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1     **Arc volcano activity driven by small-scale metasomatism**  
2                                   **of the magma source**

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29 Abstract

30

31 Arc volcanism arises from the release of fluids from descending slab, which lowers mantle  
32 solidus and enables melting. Mantle metasomatism plays an important role in magma  
33 production and arc volcanic activity at long spatial and temporal scales. However, the episodic  
34 eruption of individual volcanoes is generally thought to be primarily regulated by the  
35 evolution and recharge of crustal magma chambers, with no established link with mantle  
36 processes. Using high-resolution isotope time-series we show here a link between the most  
37 recent 1999-2016 eruptive activity of the Tungurahua volcano in Ecuador and the small-scale  
38 metasomatic enrichment of the magma source. During the eruption the average rate of  
39 magma production varies logarithmically with the degree of source enrichment. Such a  
40 correlation points to a major control of source enrichment on magma supply and eruptive  
41 activity, which is known so far only for basaltic shield volcanoes. The data also clearly show  
42 that the 1999-2016 eruption sequence ended because of the exhaustion of the enriched  
43 source during the very last weeks. Our findings suggest that the metasomatic fertilization of  
44 the mantle wedge over short spatial and temporal scales regulates magma production, and  
45 ultimately the last eruptive cycle of Tungurahua volcano. This is also a plausible mechanism  
46 to produce long-lasting eruptions at andesitic volcanoes.

47

48

49 Volcanoes are fed by vertically extensive magmatic systems rooted at the crust-mantle  
50 boundary<sup>1</sup>. In arc setting, potentially large magma chambers may form atop these trans-  
51 crustal systems and feed volcanic eruptions. Depth, temperature, and evolution of the shallow  
52 magma reservoirs are reasonably well constrained from phase equilibria experiments<sup>2-4</sup>,  
53 mineral chemistry<sup>5-7</sup>, and the temporal-compositional trends in the erupted magmas<sup>8,9</sup>.  
54 Conversely, owing to the shallow magma chamber filter, the deep roots of arc magmatic  
55 systems are less well known. Thermal and physical models point these regions (the so-called  
56 “deep crustal hot zones”) as the main place where arc magmas accumulate, differentiate and  
57 segregate from their source<sup>10</sup>. Magma production in the mantle and deep transfer processes  
58 are also increasingly thought to control the recharge and evolution of shallow magma  
59 chambers, and ultimately volcanic activity<sup>11,12</sup>. Here we take advantage of the intense activity  
60 of Tungurahua stratovolcano (Ecuador) between 1999 and 2016, and the evidence from large  
61 scale syn-eruptive uplift of the edifice for continuous replenishment of the shallow magma  
62 chamber<sup>13,14</sup> to constrain the compositional structure of the deep magma source by means of  
63 isotope analysis. We use regularly emitted glassy ash samples which, unlike scarce lavas and  
64 other eruptive products, provide a continuous record of magma composition and allow us to  
65 reconstruct the small-scale distribution of melts in the volcano source and their progressive  
66 extraction during the eruptive sequence. The high-resolution isotope record provides clues as  
67 to how deep processes control the secular activity of the Tungurahua volcano.

68

## 69 **Tungurahua volcano**

70 Tungurahua is an andesitic stratovolcano located in Ecuador along the eastern Andean  
71 Cordillera (Fig. 1a). Since 0.3 Ma the volcano produced mainly basaltic andesite and andesite,  
72 and occasionally dacite, with no compositional evolution through time<sup>15,16</sup>. Since 1532 the

73 volcano showed a frequent activity, with main eruptive phases in 1641-1646, 1773-1781,  
74 1886-1888, 1916-1918 and 1999-2016<sup>17,18</sup>. Extensive multi-disciplinary study of the last 1999-  
75 2016 eruptive sequence allowed the identification of four stages (Fig. 2a): Stage 1 (1999-2005)  
76 started quite intensely with VEI2-3 eruptions in 1999 and 2001<sup>19</sup> and the activity generally  
77 decreased through time<sup>20</sup>. Magma transfer and accumulation resulted in large-scale inflation  
78 of the volcano and passive SO<sub>2</sub> degassing<sup>21,13</sup>. Between January and August 2006 (stage 2), the  
79 injection of volatile-rich andesitic magma into a partially degassed magma chamber triggered  
80 subplinian eruptions that produced Pyroclastic Density Currents (PDC) and lava flows<sup>22-24</sup>.  
81 Following the 2006 paroxysm, the activity generally decreased between 2007 and mid 2009  
82 (stage 3), especially after the February 2008 event that produced PDCs and small lava flows.  
83 The activity became less continuous between 2010 and 2016 (stage 4), decreasing first until  
84 2012, before increasing markedly from mid-2013 to mid-2014. Then the activity strongly  
85 decreased before a last burst between December 2015 and March 2016. The eruptive activity  
86 ended suddenly in March 2016 and only cold fumaroles occurred at the summit from 2017.  
87 The less continuous activity after 2009 was interpreted as sequential plugging of the system  
88 possibly due to lower magma feeding rate<sup>25</sup>. Although few isotope data from 2006 lavas were  
89 previously reported<sup>16</sup>, no study so far investigated isotope variations over the eruptive  
90 sequence, and generally the role of magma source composition. In particular, the deep source  
91 processes responsible for the onset and sudden end of this major eruptive episode remain  
92 poorly known. Between November 1999 and March 2016, Tungurahua ash samples were  
93 collected monthly to daily at the Choglontus station located below the main plume dispersion  
94 axis (W-SW) and far from main roads (Figs. 1b and 2b). Ninety-eight samples were analyzed  
95 for Pb and Sr isotope compositions (see Methods).

96

97 **Mass balance model**

98 Lead and strontium isotope compositions of ash samples are reported in supplementary Table  
99 S1 and plotted against time in Fig. 3. The isotope compositions of ashes show rapid variations,  
100 but no clear temporal trend over the 1999-2016 eruption sequence, beyond the decrease of  
101  $^{87}\text{Sr}/^{86}\text{Sr}$  and  $^{206}\text{Pb}/^{204}\text{Pb}$ , and the increase of  $^{207}\text{Pb}/^{206}\text{Pb}$  during the last six months. The  
102 variations of raw isotopic ratios have no simple interpretation because each isotope ratio is  
103 influenced by more than two sources in Ecuador volcanism<sup>26</sup>. However, the three isotope  
104 dimensions of the Pb system can be used to disentangle the different magma sources. Three  
105 main components contribute to the Pb isotopic composition of Tungurahua magmas<sup>16</sup>: (1)  
106 Depleted mantle. (2) A chemically enriched component derived from slab dehydration and/or  
107 melting. This component may occur as pyroxenite veins in the sub-arc mantle<sup>27,28</sup>, or as later  
108 products of melt-rock reactions, such as clinopyroxene-and/or amphibole-bearing cumulates  
109 in the lower crust<sup>29,30</sup>. This component will be referred to as enriched deep component given  
110 the uncertainty on its exact nature and location. It will also be called “fertile” due to its lower  
111 melting point. (3) Upper continental crust assimilated by mantle derived magmas. The later  
112 crustal component must itself include a radiogenic and an unradiogenic end-member to  
113 account for the large Pb isotope heterogeneity of basement rocks<sup>16</sup>. Although magma  
114 components mix at the liquid state, mass balance calculation and results will refer to parental  
115 rocks to circumvent the uncertainty related to melting. All compositions used in the model are  
116 given in Table 1 (see Methods). The mass-balance equations were solved in three-dimension  
117  $^{204}\text{Pb}$ -normalized isotope space using matrix formulation (see Methods).

118

119 **Magma source components versus eruptive activity**

120 The cumulative volume of magma and derived magma production rate (see Methods) are  
121 plotted against time (Fig. 4a, b) together with the computed mass fractions of the four source  
122 components (Fig. 4c-f). A major observation is that the depleted mantle and the enriched deep  
123 component show antithetic temporal variations related to the main eruption stages. For  
124 instance, the contribution of the enriched deep component peaked during the 2006 eruption  
125 paroxysm (up to 70%), before decreasing between 2007 and mid-2009 (Stage 3), down to 23%  
126 in June 2009, as the eruption activity decreased (Fig. 4d). The contribution of the enriched  
127 deep component increased again during early stage 4, reaching 60% in August 2012 while the  
128 activity had become again nearly continuous (between November 2011 and September 2012).  
129 It stayed elevated (about 50%) during the following period of high activity (mid-2013 – mid-  
130 2014) before decreasing at the end of the eruptive sequence, in particular during the last six  
131 months (Fig. 4d). Remarkably, the fraction of the enriched deep component was the lowest  
132 (12.6%) during the very last weeks of the sequence (February 27 to March 14, 2016), strongly  
133 suggesting that the 1999-2016 eruption sequence of Tungurahua ended because of the  
134 exhaustion of the enriched component in the mantle source. A corollary is that the whole  
135 1999-2016 eruption sequence must result from the melting of a small fertile domain, whether  
136 in the crust or in the mantle. On the other hand, the highly correlated ( $r^2=0.88$ ) contributions  
137 of the two crustal components do not show clear temporal trends, but generally more  
138 variability during the first two years of the sequence, and between 2006 and 2009 (Fig. 4e, f).  
139 To evaluate statistically the apparent relation between magma source composition and  
140 eruption activity, we have analyzed the correlations between the deep source components  
141 and the rate of deep magma supply. Figure 5 shows that magma supply rate correlates  
142 positively ( $r^2=0.61$ ) with the enriched deep component and negatively ( $r^2=0.62$ ) with the  
143 depleted mantle component. Moreover, it is important to note that linear variations of

144 magma source composition result in logarithmic variations of magma production rate,  
145 demonstrating that even small variations of mantle fertility have a strong influence on magma  
146 supply and eruptive activity. For example, over the course of the 1999-2016 eruption  
147 sequence, a factor of 3 variation of the enriched component resulted in a factor of 300  
148 variations of magma production rate (Fig. 5a). Although the correlations shown in Fig. 5 are  
149 highly significant, they nevertheless show some scatter of the data. This scatter most likely  
150 results from magma chamber and conduit processes occasionally uncoupling source and  
151 surface processes. Such decoupling is especially apparent at the very beginning of the eruption  
152 (1999-2001) (Fig. 4 b-d), where high magma production rate likely resulted from the  
153 depressurization of the magma reservoir, whereas magma composition likely reflected mixing  
154 between old unerupted magmas from the last 1916-1918 eruption stored in the shallow  
155 reservoir and new magmas rising from the mantle. During stage 2 and 3, the tight coupling  
156 between eruption activity and source composition are consistent with steady-state supply and  
157 open-vent conditions. Conversely, during stage 4 the increase of the fertile component  
158 predated the peak of surface activity (July 2013 – April 2014) possibly because the plugging of  
159 the conduit<sup>25</sup> increased magma storage and mixing before eruption.

160

## 161 **Implications**

162 Arc volcanism arises from the input of slab fluids that changes mantle composition, lowers its  
163 solidus and enables partial melting. In Ecuador, the variations of mantle metasomatism in  
164 space and time are responsible for the compositional variations of magmas across<sup>26,30-32</sup> and  
165 along<sup>26,28</sup> the arc, and the chemical evolution of long-lived volcanic complexes<sup>33,34</sup>. Using high-  
166 resolution Pb isotope time-series we show here for the first time a tight link between small  
167 scale metasomatic enrichment of the magma source, and the short-term eruptive activity of



168 a volcano. This link is especially apparent after the early draining of the shallow magma  
169 reservoir (1999-2001 magma surge) once the volcano is directly fed from the deep magma  
170 source (Fig. 4). Such a link between magma source fertility and eruptive activity is known for  
171 basaltic shield volcanoes, which are connected to their mantle sources by relatively simple  
172 feeding systems<sup>35,36</sup>, but is unexpected for Tungurahua stratovolcano that includes several  
173 zones of magma storage between 1 and 15 km depth<sup>13,14,22,37,38</sup>. In the context of continental  
174 arc volcanism, the direct connection between mantle or lower-crust and eruption at the  
175 surface has two major requirements. First, a process must convert the necessarily slow  
176 melting processes into high melt flux to the shallow magma chamber. Reactive melt transport  
177 has been shown to meet this requirement beneath mid-ocean ridges<sup>39</sup> as well as in arc  
178 setting<sup>40</sup>. This process is fundamentally based on the chemically unstable nature of primary  
179 melts. In arc setting, primary melts with high silica and water contents dissolve olivine and  
180 crystallize pyroxene<sup>41</sup>. Pyroxene is subsequently replaced by amphibole as the melt-rock  
181 reaction proceeds near the mantle-crust boundary<sup>29</sup>. The dissolution-precipitation reactions  
182 increase locally the porosity and permeability of the mantle and lead to the formation of melt  
183 channels. Keller and Katz<sup>40</sup> showed that dissolution of fertile material and magma transport  
184 in such channels are enhanced by up to an order of magnitude. Very importantly, the  
185 formation of melt channels is an unstable process that bridges the gap between the slow  
186 melting of the mantle and the fast melt extraction required to feed sustained volcanic  
187 eruptions. Rawson et al.<sup>42</sup> suggested that reactive channels are likely to develop in the distal  
188 parts of the mantle wedge where more stable thermal and rheological conditions prevail. The  
189 location of Tungurahua volcano along the eastern Cordillera, 330 km away from the  
190 subduction trench, could favor the development of reactive melt channels. The high magma  
191 productivity of Tungurahua volcano might also be related to its location above a major fault

192 in the subducting plate (The Grijalva Fracture Zone) (Fig. 1a), because serpentinized  
193 peridotites exhumed near fracture zones may deliver large amounts of volatiles to the sub-arc  
194 mantle and enhance flux melting<sup>43</sup>.

195 The second requirement is that the inflow of mantle derived magmas is not totally absorbed  
196 by the magma storage system in the crust, so that part of the magma flow directly feeds the  
197 eruptions. In support of such a process, large-scale uplift of the Tungurahua edifice between  
198 2003 and 2014 indicates continuous replenishment of the magma reservoir during the  
199 eruption sequence, with slightly more than half of the magma supplied to the reservoir  
200 reaching the surface<sup>13,14</sup>. Thus, Tungurahua seems to belong to the few stratovolcanoes from  
201 the Central and South American arcs that are directly connected to their mantle source,  
202 together with Irazu volcano in Costa Rica<sup>44</sup> and Ollagüe volcano in Chile<sup>45</sup>. In such a situation,  
203 extensive crustal contamination of Tungurahua magmas<sup>16,26</sup> is at first glance paradoxical  
204 because crustal assimilation is generally thought to be related to magma storage and  
205 differentiation in the crust. The extensive contamination of Tungurahua andesitic magmas can  
206 be reconciled with the evidence for short residence time in the crust if the magmas are  
207 superheated during rapid ascent<sup>45,46</sup>. In support of this hypothesis, the absence of amphibole  
208 in Tungurahua andesites, together with thermometric and experimental constraints suggest  
209 that the magmas evolved at temperatures higher than 950–1000 °C<sup>22,37</sup>. The time scale of  
210 magma ascent is not constrained here, but the residence time of magma in the reservoir must  
211 be comparable to the time scale of geochemical fluctuations<sup>47</sup>, on the order of years (Fig. 3).

212 The view that emerges for Tungurahua is that of a more dynamic magmatic system compared  
213 to the deep crustal hot zones<sup>10</sup> which require timescale of 10<sup>5</sup>–10<sup>6</sup> years for thermal  
214 incubation at depth, and 10–10<sup>3</sup> years for magmatic differentiation from basalt to andesite.  
215 One possible solution is that the trans-crustal plumbing system is a much more dynamic

216 environment than expected, with probably “active” zones with rapid magma ascent and other  
217 zones dominated by magma accumulation and differentiation. In this frame, Tungurahua is a  
218 good candidate to join the growing family of arc volcanoes, where seismic tomography<sup>48</sup>,  
219 volatiles diffusion along crystal embayments<sup>49</sup>, and Ra-Th isotope disequilibria<sup>50</sup> indicate ultra-  
220 fast magma ascent in well-developed melt channels. A major outcome of this study is that  
221 eruption of the Tungurahua volcano requires at least 12% of fertile lithology in its source.  
222 Below 12%, melt production is likely too low to sustain magma eruption at the surface, as  
223 illustrated by the exhaustion of the fertile component a few weeks before the end of the 1999-  
224 2016 eruption sequence. Conversely, when the fraction of the fertile lithology reaches 60-  
225 70%, reactive channeling instabilities allow high melt supply to the volcano, and trigger  
226 eruption paroxysm, such as in 2006. In support of such process, magma production increases  
227 logarithmically with the degree of source enrichment (Fig. 5). The nature and location of the  
228 fertile source lithology (i.e., the enriched deep component) remain uncertain, although its  
229 isotopic signature requires a genetic link with the subducting oceanic crust. In any case, the  
230 fact that the fractions of the enriched deep component and the depleted mantle are strictly  
231 complementary (Fig. 4c,d) points to a mantle origin for the isotopic signal correlating with  
232 volcanic activity. In such situation, our preferred interpretation is that metasomatic  
233 fertilization of the mantle wedge over short spatial and temporal scales regulates magma  
234 production rates and ultimately the secular activity of the volcano. Small-scale variations of  
235 mantle metasomatism in southern Ecuador is independently evidenced by olivine-hosted melt  
236 inclusions<sup>28</sup>, and could be related to variations of the thermal structure and composition of  
237 the subducting plate in the vicinity of the Grijalva Fracture Zone.

238 This work confidently resolves a link between the short-term activity of an arc volcano and  
239 the small-scale heterogeneity of its magma source. This observation leaves some open

240 questions regarding the geometry of the melting source and how melting and melt extraction  
241 occur. For instance, the episodic activity of arc volcanoes could reflect the melting of individual  
242 fertile veins forming some kind of fractal tree in the mantle wedge. In such case, the spatial  
243 distribution and the size of the veins, and the re-organization of melt channels driven by the  
244 slow decompression of the mantle, will control melt production and volcano supply.  
245 Alternatively, if each volcano is fed by a main melt channel in the mantle wedge<sup>51</sup>, short-term  
246 variations in fluid delivery from the slab and related flux melting are expected to govern melt  
247 production and volcano supply.

248 On a more practical point of view, the results of this work suggest that volcano monitoring  
249 techniques should be designed to track the deeper process of the magmatic plumbing system.  
250 For instance, monitoring the isotopic composition of juvenile ashes during a long-lived  
251 eruption, or an eruption sequence, would provide information on how the fertility of the  
252 magma source evolves as the eruption proceeds. Such information could be used to forecast  
253 eruption paroxysm, major changes of activity, or eruption end. The volatility of Pb in magmatic  
254 systems also raises the possibility to probe the hidden magmatic evolution of quiescent  
255 volcanoes from the Pb isotope composition of volcanic gases<sup>52</sup>.

256

257 **Figure legends**

258

259 Fig. 1: Location maps. (a) Regional map showing the location of the Tungurahua volcano (star)  
260 along the eastern Cordillera, other main volcanoes of the arc (triangles), the oceanic Grijalva  
261 Fracture Zone and its projection on land (dashed line). (b) Map of Tungurahua volcano  
262 showing the sampling location.

263

264 Fig. 2: History and sampling of the 1999-2016 eruption sequence of the Tungurahua volcano.  
265 (a) Periods of eruption and inactivity are shown in black and white, respectively. The four main  
266 eruption stages (S1-S4) are indicated<sup>25,53</sup>. (b) Ash sampling during the sequence.

267

268 Fig. 3: Pb and Sr isotopic composition of ash samples plotted against time. Error bars (S.D.) are  
269 within symbol size. The four main eruption stages (S1-S4) are indicated<sup>25,53</sup>.

270

271 Fig. 4: Temporal evolution of eruptive activity and magma source components. The four main  
272 eruption stages (S1-S4) are indicated<sup>25,53</sup>. (a) Cumulative volume of magma. (b) Rate of deep  
273 magma supply estimated by calculating the slope of the cumulative volume emission curve  
274 over a moving two-year window (see Methods). The high magma production rate during the  
275 first years (1999-2001) results from the depressurization of the plumbing system (shaded  
276 area), whereas subsequent eruptive activity can be related to melt production at depth. (c-f)  
277 Mass fractions of magma source components are plotted versus time: (c) Depleted mantle.  
278 (d) Slab-derived enriched deep component, located either in the mantle or in the lower crust.  
279 (e&f) Upper crustal components including Pb-poor radiogenic end-member and Pb-rich

280 unradiogenic end-member. Component compositions and model equations are given in the  
281 methods.

282

283 Fig. 5: Rate of deep magma supply plotted against magma source components. The rate of  
284 deep magma supply was estimated by calculating the slope of the cumulative volume emission  
285 curve over a moving two-year window (see Methods). The rate of deep magma supply is  
286 plotted against (a) the mass fraction of the enriched deep component, and (b) the mass  
287 fraction of the depleted mantle component. Linear regressions and correlation coefficients at  
288 95% confidence level are indicated.

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440

441 **Author contribution**

442 I.V. designed the study, coordinated the analyses and wrote the paper

443 N.S. performed the isotopic analyses and wrote the paper

444 P.S. designed the study and contributed to data interpretation

445 B.B. collected samples and contributed to data interpretation

446 F.N. and S.H. contributed to the discussions and interpretation of the data

447 D.A. supervised Sr isotope measurements

448 M.G. supervised Pb isotope measurements

449

450 **Supplementary information**

451 Supplementary Table S1

452

453 **Competing interests**

454 The authors declare no competing interests.

455

456 **Methods**

457

458 **Samples.** Ash fallouts were sampled using ash collectors before 2010 and an ashmeter device  
459 subsequently<sup>54</sup>. The 1999-2016 ash samples are dominantly juvenile. They include black and  
460 brown scoria, 100 to 600  $\mu\text{m}$  in size, larger beige pumices (1000 to 5000  $\mu\text{m}$ ) and glass shards.  
461 Ashes matrix is glassy to micro-crystallized and shows a uniform mineralogical assemblage  
462 (plagioclases + orthopyroxenes + clinopyroxenes + Fe-Ti oxides  $\pm$  olivines). These observations  
463 are consistent with previous descriptions<sup>22,23,53</sup>.

464

465 **Analytical methods.** All chemical analyses were performed at the Laboratoire Magmas et  
466 Volcans (Clermont-Ferrand, France) between 2018 and 2020. Lead and Sr isotope  
467 compositions were measured on 5 mg of juvenile glassy fragments that were hand-picked  
468 under a binocular microscope and washed in deionized water. Following sample dissolution  
469 with  $\text{HNO}_3$ -HF, Pb and Sr were separated using polyethylene columns filled with 50  $\mu\text{L}$  of  
470 Sr.Spec extraction chromatographic material<sup>55</sup>. Full procedural blanks were  $2.7 \pm 1.1$  ng for Sr  
471 ( $n=13$ ) and  $19 \pm 15$  pg for Pb ( $n=9$ ), which are negligible compared to the amounts of Sr (2.1-  
472 3.7  $\mu\text{g}$ ) and Pb (40-75 ng) extracted from the ash samples. Lead isotope compositions were  
473 measured on a Thermo Fisher Scientific Neptune Plus MC-ICP-MS coupled to an Aridus II  
474 desolvating nebulizer system. The Pb instrumental mass fractionation was corrected during  
475 measurement by doping the Pb solutions with the Tl NBS997 standard. The NBS981 standard  
476 was measured between every two samples to correct Pb ratios for instrumental drift and the  
477 slight difference in mass fractionation between Pb and Tl. Data were normalized to the  
478 NBS981 standard using the values of Todt et al.<sup>56</sup> ( $^{206}\text{Pb}/^{204}\text{Pb}=16.9356$ ,  $^{207}\text{Pb}/^{204}\text{Pb}=15.4891$ ,  
479  $^{208}\text{Pb}/^{204}\text{Pb}=36.7006$ ). Repeated analysis of the NBS981 standard every two samples yielded

480 average intra-session reproducibility of 115 ppm (2 S.D.) for the  $^{206}\text{Pb}/^{204}\text{Pb}$  ratio. Strontium  
 481 isotope compositions were measured on a Thermo Fisher Scientific Triton Thermal Ionization  
 482 Mass Spectrometer (TIMS) and were corrected for mass fractionation using  $^{86}\text{Sr}/^{88}\text{Sr}=0.1194$ .  
 483 Repeated measurement of the NBS SRM 987 standard during the course of the study yielded  
 484  $^{87}\text{Sr}/^{86}\text{Sr}=0.710232 \pm 8$  (2 S.D.). Sample data were renormalized to the accepted value of  
 485 0.710245. Repeated measurement (duplicate dissolution) of rock reference materials yielded  
 486  $^{87}\text{Sr}/^{86}\text{Sr}= 0.705004 \pm 14$ ,  $^{206}\text{Pb}/^{204}\text{Pb} = 18.756 \pm 0.005$ ,  $^{207}\text{Pb}/^{204}\text{Pb} = 15.615 \pm 0.004$ ,  
 487  $^{208}\text{Pb}/^{204}\text{Pb} = 38.708 \pm 0.026$  for BCR2 basalt (n=5), and  $^{87}\text{Sr}/^{86}\text{Sr}= 0.703532 \pm 13$ ,  $^{206}\text{Pb}/^{204}\text{Pb}$   
 488  $= 18.301 \pm 0.005$ ,  $^{207}\text{Pb}/^{204}\text{Pb} = 15.536 \pm 0.007$ ,  $^{208}\text{Pb}/^{204}\text{Pb} = 38.249 \pm 0.024$  for JA-1 andesite  
 489 (n=6).

490

491 **Mixing model.** The compositions of the four end-member components used in the mixing  
 492 model are given in Table1.

Table 1

	Depleted mantle	Enriched deep component (arc pyroxenite)	High U/Pb Pb poor crust	Low U/Pb Pb rich crust
Pb (ppm)	0.018	0.06	9	17
$^{206}\text{Pb}/^{204}\text{Pb}$	18.40	19.10	19.20	18.79
$^{207}\text{Pb}/^{204}\text{Pb}$	15.50	15.56	15.72	15.66
$^{208}\text{Pb}/^{204}\text{Pb}$	38.00	38.70	39.22	38.60

493

494 The Pb concentration of the depleted mantle is from ref. <sup>57</sup>. The Pb content of the enriched  
 495 deep component is assumed to be as in clinopyroxenes crystalizing early in arc magmatism<sup>29</sup>.



496 Lead concentrations of basement rocks are from Nauret et al.<sup>16</sup>. Lead isotopic compositions  
 497 of the four components are also from Nauret et al.<sup>16</sup>, with slight modification of the crustal  
 498 compositions. This fine tuning is needed to model the most contaminated samples with high  
 499 <sup>207</sup>Pb/<sup>204</sup>Pb. The Andean continental crust being highly heterogeneous, it is possible that the  
 500 crustal rocks assimilated during the 1999-2016 eruption of the Tungurahua volcano are not  
 501 exactly the same as the five basement samples analyzed by Nauret et al.<sup>16</sup>. The mass fractions  
 502 (x) of the four components are quantified in the <sup>206</sup>Pb/<sup>204</sup>Pb —<sup>207</sup>Pb/<sup>204</sup>Pb—<sup>208</sup>Pb/<sup>204</sup>Pb  
 503 isotope space by solving the mass balance equation system:

504

$$505 \text{ Pb}_1 \cdot x_1 \cdot [(64)_s - (64)_1] + \text{Pb}_2 \cdot x_2 \cdot [(64)_s - (64)_2] + \text{Pb}_3 \cdot x_3 \cdot [(64)_s - (64)_3] + \text{Pb}_4 \cdot x_4 \cdot [(64)_s - (64)_4] = 0$$

$$506 \text{ Pb}_1 \cdot x_1 \cdot [(74)_s - (74)_1] + \text{Pb}_2 \cdot x_2 \cdot [(74)_s - (74)_2] + \text{Pb}_3 \cdot x_3 \cdot [(74)_s - (74)_3] + \text{Pb}_4 \cdot x_4 \cdot [(74)_s - (74)_4] = 0$$

$$507 \text{ Pb}_1 \cdot x_1 \cdot [(84)_s - (84)_1] + \text{Pb}_2 \cdot x_2 \cdot [(84)_s - (84)_2] + \text{Pb}_3 \cdot x_3 \cdot [(84)_s - (84)_3] + \text{Pb}_4 \cdot x_4 \cdot [(84)_s - (84)_4] = 0$$

$$508 x_1 + x_2 + x_3 + x_4 = 1$$

509

510 where (64), (74) and (84) are for <sup>206</sup>Pb/<sup>204</sup>Pb, <sup>207</sup>Pb/<sup>204</sup>Pb and <sup>208</sup>Pb/<sup>204</sup>Pb isotope ratios,  
 511 respectively; Pb is for lead concentration, subscript S is for sample, and subscripts 1-4 are for  
 512 the four mixing components reported in Table 1. The equation system can be written as the  
 513 matrix product:

514

$$\begin{pmatrix} \text{Pb}_1 \cdot [(64)_s - (64)_1] & \text{Pb}_2 \cdot [(64)_s - (64)_2] & \text{Pb}_3 \cdot [(64)_s - (64)_3] & \text{Pb}_4 \cdot [(64)_s - (64)_4] \\ \text{Pb}_1 \cdot [(74)_s - (74)_1] & \text{Pb}_2 \cdot [(74)_s - (74)_2] & \text{Pb}_3 \cdot [(74)_s - (74)_3] & \text{Pb}_4 \cdot [(74)_s - (74)_4] \\ \text{Pb}_1 \cdot [(84)_s - (84)_1] & \text{Pb}_2 \cdot [(84)_s - (84)_2] & \text{Pb}_3 \cdot [(84)_s - (84)_3] & \text{Pb}_4 \cdot [(84)_s - (84)_4] \\ 1 & 1 & 1 & 1 \end{pmatrix} \cdot \begin{pmatrix} x_1 \\ x_2 \\ x_3 \\ x_4 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix}$$

515

516 or :

517

518  $A \cdot B = C$

519

520 the solution matrix B is obtained from

521

522  $B = A^{-1} \cdot C$

523

524 where  $A^{-1}$  is the inverse matrix of A.

525 We stress that the temporal trends inferred from the model (Fig. 4) do not depend on the

526 precise composition of the end-member components used (Table 1). In support of this idea,

527 consistent trends can be inferred from the raw  $^{207}\text{Pb}/^{206}\text{Pb}$  and  $^{207}\text{Pb}/^{204}\text{Pb}$  ratios (Fig. 3b,d),

528 which dominantly record mantle composition and crustal processes, respectively<sup>16,26</sup>.

529

530

531 **Rate of deep magma supply.** The cumulative volume of magma was determined using ash

532 fallout data from ref. <sup>20</sup> for the 1999-2004 period, and ref. <sup>58</sup> for the 2006-2014 period. Ash

533 volumes were converted into magma volumes assuming the density of magma is 2.5 times

534 that of ash deposit. For the August 2006 paroxysmal eruption, we used the complete mass

535 quantification of ref. <sup>59</sup>. For the 2014-2016 period, our ash fallout measurements correspond

536 to  $9.4 \times 10^4 \text{ m}^3$  (09/03/2014-06/22/2015),  $8.7 \times 10^4 \text{ m}^3$  (06/22/2015-09/05/2015),  $8.7 \times 10^5 \text{ m}^3$

537 (09/05/2015-12/08/2015) and  $4.2 \times 10^5 \text{ m}^3$  (02/26/2016-03/15/2016) of magma (B.B.

538 unpublished data and IG-EPN report available at:

539 <https://www.igepn.edu.ec/servicios/noticias/1463-erupcion-de-febrero-marzo-2016-del->

540 volcan). We also included in the calculation the 2010 and 2014 lava flows ( $1 \times 10^6 \text{ m}^3$  each  
541 according to ref. <sup>60</sup> and B.B. unpublished data) and the accumulation of ballistic material in  
542 the summit crater ( $3.4 \times 10^6 \text{ m}^3$  between 2008 and 2012 and  $0.8 \times 10^6 \text{ m}^3$  between 2012 and  
543 2016, which translates into  $2.1 \times 10^6 \text{ m}^3$  and  $0.5 \times 10^6 \text{ m}^3$  of magma, respectively, according to  
544 ref.<sup>59</sup>). Summit crater data are from D. Yaguana 2020 engineering thesis  
545 (<http://bibdigital.epn.edu.ec/handle/15000/21245>). The cumulative volume emission curve  
546 was built with a time resolution of one point per month (Fig. 4a). The rate of deep magma  
547 supply (Fig. 4b) was estimated by calculating the slope of the cumulative volume emission  
548 curve over a moving two-year window (linear regression through 24 data points). A two-year  
549 window was selected because it corresponds to the timescale of source component  
550 fluctuations (Fig. 4c,d). Such a relatively wide time-window is expected to smooth monthly  
551 variations of shallow magma supply, which has varied by a factor of  $10^6$  over the course of the  
552 1999-2006 eruption<sup>20</sup>. The calculated rate of magma supply was assigned to the middle of the  
553 time window.

554

## 555 **Data availability**

556 All data generated during this study are included in this published article (and its  
557 supplementary information files)

558

## 559 **Methods references**

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