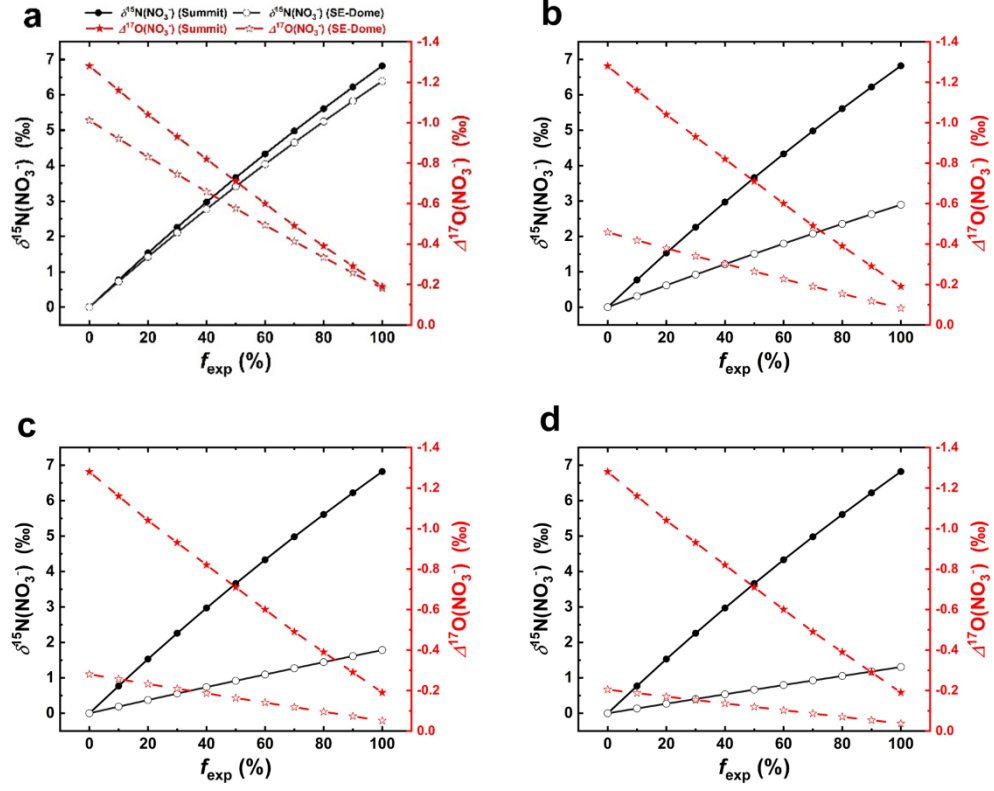
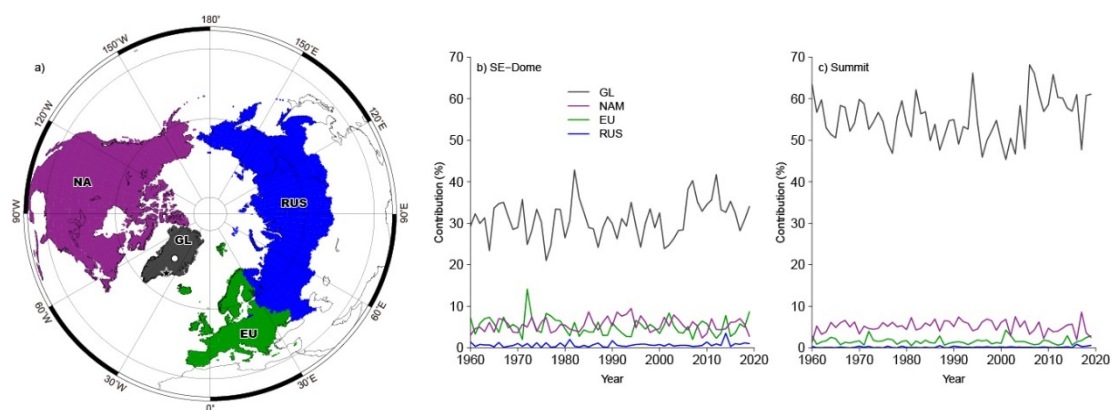


**Figure S1.** The seasonal change of observed  $\text{NO}_3^-$  isotopes and concentration SE-Dome ice core. (a)  $\delta^{15}\text{N}(\text{NO}_3^-)$  (1959-2014), (b)  $\delta^{15}\text{N}(\text{NO}_3^-)$  (1959-1974), (c)  $\delta^{15}\text{N}(\text{NO}_3^-)$  (1975-1980 & 1995-2014), (d)  $\delta^{15}\text{N}(\text{NO}_3^-)$  (1981-1993), (e)  $\Delta^{17}\text{O}(\text{NO}_3^-)$  (1959-2014), (f)  $\Delta^{17}\text{O}(\text{NO}_3^-)$  (1959-1980 & 1995-2014), (g)  $\Delta^{17}\text{O}(\text{NO}_3^-)$  (1981-1994) and (h) the  $\text{NO}_3^-$  flux ( $\text{mmol m}^{-2} \text{yr}^{-1}$ ).



**Figure S2.** The sensitivity of the changes in  $\delta^{15}\text{N}(\text{NO}_3^-)$  and  $\Delta^{17}\text{O}(\text{NO}_3^-)$  of the ice-core nitrate to  $f_{\text{exp}}$  with different snow accumulation rate ( $A$ ). (a)  $A = 0.25 \text{ m w e a}^{-1}$  which equals to that in Summit; (b)  $A = 0.6 \text{ m w e a}^{-1}$ ; (c)  $A = 1.01 \text{ m w e a}^{-1}$ ; (d)  $A = 1.4 \text{ m w e a}^{-1}$ .



**Figure S3.** Time series of regional contributions of air mass origins for SE-Dome (b) and Summit 39 (c). The regions are shown at the top. Greenland in gray, North America in purple (Canada and the 40 U.S.), Europe (EU) in green, Russia in blue.

**Table S1** The samples and corresponding years

Year	Seasonal	$\delta^{18}\text{O}$	$\Delta^{17}\text{O}$	$\delta^{15}\text{N}$	Year	Seasonal	$\delta^{18}\text{O}$	$\Delta^{17}\text{O}$	$\delta^{15}\text{N}$
1959	summer	79	27.4	0.5	1988	spring	88.5	32.8	-3.4
1959	winter	76.6	26.5	-1.2	1988	summer	86.7	33.4	-6.2
1960	summer	83.1	30	-4.3	1988	autumn	85.5	34	-7.9
1961	winter	76.5	27.2	-1.6	1988	winter	87.5	33.9	-6.1
1961	summer	80.1	27.7	-0.5	1989	spring	80.9	29	-2.4
1962	winter	78.1	27.5	-0.5	1989	summer	86.8	34.5	-8.6
1962	summer	77.7	28.4	1.4	1989	autumn	80.9	30.3	-1.3
1963	winter	87.1	33.9	-3.4	1989	winter	83.9	31.9	-5.3
1963	summer	76.6	27.8	2	1990	spring	74	26.2	-3.1
1964	winter	85.9	33.4	-2	1990	summer	77.8	30	-8.3
1964	summer	76.5	26.7	0.1	1990	autumn	86	32.6	-9.7
1965	winter	79.8	31.5	-4.4	1991	spring	74.7	28	-1.6
1965	summer	80.7	30.2	2.5	1991	summer	70.4	25.9	-0.5
1966	winter	77	30.7	-6	1991	autumn	84.3	32.5	-9.1
1966	summer	73	26.9	-0.3	1991	winter	83.7	32.8	-6.4
1967	winter	80.3	31.1	-7.4	1992	spring	79.3	29.5	4.8
1967	summer	73.7	26.6	-1.4	1992	summer	78.6	28.1	-6.3
1968	winter	79.8	30.4	-4.1	1992	autumn	82.8	32	-7.9
1968	summer	76	27.4	-1.8	1993	summer	87	34.2	-8.6
1969	winter	80.9	30.9	-5.7	1993	autumn	85.6	34.5	-9.8
1969	summer	73	26.4	-0.3	1993	winter	84.7	32.6	-10
1970	winter	83.9	32	-4.3	1994	spring	76	27.8	-0.9
1970	summer	75.8	27.2	-1.8	1994	summer	78.9	28.2	-4
1971	winter	82.3	32.1	-7	1994	winter	82.4	29.5	-4.4
1971	summer	75.8	27.4	0.8	1994	autumn	83.4	31.2	-9.1
1972	winter	82.3	32.7	-9	1995	summer	83.1	31.7	-7.6
1972	summer	74.3	26.6	-7.2	1996	winter	78.1	29.4	-5.7
1973	winter	78.9	30.4	-6.2	1996	summer	77.3	28.1	-1.9
1973	summer	75.7	27.6	-4.1	1997	winter	82.7	32.7	-9.2
1974	winter	79.2	30.4	-7.1	1997	summer	78	28.2	-0.5
1974	summer	75.6	27.7	-5.8	1998	winter	82.3	32.2	-9.7
1975	winter	79.7	31.8	-12.4	1998	summer	77.2	27.6	-1.6
1975	summer	80.6	28.9	-0.3	1999	winter	83.1	31.6	-4.8
1976	winter	80.9	30.4	-8.3	1999	summer	78.4	28.7	-1.2
1976	summer	75.4	26.9	-3	2000	winter	83	32.3	-8.6
1977	winter	81.5	31.4	-12.8	2000	summer	74.9	27.6	-3
1977	summer	77.8	28.2	-3.6	2001	winter	83.7	32.9	-8.4
1978	winter	87.2	32.5	-13.3	2001	summer	78.3	29.5	-3.8
1978	summer	76.1	27.9	-2	2002	winter	81.1	31.1	-9
1979	winter	85.5	33	-9.2	2002	summer	74.1	26.7	-4.2
1979	summer	76.6	27.8	-4.7	2003	winter	80.9	31.8	-9.7
1980	winter	84.1	32.9	-8.6	2003	summer	76.9	28.4	-3.2

Year	Seasonal	$\delta^{18}\text{O}$	$\Delta^{17}\text{O}$	$\delta^{15}\text{N}$	Year	Seasonal	$\delta^{18}\text{O}$	$\Delta^{17}\text{O}$	$\delta^{15}\text{N}$
1980	summer	74.6	26.7	-3.7	2004	winter	81.7	31.4	-6
1981	winter	80.7	31	-7.2	2004	summer	76.6	27.8	-2.2
1981	spring	77.5	28.8	-4.6	2005	winter	82.7	31.3	-5.4
1981	summer	75.1	27.3	-2.4	2005	summer	81.7	30.9	-5.2
1981	autumn	84.7	33.9	-12.6	2006	winter	80.2	30.1	-2.2
1981	winter	74.6	26.8	-2.1	2006	summer	76.9	27.9	-1.9
1982	Spring	76.1	28.2	-4.4	2007	winter	82.2	32.1	-6.8
1982	summer	81.3	31.9	-7.5	2007	summer	75.7	28.8	-3.5
1982	autumn	83.2	32.1	-9.5	2008	winter	82.6	33.5	-9.1
1982	winter	72.3	26.8	-4.6	2008	summer	77.1	28.8	-3.7
1983	Spring	72.1	26.8	-3.2	2009	winter	78.1	31.6	-9
1983	summer	80.1	28.9	-2.3	2009	summer	71.7	27.3	-3.6
1983	autumn	87	33.6	-6.1	2010	winter	80.9	32.8	-7.3
1983	winter	86.4	34.1	-10.3	2010	summer	74.7	28.1	-4
1984	Spring	76.3	27.8	-6.8	2011	winter	81.9	32.5	-5
1984	Summer	73.7	27.6	-3.9	2011	summer	70.2	26.2	-2.4
1984	autumn	83.1	31.5	-3.3	2012	winter	82.8	33.6	-7.7
1984	winter	82.7	32.3	-9.4	2012	summer	73.6	27.2	-1.4
1985	Spring	71.5	27.6	-5.5	2013	winter	75.1	29.8	-8.4
1985	summer	73.7	26.7	-2	2013	summer	73.7	26.8	-1.6
1985	autumn	82.8	31.8	-6.2	2014	winter	65.1	24.6	-6.3
1985	winter	86.2	34.4	-7.4	2014	summer	67	24.2	-3.9
1986	Spring	70	26.2	-5.3	2015	winter	82.1	32.4	-7.5
1986	summer	83.7	31.4	-4.2					
1986	autumn	89.6	35	-11.4					
1987	spring	81.4	32.7	-13.5					
1987	summer	77.4	28.2	1.3					
1987	autumn	83.4	32.9	-11.9					
1987	winter	90.4	35.1	-10.1					
1988	spring	88.5	32.8	-3.4					
1988	summer	86.7	33.4	-6.2					
1988	autumn	85.5	34	-7.9					
1988	winter	87.5	33.9	-6.1					

**Table S2** Parameters used for calculating  $f_{\text{exp}}$  in Summit and SE-Dome ice core

Parameters	Parameters	Description	Value	Unit	Data origin
Summit	$L$	Summer boundary layer height	3.50E+05	m	Honrath et al., 2002
	$V_d$	The dry deposition velocity of $\text{HNO}_3$	0.0063	$\text{m s}^{-1}$	Björkman et al., 2013
	$k[\text{OH}]$	The rate constant for the $\text{NO}_2 + \text{OH}$ reaction	1.09373E-11		Atkinson et al., 2004
	$V_h$	Mean horizontal wind speed	5	$\text{m s}^{-1}$	Mcdowell et al., 2020
	$[\text{OH}]$	Measured average OH radical concentration in summer	6.30E+06	$\text{mloec cm}^{-3}$	Sjostedt et al., 2006
	$H$	Horizontal characteristic	156	m	Characteristic length of summit of the Greenland ice cap; Honrath et al., 2002
SE-Dome	$L$	Summer boundary layer height	Same as Summit	m	-
	$V_d$	The dry deposition velocity of $\text{HNO}_3$	Same as Summit	$\text{m s}^{-1}$	
	$k[\text{OH}]$	The rate constant for the $\text{NO}_2 + \text{OH}$ reaction	1.13E-11		ERA5(Hersbach et al., (2020); Khalzan et al., (2022))
	$V_h$	Mean horizontal wind speed	5.2	$\text{m s}^{-1}$	
	$[\text{OH}]$	Measured average OH radical concentration in summer	1.02E+06	$\text{mloec cm}^{-3}$	
	$H$	Horizontal characteristic	235.31	m	

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