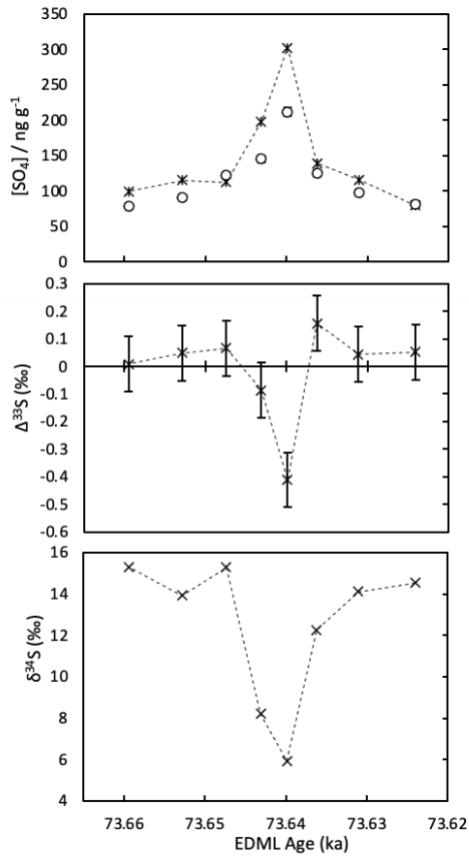
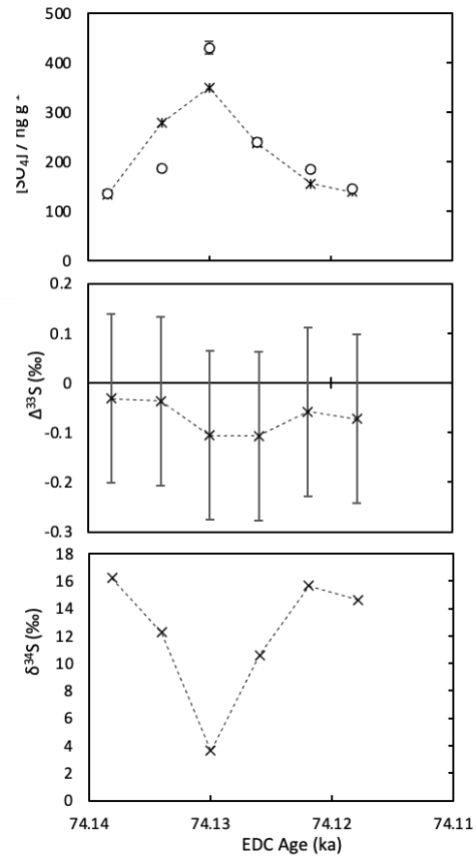


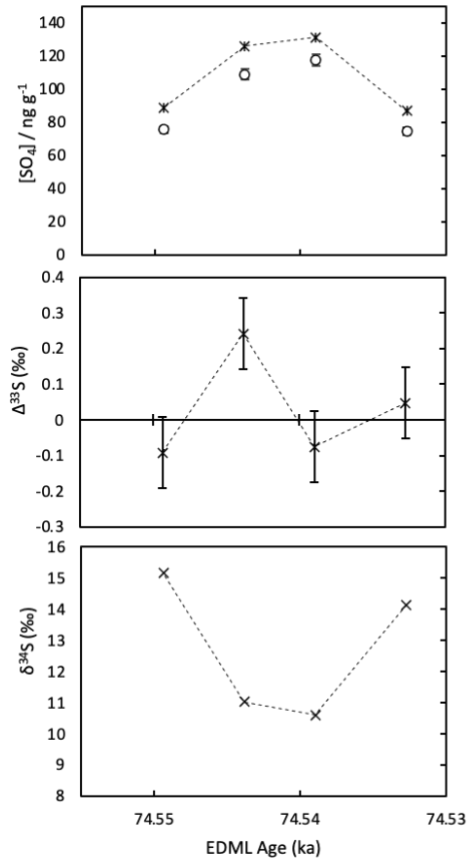
1. Pre-T1



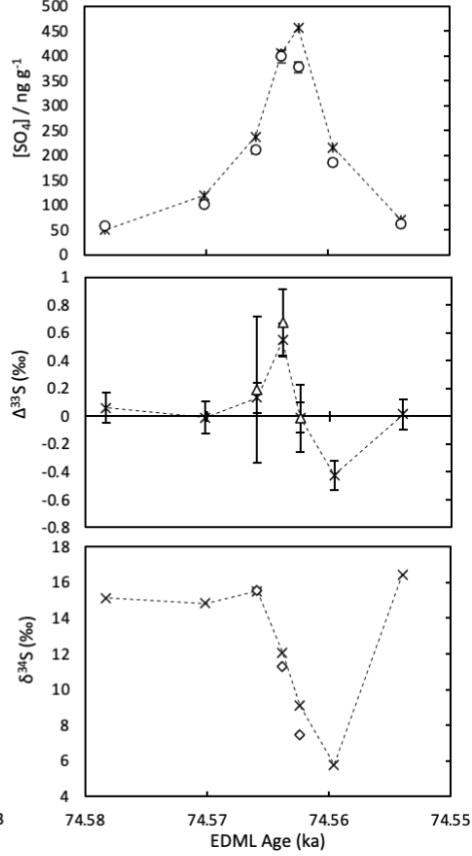
2. T4b



3. T5



4. T6



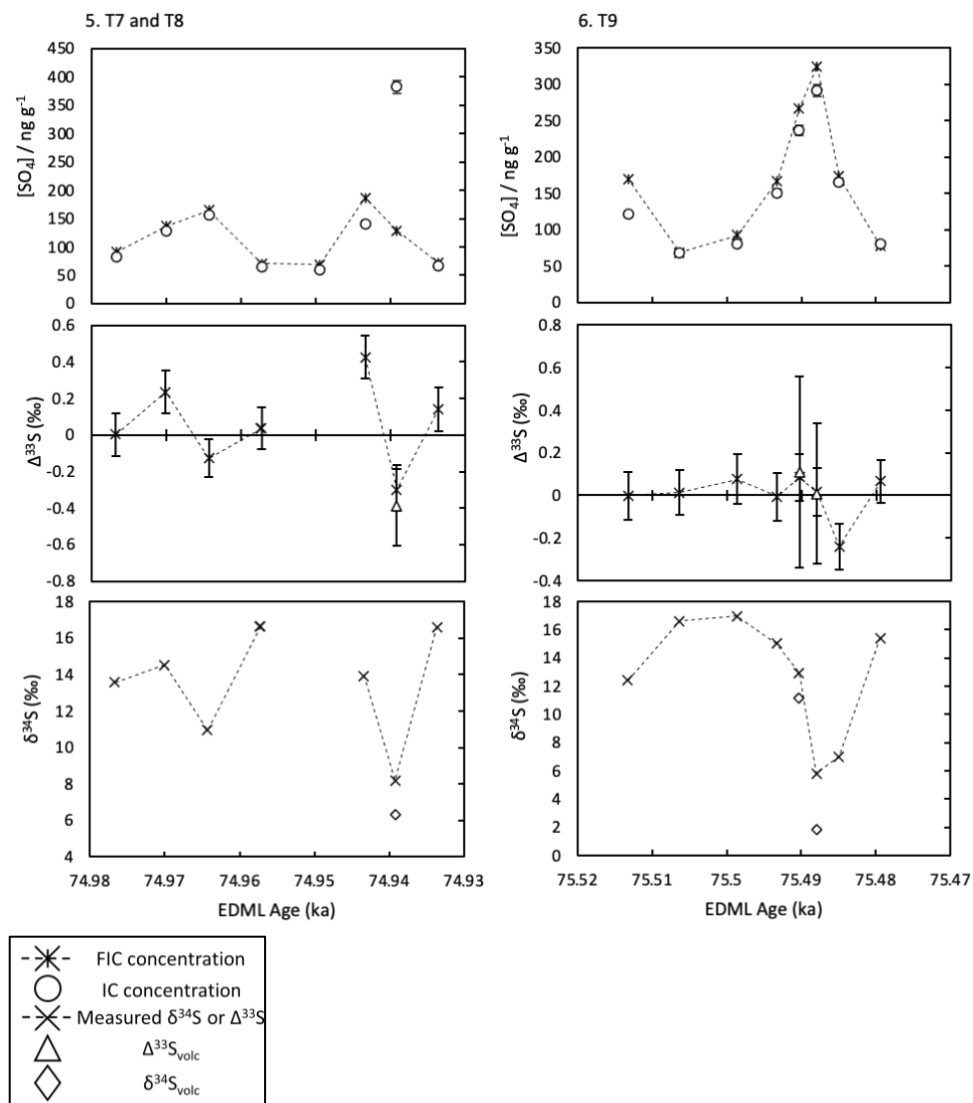


Figure S1–6. Sulfate concentration measurements and sulfur isotopic composition data for 7 of the Toba candidates. For sulfate concentration crosses represent fast ion chromatography measurements (FIC) and open symbols are ion chromatography (IC) measurements. Measured isotopic ratios are shown by crosses and open symbols are data points with $f_{\text{volc}} \geq 0.65$ and have been background corrected. All Error bars indicate 2σ for measured ratios and 1σ for background corrected ratios, where error bars are not visible the error is smaller than the symbol. All ages are on the AICC2012 age model (Veres et al., 2013).

Table S1. Summary of IC [S], total sulfate deposition, blank corrected $\Delta^{33}\text{S}_{\text{measured}}$ and $\Delta^{33}\text{S}_{\text{volc}}$ data for 14 peaks measured in this study. The error on IC sulfate concentration data is 5%. $\Delta^{33}\text{S}_{\text{meas}}$ is reported with 2σ errors and $\Delta^{33}\text{S}_{\text{volc}}$ with 1σ .

Core	Toba Candidate	Max [SO ₄] (ng g ⁻¹)	Total Deposition (mg m ⁻²)	Max +ve $\Delta^{33}\text{S}_{\text{meas}}$ (‰)	Max -ve $\Delta^{33}\text{S}_{\text{meas}}$ (‰)	Maximum $\Delta^{33}\text{S}_{\text{volc}}$ (‰)
EDML	Pre-T1	212	44.0	+0.16 (± 0.10)	-0.41 (± 0.10)	No samples with $f_{\text{volc}} \geq 0.65$
EDC	T1	751	54.3	-	-3.64 (± 0.17)	-4.75 (± 0.51)
EDML	T1	306	87.7	+0.98 (± 0.10)	-2.23 (± 0.10)	-3.08 (± 0.50)
EDC	T2	748	46.5	+0.57 (± 0.17)	-2.42 (± 0.17)	-3.41 (± 0.28)
EDML	T2	1161	424.3	+0.61 (± 0.31)	-1.40 (± 0.11)	-1.72 (± 0.24)
EDC	T3	1178	133.5	+0.42 (± 0.32)	-0.78 (± 0.44)	-0.93 (± 0.52)
EDML	T3	394	103.3	+0.95 (± 0.10)	-0.06 (± 0.10)	+0.99 (± 0.57) $f_{\text{volc}} = 0.63$
EDML	T4	990	239.5	+0.10 (± 0.12)	-0.40 (± 0.10)	-0.45 (± 0.15)
EDC	T4b	431	26.5	-	-0.11 (± 0.17)	-0.13 (± 0.13)
EDML	T5	117	32.7	+0.24 (± 0.10)	-0.08 (± 0.10)	No samples with $f_{\text{volc}} \geq 0.65$
EDML	T6	399	95.8	+0.55 (± 0.12)	-0.42 (± 0.11)	+0.67 (± 0.24)
EDML	T7	383	27.5	+0.43 (± 0.12)	-0.30 (± 0.11)	-0.39 (± 0.22)
EDML	T8	157	33.3	+0.23 (± 0.12)	-0.13 (± 0.12)	No samples with $f_{\text{volc}} \geq 0.65$
EDML	T9	291	86.1	+0.08 (± 0.11)	-0.24 (± 0.11)	All values within 2sd of 0

Table S2. Comparison of maximum positive and negative volcanic sulfur MIF signals in both this study and previous studies. See Fig. 1 for Toba candidate ages. A: Arctic, AN: Antarctic core. †These dates are taken from Sigl et al., (2015).

Event (if known)	Date C. E. (year)	Core (Location)	Maximum $\Delta^{33}\text{S}_{\text{volc}}$ (‰)		Study
			Positive	Negative	
Pinatubo	1991	South Pole (AN)	+0.67 (\pm 0.16)	-	Savarino et al., 2003
Samalas	1257†	South Pole (AN)	-	-0.5 (\pm 0.16)	
Pinatubo	1991	Dome C (AN)	+1.19 (\pm 0.12)	-1.03 (\pm 0.12)	Baroni et al., 2007
Agung	1963	Dome C (AN)	+1.41 (\pm 0.12)	-0.81 (\pm 0.12)	
Tambora	1815	South Pole (AN)	+0.20 (\pm 0.12)	-0.60 (\pm 0.12)	Cole-Dai et al., 2009
-	1458†	South Pole (AN)	+0.60 (\pm 0.12)	-0.55 (\pm 0.26)	
Samalas	1257†	Dome C (AN)	+2.22 (\pm 0.06)	-1.21 (\pm 0.28)	Gautier et al., 2018
Ruang	1597	Dome C (AN)	+2.29 (\pm 0.34)	-	Gautier et al., 2019
Tambora	1815	Tunu (A)	+2.16 (\pm 0.54)	-0.85 (\pm 0.33)	Burke et al., 2019
Samalas	1257	Tunu (A)	+2.79 (\pm 0.79)	-2.47 (\pm 0.58)	
Toba	T1	EDC (AN)	-	-4.75 (\pm 0.51)	This study
Toba	T1	EDML (AN)	+1.34 (\pm 0.36)	-3.08 (\pm 0.50)	
Toba	T2	EDC (AN)	-	-3.41 (\pm 0.28)	
Toba	T2	EDML (AN)	+0.74 (\pm 0.40)	-1.72 (\pm 0.24)	
Toba	T3	EDC (AN)	+0.62 (\pm 0.15)	-0.93 (\pm 0.52)	
Toba	T3	EDML (AN)	+0.99 (\pm 0.57)	-	

Table S3. Summary of the $\Delta^{33}\text{S}_{\text{volc}}$ vs $\delta^{34}\text{S}_{\text{volc}}$ York et al., (2004) regression data for this study including each core individually and eruptions where 3 or more data points are available with $f_{\text{volc}} \geq 0.65$.

	Slope	Intercept	R ²
All stratospheric data	0.106 \pm 0.003	-0.464 \pm 0.034	0.964
EDML	0.133 \pm 0.010	-0.634 \pm 0.065	0.863
EDC	0.104 \pm 0.003	-0.413 \pm 0.035	0.964
T1 EDC	0.082 \pm 0.009	-0.896 \pm 0.366	0.972
T2 EDML	0.137 \pm 0.015	-0.651 \pm 0.096	0.954
T2 EDC	0.102 \pm 0.004	-0.813 \pm 0.055	0.975
T3 EDC	0.092 \pm 0.056	-0.343 \pm 0.056	0.936
T6 EDML	0.081 \pm 0.063	-0.485 \pm 0.647	-0.659
T4 EDML	0.037 \pm 0.018	-0.307 \pm 0.099	0.183

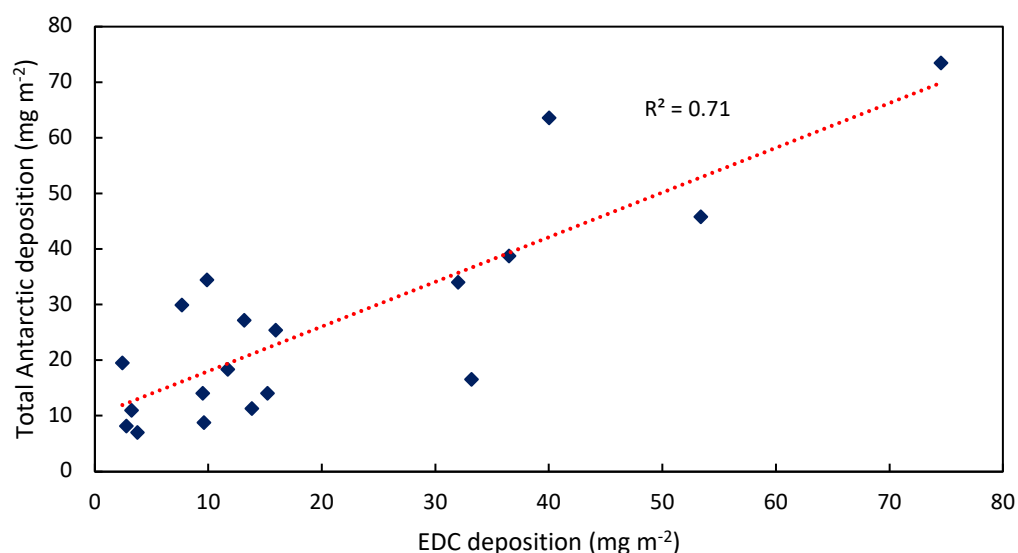


Figure S7. Comparison of sulfate deposition in EDC to total Antarctic deposition in corresponding events in Sigl et al., (2015).

Table S3. Sulfate deposition for Common Era events for EDC and Antarctic total. Antarctic depositions are from Sigl et al., (2015) with ages calculated with the WD2014 age model. EDC ages are determined from the AICC2012 age model.

Sigl et al., 2015 date (CE)	EDC date (CE)	Sulfate deposition (mg m ⁻²)	
		Dome C	Antarctic total
1815	1824	53.40	45.77
1809	1818	15.95	25.41
1674	1673	2.82	8.11
1458	1453	40.01	63.64
1277	1276	7.67	29.92
1258	1254	74.54	73.42
1172	1172	2.45	19.47
1039	1036	3.27	10.94
745	741	9.65	8.79
714	705	3.76	7.01
682	672	36.49	38.71
575	569	32.03	34.06
540	540	9.90	34.44
435	429	13.19	27.21
304	298	33.19	16.60
266	259	13.86	11.27
236	232	15.20	14.07
206	199	9.53	14.03
170	163	11.73	18.39

Table S4. Concentration and sulfur isotopic composition of procedural blanks measured at the University of St Andrews.

	nmol	$\delta^{34}\text{S}$ (‰)	1σ
	0.20	4.28	0.94
	0.36	6.24	0.60
	0.33	3.66	0.40
	0.16	2.73	0.74
	0.21	3.68	0.50
	0.23	4.59	0.46
	0.22	3.53	0.46
	0.24	2.93	0.44
	0.24	4.06	0.46
	0.22	3.81	0.44
	0.29	4.11	0.38
	0.18	4.35	0.54
	0.23	3.27	0.46
	0.22	3.08	0.39
	0.26	4.84	0.42
	0.33	4.58	0.38
	0.25	5.89	0.43
	0.14	8.17	1.06
Average	0.24	4.32	
2 s.d.	0.12	2.67	

Table S5. Sulfur isotopic composition of Switzer Falls consistency standard measured at the University of St Andrews.

	$\delta^{34}\text{S}$ (‰)	1 σ	$\delta^{33}\text{S}$ (‰)	1 σ	$\Delta^{33}\text{S}$ (‰)	1 σ
	4.13	0.02	2.10	0.06	-0.02	0.05
	4.14	0.02	2.20	0.05	0.06	0.09
	4.15	0.02	2.16	0.05	0.03	0.04
	4.13	0.02	2.11	0.05	-0.02	0.09
	4.14	0.02	2.07	0.05	-0.06	0.05
	4.16	0.02	2.12	0.06	-0.02	0.09
	4.12	0.02	2.15	0.05	0.03	0.11
	4.12	0.02	2.14	0.05	0.02	0.07
	4.18	0.02	2.24	0.06	0.09	0.12
	4.11	0.02	2.16	0.05	0.05	0.04
	4.21	0.02	2.19	0.05	0.03	0.04
	4.26	0.02	2.28	0.07	0.09	0.08
	4.16	0.03	1.99	0.11	-0.15	0.10
	4.24	0.02	1.96	0.07	-0.22	0.07
	4.22	0.02	2.07	0.07	-0.10	0.06
	4.24	0.02	1.99	0.07	-0.19	0.10
	4.30	0.02	2.16	0.06	-0.05	0.06
	4.12	0.03	2.14	0.07	0.02	0.13
	4.09	0.02	2.13	0.07	0.02	0.08
	4.12	0.02	2.17	0.07	0.05	0.09
	4.18	0.02	2.02	0.07	-0.13	0.08
	4.15	0.02	2.19	0.06	0.06	0.06
	4.15	0.02	2.21	0.08	0.07	0.11
	3.90	0.02	1.92	0.07	-0.09	0.08
	4.07	0.02	2.02	0.07	-0.08	0.07
Average	4.15		2.12		-0.02	
2 s.d.	0.15		0.18		0.17	

References

- Baroni, M., Thiemens, M. H., Delmas, R. J., and Savarino, J.: Mass-Independent Sulfur Isotopic Compositions in Stratospheric Volcanic Eruptions, *Science*, 315, 84–87, <https://doi.org/10.1126/science.1131754>, 2007.
- Burke, A., Moore, K. A., Sigl, M., Nita, D. C., McConnell, J. R., and Adkins, J. F.: Stratospheric eruptions from tropical and extra-tropical volcanoes constrained using high-resolution sulfur isotopes in ice cores, *Earth Planet. Sci. Lett.*, 521, 113–119, <https://doi.org/10.1016/j.epsl.2019.06.006>, 2019.
- Cole-Dai, J., Ferris, D., Lanciki, A., Savarino, J., Baroni, M., and Thiemens, M. H.: Cold decade (AD 1810–1819) caused by Tambora (1815) and another (1809) stratospheric volcanic eruption, *Geophys. Res. Lett.*, 36, 1–6, <https://doi.org/10.1029/2009GL040882>, 2009.

Gautier, E., Savarino, J., Erbland, J., and Farquhar, J.: SO₂ Oxidation Kinetics Leave a Consistent Isotopic Imprint on Volcanic Ice Core Sulfate, *J. Geophys. Res. Atmos.*, 123, 9801–9812, <https://doi.org/10.1029/2018JD028456>, 2018.

Gautier, E., Savarino, J., Hoek, J., Erbland, J., Caillon, N., Hattori, S., Albarede, F., Farquhar, J., and Yoshida, N.: 2600-years of stratospheric volcanism through sulfate isotopes, *Nat. Commun.*, 10, 1–7, <https://doi.org/10.1038/s41467-019-08357-0>, 2019.

Savarino, J., Romero, A., Cole-Dai, J., Bekki, S., and Thiemens, M. H.: UV induced mass-independent sulfur isotope fractionation in stratospheric volcanic sulfate, *Geophys. Res. Lett.*, 30, 2131, <https://doi.org/10.1029/2003GL018134>, 2003.

Sigl, M., Winstrup, M., McConnell, J. R., Welten, K. C., Plunkett, G., Ludlow, F., Büntgen, U., Caffee, M., Chellman, N., Dahl-Jensen, D., Fischer, H., Kipfstuhl, S., Kostick, C., Maselli, O. J., Mekhaldi, F., Mulvaney, R., Muscheler, R., Pasteris, D. R., Pilcher, J. R., Salzer, M., Schüpbach, S., Steffensen, J. P., Vinther, B. M., and Woodruff, T. E.: Timing and climate forcing of volcanic eruptions for the past 2,500 years., *Nature*, 523, 543–9, <https://doi.org/https://doi.org/10.1038/nature14565>, 2015.

Veres, D., Bazin, L., Landais, A., Toyé Mahamadou Kele, H., Lemieux-Dudon, B., Parrenin, F., Martinerie, P., Blayo, E., Blunier, T., Capron, E., Chappellaz, J., Rasmussen, S. O., Severi, M., Svensson, A., Vinther, B., and Wolff, E. W.: The Antarctic ice core chronology (AICC2012): An optimized multi-parameter and multi-site dating approach for the last 120 thousand years, *Clim. Past*, 9, 1733–1748, <https://doi.org/10.5194/cp-9-1733-2013>, 2013.

York, D., Evensen, N. M., Martínez, M. L., and De Basabe Delgado, J.: Unified equations for the slope, intercept, and standard errors of the best straight line, *Am. J. Phys.*, 72, 367–375, <https://doi.org/10.1119/1.1632486>, 2004.