

1 Variability of sulfate signal in ice-core records based on five replicate cores

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7

8 **Abstract**

9 Current volcanic reconstructions based on ice core analysis have significantly improved over
10 the past few decades by incorporating multiple core analysis with high temporal resolution
11 from different parts of the Polar Regions. Regional patterns of volcanic deposition are based
12 on composite records, built from cores taken at both poles. However, in many cases only a
13 single record at a given site is used for these reconstructions. This assumes that transport and
14 regional meteorological patterns are the only source of the dispersion of the volcanic-products.
15 Here we evaluate the local scale variability of a sulfate profile in a low accumulation site
16 (Dome C, Antarctica), in order to assess the representativeness of one core for such
17 reconstruction. We evaluate the variability with depth, statistical occurrence, and sulfate flux
18 deposition variability of volcanic eruptions detected on 5 ice cores, drilled 1 meter away from
19 each other. Local scale variability, essentially attributed to snow drift and surface roughness
20 at Dome C, can lead to a non-exhaustive record of volcanic events when a single core is used
21 as the site reference with a bulk probability of 30 % of missing volcanic events and almost
22 60 % uncertainty on the volcanic flux estimation. Averaging multiple records almost erases
23 the probability of missing volcanic events and can reduce by half the uncertainty pertaining to
24 the deposition flux.

25

26 **Introduction**

27 When a large and powerful volcanic eruption occurs, the energy of the blast is sufficient to inject
28 megatons of material directly into the upper atmosphere [Robock, 2000]. While ashes and pyroclastic
29 materials fall rapidly to the ground because of gravity, gases remain over longer time scales. Among
30 gases, SO₂ is of a particular interest due to its conversion to tiny sulfuric acid aerosols, which can
31 potentially impact the radiative budget of the atmosphere [Rampino and Self, 1982; Timmreck, 2012].
32 In the troposphere a combination of turbulence, cloud formation, rainout and downward transport are
33 efficient processes that clean the atmosphere of sulfuric acid, and volcanic sulfuric acid layers rarely
34 survive more than a few weeks, limiting their impact on climate. The story is different when volcanic
35 SO₂ is injected into the stratosphere. There, the dry, cold and stratified atmosphere allows sulfuric acid
36 layers to remain for years, slowly spreading an aerosols blanket around the globe. The tiny aerosols
37 then act as efficient reflectors and absorbers of incoming solar radiations, significantly modifying the
38 energy balance of the atmosphere [Kiehl and Briegleb, 1993] and the ocean [Gleckler et al., 2006;
39 Miller et al., 2012; Ortega et al., 2015]. With a lifetime of 2 to 4 years, these aerosols of sulfuric acid
40 ultimately fall into the troposphere where they are removed within weeks.

41 In Polar Regions, the deposition of the sulfuric acid particles on pristine snow will generate an acidic
42 snow layer, enriched in sulfate. The continuous falling of snow, the absence of melting and the ice
43 thickness make the polar snowpack the best records of the Earth's volcanic eruptions. Hammer [1977]
44 was the first to recognize the polar ice propensity to record such volcanic history. Built on the seminal
45 work of Hammer et al., a paleo-volcanism science developed around this discovery with two aims.

46 The first relies on the idea that the ice record can reveal past volcanic activity and, to a greater extent,
47 its impact on Earth's climate history [Robock, 2000; Timmreck, 2012]. Indeed, at millennium time
48 scale, volcanoes and the solar activity are the only two recognized natural climate forcings [Stocker et
49 al., 2013]. Based on ice records, many attempts are made to extract the climate forcing induced by a
50 volcanic eruption [Crowley and Unterman, 2013 ; Gao et al., 2008; Gao et al., 2007; Sigl et al., 2013;
51 Sigl et al., 2014; Zielinski, 1995]. However, such an approach is inevitably prone to large uncertainty

52 pertaining to the quality of the ice record and non-linear effects between deposition fluxes and source
53 emissions [Pfeiffer *et al.*, 2006].

54 The second aim of the paleo-volcanism is to provide an absolute dating scale when clear volcanic
55 events in differently located ice cores can be unambiguously attributed to the same dated event [Severi
56 *et al.*, 2007]. The time synchronization of different proxy records is possible, allowing study of the
57 phasing response of different environmental parameters to climate perturbation [Ortega *et al.*, 2015;
58 Sigl *et al.*, 2015] or estimating the snow deposition over time [Parrenin *et al.*, 2007]. Whatever the
59 intent, paleo-volcanism should rely on robust and statistically relevant ice core records.

60 Work to establish a volcanic index, undertaken to date, has assumed volcanic event are clearly
61 identified, without any false signal from background variations induced by other sulfur sources (eg
62 marine, anthropogenic, etc.). Seasonal layer counting is used whenever possible, bi-polar comparison
63 of ice sulfate records has become the method of choice to establish an absolute dated volcanic index
64 [Langway *et al.*, 1988]. Both known and unknown events can be used to synchronize different cores.
65 However, only a limited number of peaks, with characteristic shape or intensity, and known to be
66 associated with a dated eruption, can be used to set a reliable time scale [Parrenin *et al.*, 2007]. This
67 restriction is partly fueled by the poor and/or unknown representativeness of most volcanic events
68 found in ice cores. Most of the time, a single core is drilled at a given site and used for cross
69 comparison with other sites. This approach is clearly insufficient for ambiguous events.

70 At a large scale, sulfate deposition is highly variable in space and mainly associated with atmospheric
71 transport and precipitation patterns. At a local scale (ca. 1m), variability can emerge from post-
72 deposition processes. While sulfate is a non-volatile species supposed to be well preserved in snow,
73 spatial variability is induced by drifted snow, wind erosion leading to surface roughness
74 heterogeneities [Libois *et al.*, 2014]. These effects are amplified in low accumulation sites where most
75 of the deep drilling sites are performed [EPICA-community-members, 2004; Jouzel, 2013; Lorius *et al.*,
76 1985]). To the best of our knowledge, one single study has used multiple drillings at a given site to
77 analyze the representativeness of the ice core record [Wolff *et al.*, 2005]. This study took advantage of
78 the two EPICA cores drilled at Dome C, 10 m apart (Antarctica, 75°06'S, 123°21'E, elevation 3220 m,

79 mean annual temperature -54.5°C) [*EPICA-community-members*, 2004] to compare the dielectric
80 profile (DEP) along the 788 m common length of the two cores. For the two replicate cores, statistical
81 analysis showed that up to 50 % variability in the pattern of any given peak was encountered as a
82 consequence of the spatial variability of the snow deposition. The authors concluded that ice-core
83 volcanic indices from single cores at such low-accumulation sites couldn't be reliable and what was
84 required was a network of close-spaced records. However, as mentioned in Wolff's conclusion, this
85 statistical study relied only on two records. Additionally, DEP signals are known to be less sensitive
86 than sulfate signals for volcanic identification, and more accuracy is expected by comparing sulfate
87 profiles. The authors thus encouraged conducting a similar study on multiple ice cores to see if the
88 uncertainty could be reduced.

89 In the present study we took advantage of the drilling of 5 ice cores at Dome C, initially intended for
90 the analysis of sulfur isotopes of the volcanic sulfate. Putting aside the number of records, our
91 approach is similar in many points to Wolff's work. However, it has the advantage of relying on highly
92 resolved sulfate profiles. In addition, the spatial scale is slightly smaller as the 5 cores were drilled 1-
93 meter apart. The comparison of 5 identically processed cores is a chance to approach the
94 representativeness of a single core reconstruction at a low accumulation site, the most prone to spatial
95 variability. Therefore new constraints on variability of sulfate deposition recorded by spatial
96 heterogeneity in such sites are expected from the present work. Even if recent publications [*Sigl et al.*,
97 2014], underline the need of using multiple records in low accumulation sites, to overcome the spatial
98 variability issue, such records are not always available. This lack of records adds uncertainty in the
99 volcanic flux reconstruction based on polar depositional pattern. Our study should help to better
100 constrain the error associated with local scale variability, and ultimately, the statistical significance of
101 volcanic reconstructions. The present study discusses the depth shift, occurrence of events and
102 deposition flux variability observed in the 5 cores drilled.

103

104 **Experimental setup and Methods**

105 **Core drilling**

106 The project “VolSol”, initiated in 2009, aimed at constraining the estimation of the natural part of
107 radiative forcing, composed of both volcanic and solar contributions using ice core records of sulfate
108 and Beryllium-10. In order to build a robust volcanic index including a discrimination of stratospheric
109 events based on sulfur isotopic ratios [Baroni *et al.*, 2008; Savarino *et al.*, 2003], 5 x 100 m-firn cores
110 (dia. 10 cm) were drilled in 2010/2011 along a 5 m straight line, and spaced approximately 1 m apart.
111 The drilling took place at the French-Italian station Concordia (Dome C, Antarctica, 75°06’S,
112 123°21’E, elevation 3220 m, mean annual temperature -54.5°C) more precisely between Concordia
113 station and EDC drilling tent (300m west of the EDC drilling tent). At this site, the mean annual snow
114 accumulation rate is about 25 kg m⁻² y⁻¹, leading to an estimated time-period covered by the cores of
115 2500 years. Cores were logged and bagged in the field, and temporarily stored in the underground core
116 buffer (- 50 °C) before analysis. The unusual number of ice core drilled at the same place was driven
117 by the amount of sulfate necessary to conduct the isotopic analysis. However, this number of replicate
118 cores drilled 1m apart offers the opportunity to question the representativeness of a volcanic signal
119 extracted from a single core per site.

120

121 **Sampling, Resolution and IC Analyses**

122 Analyses were directly performed on the field during two consecutive summer campaigns. Thirty
123 meters were analyzed in 2011, the rest was processed the following year. The protocol was identical
124 for each core and the steps followed were:

- 125 - Decontamination of the external layer by scalpel scrapping
- 126 - Longitudinal cut with a band saw of a 2 cm stick of the most external layer
- 127 - Sampling of the ice stick at a 2 cm-resolution (ca. 23 600 samples)
- 128 - Thawing the samples in 50 ml centrifuge tubes, and transfer in 15 ml centrifuge tubes positioned in
129 an autosampler
- 130 - Automatic analysis with a Metrohm IC 850 in suppressed mode (NaOH at 7 mM, suppressor H₂SO₄
131 at 50 mM, Dionex AG11 column), in a fast IC configuration (2 min run) with regular calibration
132 (every 60 samples) using certified sulfate reference solution (Fisher brand, 1000 ppm certified).

133 Due to the fragility of snow cores, the first 4 m were only analyzed on a single core (Figure 1). We
134 will thus not discuss the variability of the Pinatubo and Agung eruptions present in these first 4 meters.
135 Concentration data are deposited in the public domain and made freely available in NOAA National
136 Climatic Data center.

137

138 **Peaks discrimination method**

139 As with most algorithms used for peak detection, the principle is to detect anomalous sulfate
140 concentration peaks from a background noise (stationary or not), which could potentially indicate a
141 volcanic event. The estimation of the background value should therefore be as accurate as possible.
142 Using core 2 as our reference core, we observed a background average value stationary and close to 85
143 ppb \pm 30 ppb (1σ) at Dome C during the 2,500 years of the record. However, the variability is
144 sufficient enough to induce potential confusion on detection of small peaks. Therefore, a stringent
145 algorithm using PYTHON language (accessible on demand) was developed to isolate each possible
146 peak. The algorithm treats the full ice record by 1-meter section (ca. 45-50 samples). For each meter, a
147 mean concentration (m) and standard deviation (σ) is calculated regardless of the presence or not of
148 peaks in the section. Then, every value above the $m + 2\sigma$ is removed from the 1-meter dataset. A new
149 mean and standard deviation is calculated and the same filtration is applied. Iteration runs until no
150 more data above $m + 2\sigma$ is found. At that point, m represents the background mean concentration.
151 The process runs for each 1-m section, starting from the surface sample and until the end of the core.
152 Then, each 1-meter dataset is shifted by one sample; the process is reset and the peak detection run
153 again on each new 1-m dataset. Sample shift is applied until the last sample of the first 1-meter section
154 is reached so that no bias is introduced by the sampling scheme. Every concentration data point is thus
155 compared approximately with its 100 neighbor data (50 of each side). Each data point isolated by the
156 algorithm is further tested. To be considered as a point belonging to a potential volcanic peak, the data
157 should be detected in a given core (i.e. for being above the $m + 2\sigma$ final threshold) in at least 50 % of
158 the 50 runs. Additionally, the point has to be part of at least three consecutive points passing the same
159 50 % threshold detection. This algorithm was applied individually on each core, giving 5 different

160 lists of peak. In total, 54, 51, 47, 50 and 47 peaks were detected on core 1, 2, 3, 4 and 5, respectively.
161 A manual detection is then required if one wants to build a more accomplished volcanic record from
162 several profiles, which must be based on shape criteria, and not only statistical criteria. However, in
163 the scope of this paper, no manual sorting was applied, so that the statistical assessment could rely on
164 more objective criteria (the number of occurrences).

165

166 **Core synchronization and dating**

167 Core 1 was entirely dated with respect to the recently published volcanic ice core database [Sigl *et al.*,
168 2015] using *Analyseries* 2.0.8 software (<http://www.lsce.ipsl.fr/Phocea/Page/index.php?id=3>), and
169 covers the time period of -588 to 2010 CE. Figure 2 shows the age-depth profile obtained for this core.
170 A total of 13 major volcanic eruptions well dated were used as time markers to set a time scale (bold
171 date in Table 1). Core 1 was entirely dated through linear interpolation between those tie points. Dated
172 core 1 was then used as a reference to synchronize the remaining 4 cores, using the same tie points and
173 10 additional peaks (non-bold date in Table 1), presenting characteristic patterns common to each core.
174 In total, 23 points were therefore used to synchronize the cores.

175

176 **Composite building from the 5 ice cores**

177 Through the routine described above, the five cores are depth-synchronized using the 23 tie points and
178 other potential volcanic events in each core cores are detected independently. Therefore, the number
179 of peaks detected in each core is different (between 47 and 54) and their depth (with the exception of
180 the tie points used) is slightly different to each other cores due to sampling scheme and position of the
181 maximum concentration. After correcting the depth shift between cores, a composite profile was built
182 by summing all the peaks identified in the 5 cores. In this composite, sulfate peaks from different
183 cores are associated to a same event as soon as their respective depth (corresponding to the maximum
184 concentration) are included in a 20cm depth window. This level of tolerance is consistent with the
185 dispersion in width and shape of peaks observed. A number of occurrences is then attributed to each

186 sulfate peak, reflecting the number of time it has been detected in the 5 cores dataset (Figure 4).

187

188 **Results and Discussions**

189 **Depth offset between cores**

190 Depth offsets between cores are the result of the surface roughness at the time of drilling, variability in
191 snow accumulation, heterogeneous compaction during the burying of snow layers and logging
192 uncertainty. This aspect has been discussed previously, over a similar time-scale (Wolff et al. 2005),
193 and over a longer time-scale (Barnes et al. 2006) in Dome C. Surface roughness, attributed to wind
194 speed, temperatures and accumulation rate, is highly variable in time and space. These small features
195 hardly contribute to the depth offset on a larger spatial scale, in which case glacial flow can control the
196 offset between synchronized peaks, as it seems to be the case in South pole site (Bay et al. 2010).
197 However, in Dome C, and at the very local spatial scale we are considering in the present work,
198 roughness is significant regarding to the accumulation rate. It is therefore expected that synchronized
199 peaks should be found at different depths. The offset trend fluctuates with depth, due to a variable
200 wind speed (Barnes et al. 2006). To estimate the variability in the depth shift for identical volcanic
201 events, we used the tie points listed in Table 1. For each peak maximum, we evaluate the depth offset
202 of core 1, 3, 4 and 5, with respect to core 2. To avoid logging uncertainty due to poor snow
203 compaction in the first meters of the cores and surface roughness at the time of the drilling, we used
204 the UE 1809 depth in core 2 (13.30 m) as a depth reference horizon from which all other depth cores
205 were anchored using the same 1809 event. For this reason, only eruptions prior to 1809 were used to
206 evaluate the offset variability, that is 18 eruptions instead of the 23 used for the core synchronization.
207 Figure 5, shows the distribution of depth shift of the cores with respect to core 2. While the first 40 m
208 appear to be stochastic in nature, a feature consistent with the random local accumulation variations
209 associated with snow drift in Dome C site, it is surprising that at greater depth, offset increases (note
210 that the positive or negative trends are purely arbitrary and depends only on the reference used, here
211 core 2). The maximum offset, obtained between core 3 and 5 is about 40 cm. Such accrued offsets

212 with depth were also observed by *Wolff et al.*, [2005] and were attributed to the process of logging
213 despite the stringent guidelines used during EPICA drilling. Similarly, discontinuities in the depth
214 offset, observed by *Barnes et al.*, [2006] were interpreted as resulting from logging errors. As no
215 physical processes can explain a trend in the offsets, we should also admit that the accrued offset is
216 certainly the result of the logging process. In the field, different operators were involved but a
217 common procedure was used for the logging. Two successive cores extracted from the drill were
218 reassembled on a bench to match the non uniform drill cut and then hand sawed meter by meter to get
219 the best precise depth core, as neither the drill depth recorder nor the length of the drilled core section
220 can be used for establishing the depth scale. This methodology involving different operators should
221 have randomized systematic errors but obviously this was not the case. Despite the systematic depth
222 offset observed, synchronization did not pose fundamental issues as the maximum offset in rescaled
223 profiles never exceeds the peak width (ca. 20 cm) thank to the 10 possible comparisons when pair of
224 core are compared. Confusion of events or missing of events are thus very limited in our analysis (see
225 next section).

226

227 **Variability in events occurrence**

228 The variability in events occurrence in the 5 ice cores has been evaluated through the construction of a
229 composite record (Figure 4) and the counting of events in each core as described in the method. By
230 combining the five ice cores, we listed a total amount of 91 sulfate peaks (Pinatubo and Agung not
231 included), which are not necessarily from volcanic sources. Some peaks can be due to post deposition
232 effects affecting the background deposition, or even contamination. When it comes to defining a
233 robust volcanic index, peak detection issues emerge. Chances to misinterpret a sulfate peak and assign
234 it, by mistake, to a volcanic eruption, as well as chances to miss a volcanic peak, can be discussed
235 through a statistic analysis conducted on our five cores.

236 We try to evaluate to what extent multiple cores comparison facilitates the identification of volcanic
237 peaks, among all sulfate peaks that can be detected in a core. To do so, we assumed that a peak is of
238 volcanic origin as soon as it is detected at least in two cores. In other words, the probability to have

239 two non-volcanic peaks synchronized in two different cores is nil. It is expected that combining an
240 increasing number of cores will increasingly reveal the real pattern of the volcanic events. All possible
241 combinations from 2 to 5 cores comparison were analyzed, totalizing 26 possibilities for the entire
242 population. The results for each comparison were averaged, giving a statistic on the average number of
243 volcanic peaks identified per number of cores compared. The results of the statistical analysis are
244 presented in Figure 6. As expected, in a composite made of 1 to 5 cores, the number of sulfate peaks
245 identified as volcanic peaks (for being detected at least twice) increases with the number of cores
246 combined in the composite. Thus, while only 30 peaks can be identified as volcanic from a two cores
247 study, a study based on 5 cores can yields 62 such peaks. The 5-cores comparison results in the
248 composite profile given in Figure 4a. The initial composite of 93 peaks is reduced to 64 volcanic
249 peaks (Pinatubo and Agung included) after removing the single peaks (Figure 4b). Each characteristic
250 of the retained peaks is given in Table 2. The main conclusion observing the final composite record is
251 that only 17 of the 64 peaks were detected in all of the 5 cores and 68 % of all peaks were at least
252 present in two cores. At the other side of the spectrum, 2-cores analysis reveals that only 33 % (30
253 peaks on average) of the peaks are identified as possible eruptions. Two cores comparison presents
254 still a high risk of not extracting the most robust volcanic profile at low accumulation sites, a
255 conclusion similar to *Wolff et al.*, [2005]. Surprisingly, it can also be noticed that this 5-core
256 comparison doesn't results in an asymptotic ratio of identified volcanic peaks, suggesting that 5 cores
257 are not sufficient either to produce a full picture. High accumulation sites should be prone to less
258 uncertainty; however, this conclusion remains an a priori that still requires a confirmation.

259 Large and small events are not equally concerned by those statistics. Figure 7 shows that the
260 probability of presence is highly dependent on peak flux and the chance to miss a small peak
261 (maximum flux in the window $[f + 2\sigma : f + 5\sigma]$, f being the background average flux) is much higher
262 than the chance to miss a large one (maximum flux above $f + 8\sigma$). However, it is worth noticing that
263 major eruptions can also be missing from the record, as it has already been observed in other studies
264 [*Castellano et al.*, 2005; *Delmas et al.*, 1992]. The most obvious example in our case is the Tambora
265 peak (1815 AD), absent in 2 of our 5 drillings, while presenting an intermediate to strong signal in the

266 others (Figure 8). The reason for the variability in event occurrence has been discussed already by
267 *Castellano et al.*, [2005]. In the present case of close drillings, long-range transport and large-scale
268 meteorological conditions can be disregarded due to the small spatial scale of our study; the snow drift
269 and surface roughness is certainly the main reasons for missing peaks. The fact that two close events
270 as UE 1809 and Tambora are so differently recorded indicates how punctual, in time and space post-
271 depositional effects can affect the recording of eruptions.

272

273 **Variability in signal strength**

274 To compare peak height variability, detected peaks were corrected by subtracting the background from
275 peak maximum. We considered C_i/C_{mean} variations, C_i being the SO_4^{2-} maximum concentration in core
276 i (1 to 5), and C_{mean} being the mean of those concentration for the event i . For concentration values,
277 positive by definition, the log-normal distribution is more appropriate; geometric means and geometric
278 standard deviations were used, as described by *Wolff et al.*, [2005] (Table 3). In our calculation, the
279 geometric standard deviation based on 2 cores is 1.35; in other words, maximum concentrations are
280 uncertain by a factor 1.35. This factor is slightly lower than the one obtained in *Wolff et al.*, [2005]
281 (1.5). Our cores are drilled closer (one meter from each others, instead of 10 m for *Wolff et al.*), which
282 might slightly reduce the uncertainty. The peaks height variability obtained by averaging 5 cores
283 (1.21), matches *Wolff et al.* forecast. Based on a 50 % uncertainty on 2 cores, *Wolff et al.* predicted a
284 20 % uncertainty on a 5 cores study (consistent with a reduction of the standard deviation by a factor
285 of $1/\sqrt{n}$, by averaging n values). Comparing the peaks maxima enables us to compare our study with
286 *Wolff's* study, also based on peaks maxima. However, in our case, comparing maxima induces a bias
287 related to the sampling method: with a two centimeters resolution on average, peak's height is directly
288 impacted by the cutting, which tends to smooth the maxima. Comparing the total sulfate deposited
289 during the event is more appropriate. Proceeding on a similar approach, but reasoning on mass of
290 deposited sulfate rather than maximum concentration, the obtained variability is higher than
291 previously: 41 % uncertainty on volcanic deposited sulfate mass, on a 5-cores study (F_i/F_{mean} , F_i being
292 the mass flux of peak i), and 56 % uncertainty on a 2-cores comparison (F_i/F_1). The difference in the

293 signal dispersion between the two approaches rests on the fact that peak maximum has a tendency to
294 smooth the concentration profile as a consequence of the sampling strategy. This artifact is suppressed
295 when the total mass deposited is considered. In any case, uncertainty seems to be significantly reduced
296 when comparing 5 cores instead of 2.

297

298 **Conclusion:**

299 This study confirms in many ways previous work on multiple drilling variability [*Wolff et al.*, 2005].

300 As already discussed, peaks flux uncertainty can be significantly reduced (56 % to 41 %) by averaging

301 5 ice-cores signals instead of 2. A 5-cores composite profile has been built using the criteria that a

302 peak is considered as volcanic if present at least in two cores. We observed that the number of

303 volcanic peaks listed in a composite profile increases with the number of cores considered. With 2

304 cores, only 33 % of the peaks present in the composite profile are tagged as volcanoes. This

305 percentage increases to 68 % with 5 cores. However, we did not observe an asymptotic value, even

306 with 5 cores drilled. A record based on a single record in a low accumulation site is therefore very

307 unlikely to be a robust volcanic record. Of course, peaks presenting the largest flux are more likely to

308 be detected in any drilling, but the example of the Tambora shows that surface topography is variable

309 enough to erase even the most significant signal, although rarely. This variability in snow surface is

310 evidenced in the depth offset between two cores drilled less than 5 meters from each other, as peaks

311 can easily be situated 40 cm apart.

312 In low accumulation sites such as Dome C, where surface roughness can be on the order of the snow

313 accumulation and highly variable, indices based on chemical records should be considered with

314 respect to the time-scale of the proxy studied. Large time-scale trends are faintly sensitive to this effect.

315 On the contrary, a study on episodic events like volcanic eruptions or biomass burning, with a

316 deposition time in the order of magnitude of the surface variability scale should be based on a

317 multiple-drilling analysis. A network of several cores is needed to obtain a representative record, at

318 least in terms of recorded events. However, although lowered by the number of cores, the flux remains

319 highly variable, and still uncertain by a factor of 1.4 with 5 cores. This point is particularly critical in

320 volcanic reconstructions that rely on the deposited flux to estimate the mass of aerosols loaded in the
321 stratosphere, and to a larger extent, the climatic forcing induced. Recent reconstructions largely take
322 into account flux variability associated with regional pattern of deposition, but this study underlines
323 the necessity of not neglecting local scale variability in low accumulation sites. Less variability is
324 expected with higher accumulation rate, but this still has to be demonstrated. Sulfate flux is clearly
325 one of the indicators of the eruption strength, but due to transport, deposition and post-deposition
326 effects, such direct link should not be taken for granted.

327 With such statistical analysis performed systematically at other sites, we should be able to reveal even
328 the smallest imprinted volcanoes in ice cores, extending the absolute ice core dating, the
329 teleconnection between climate and volcanic events and improving the time-resolution of mass
330 balance calculation of ice sheets.

331

332

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442 **Table 1** – Tie points used to set the time scale and synchronize the cores. Volcanic events are
443 named "Ev x" if they are not assigned to a well-known eruption. Dating of the events is based
444 on *Sigl et al.*, [2015].

445

| Eruption | core 1 | core 2 | core 3 | core 4 | core 5 | date of deposition |
|-----------|--------|--------|--------|--------|--------|--------------------|
| Surface | 0 | 0 | 0 | 0 | 0 | 2010 |
| Pinatubo | 1.53 | | | | | 1992 |
| Krakatoa | 8.82 | 8.92 | 8.67 | 8.71 | 8.63 | 1884 |
| Cosiguina | 11.98 | 11.83 | 11.65 | 11.62 | 11.46 | 1835 |
| Tambora | 12.85 | | | 12.6 | 12.57 | 1816 |
| UE 1809 | 13.33 | 13.3 | 13.04 | 13.08 | 12.98 | 1809 |
| ev 7 | 15.98 | 15.93 | 15.66 | 15.67 | 15.52 | 1762 |
| Serua/UE | 19.29 | 19.22 | 18.93 | 18.94 | 18.78 | 1695 |
| Ev 10 | 21.87 | 21.74 | 21.53 | 21.48 | 21.4 | 1646 |
| kuwae | 30.18 | 30.04 | 29.92 | 29.85 | 29.73 | 1459 |
| ev 16 - A | 37.35 | 37.29 | 37.17 | 37.04 | 36.91 | 1286 |
| ev 16 - B | 37.77 | 37.77 | 37.62 | 37.52 | 37.4 | 1276 |
| ev 16 - C | 38.1 | 38.04 | | 37.78 | | 1271 |
| Samalas | 38.49 | 38.46 | 38.28 | 38.2 | 38.09 | 1259 |
| ev 17 | 39.59 | 39.56 | 39.46 | 39.36 | 39.2 | 1230 |
| ev 18 | 41.87 | 41.83 | 41.7 | 41.6 | 41.41 | 1172 |
| ev 22 | 50.26 | 50.3 | 50.2 | 50.11 | 49.87 | 9599 |
| ev 27 | 60.77 | 60.72 | 60.66 | | 60.27 | 684 |
| ev 31 | 65.72 | 65.74 | 65.68 | 65.6 | 65.25 | 541 |
| ev 35 | 76.06 | 76.13 | 76 | 75.94 | 75.64 | 235 |
| ev 46 | 90.42 | 90.53 | 90.36 | 90.41 | 89.95 | -214 |
| ev 49 | 97.15 | 97.16 | 97.19 | 97.22 | 96.74 | -426 |
| ev 51 | 100.16 | 100.19 | | 100.22 | 99.7 | -529 |

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448 **Table 2** – Sulfate peak (maximum concentration, in ng.g^{-1} , and flux of volcanic sulfate
449 deposited, in kg.km^{-2}) considered as volcanic eruptions based on the statistical analysis of the
450 5 cores. Flux is calculated by integrating the peak, using the density profile obtained during
451 the logging process. Volcanic flux values are corrected from background sulfate (calculated
452 separately for each sulfate peak). 0 stands for non-detected events in the cores. Agung
453 (3.77m) and Pinatubo (1.52m) were not included in the statistical analysis because they were
454 analyzed only in core one and thus are marked as not applicable (N/A). The estimation of the
455 average volcanic flux is calculated considering detected peaks only (non detected peaks are
456 not included in this estimation). The relative error on the flux (estimated as 10%) takes into
457 account the IC measurement relative standard deviation (below 4% based on standards runs),
458 the error on firn density (relative error estimated as 2%) and the error on samples time length
459 (10%). The last column displays data obtained from Castellano *et al.* (2005), for identical
460 volcanic peaks. For similar peaks Castellano's flux generally falls into the average flux + 40%
461 uncertainty, sometimes exceeding this value.

| Peak depth (m) | date (year) | core 1 | | core 2 | | core 3 | | core 4 | | core 5 | | average* | | | Castellano <i>et al.</i> , 2005 | |
|----------------|-------------|----------------------------------|------------|----------------------------------|------------|----------------------------------|------------|----------------------------------|------------|----------------------------------|------------|----------------------------------|------------|-----------|----------------------------------|------------|
| | | [SO ₄ ²⁻] | Volc. flux | [SO ₄ ²⁻] | Volc. flux | [SO ₄ ²⁻] | Volc. flux | [SO ₄ ²⁻] | Volc. flux | [SO ₄ ²⁻] | Volc. flux | [SO ₄ ²⁻] | Volc. flux | 1σ (flux) | [SO ₄ ²⁻] | Volc. flux |
| 1.52 | 1992 | 188 | 5.0 | N/A | N/A | N/A | N/A | N/A | N/A | N/A | N/A | 188 | 5.0 | 0.5 | 313 | 11 |
| 3.77 | 1964 | 207 | 5 | N/A | N/A | N/A | N/A | N/A | N/A | N/A | N/A | N/A | N/A | N/A | 362 | 8 |
| 6.24 | 1929 | 0 | 0.0 | 164 | 1.3 | 0 | 0.0 | 132 | 1.1 | 0 | 0.0 | 148 | 1.2 | 0.1 | | |
| 8.59 | 1891 | 0 | 0.0 | 0 | 0.0 | 0 | 0.0 | 134 | 1.3 | 117 | 0.9 | 126 | 1.1 | 0.1 | 140 | 3.1 |
| 8.92 | 1885 | 232 | 8.1 | 262 | 8.8 | 236 | 10.5 | 240 | 10.2 | 216 | 7.7 | 237 | 9.1 | 0.9 | 289 | 9.3 |
| 11.83 | 1839 | 220 | 7.7 | 173 | 5.4 | 190 | 4.9 | 177 | 5.5 | 173 | 4.0 | 187 | 5.5 | 0.6 | | |
| 12.08 | 1834 | 0 | 0.0 | 0 | 0.0 | 144 | 2.5 | 0 | 0.0 | 137 | 1.3 | 140 | 1.9 | 0.2 | | |
| 12.91 | 1816 | 455 | 13.1 | 0 | 0.0 | 0 | 0.0 | 188 | 1.8 | 307 | 6.0 | 317 | 7.0 | 0.7 | 606 | 39.3 |
| 13.3 | 1809 | 436 | 16.6 | 291 | 10.5 | 392 | 12.7 | 408 | 16.3 | 461 | 13.4 | 398 | 13.9 | 1.4 | 271 | 10.2 |
| 15.93 | 1762 | 176 | 2.7 | 248 | 6.7 | 201 | 3.4 | 0 | 0.0 | 0 | 0.0 | 208 | 4.2 | 0.4 | 174 | 4.5 |
| 19.29 | 1695 | 287 | 13.4 | 0 | 0.0 | 168 | 9.2 | 194 | 7.3 | 0 | 0.0 | 217 | 10.0 | 1.0 | 185 | 8.8 |
| 20.3 | 1674 | 261 | 7.8 | 0 | 0.0 | 0 | 0.0 | 196 | 4.3 | 178 | 2.3 | 212 | 4.8 | 0.5 | 142 | 5.3 |
| 20.7 | 1666 | 0 | 0.0 | 0 | 0.0 | 0 | 0.0 | 123 | 1.6 | 149 | 2.4 | 136 | 2.0 | 0.2 | | |
| 21.74 | 1646 | 257 | 10.1 | 249 | 10.3 | 259 | 13.2 | 282 | 17.5 | 257 | 13.2 | 261 | 12.8 | 1.3 | | |
| 22.72 | 1625 | 181 | 4.8 | 146 | 2.7 | 141 | 2.9 | 0 | 0.0 | 0 | 0.0 | 156 | 3.5 | 0.3 | 175 | 8.0 |
| 23.77 | 1600 | 225 | 10.6 | 0 | 0.0 | 170 | 2.5 | 0 | 0.0 | 0 | 0.0 | 197 | 6.5 | 0.7 | 194 | 13.4 |
| 25.78 | 1557 | 144 | 2.1 | 0 | 0.0 | 0 | 0.0 | 148 | 2.2 | 0 | 0.0 | 146 | 2.1 | 0.2 | | |
| 30 | 1459 | 496 | 33.2 | 442 | 31.1 | 422 | 31.6 | 543 | 37.2 | 559 | 36.9 | 493 | 34.0 | 3.4 | 399 | 31.7 |
| 30.56 | 1449 | 0 | 0.0 | 143 | 1.8 | 131 | 2.8 | 0 | 0.0 | 0 | 0.0 | 137 | 2.3 | 0.2 | | |
| 31.83 | 1417 | 0 | 0.0 | 0 | 0.0 | 0 | 0.0 | 155 | 2.6 | 148 | 2.6 | 151 | 2.6 | 0.3 | | |
| 33.51 | 1377 | 0 | 0.0 | 0 | 0.0 | 140 | 2.3 | 0 | 0.0 | 162 | 5.4 | 151 | 3.9 | 0.4 | | |
| 34.85 | 1348 | 273 | 12.4 | 288 | 14.2 | 209 | 7.9 | 303 | 18.3 | 269 | 13.2 | 268 | 13.2 | 1.3 | 211 | 10.4 |
| 37.29 | 1286 | 325 | 18.3 | 324 | 16.1 | 373 | 17.1 | 347 | 14.8 | 458 | 30.7 | 365 | 19.4 | 1.9 | 258 | 22.4 |
| 37.77 | 1276 | 563 | 28.9 | 605 | 40.4 | 570 | 28.8 | 525 | 26.3 | 497 | 21.6 | 552 | 29.2 | 2.9 | 304 | 20.5 |

| | | | | | | | | | | | | | | | | |
|--------|-------------|------|------|------|------|-----|------|------|------|------|-------|------|------|-----|-----|------|
| 38.04 | 1271 | 205 | 4.1 | 180 | 3.1 | 0 | 0.0 | 235 | 5.1 | 0 | 0.0 | 206 | 4.1 | 0.4 | | |
| 38.46 | 1259 | 1086 | 59.7 | 1022 | 63.8 | 928 | 61.4 | 1030 | 78.5 | 1428 | 104.8 | 1099 | 73.6 | 7.4 | 637 | 60.4 |
| 39.25 | 1239 | 0 | 0.0 | 0 | 0.0 | 132 | 2.6 | 147 | 2.4 | 151 | 2.7 | 143 | 2.5 | 0.3 | | |
| 39.56 | 1230 | 268 | 17.8 | 260 | 16.8 | 279 | 15.6 | 315 | 18.7 | 320 | 16.7 | 288 | 17.1 | 1.7 | 337 | 25.2 |
| 41.17 | 1191 | 0 | 0.0 | 216 | 4.2 | 247 | 12.9 | 0 | 0.0 | 241 | 7.3 | 235 | 8.1 | 0.8 | 227 | 18.0 |
| 41.83 | 1172 | 437 | 30.9 | 401 | 29.4 | 377 | 25.2 | 378 | 23.3 | 433 | 29.4 | 405 | 27.6 | 2.8 | 311 | 20.8 |
| 44.4 | 1111 | 186 | 5.3 | 0 | 0.0 | 243 | 5.4 | 225 | 9.7 | 195 | 6.2 | 212 | 6.7 | 0.7 | | |
| 44.87 | 1099 | 174 | 2.5 | 0 | 0.0 | 0 | 0.0 | 153 | 2.4 | 0 | 0.0 | 163 | 2.5 | 0.2 | | |
| 45.81 | 1075 | 129 | 1.6 | 144 | 2.3 | 0 | 0.0 | 0 | 0.0 | 0 | 0.0 | 137 | 2.0 | 0.2 | | |
| 47.15 | 1041 | 187 | 3.6 | 193 | 3.6 | 217 | 4.4 | 0 | 0.0 | 203 | 6.2 | 200 | 4.5 | 0.4 | | |
| 47.5 | 1031 | 192 | 7.0 | 163 | 5.0 | 166 | 3.1 | 0 | 0.0 | 198 | 4.5 | 180 | 4.9 | 0.5 | | |
| 48 | 1018 | 0 | 0.0 | 155 | 3.2 | 168 | 2.8 | 0 | 0.0 | 0 | 0.0 | 161 | 3.0 | 0.3 | | |
| 49.63 | 976 | 132 | 2.0 | 0 | 0.0 | 139 | 2.5 | 0 | 0.0 | 0 | 0.0 | 135 | 2.2 | 0.2 | | |
| 50.3 | 959 | 209 | 8.2 | 256 | 15.6 | 236 | 12.6 | 220 | 11.9 | 227 | 12.1 | 230 | 12.1 | 1.2 | | |
| 52.49 | 902 | 254 | 3.9 | 0 | 0.0 | 215 | 4.8 | 184 | 5.9 | 233 | 7.7 | 222 | 5.6 | 0.6 | | |
| 54.35 | 852 | 0 | 0.0 | 0 | 0.0 | 0 | 0.0 | 155 | 2.3 | 249 | 5.2 | 202 | 3.7 | 0.4 | | |
| 55.65 | 819 | 184 | 8.8 | 193 | 7.3 | 191 | 6.7 | 181 | 7.1 | 249 | 5.2 | 200 | 7.0 | 0.7 | | |
| 58.26 | 749 | 155 | 3.2 | 202 | 3.4 | 0 | 0.0 | 201 | 6.6 | 0 | 0.0 | 186 | 4.4 | 0.4 | | |
| 60.72 | 684 | 287 | 12.9 | 216 | 14.0 | 243 | 7.8 | 0 | 0.0 | 230 | 4.9 | 244 | 9.9 | 1.0 | | |
| 64.49 | 577 | 528 | 36.0 | 0 | 0.0 | 430 | 25.8 | 367 | 21.4 | 393 | 23.3 | 430 | 26.6 | 2.7 | | |
| 65.74 | 541 | 287 | 19.1 | 274 | 12.7 | 283 | 20.5 | 306 | 21.5 | 304 | 16.3 | 291 | 18.0 | 1.8 | | |
| 68.41 | 465 | 132 | 2.9 | 0 | 0.0 | 182 | 4.4 | 0 | 0.0 | 0 | 0.0 | 157 | 3.7 | 0.4 | | |
| 69.41 | 436 | 194 | 10.7 | 168 | 3.8 | 0 | 0.0 | 207 | 11.1 | 233 | 9.1 | 201 | 8.7 | 0.9 | | |
| 72.38 | 352 | 0 | 0.0 | 172 | 4.7 | 203 | 5.3 | 0 | 0.0 | 188 | 5.8 | 188 | 5.3 | 0.5 | | |
| 73.13 | 331 | 0 | 0.0 | 169 | 4.1 | 152 | 2.8 | 0 | 0.0 | 0 | 0.0 | 160 | 3.5 | 0.3 | | |
| 73.95 | 304 | 0 | 0.0 | 0 | 0.0 | 171 | 3.7 | 190 | 5.7 | 0 | 0.0 | 180 | 4.7 | 0.5 | | |
| 76.13 | 235 | 205 | 12.1 | 258 | 20.0 | 237 | 21.7 | 287 | 23.8 | 262 | 13.0 | 250 | 18.1 | 1.8 | | |
| 77.17 | 206 | 179 | 5.4 | 206 | 15.4 | 211 | 12.5 | 219 | 13.2 | 272 | 13.5 | 217 | 12.0 | 1.2 | | |
| 78.31 | 172 | 250 | 15.3 | 0 | 0.0 | 156 | 4.3 | 203 | 5.4 | 219 | 7.7 | 207 | 8.2 | 0.8 | | |
| 79.98 | 125 | 165 | 4.4 | 187 | 3.7 | 0 | 0.0 | 162 | 3.2 | 167 | 3.3 | 170 | 3.7 | 0.4 | | |
| 84.5 | -4 | 202 | 9.8 | 199 | 7.7 | 222 | 5.0 | 0 | 0.0 | 188 | 7.9 | 203 | 7.6 | 0.8 | | |
| 85.44 | -37 | 0 | 0.0 | 155 | 4.4 | 0 | 0.0 | 0 | 0.0 | 240 | 8.6 | 197 | 6.5 | 0.7 | | |
| 87.89 | -128 | 236 | 11.2 | 212 | 9.6 | 270 | 12.9 | 244 | 12.1 | 0 | 0.0 | 241 | 11.4 | 1.1 | | |
| 89.28 | -173 | 0 | 0.0 | 0 | 0.0 | 0 | 0.0 | 190 | 5.6 | 164 | 3.7 | 177 | 4.7 | 0.5 | | |
| 90.53 | -214 | 276 | 18.8 | 286 | 26.1 | 278 | 16.5 | 296 | 18.1 | 241 | 6.9 | 275 | 17.3 | 1.7 | | |
| 91.72 | -251 | 0 | 0.0 | 0 | 0.0 | 0 | 0.0 | 227 | 10.4 | 244 | 12.5 | 236 | 11.4 | 1.1 | | |
| 94.83 | -347 | 0 | 0.0 | 191 | 4.6 | 198 | 5.9 | 216 | 8.7 | 0 | 0.0 | 201 | 6.4 | 0.6 | | |
| 97.16 | -426 | 331 | 22.6 | 228 | 15.4 | 403 | 35.2 | 436 | 48.5 | 675 | 75.0 | 414 | 39.3 | 3.9 | | |
| 97.31 | -431 | 0 | 0.0 | 131 | 2.9 | 0 | 0.0 | 0 | 0.0 | 0 | 0.0 | 65 | 1.5 | 0.1 | | |
| 100.19 | -529 | 219 | 12.1 | 224 | 6.6 | 0 | 0.0 | 247 | 15.9 | 235 | 7.7 | 231 | 10.6 | 1.1 | | |

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470 **Table 3** – Statistics on sulfate signal for identical peaks in core 1, 2, 3, 4 and 5. Geometric
 471 standard deviations are calculated on peaks heights (i.e maximum concentration reached, in
 472 ng.g⁻¹) and on peaks sulfate flux (i.e total mass of volcanic sulfate deposited after the
 473 eruption). Background corrections are based on background values calculated separately for
 474 each volcanic event.

| Study | Number of compared cores | Geom. std deviation based on maximum concentration | Geom std deviation based on deposition flux |
|------------------|--------------------------|--|---|
| Wolff and others | 2 | 1.5 | |
| This study | 2* | 1.35 | 1.56 |
| This study | 5 | 1.21 | 1.41 |

475 * : C_x/C_1 , with x=2,3,4,5

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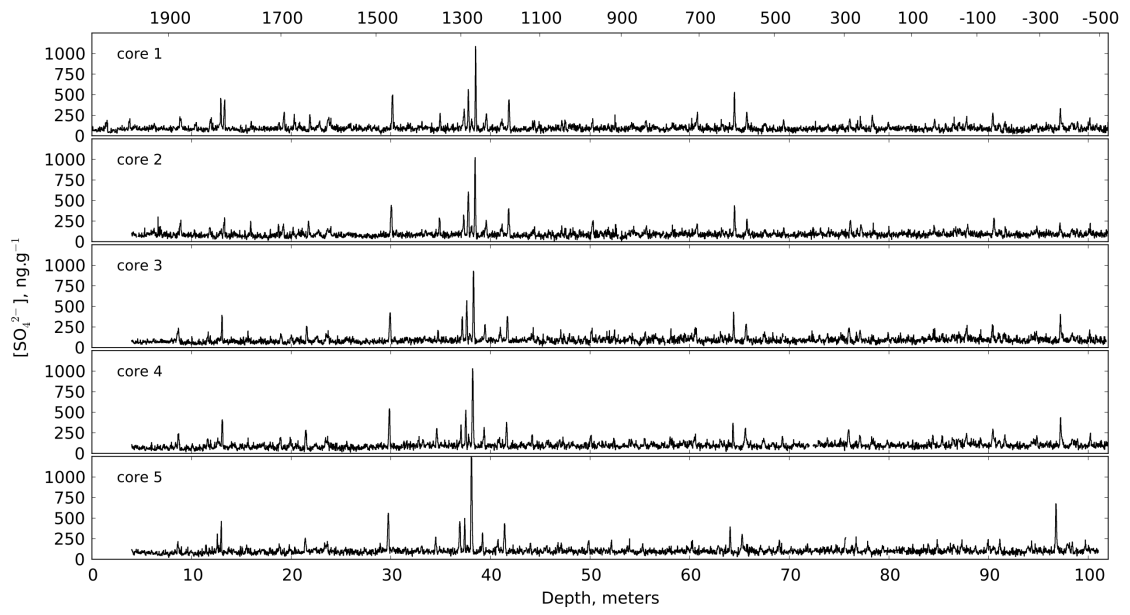
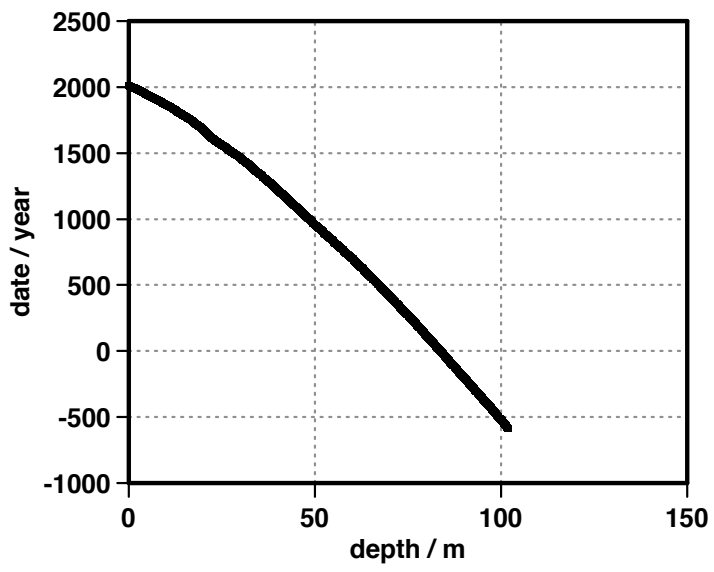


Figure 1 - Sulfate profiles on the 5 replicate cores obtained during a drilling operation at Dome C – Antarctica in 2011.

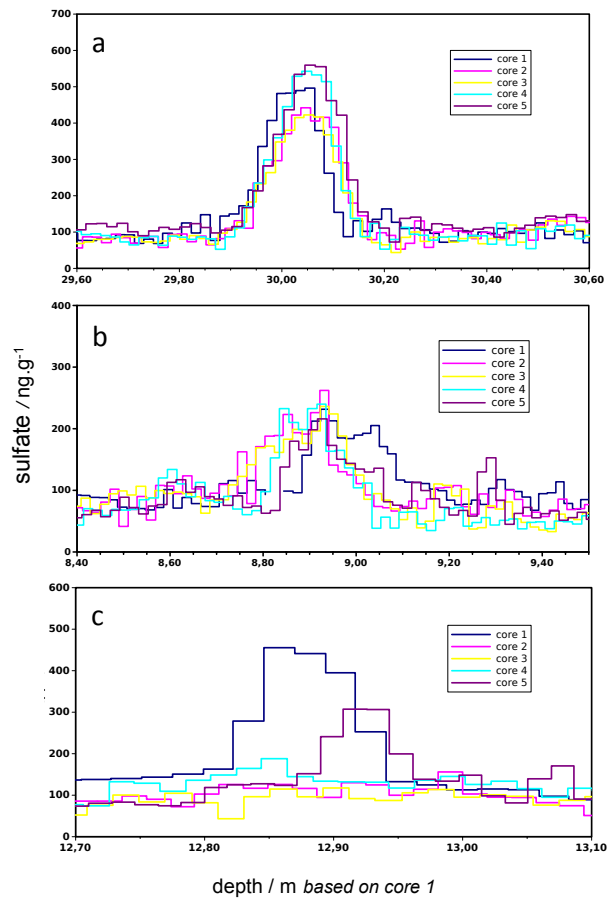


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512 **Figure 2** - Age versus depth in core 1 drilled in 2011 CE, Dome C – Antarctica

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516 **Figure 3** –Kuwae (a, top), Krakatoa (b, middle) and Tambora (c, bottom) sulfate

517 concentration profiles after depth synchronization. All peaks are within a 20 cm uncertainty,

518 enabling to clearly attribute each occurrence to a single event.

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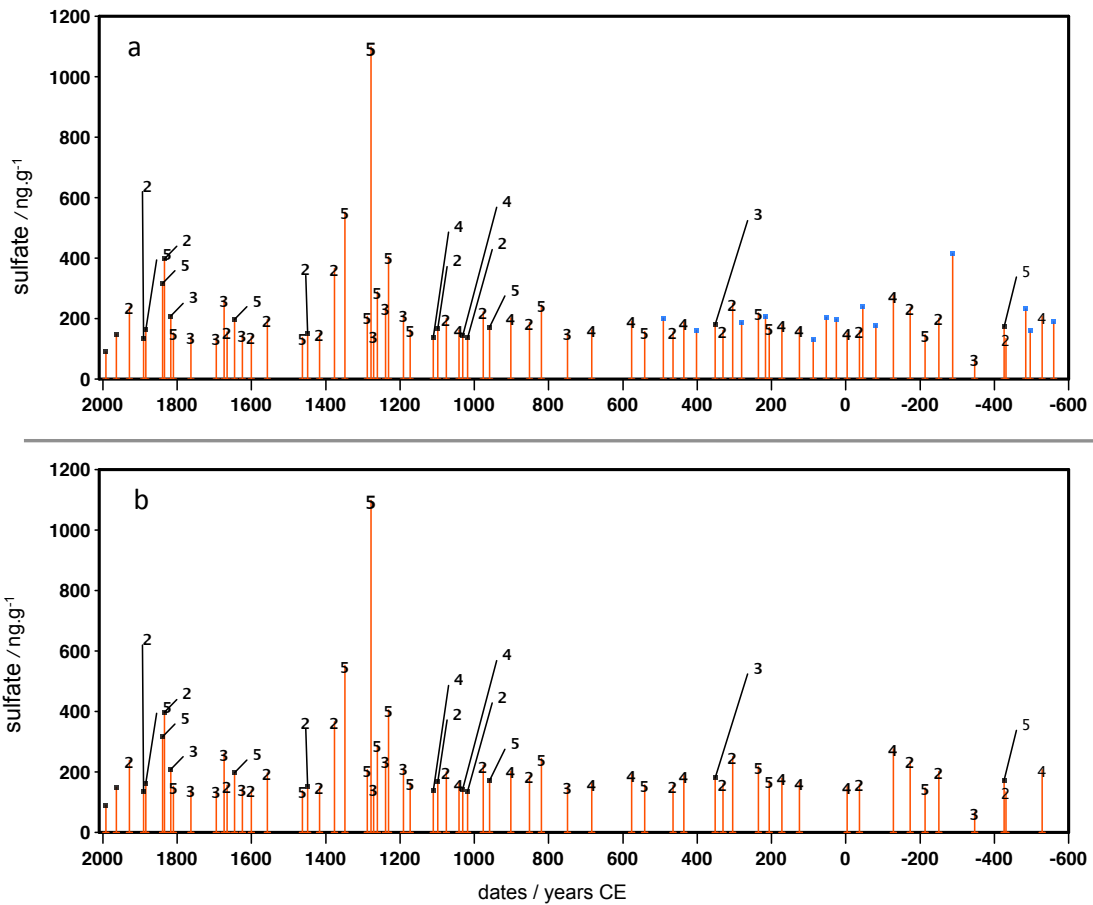
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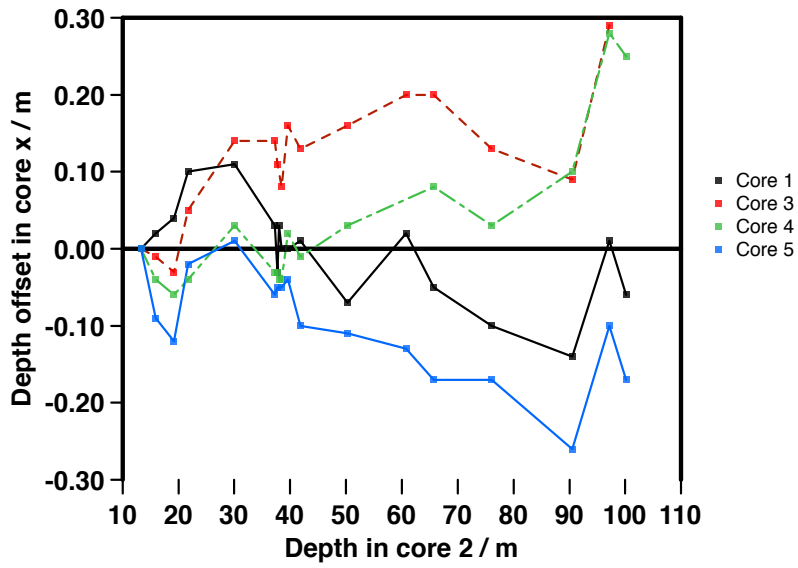


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531 **Figure 4** – a) Composite sulfate peak profile deduced from our statistical analysis of the 5
532 cores using our detection peak and synchronization algorithms (see text). The numbers
533 indicate the number of time a common peak is found in the cores. Unnumbered peaks, peaks
534 found only in single core. b) same as a) without the single detected peaks. All the remaining
535 peaks are considered as volcanic eruptions. See Table 2 for details.

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540 **Figure 5** – Depth offset of 18 common and well-identified volcanic events in cores 1, 3, 4 and

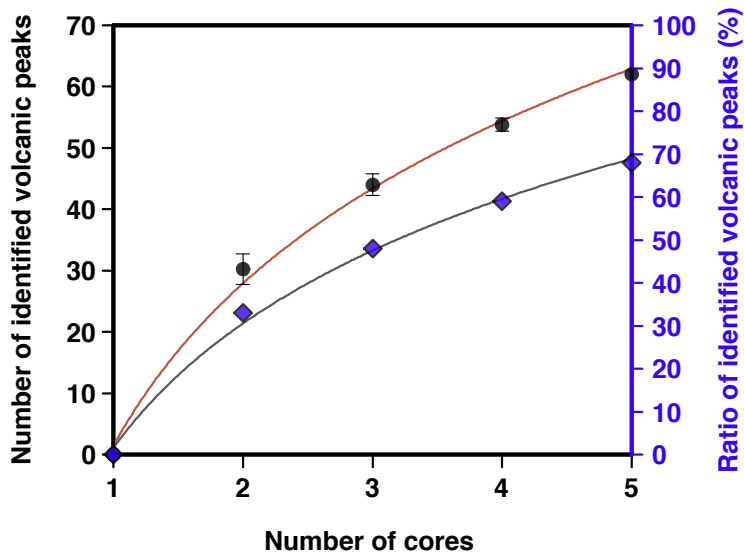
541 5 relative to core 2. To overcome offset due to the drilling process and poor core quality on

542 the first meters, UE 1809 (depth ca. 13 m) is taken as the origin and horizon reference.

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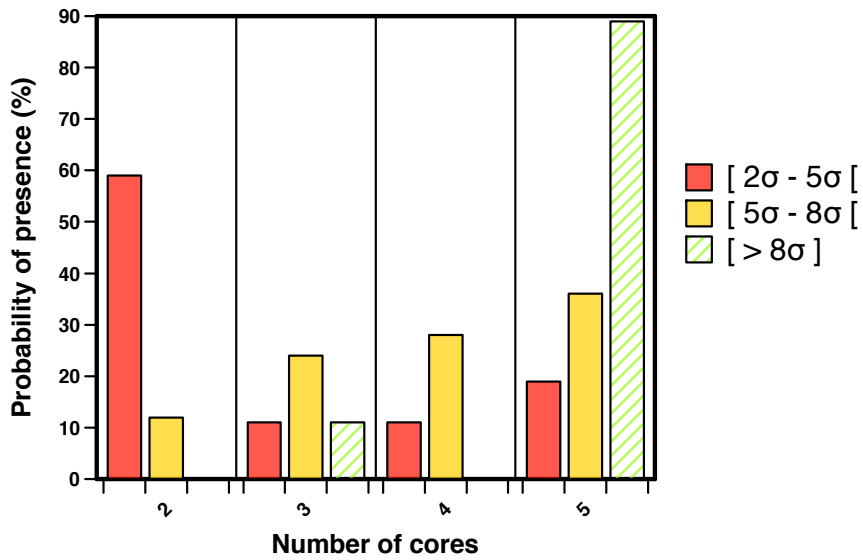
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548 **Figure 6** – Black dots with red line (left axis) represent the number of sulfate peaks that can
549 be identified as volcanic peaks in a composite profile, made of n cores (with n ranging from 1
550 to 5). A sulfate peak appearing simultaneously in at least two cores is considered to be a
551 volcanic peak. Blue diamonds represent the ratio of identified volcanic peaks, i.e the number
552 of identified volcanic peaks (plotted on the left axis), relatively to the total number of sulfate
553 peaks (no discrimination criteria) in a composite made of 5 cores. In our case, the 5 ice-cores
554 composite comprises 91 sulfate peaks (Agung and Pinatubo excluded). With two cores, only
555 33% of them would be identified as being volcanic peaks (detected in both cores), while 68%
556 of them can be identified as volcanic events using 5 cores.

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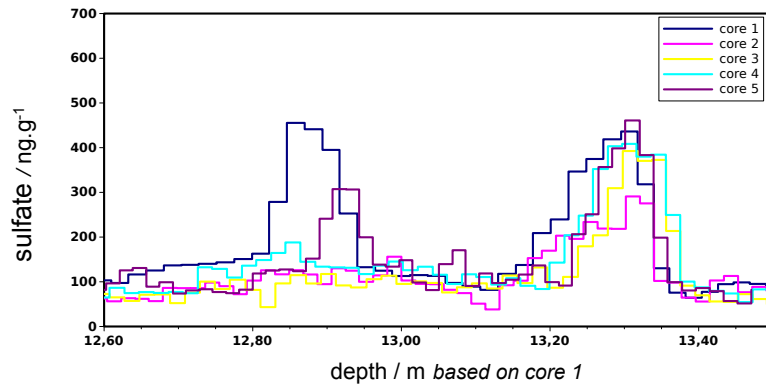


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Figure 7 - Peaks probability to be detected in 2, 3, 4 or 5 cores, as function of their flux. The three categories of flux are defined by peaks flux value, relatively to the average flux, and quantified by x time (2, 5 and 8) the flux standard deviation, calculated for a 30 ppb standard deviation in concentrations.

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585 **Figure 8:** Close look at UE 1809 and Tambora (1815) events showing the absence of the
586 Tambora event in 2 out of the 5 cores. This figure illustrates the possibility of missing major
587 volcanic eruptions when a single core is used.

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589 **SOM**

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591 1. Gfeller *et al.* (2014) approach on Dome C 5 cores: calculation of the representativeness

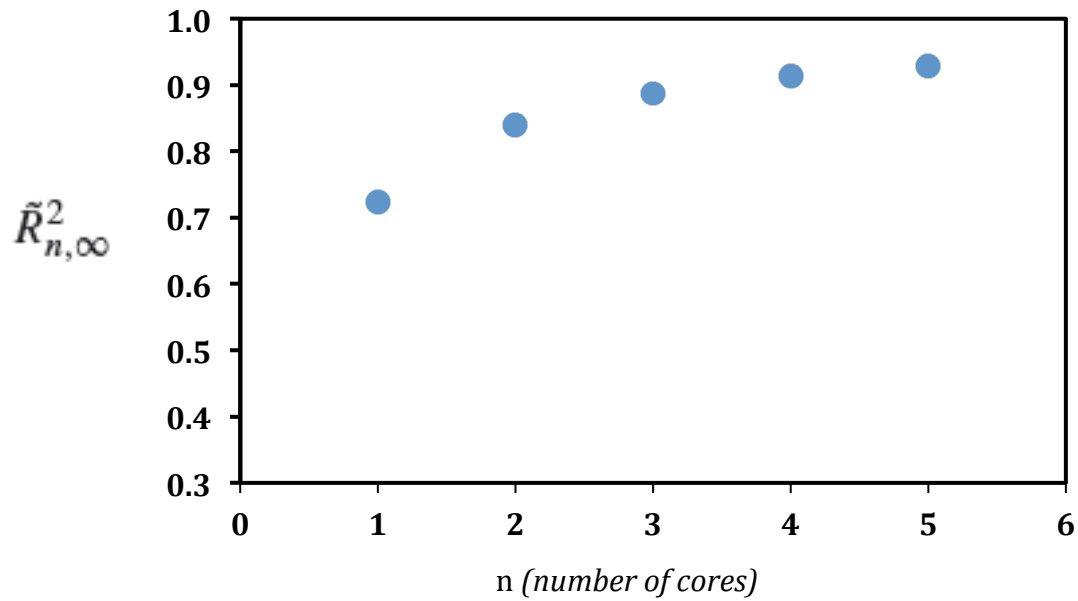
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| n (number of cores) | 1 | 2 | 3 | 4 | 5 |
|---|-------------|-------------|-------------|-------------|-------------|
| $\tilde{R}_{n,\infty}^2 \text{SO}_4^{2-}$ | 0.72 | 0.84 | 0.89 | 0.91 | 0.93 |

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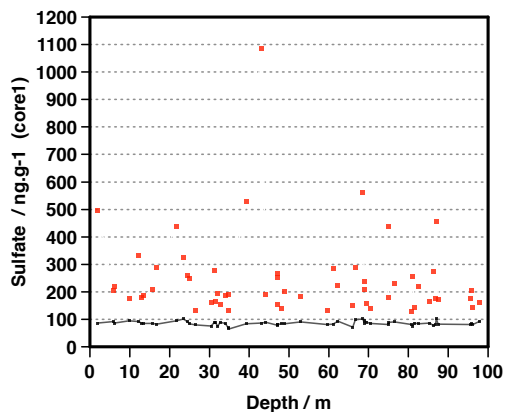
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599 Figure S1: Representativeness of sulfate in the cores depending on the number of cores n (based on

600 Gfeller et al., 2014 approach).

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603 Figure S2 - Variation of the background along depth in core 1, red dots are detected peaks, the dark

604 line stands for the background concentration.

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