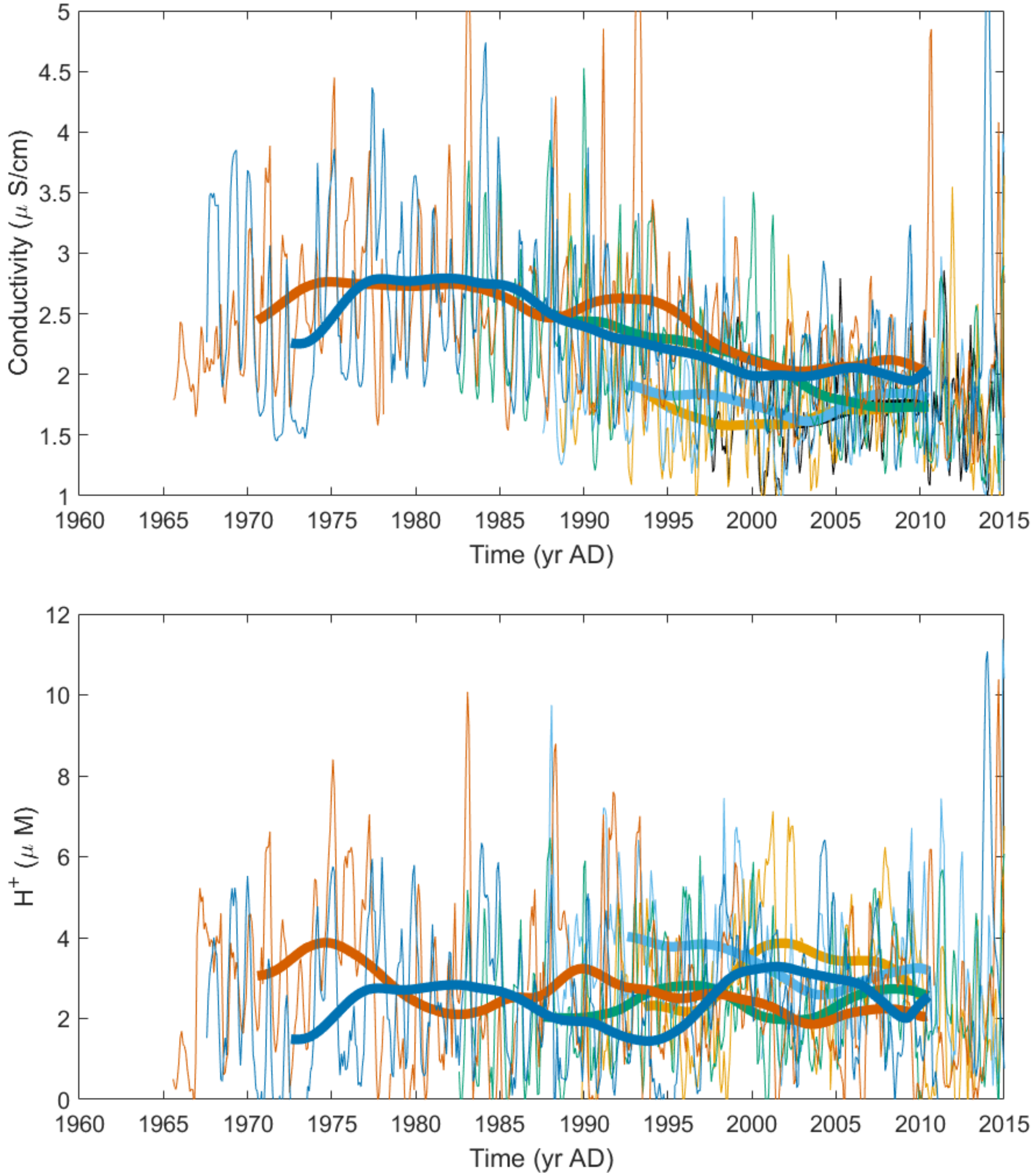

Figure S8: Top) Conductivity and Bottom) Acidity in the T2015-A2(yellow), T2015-A3 (light blue), T2015-A4 (green), T2015-A5 (orange) and T2015-A6 (darker blue) shown in monthly resolution (thin lines) and in a 5 year running average (thick lines). Note the data for A1 was not in sufficient quality for the acidity.

Table S5: Pearson correlations for excess ammonium between the 5 shallow traverse cores. Excess ammonium is defined as excess compared to a 5 year running average. In bold if $p<0.01$. Bold and italic if $p<0.001$.

R^2	T2015-A1	T2015-A2	T2015-A3	T2015-A4	T2015-A6
T2015-A1	***1.00***	***0.79***	***0.82***	***0.71***	***0.69***
T2015-A2		***1.00***	**0.56**	0.41	***0.64***
T2015-A3			***1.00***	**0.54**	**0.56**
T2015-A4				***1.00***	***0.56***
T2015-A6					***1.00***

Table S6: Dust fluxes and trends in dust fluxes for the period 1988-2015 for each of the 6 shallow firn cores which cover the full period. Small (1.25-2.9 µm), intermediate (2.9-8.13 µm) and large (8.13-10.5 µm) refers to the dust sizes as analyzed by the Abakus instrument. P values given in parenthesis, when significant $p<0.1$ in bold and when $p<0.01$ in bold and italic. Note sample size only N=18.

	Dust Flux (1988-2015) [mg/kg/yr]				**Dust flux trend 1988-2015** [mg/kg/yr2] (p-value)			
Firn core	**Total**	**Small**	**Intermed**	**Large**	**Total**	**Small**	**Intermed**	**Large**
T2015-A2	35.50	6.33	16.48	3.37	**0.71** **(0.03)**	0.02 (0.84)	0.22 (0.20)	***0.12*** ***(0.00)***
T2015-A3	17.60	4.61	8.91	0.9	***0.40*** ***(0.00)***	**0.08** **(0.04)**	***0.27 (0.00)***	***0.03*** ***(0.00)***
T2015-A4	50.03	8.54	22.20	5.34	0.72 (0.29)	0.19 (0.13)	0.51 (0.12)	0.02 (0.79)
T2015-A5	50.01	8.40	21.76	5.57	**1.21** **(0.02)**	0.06 (0.54)	**0.48 (0.02)**	***0.21*** ***(0.00)***
T2015-A6	17.09	4.87	8.65	0.52	-0.19 (0.30)	-0.04 (0.46)	-0.08 (0.45)	**-0.02** **(0.09)**

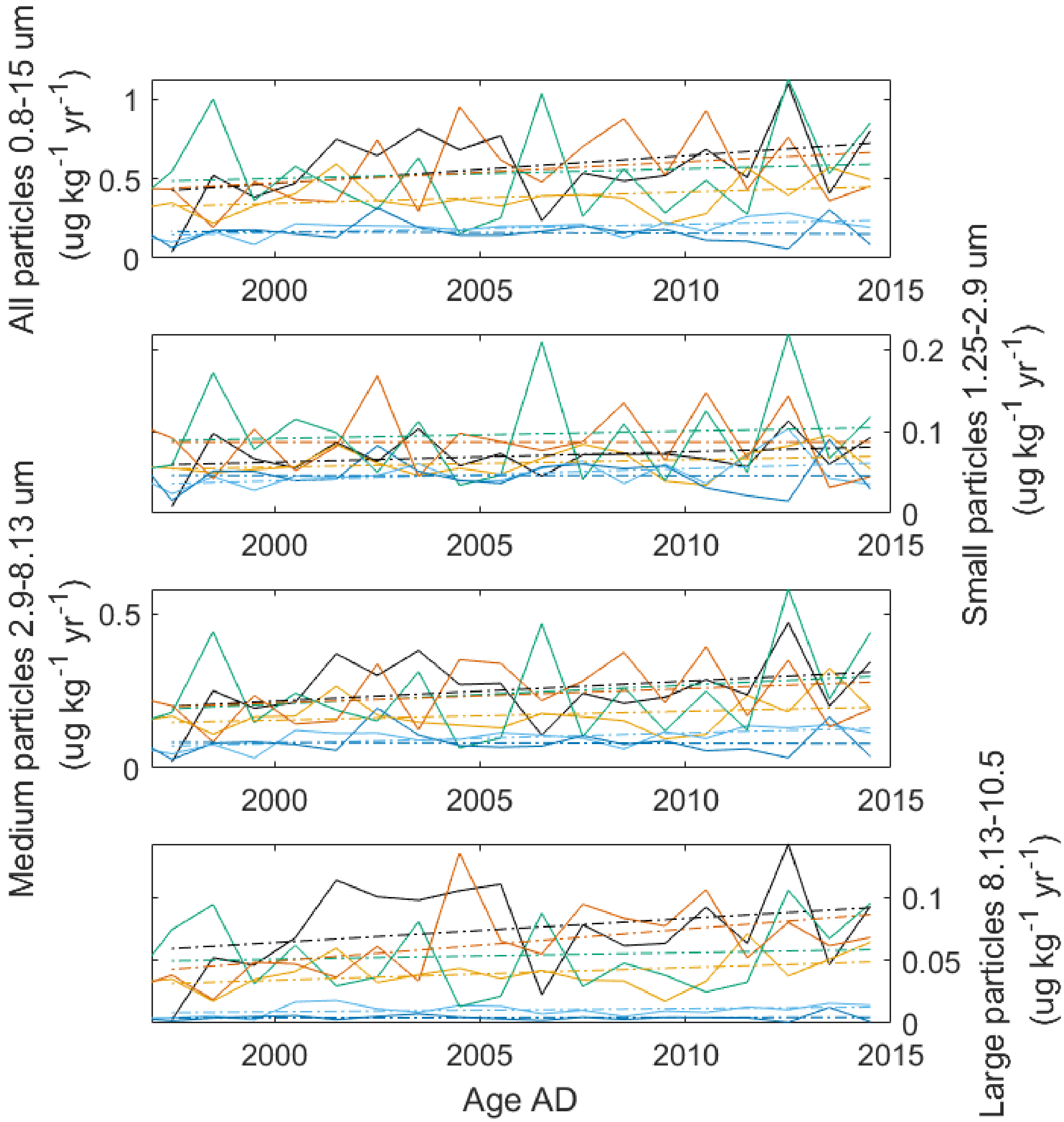

Figure S9: Insoluble dust particle fluxes (full lines) determined from the Abakus instrument by assumptions of perfect spherical particles and a weight of 2400 kg/m^3. In dashed is shown the trends since 1998. From the top total 0.8-15 um, Small particles 1.25-2.9 um, Medium particles 2.9-8.13 um and Large particles 8.13-10.5. The colors reflect the firn cores T2015-A2 (yellow), T2015-A3 (light blue), T2015-A4 (green), T2015-A5 (orange) and T2015-A6 (darker blue).

Table S7: Dust flux trends in percentage for the period 1998-2015 (left) and 1988-2015 (right) for each of the 6 shallow firn cores. Small (1.25-2.9 µm), intermediate (2.9-8.13 µm) and large (8.13-10.5 µm) refers to the dust sizes as analyzed by the Abakus instrument. P values p<0.1 in bold and when p<0.01 in bold and italic. Note sample size only N=18 and 28 respectively.

	Dust Flux trend 1998-2015 [%]				**Dust flux trend 1988-2015** [%]			
Firn core	**Total**	**Small**	**Intermed**	**Large**	**Total**	**Small**	**Intermed**	**Large**
T2015-A1	**3.03**	1.81	2.53	2.53	**-**	-	-	-
T2015-A2	**1.86**	1.44	1.66	**2.52**	**2.02**	0.24	1.34	***3.65***
T2015-A3	***2.76***	***3.03***	***3.37***	2.48	***2.29***	**1.82**	***3.05***	***3.64***
T2015-A4	1.13	0.94	2.52	0.98	1.45	2.25	2.32	0.38
T2015-A5	2.38	0.01	1.95	***3.96***	**2.42**	0.67	**2.23**	***3.65***
T2015-A6	-0.55	-0.05	-0.27	0.59	-1.11	-0.82	-0.96	**-2.90**