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1 **Eruptive chronology of Tungurahua volcano (Ecuador) revisited based on new K-Ar**  
2 **ages and geomorphological reconstructions**

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14 .  
15 **ABSTRACT**

16 This study focuses on the evolution through time of Tungurahua volcano (Ecuador), and  
17 provides new information regarding its history. Eighteen new K-Ar ages constrain its  
18 construction and the activity of its three successive edifices. We show that the volcano is  
19 much younger than expected. Indeed, the older edifice activity only began around  $293 \pm 10$   
20 ka, and ended at  $79 \pm 3$  ka. After  $\sim 50$  ka of quiescence, the second edifice started growing at  
21  $29 \pm 2$  ka after a major sector collapse, and itself collapsed at  $\sim 3$  ka. Since then, the third  
22 edifice filled the amphitheatre and is still active. Together with numerical reconstructions of  
23 the morphology of the three edifices flanks before erosion, these new ages allow us to  
24 quantify the magmatic productivity rates during their construction, from  $0.6 \pm 0.3$  and  
25  $0.9 \pm 0.2$  km<sup>3</sup>/ka for the two older edifices to  $2.5 \pm 1.0$  km<sup>3</sup>/ka for the youngest, as well as an

26 erosion rate of  $0.2 \pm 0.1 \text{ km}^3/\text{ka}$ , occurring since the end of Tungurahua I construction. Major  
27 and trace element contents of lavas from the three edifices display rather similar trends.  
28 Combined with our new ages, the magmatic signature through time does not seem to have  
29 been significantly affected either by the sector collapses experienced by the volcano, or by  
30 changes of the deep magmatic source. Finally, our results show that the K-Ar dating method  
31 by the unspiked Cassinot-Gillot technique performed on groundmass can be successfully  
32 applied to lava flows older than the Holocene, while the uncertainties related to younger units  
33 can prevent an accurate age determination. Particularly, this method can be applied to  
34 Quaternary volcanoes from the Ecuadorian arc, with many of them remaining without  
35 knowledge of the timing of their past activity.

36

37 *Keywords:*

38 Ecuador

39 Tungurahua volcano

40 K-Ar dating

41 Geomorphology

42 Geochemistry

43

## 44 **1. Introduction**

45 The Ecuadorian arc presents a remarkably dense volcanism, composed of about 85  
46 Quaternary volcanic edifices (Hall et al., 2008; Bernard and Andrade, 2011). Twenty-five of  
47 them are potentially active or presently erupting, such as Sangay, Cotopaxi, Reventador and  
48 Tungurahua volcanoes (Fig. 1; Bernard and Andrade, 2011). Tungurahua volcano  
49 (5023 m a.s.l.; Lat.  $01^{\circ}28'S$ ; Long.  $78^{\circ}27'W$ ) is one of the most active volcanoes in the  
50 Andean Northern Volcanic Zone with at least one eruption per century since the 13<sup>th</sup> century  
51 (Le Pennec et al., 2008, 2016). It is characterized by a strombolian to subplinian activity, and

52 experienced at least two major sector collapses during the last 30 ka (Hall et al., 1999; Le  
53 Pennec et al., 2013). Since 1999, the ongoing eruption has seriously threatened surrounding  
54 villages and infrastructures (Samaniego et al., 2008; Le Pennec et al., 2012). The recent  
55 activity is well studied and carefully monitored (e.g., Molina et al., 2005; Arellano et al.,  
56 2008; Kumagai et al., 2011; Hall et al., 2013; Bernard et al., 2014; Hidalgo et al., 2015), but  
57 the timing of its pre-Holocene history remains poorly documented. Reconstructing the past  
58 eruptive history of Tungurahua volcano is essential to better understand its growth and  
59 evolution. This study combines stratigraphic analyses, new K-Ar ages and geomorphometric  
60 reconstructions to better constrain the construction and destruction stages of Tungurahua  
61 volcano through time, and to quantify its magmatic output rates, as well as the erosion rate  
62 occurring during quiescence periods. In addition, geochemical analyses allow us to study the  
63 long-term evolution of the lavas composition. We show in particular that the initial activity of  
64 Tungurahua volcano is much younger than previously thought, and that its construction is  
65 characterized by an increase of the eruptive rate since the last 3 ka BP.

66

## 67 **2. Geological context**

68 The Andean volcanic arc originates from the subduction of the oceanic Nazca plate  
69 beneath the continental South American plate. The arc comprises four distinct zones,  
70 including the Northern Volcanic Zone (NVZ), which lies in Ecuador and Colombia. To the  
71 west of the NVZ, an oceanic plateau, the Carnegie ridge, subducts below Ecuador (Fig. 1a). It  
72 has been proposed that the subduction of this ridge may impact the thermal gradient at depth  
73 and the deep magmatic sources (e.g., Samaniego et al., 2002; Bourdon et al., 2003; Hidalgo et  
74 al., 2012; Ancellin et al., 2017), which could explain the high density of volcanoes observed  
75 in Ecuador (Martin et al., 2014). Tungurahua volcano is located in the southern part of the  
76 Ecuadorian volcanic arc, on the Eastern Cordillera (Fig. 1b). Constructed upon the

77 Palaeozoic-Mesozoic metamorphic basement (Aspden and Litherland, 1992), this andesitic  
78 stratovolcano is surrounded by the Puela (S), Chambo (W) and Pastaza (N) rivers (Fig. 2).  
79 The upper part of the edifice presents steep flanks with 40° slopes, while ~20-25° slope  
80 plateaux incised by deep valleys constitute its lower part, with a progressive change in  
81 steepness. The morphology of the eastern flank shows features of glacial erosion (u-shaped  
82 valleys; Clapperton, 1990; Heine, 2000), as well as deep fluvial incisions due to the tropical  
83 precipitation regime. It contrasts with the western flank which is smoothed by the recent  
84 volcanic deposits. The internal structure and plumbing system of Tungurahua volcano are not  
85 well known. Nevertheless, petrological, melt inclusions, seismic tomography and  
86 interferometric studies suggest the presence of a subvertical upper conduit located above a  
87 shallow magmatic reservoir, supplied by mafic magmas from a deeper reservoir, located  
88 around 5 and 8.5 km below the summit, respectively (Molina et al., 2005; Samaniego et al.,  
89 2011; Kumagai et al., 2011; Champenois et al., 2014; Myers et al., 2014).

90 Previous works showed that Tungurahua volcano was built in three construction stages,  
91 namely Tungurahua I, II and III, and experienced two major sector collapse events (Hall et al.,  
92 1999; Le Pennec et al., 2013, 2016). Inclined plateaux of the northern, eastern and southern  
93 flanks essentially belong to Tungurahua I activity, which started during the Pleistocene, based  
94 on two available whole-rock K-Ar ages of  $770 \pm 5$  and  $350 \pm 4$  ka (Barberi et al., 1988). The  
95 western part of Tungurahua I was destroyed by a first sector collapse,  $^{14}\text{C}$  dated at  
96  $33.1 \pm 0.2$  ka BP (Le Pennec et al., 2013), which might have been triggered by a silicic  
97 cryptodome intrusion (Le Pennec et al., 2006). The resulting amphitheatre volume was  
98 estimated at  $10 \text{ km}^3$  (Bustillos, 2008), but the collapse scar location is not accurately  
99 identified in the present-day edifice morphology. Tungurahua II lavas and deposits then filled  
100 the amphitheatre, before being destroyed in turn by a new western sector collapse  $^{14}\text{C}$  dated at  
101  $2955 \pm 90$  BP (Hall et al., 1999) and  $2960 \pm 30$  BP (Le Pennec et al., 2013). It was triggered

102 by a new viscous intrusion or magma mixing occurring in the chamber (Hall et al., 1999; Le  
103 Pennec et al., 2006, 2013). The avalanche deposits associated with this second event cover  
104 between 23 and 80 km<sup>2</sup>, for a collapse volume estimated between 2.8 and 6 km<sup>3</sup> (Bustillos,  
105 2008 and Hall et al., 1999 respectively). However, the area and the volume obtained by Hall  
106 et al. (1999) are necessarily higher because they mapped as a single sector collapse the  
107 deposits of the two sector collapses of Tungurahua, and the one experienced by Huisla  
108 volcano (Fig. 2; Bustillos, 2008). The southern location of the scar boundary is well delimited  
109 in the morphology, whereas the northern part was first proposed along the Pondoia plateau  
110 (Hall et al., 1999), and recently shifted to the west based on geomorphological and  
111 seismological studies (Molina et al., 2005; Le Pennec et al., 2006; Fig. 2). Due to this collapse  
112 and continuous erosion, remnants of Tungurahua II mainly crop out on the eastern summit  
113 area and in the southwestern valleys (i.e. the Tiacos lava sequences of Hall et al., 1999).  
114 Products from the younger and still active Tungurahua III cone have then filled the  
115 amphitheatre left by the second sector collapse (Hall et al., 1999; Le Pennec et al., 2006,  
116 2013). Lavas and pyroclastic flow deposits, as well as tephra fall and epiclastic layers  
117 displaying basaltic andesite to dacitic composition make up the present western flank. Eight  
118 pre-Columbian eruptive periods were identified, and five major eruptions occurred during  
119 historical times: AD 1640-1641, 1773-1776, 1886-1888, 1916-1918 and 1999-present, with  
120 therefore a recurrence rate of about one pyroclastic flow-forming event per century (Hall et  
121 al., 1999; Le Pennec et al., 2008, 2016).

122         Given that Tungurahua III activity has already been well described and <sup>14</sup>C dated (Hall  
123 et al., 1999; Le Pennec et al., 2006, 2008, 2013, 2016), in this study we focus on the two older  
124 edifices, Tungurahua I and II.

125

### 126 **3. Materials and methods**

### 127 3.1. Sampling

128 Twenty-four fresh hand-size samples of lava flows were collected during several field  
129 trips from 2013 to 2016 (Fig. 3a). Geographical coordinates of the samples are provided in  
130 Table 1 and the associated KMZ file is available in Appendix C. In order to constrain the  
131 onset of the volcano activity, seven samples were taken from the basal flows, close to the  
132 contact with the metamorphic basement (PS-102, PS-106, PS-110, 16EQ15, 16EQ12, PS-118  
133 and 16EQ26). Sample 16EQ12 was collected from a breccia containing monogenic blocks.  
134 Seven lavas were sampled in the uppermost part of Pondoá, Runtún, Ulba and Patacocha  
135 plateaux (Fig. 2). Because of their position in the stratigraphic sequence of these plateaux, we  
136 infer that they were emplaced late in Tungurahua I history (PS-116, PS-99, 16EQ16, 16EQ17,  
137 16EQ25, TUNG 3, TUNG4). Seven lavas of Tungurahua II were sampled along Ulba valley  
138 (PS-111, 16EQ19), in Las Juntas and Juive Grande areas (PS-107, PS-109), and near  
139 Palitahua waterfalls (Fig. 2; PS-117, PS-119, 16EQ13). Two recent lavas were also sampled  
140 to compare our dating method with published  $^{14}\text{C}$  ages: a dacitic flow near Cusúa (16EQ36),  
141 which belongs to the beginning of Tungurahua III activity (Hall et al., 1999), and the Baños  
142 lava flow (16EQ37). In addition, a massive lava flow erupted from an unknown source and  
143 exposed between the Pastaza valley and Vizcaya village was also sampled (16EQ20). The aim  
144 was to determine whether it could be contemporaneous with Tungurahua volcano activity.

145

### 146 3.2. K-Ar dating

147 The potassium-argon (K-Ar) dating method was applied here by using the unspiked  
148 Cassinol-Gillot technique (Cassinol and Gillot, 1982), which was developed for Quaternary  
149 volcanics with low radiogenic argon ( $^{40}\text{Ar}^*$ ) contents (Gillot et al., 2006). It has been shown to  
150 be especially suitable for dating young subduction lavas with low K and high Ca contents, as  
151 is the case for instance in South America and in the Lesser Antilles (e.g., Samper et al., 2009;

152 Germa et al., 2011). The technique relies on the detection of a very small difference between  
153 the isotopic  $^{40}\text{Ar}/^{36}\text{Ar}$  ratio extracted from the sample and the atmospheric ratio. The  
154 radiogenic argon content, which derived from the radioactive decay of  $^{40}\text{K}$ , together with the  
155 potassium content of the sample and the  $^{40}\text{K}$  decay constant (Steiger and Jäger, 1977), allow  
156 determining the age of the lava sample.

157

### 158 *3.2.1. Sample preparation*

159 The groundmass is the last phase to crystallize when lavas cool. It is thus enriched in  
160 incompatible elements, including potassium, and in equilibrium with the atmosphere. Hence  
161 the initial argon isotopic ratios in the groundmass are atmospheric, and it is devoid of  
162 radiogenic argon ( $^{40}\text{Ar}^*$ ). On the other hand, phenocrysts can carry inherited  $^{40}\text{Ar}^*$ , that might  
163 bias ages significantly (e.g., Harford et al., 2002; Samper et al., 2008). We therefore carried  
164 out our measurements on the groundmass following a careful mineralogical separation.  
165 Observation of thin sections (available on [Appendix A](#)) helped us to attest of the samples  
166 freshness, and to determine the size fraction. The selected samples were crushed and sieved at  
167 80-125  $\mu\text{m}$ , 80-160  $\mu\text{m}$ , or 125-250  $\mu\text{m}$ , then ultrasonically washed with a 10% nitric acid  
168 solution and deionized water. Heavy liquids, mainly bromoform, were used to isolate the  
169 groundmass by removing heavy mafic minerals, and light weathered fractions, if present.  
170 Finally, the separation was refined with a magnetic separator, which removed non-magnetic  
171 plagioclases remaining within the magnetic groundmass, as their respective ranges of density  
172 overlap. The obtained fractions of groundmass, which amount to only a few percent of the  
173 initial sample, have a narrow density range, with a typical value of 2.71. Their homogeneity  
174 was checked under a binocular magnifier, and both potassium and argon measurements were  
175 carried out at the Laboratoire GEOPS of Université Paris-Sud (Orsay, France).

176



177 3.2.2. Potassium and argon measurements

178 Potassium was measured with an Agilent 240 Series AA flame absorption spectrometer,  
179 after dissolving samples with an acid attack. Results were corrected with reference values of  
180 MDO-G (K = 3.510%; Gillot et al., 1992) and BCR-2 (K = 1.481%; Raczek et al., 2001)  
181 standards.

182 Argon was measured using a mass spectrometer similar to the one described by Gillot  
183 and Cornette (1986). Very stable conditions reached during the measurements rendered the  
184 use of the  $^{38}\text{Ar}$  spike, needed for the conventional K-Ar technique, unnecessary. Gases  
185 extracted by fusing the sample were purified with high temperature Ti foam and SAES  
186 getters. An air pipette containing a known amount of  $^{40}\text{Ar}$  atoms was used to calibrate the  
187 mass spectrometer. Another air pipette, with the same  $^{40}\text{Ar}$  signal as the sample to be dated,  
188 allows an accurate atmospheric correction to be performed.

189 The accuracy of this correction and of the  $^{40}\text{Ar}$  calibration are routinely checked by  
190 analyses of international standards samples measurements, such as ISH-G (1301-1302 AD;  
191 Gillot et al., 1992) and HD-B1 ( $24.21 \pm 0.32$  Ma; Hess and Lippolt, 1994).

192 The total relative age uncertainty ( $\sigma_{\text{tot}}$ ) is calculated using the following formula:

193 
$$\sigma_{\text{tot}} = \sqrt{\sigma_K^2 + \sigma_{^{40}\text{Ar}_{\text{cal}}}^2 + \sigma_{^{40}\text{Ar}^*}^2}$$

194 The relative uncertainties of the potassium content ( $\sigma_K$ ) and the  $^{40}\text{Ar}$  calibration ( $\sigma_{^{40}\text{Ar}_{\text{cal}}}$ ) are  
195 both evaluated at 1%. The uncertainty of radiogenic argon ( $\sigma_{^{40}\text{Ar}^*}$ ) depends on the radiogenic  
196 content of the sample. For young samples, this uncertainty is larger because of their high  
197 atmospheric and low radiogenic argon contents. The  $^{40}\text{Ar}^*$  detection limit of our  
198 measurements is presently at 0.1% (Quidelleur et al., 2001).

199 All potassium and argon measurements were performed at least twice in order to check  
200 their reproducibility within uncertainty. Final ages and uncertainties were calculated by

201 averaging each analysis, weighted by their  $^{40}\text{Ar}^*$  content. However, for the youngest samples  
202 where  $^{40}\text{Ar}^*$  content is lower than 0.1%, ages were calculated using an unweighted average.  
203 Ages obtained here are given at the 1  $\sigma$  confidence level (Table 1).

204

### 205 *3.3. Geochemical analyses*

206 Major and trace element whole-rock contents of all samples were measured at the  
207 Laboratoire Géosciences Océan of the Université de Bretagne Occidentale (Brest, France).  
208 Agate-crushed powders of the twenty-four samples were analysed by ICP-AES (Inductively  
209 Coupled Plasma - Atomic Emission Spectrometry), following the analytical procedure  
210 detailed in Cotten et al. (1995). Relative uncertainties are  $\leq 2\%$  for major elements and  $\leq 5\%$   
211 for trace elements. Major elements data were recalculated to a total of 100% on a water free-  
212 basis and together with trace elements are provided in Table 2. Our geochemical database was  
213 completed by seventy-two unpublished major and trace elements analyses of samples from  
214 the three edifices and blocks from the sector collapse deposits (Hall et al., 1999; Bustillos,  
215 2008), which are presented in Appendix B.

216

### 217 *3.4. Numerical reconstructions of past morphology*

218 Numerical reconstructions of palaeotopographies were used to model the morphology of  
219 the volcano during its main phases of construction, erosion and collapse. They provided  
220 information regarding evolution of the magmatic productivity through time, and the erosion  
221 rates, which are influenced by precipitation and glacial-interglacial alternating periods. The  
222 present topographic data are extracted from a 30-m SRTM Digital Elevation Model (DEM).  
223 The reconstruction method used here was developed in Lahitte et al. (2012) and improved in  
224 Germa et al. (2015). It is summarized below.

225 Past Tungurahua topographic models are constrained from points selected from the  
226 present-day topography, as belonging to non-eroded surfaces. In general, they are from crests,  
227 upper parts of plateaux, planeze surfaces or recent deposits (Fig. 4a and b). ShapeVolc, a  
228 specifically developed method, is used for modelling the different palaeotopographies based  
229 on these points (Lahitte et al., 2012). This method first finds a global model, by defining the  
230 best summit location and the parameters of the function that define a revolution surface,  
231 which best fit the selected points following a least-squares method adjustment. Thereby,  
232 Tungurahua I reconstruction displays a conical shape with linear flank profiles and an  
233 elliptical base, while Tungurahua II and III have an exponential shape with a concave profile  
234 (Fig. 4c). The software then adjusts the first-order modelling by modifying locally the  
235 parameters of the global function, in order to take into account local elevation variations, i.e.  
236 the irregularity of the surface of each stage of construction (Lavigne et al., 2013; Germa et al.,  
237 2015). These second-order surfaces correspond to our final construction models. The  
238 evolution of Tungurahua I altitude after its erosion is calculated by lowering each elevation of  
239 the model as a linear function of time, between the model elevation and the present  
240 topography. Moreover, as the geometry of the depressions of the collapses is not precisely  
241 known, we modelled them by an interpolation between points currently located on the edge of  
242 the scars and two profiles forming the possible bottom surface represented by the  
243 discontinuous curves in Fig. 4a, located so as to respect the collapse volumes estimates,  
244 respectively of  $\sim 10$  and  $\sim 2.8 \text{ km}^3$ , based on the deposits located in the Río Chambo valley  
245 (Hall et al., 1999; Bustillos, 2008).

246 Finally, the reconstruction of pre-Tungurahua topography is obtained by selecting DEM  
247 points which belong to the metamorphic basement outcropping around the volcano, in order  
248 to extrapolate them under the present volcano surface. This palaeosurface is modelled  
249 applying a Local Polynomial Interpolation method with a resolution of 100 m, using ArcGis

250 software. The model is therefore smooth, since we cannot constrain the exact morphology of  
251 the basement 300 ka ago, which is currently overlaid by Tungurahua products.

252

### 253 3.5. Magmatic productivity and erosion rates

254 The volume ( $V$ ), either constructed ( $Vc$ ) or eroded ( $Ve$ ), corresponds to the integration  
255 of the difference between the upper and the lower surface, over the area of the edifice  
256 considered. In this fashion, the eroded volume is the difference between the modelled surface  
257 and the present topography ( $\Delta z$ ). The deposits volume for each edifice corresponds to the  
258 difference between the surface elevation modelled at the end of their constructions and the  
259 one which preceded the edifice construction. Erosion that occurred during the Tungurahua I  
260 and II construction lowered the apparent volume of emitted magma. To take into account this  
261 erosion, we added to the raw volume the eroded volume estimated by considering that the  
262 erosion rate calculated for the last 80 ka was rather constant through time. The volume  
263 uncertainty ( $\sigma_V$ ) is calculated using the following formula:

$$264 \quad \sigma_V = V \cdot \frac{\sigma_z}{\Delta z}$$

265 where the uncertainty of the elevation ( $\sigma_z$ ) is defined as  $\sigma_z = \sqrt{\sigma_{i\ surf}^2 + \sigma_{f\ surf}^2}$ ,  
266 where the uncertainties of the initial ( $\sigma_{i\ surf}$ ) and the final ( $\sigma_{f\ surf}$ ) surface elevation are  
267 estimated to 200 m for the interpolated surface before Tungurahua construction, 50 m for our  
268 surface models, and null for the present elevation. These uncertainties are obtained from the  
269 standard deviation of the elevation dispersion of the points selected to construct the  
270 palaeotopographies around the surface model, and calculated from the error map of the  
271 kriging interpolation method for the pre-Tungurahua surface (Ricci et al., 2015a).

272 Together with our new K-Ar ages, these volumes, divided by the period of activity or  
273 quiescence ( $\Delta T$ ), allow us to calculate the apparent magmatic productivity ( $MPR$ ) and erosion  
274 ( $ER$ ) rates of Tungurahua volcano, with  $MPR = \frac{Vc}{\Delta T}$ , and  $ER = \frac{Ve}{\Delta T}$ . As the parameters

275 involved in these ratios are independent, the uncertainty of the  $\frac{\sigma_R}{R}$  ratio is then defined as the  
276 square root of the quadratic sum of the relative uncertainties of both stage duration ( $\sigma_T$ ) and  
277 volume ( $\sigma_V$ ). Consequently, the uncertainty of the magmatic productivity or erosion rate ( $\sigma_R$ )  
278 is calculated using the following formula:

$$279 \quad \sigma_R = R \cdot \sqrt{\left(\frac{\sigma_V}{V}\right)^2 + \left(\frac{\sigma_T}{\Delta T}\right)^2} \text{ where } R \text{ is the considered rate.}$$

280 The uncertainty on the duration is estimated from K-Ar ages uncertainty, which  
281 constrains the timing of each edifices construction and erosion. Consequently, because of the  
282 relatively good age precision,  $\sigma_R$  is mainly dominated by the volume uncertainty. Moreover,  
283 this technique of calculation based on numerical reconstructions is more accurate than  
284 traditional methods, which generally use approximate volumes. We also present the minimum  
285 and maximum values of emission rates, considering the upper and lower values of both  
286 volume and time period. However, we chose to only use the previous calculation method for  
287 discussion and comparisons, since it provides a more realistic result and takes into account the  
288 Gaussian distribution of the data. Results of the volumes, magmatic productivity and erosion  
289 rates calculations are provided in [Table 3](#).

290

## 291 **4. Results**

### 292 *4.1. Petrography of the samples*

293 There is no clear mineralogy difference between the lava flows from the three edifices.  
294 Most of the Tungurahua lava flow samples have porphyric textures with a variable amount of  
295 plagioclase, orthopyroxene, clinopyroxene and olivine phenocrysts, except samples of  
296 Vizcaya lava flow (16EQ20) and the lower Runtún plateau (PS-110 and PS-106), which  
297 present a vitreous texture with scarce crystals ([Appendix A](#)). Sample PS-119, from the  
298 southern flank, also present some amphibole crystals, but none of our lava flow sample

299 contains biotite crystals. The groundmass contains plagioclase, pyroxene and Fe-Ti oxide  
300 microcrysts separated by volcanic glass, and mainly represents about 50-70% of the rock  
301 volume. Moreover, some samples present color variations of their groundmass, such as  
302 16EQ15 and 16EQ17 from the Ulba plateau, 16EQ19 and PS-111 from the bottom of Ulba  
303 Valley, PS-99 from the Runtún upper part, as well as PS-117 and PS-118 from the southern  
304 flank (Fig. 3). These variations correspond to more or less differentiated magmas and display  
305 that magma mixing processes occurred.

306

#### 307 4.2. K-Ar dating

308 K-Ar ages obtained in this study are presented in Table 1 and Fig. 3 and 5. Six samples  
309 were not considered because they were either altered (16EQ12) or too contaminated by  
310 atmospheric argon (16EQ13, 16EQ19, 16EQ36, TUNG3) to yield a meaningful age.  
311 Moreover, as the base and the top of Ulba sequence are well constrained by samples 16EQ15  
312 and 16EQ17 respectively, dating sample 16EQ16 was not necessary, since it is an  
313 intermediate lava located between the other two samples. Eighteen samples were successfully  
314 dated, with the K content of the groundmass ranging between 1.122% and 3.253%, and the  
315 maximum radiogenic argon content reaching 13.78%. These new data allow us to refine the  
316 timing of activity of Tungurahua volcano. The oldest edifice is well constrained by eleven  
317 ages, and was built between  $293 \pm 10$  and  $79 \pm 3$  ka. Our ages obtained for the lavas from  
318 Ulba, Pondoá and Runtún plateaux range between  $92 \pm 1$  and  $79 \pm 3$  ka, and suggest a gap of  
319 activity between 76 ka and the ~33 ka first sector collapse event (Fig. 5; Le Pennec et al.,  
320 2013). The construction of Tungurahua II started at least  $29 \pm 2$  ka (PS-117), therefore  
321 quickly after the first sector collapse. Similarly than for Tungurahua I, we have no evidence of  
322 activity between 27 and 15 ka. The five youngest ages are below 15 ka, but their relatively  
323 large uncertainties prevent us to determine whether they belong to Tungurahua II, or are

324 younger than 3 ka and are part of Tungurahua III. Only lava from the sample PS-109 clearly  
325 belongs to Tungurahua II. We propose that samples PS-107, PS-111 and PS-119 are related to  
326 Tungurahua II activity, while the sample of Baños lava flow (17EQ37) was emitted in the  
327 early stage of Tungurahua III construction, based on their stratigraphic relationships, further  
328 described in the discussion. Finally, the Vizcaya lava flow (16EQ20) dated at  $253 \pm 5$  ka, is  
329 coeval with the early construction of Tungurahua I.

330

### 331 *4.3. Whole-rock geochemical analyses*

332 The major and trace element contents for all samples are presented in [Table 2](#). Loss on  
333 ignition (L.O.I) measurements are low, with a maximum value of 1.15%, which attests for  
334 samples freshness and their low content in volatiles.

335

#### 336 *4.3.1. Major elements*

337 Tungurahua lava flows show typical trends of subduction-related volcanic rocks, and  
338 are similar to the previous analyses from its different units and the published data from the  
339 Eastern Cordillera ([Fig. 6](#)). Indeed, most samples fall within basaltic andesite and andesite  
340 fields in the  $K_2O$  vs.  $SiO_2$  diagram ([Peccerillo and Taylor, 1976; Fig. 6](#)), and two samples are  
341 dacites (16EQ17 and 16EQ20). Silica content ranges between 53.3 and 67.0 wt.%, and  $K_2O$   
342 content between 1.3 and 3.6 wt.%. Consequently, rocks from Tungurahua belong to the  
343 medium-K calc-alkaline series, except for some samples from Tungurahua I, which are more  
344 alkaline-rich and belong to the high-K calc-alkaline series.

345 Variation diagrams, shown as a function of Th (as a differentiation index), also show  
346 typical trends of subduction zone volcanism ([Fig. 7](#)). Th is a highly incompatible element,  
347 which is not fractionated during partial melting or fractional crystallization process. The  
348 decrease of  $MgO$ ,  $Fe_2O_3$ ,  $Al_2O_3$ ,  $CaO$  and  $TiO_2$  ([Fig. 7a, b, c and d](#)) suggests fractional

349 crystallization of plagioclase and Fe-Ti oxide phenocrysts present in the samples, sometimes  
350 along with pyroxene and olivine crystals (Appendix A). Magmas are not evolved enough to  
351 loose  $K_2O$  and  $Na_2O$  through incorporation into crystals, and hence the alkali content  
352 increases as the fractional crystallization proceeds (Fig. 7e and f).

353

#### 354 4.3.2. Trace elements

355 Spider diagrams of trace elements, normalized to the primitive mantle (Sun and  
356 McDonough, 1989; Fig. 8a), shows enrichment of Large-Ion Lithophile Elements (LILE; Rb,  
357 Ba, and K) and depletion of High-Field Strength Elements (HFSE; Nb, Ti and Y), also typical  
358 of subduction magmas. These signatures are also found on the Rare Earth Elements (REE)  
359 diagram normalized to chondrites values (Sun and McDonough, 1989; Fig. 8b), with an  
360 enrichment in Light REE (LREE; to the left) compared to Heavy REE (HREE, to the right),  
361 without a negative Eu anomaly.

362 Overall, lavas from Tungurahua I have higher REE contents, which can be linked with  
363 its high-K signature (Fig. 6). In the Sr/Y versus Y diagram (Fig. 9e), most of the rocks plot in  
364 the classical calc-alkaline arc lavas field, but some lavas are more depleted in Y and tend  
365 toward an “adakitic” signature. However, all samples are too enriched in Yb and too depleted  
366 in La to reach the “adakitic” domain in the La/Yb versus Yb diagram (Fig. 9f). Moreover, the  
367 Sr/Y ratio decreases as the fractional crystallization proceeds, whereas La/Yb and Th are  
368 positively correlated (Fig. 9c and d, respectively). Tungurahua I lavas are rather scattered,  
369 particularly for Pondoá basal unit and younger plateau lavas, which differ significantly from  
370 the general trend (Fig. 7). Indeed, old lavas of Pondoá, including PS-102, show a high  
371 enrichment in all incompatible elements and REE, notably Y and Yb (Fig. 8; Fig. 9e and f).  
372 Overall, all samples are porphyritic except one from the upper part of Ulba plateau (16EQ17),  
373 which is almost entirely crystallized and presents a trachytic texture (Appendix A). It is the



374 most evolved sample of plateau lavas, and presents high enrichment in the most incompatible  
375 elements and REE (Fig. 7; Fig. 8; Fig. 9).

376 Tungurahua II and III lavas compositions are rather homogeneous, and major and trace  
377 element contents do not allow us to clearly discriminate between these two different edifices.  
378 However, it can be noticed that sample PS-117, which corresponds to the beginning of  
379 Tungurahua II activity shortly after the first sector collapse event, is the most depleted in  
380 HFSE and HREE, notably in Nb, Y and Yb (Fig. 8; Fig. 9e and f). Moreover, the composition  
381 of the “Cusúa dacite” sample (16EQ36) is slightly less differentiated than the one of Hall et  
382 al. (1999), with 62.2 wt.% of SiO<sub>2</sub>. Finally, the Vizcaya lava flow (16EQ20) differs  
383 significantly from Tungurahua lavas for most major elements (Fig. 6; Fig. 7) and for some  
384 trace elements (Fig. 9b).

385

#### 386 4.4. Geomorphological reconstructions

387 Palaeotopographical reconstructions were used to propose a temporal evolution of  
388 Tungurahua volcano morphology (Fig. 10; Table 3).

389 The morphology of the metamorphic basement suggests that a north-south crest was  
390 present under the present edifice before its construction (Fig. 10a), extending the one  
391 currently found south of the volcano. As the volcano grew, Río Chambo and Pastaza  
392 palaeovalleys (Fig. 2), which were present initially, were shifted westward and northward.

393 Overall, the present Tungurahua edifice presents a conic morphology, with a rather  
394 circular base and a concave profile. According to our extrapolation of the basement and the  
395 present topography, the present volume of this volcano is  $108 \pm 30 \text{ km}^3$  (Table 3), with a basal  
396 surface of about 150 km<sup>2</sup>. Given our maximum age of  $293 \pm 10 \text{ ka}$ , the apparent production  
397 rate is  $0.4 \pm 0.1 \text{ km}^3/\text{ka}$ .

398 In detail, Tungurahua I was a volcano with a rather flat summit, which peaked at a  
399 minimum elevation of 4500 m (Fig. 10b), 2 km southeast to the present summit. Based on our  
400 numerical reconstructions, the volume of Tungurahua I at the end of its construction was  
401  $56 \pm 33 \text{ km}^3$ . Considering that the erosion rate affecting its flanks during its growth was  
402 similar to the erosion rate calculated since the end of its construction, we obtain an eroded  
403 volume of  $68 \pm 41 \text{ km}^3$  between ~293 and 81 ka. This volume is added to the first volume of  
404  $56 \pm 33 \text{ km}^3$ , which provides a total volume of  $124 \pm 74 \text{ km}^3$  of products emitted during the  
405 ~212 ka of Tungurahua I construction (Table 3). As a result, the magmatic productivity rate of  
406 this first edifice was  $0.6 \pm 0.3 \text{ km}^3/\text{ka}$ . A quiescence period favoured the erosion of  
407 Tungurahua I flanks between 80 and 33 ka (Fig. 10c). Then, the sudden loss of  $\sim 10 \text{ km}^3$  of  
408 deposits (Bustillos, 2008) during the first western sector collapse at ~33 ka (Fig. 10d; Table  
409 3) may have favoured the rise of magma (e.g., Hildenbrand et al., 2004) and the rapid  
410 construction of Tungurahua II within the resulting amphitheatre (Fig. 10e). This second  
411 edifice culminated at about 5070 m, with a concave upward profile similar to the present  
412 volcano. Although the volume of the emitted products ( $24 \pm 4 \text{ km}^3$ ) is significantly lower than  
413 the one calculated for the first edifice, its eruptive rate was similar ( $0.9 \pm 0.2 \text{ km}^3/\text{ka}$ ) given  
414 the shorter duration of this stage, between  $29 \pm 2 \text{ ka}$  (Table 1) and  $2.96 \pm 0.03 \text{ ka}$  (Le Pennec  
415 et al., 2013). A new flank failure occurred at ~3 ka (Fig. 10f), mobilizing around  $3 \text{ km}^3$  of  
416 deposits (Bustillos, 2008) from the western summit part. Finally, the third and still active  
417 edifice was rapidly constructed in the depression left by this collapse (Fig. 10g). Tungurahua  
418 III is characterized by the lowest volume of deposits ( $7 \pm 3 \text{ km}^3$ ), but displays a relatively  
419 high eruptive rate of  $2.5 \pm 1.0 \text{ km}^3/\text{ka}$ . In addition, an erosion rate of  $0.2 \pm 0.1 \text{ km}^3/\text{ka}$  for the  
420 eastern part of Tungurahua I not affected by the collapses is obtained, considering the eroded  
421 volume of  $\sim 16 \text{ km}^3$  during ~80 ka, i.e. since the end of the first edifice construction (Table 3).  
422 As Tungurahua II collapsed rapidly after its construction without significant periods of

423 quiescence, and as Tungurahua III is still active, we cannot calculate erosion rates for these  
424 edifices.

425 Based on these new results and field observations, we propose in Fig. 3 an update of the  
426 Tungurahua geological map (Hall et al., 1999; Bustillos, 2008). Tungurahua II not only  
427 consists of the lava flow sequence of the Tiacos unit (Fig. 2), but also crops out in the  
428 northern and southwestern flanks. We also propose that the Tungurahua I collapse scar is  
429 located to the west of the Tungurahua II scar, as suggested by the location of the upper parts  
430 of the Pondoá, Runtún and Ulba plateaux.

431

## 432 **5. Discussion**

### 433 *5.1. New groundmass K-Ar ages*

#### 434 *5.1.1 Comparison with previously published ages*

435 The new K-Ar ages of Tungurahua I (Table 1 and Fig. 3b, c and d) are strikingly  
436 younger than previously published whole-rock K-Ar ages of  $770 \pm 5$  and  $350 \pm 4$  ka (Barberi  
437 et al., 1988), for samples from Tungurahua, but whose location is unknown. Such difference  
438 can be explained by the analysis of whole-rock samples that include phenocrysts, which can  
439 carry inherited radiogenic argon and/or weathering areas that have experienced loss of  
440 potassium. Both phenomena will induce older K-Ar ages. Our results highlight the importance  
441 of analysing the groundmass alone, after a thorough mineralogical separation, as previously  
442 reported in different contexts, including arc volcanism (e.g., Samper et al., 2007; Germa et al.,  
443 2011; Zami et al., 2014). Although unpublished, four  $^{40}\text{Ar}/^{39}\text{Ar}$  ages obtained at the  
444 Laboratoire Geoazur (Côte d'Azur University, Nice, France) support our K-Ar ages. Indeed,  
445 inverse isochron ages of  $27 \pm 4$  and  $104 \pm 44$  ka (M. Fornari, pers. com.) agree with our ages  
446 of  $29 \pm 2$  and  $153 \pm 14$  ka obtained for the same lava flows (PS-117 and PS-110 respectively;  
447 Table 1). In addition, two distinct lava flows sampled in the same eruptive phase than PS-99

448 and PS-118 have  $^{40}\text{Ar}/^{39}\text{Ar}$  ages of  $99 \pm 2$  and  $252 \pm 6$  ka (M. Fornari, pers. com.), which are  
449 in the range of our K-Ar ages of  $79 \pm 3$  and  $293 \pm 10$  ka, respectively (Table 1). Moreover,  
450 most samples from Tungurahua II and III are younger than 15 ka (Table 1), in agreement with  
451 their emplacement after the Last Glacial Maximum (Hall et al., 1999; Clapperton, 1990).  
452 Although they display a large relative uncertainty, our ages are also consistent with previously  
453 published  $^{14}\text{C}$  ages of less than  $\sim 3$  ka for Tungurahua III deposits (Hall et al., 1999; Le Pennec  
454 et al., 2008, 2013, 2016).

455 In this southern part of the Ecuadorian arc, within the Inter-andean valley, radiometric  
456 ages are available only for Chimborazo and Carihuairazo volcanoes (Fig. 1b). Both were built  
457 up since 230 ka, and until Holocene for Chimborazo volcano (Samaniego et al., 2012; Barba  
458 et al., 2008). Further south, the construction of the Sangay volcano, the last active edifice of  
459 the Eastern Cordillera, started around 500 ka (Monzier et al., 1999; Fig. 1b). Hence, it appears  
460 that the construction of Chimborazo, Carihuairazo, Sangay and Tungurahua volcanoes are  
461 rather coeval, which suggests a recent development of this part of the arc.

462

### 463 *5.1.2. Association between ages and volcanic phases*

464 The distribution of our new ages is fairly homogeneous on the scale of the volcano  
465 activity, apart from the end of the first edifice construction (Fig. 5). Indeed, the oldest basal  
466 lava flows of Tungurahua I have an age of 300 ka, and the edifice remained active until the  
467 emplacement of lava flows forming the upper parts of plateaus, at around 80 ka. The edifice  
468 collapsed around 33 ka (Le Pennec et al., 2013), but we do not have any sample with an age  
469 between 80 ka and the 33 ka destabilization. Tungurahua I activity might have continued  
470 between 80 and 33 ka if the lava flows were only emplaced in the summit area or in the  
471 western flank that have been destroyed by latter flank collapses, since the plateaus do not  
472 seem covered by any younger deposits. Similarly, we did not observe any activity between

473 246 ± 17 and 188 ± 7 ka (Fig. 5). Although it could be due to a sampling bias, these ~50 ka  
474 quiescence periods would be the longest experienced by the volcano. Moreover, owing to the  
475 relatively high age-uncertainty of lavas younger than 15 ka (Table 1), and the similar  
476 geochemistry observed for the two younger edifices, we cannot determine whether these lavas  
477 belong to Tungurahua II or III. Therefore, we rely on stratigraphic relationships observed in  
478 the field to associate young samples with a given eruptive phase. Sample PS-109 (7 ± 4 ka) is  
479 older than 3 ka and corresponds to Tungurahua II activity (Fig. 3), probably as well as sample  
480 PS-107 (5 ± 8 ka), which belongs to a nearby lava flow. Hall et al. (1999) associated the  
481 young lava flow at the bottom of the Ulba valley (sampled here as PS-111 and 16EQ19) to  
482 Tungurahua III activity based on petrological similarities. However, there are very few  
483 mineralogical differences between the two younger edifices (Appendix A). We associate  
484 PS-111 and 16EQ19, as well as PS-119 and 16EQ13 (southwestern flank lavas), to  
485 Tungurahua II activity (Fig. 3), because Tungurahua III deposits in the sector collapse  
486 depression are not thick enough to have overflowed into the eastern and southern valleys.  
487 Regarding the Baños lava flow (16EQ37; 6 ± 9 ka), there is no place where a clear  
488 stratigraphic relationship could be observed between the lava and the ~3 ka collapse deposits  
489 (namely the blast deposits associated with the sector collapse). Indeed, these Tungurahua II  
490 blast deposits are dated between 2640 ± 45 and 3195 ± 45 BP (<sup>14</sup>C on charcoal and carbonized  
491 twigs), crop out along the Chambo and Puela valleys (Fig. 2) and cover the northern flank of  
492 the volcano (Le Pennec et al., 2013), especially Pondoá and Runtún plateaux around Baños  
493 city (Fig. 2), but do not crop out above the Baños lava flow. The lava flow originated from a  
494 lateral vent (Fig. 3b), and was later covered by an ash deposit dated at 1470 ± 85 BP (<sup>14</sup>C on  
495 charcoal; Hall et al., 1999). Assuming that the Baños lava flow is younger than the blast  
496 deposits, we associate the Baños lava flow with the beginning of Tungurahua III activity, in  
497 agreement with Hall et al. (1999) and Le Pennec et al. (2016).

498 The Vizcaya lava flow is coeval with Tungurahua I activity ( $253 \pm 5$  ka), but its major  
499 and trace contents clearly differ from Tungurahua rocks (Fig. 7; Fig. 9). Hence, this flow  
500 cannot be associated with Tungurahua volcano and was rather erupted from an unknown  
501 volcanic centre, probably located to the north.

502 Unfortunately, the southeastern flank could not be sampled and dated in this study.  
503 Indeed, access to the Minsas plateau (Fig. 2) was not possible during the erupting period,  
504 when our last field trip took place. However, previous major and trace element analyses (Hall  
505 et al., 1999; Appendix B) indicate a composition close to those of Ulba upper part (16EQ17;  
506 Table 2). Minsas plateau lava geochemistry, morphology and erosion state therefore suggest  
507 that the flow is coeval with other Tungurahua I plateau lavas, and was erupted around 80 ka  
508 (Fig. 3).

509

## 510 *5.2. Location of the ~33 ka sector collapse scar*

511 The succession of construction stages of the volcano and the erosion prevent to  
512 accurately determine the location of the Tungurahua I collapse scar. However, morphology  
513 analysis of the DEM, and observations made during field campaigns allow us to propose that  
514 the northern part of the scar follows the western escarpment of the Pondoá plateau (Fig. 3).  
515 Indeed, a “diamond-shaped” plateau, located west of Pondoá, is made of Tungurahua I  
516 collapse deposits overlain by Tungurahua II lava flows (Fig. 11). This part of the volcano was  
517 therefore destroyed by the first collapse, but was not affected by the second one. Note that  
518 such observations are in agreement with the geological map of Hall et al. (1999). The  
519 morphology of the southwestern flank, from Patacochoa plateau to the second collapse scar,  
520 seems to constitute a single unit filling a wide valley (Fig. 3d and Fig. 11d). Indeed, lava  
521 flows cover a Tungurahua I breccia deposit, where sample 16EQ12 comes from (Fig. 3a), and  
522 no dated lava is there older than 30 ka (Fig. 3d). We therefore propose that the top of the first

523 (~33 ka) collapse scar is located on the eastern flank at around 3900 m a.s.l., where plateaux  
524 slopes change (Fig. 4) forming the lower boundary of the Tiacos unit, and located along the  
525 summit part of the Patacocha plateau (Fig. 3).

526

### 527 *5.3. Geochemical evolution of the volcanic products*

528 Overall, the lavas of the three successive edifices of Tungurahua volcano show quite  
529 similar compositions. Indeed, their silica content varies between 52 and 62 wt.%, and their  
530 Mg# between 40 and 60 (Fig. 12a), except for 16EQ17 sample, which is more differentiated  
531 than the other lavas, and presents higher content of incompatible elements (Fig. 7; Fig. 9).

532 The La/Yb ratio varies depending on the depth, composition and nature of the magmas  
533 source, fractional crystallization and/or interaction with the mantle (Moyen, 2009). Temporal  
534 evolution of the La/Yb ratio for Tungurahua samples (Fig. 12b) shows an overall decrease  
535 between 300 and 100 ka, and a significant increase toward the end of Tungurahua I activity,  
536 shown by samples younger than 100 ka. During Tungurahua II and III construction, La/Yb  
537 contents are relatively scattered, and vary within the range observed for Tungurahua I lavas.  
538 Moreover, the La/Sm ratio provides information about the degree of partial melting, and the  
539 Sm/Yb ratio is affected by the depth or nature of the magmatic source (e.g., Wilson, 1989;  
540 Rollinson, 1993). Fig. 12b shows that these ratios are relatively constant through time for  
541 Tungurahua lavas, except a high Sm/Yb content after the ~33 ka collapse (PS-117) and a  
542 relatively high La/Sm content towards the end of Tungurahua I construction (16EQ17).  
543 Overall, these geochemical evolutions seem to be mainly due to the nature of the magmas.  
544 The La/Sm and La/Yb ratios have the same trend than the SiO<sub>2</sub> content as a function of time,  
545 except for sample PS-117. The higher ratios correspond to the most differentiated lava flows,  
546 such as sample 16EQ17, which is a dacite significantly enriched in the most incompatible  
547 elements (LILE, HFSE and LREE; Table 2) compared to other andesite samples. Concerning

548 sample PS-117, the high La/Yb ratio is correlated with its high Sm/Yb ratio, due to its  
549 depletion in Dy, Er, Y and Yb (Table 2) that could be related to the heterogeneity of the  
550 mantle wedge composition. Large scale sector collapses occurring in volcanic islands may  
551 cause an increase of the partial melting rate due to the pressure reduction affecting the magma  
552 chamber (e.g., Hildenbrand et al., 2004). On the other hand, the thick crust may prevent sector  
553 collapses to affect the deep magmatic source in continental environments. In Ecuador, Robin  
554 et al. (2010) observed large geochemical variations just after two major sector collapses in  
555 Pichincha volcano lavas (Fig. 1b). The authors infer that new magma batches, associated with  
556 new magmatic cycles, may have triggered these sector collapses. We have fewer analyses for  
557 Tungurahua volcano, and they are spread over a shorter time scale. However, the  
558 geochemistry of Tungurahua lava flows is rather homogeneous for the three edifices and does  
559 not seem to be affected by the two observed collapses.

560 Several studies have focused on the evolution through time of Ecuadorian lavas  
561 geochemistry, notably for Atacazo (Hidalgo, 2006), Cayambe (Samaniego et al., 2005),  
562 Iliniza (Hidalgo et al., 2007), Mojanda-Fuya Fuya (Robin et al., 2009), and Pichincha  
563 (Samaniego et al., 2010) volcanoes (Fig. 1b). These edifices are located in the northern part of  
564 the Ecuadorian arc, in front of the Carnegie ridge (Fig. 1a). They were constructed since  
565 1.3-1.0 Ma until the Holocene, except for Mojanda-Fuya Fuya and Iliniza, which are not  
566 precisely dated yet. These edifices present a long-term evolution, between typical  
567 calc-alkaline compositions for the oldest lavas, to adakitic compositions for the most recent  
568 deposits, which is interpreted as a modification of the sources of the magmas induced by the  
569 subduction of the Carnegie ridge, and the subsequent increase of the geothermal gradient. The  
570 relatively short duration of Tungurahua activity (about 300 ka), and its location in the  
571 southern part of the arc (farther away from the Carnegie ridge than the northern volcanoes),  
572 prevent us to identify these long term deep processes, if present. This latter conclusion has



573 been corroborated by the recent work of Ancellin et al. (2017), who analysed the Sr-Nd-Pb  
574 isotopic signature of the Ecuadorian magmas and conclude that magmas from the southern  
575 part of the arc (as those of Tungurahua and Sangay volcanoes) do not display a clear  
576 subduction slab partial melting component.

577

#### 578 *5.4. Magmatic productivity and erosion rates*

##### 579 *5.4.1. Tungurahua eruptive and erosion rates*

580 The overall volume obtained for the present edifice of Tungurahua ( $108 \pm 30 \text{ km}^3/\text{ka}$ ;  
581 Table 3 and 4) is similar to the one obtained for Viejo Cayambe (Samaniego et al., 2005), and  
582 seems to correspond to a medium size compared to arc volcanoes in general (Table 4; Grosse  
583 et al., 2009). However his volume is higher than Ecuadorian Antisana (Hall et al., 2017; Table  
584 4), Imbabura (Le Pennec et al., 2011) and Chimborazo (Samaniego et al., 2012) volcanoes,  
585 which are more eroded or younger, and is lower than Rucu Pichincha (Robin et al., 2010) and  
586 Mojanda ( $> 140 \text{ km}^3$ ; Robin et al., 1997) volcanoes, located more to the north (Fig. 1b), and  
587 significantly older. Our palaeotopographical reconstructions suggest that Tungurahua I was a  
588 volcano with lower sloping upper flanks, while Tungurahua II and III have an exponential  
589 shape with a concave profile and steeper slopes (Fig. 4; Fig. 10). The nature of lavas  
590 composing these three edifices is very similar and cannot explain this morphological  
591 difference. Tungurahua I has been subjected to erosion longer than Tungurahua II and III, and  
592 the erosion mainly affected steep slopes of areas near the summit. Less eroded parts, in  
593 particularly flanks with low slopes, constitute the present planezes. Our model of  
594 Tungurahua I morphology is based on the surface of these planezes, and it does not exclude  
595 the possibility that Tungurahua I could have had a similar concave profile as the present  
596 edifice at the end of its construction. Assuming that the activity of Tungurahua I ended  
597  $\sim 80 \text{ ka}$ , we determined the erosion rate by calculating the volume loss between our

598 topographic model at 80 ka and the present-day topography. This was performed only  
599 considering the eastern flank, where the remnants of Tungurahua I not affected by sector  
600 collapses crop out, and yielded a volume of  $16 \pm 4 \text{ km}^3$ , corresponding to an erosion rate of  
601  $0.2 \pm 0.1 \text{ km}^3/\text{ka}$  (Table 3). Assuming the long term erosion rate is constant over time, we  
602 obtain a total eroded volume of  $68 \pm 41 \text{ km}^3$  during the construction of Tungurahua I, between  
603 293 and 81 ka (PS-118 and PS-116 respectively; Table 1 and 3). Based on our models, the  
604 volume of Tungurahua I corresponds to  $56 \pm 33 \text{ km}^3$  of deposits. In order to take into account  
605 the erosion that affected the edifice during its construction phase, we have added the  
606  $68 \pm 41 \text{ km}^3$  eroded volume to calculate the volume of emitted products and the magmatic  
607 productivity rate. Our result of  $0.6 \pm 0.2 \text{ km}^3/\text{ka}$  (Table 3) thus corresponds to a magmatic  
608 production volume of  $124 \pm 74 \text{ km}^3$  during a period of 212 ka. However, these values should  
609 be considered with caution, as the basement elevation under Tungurahua volcano is not well  
610 constrained, and the constant erosion rate applied over the whole construction period was  
611 most probably variable through time, as a function of the climatic conditions and the nature of  
612 outcropping volcanic products. Moreover, we consider that the deep valleys have been dug by  
613 erosion, which may have occurred mainly before 30 ka since Tungurahua II lavas outcrop at  
614 the bottom of these valleys (Fig. 3). However, these lavas are younger than 15 ka (e.g. sample  
615 PS-111), and we cannot quantify the volume of deposits eroded by the Last Glacial  
616 Maximum, which occurred between 25 and 15 ka in this region (Clapperton, 1990; Heine,  
617 2000). Furthermore, tephra deposits can easily erode when they are not covered with lava  
618 flows. It is possible that such deposits existed, but it is difficult to estimate and take into  
619 account this volume in our calculations of erosion rates. Similarly, from a calculated volume  
620 of  $21 \pm 4 \text{ km}^3$  obtained for Tungurahua II, added with a volume of  $4 \pm 2 \text{ km}^3$  for erosion, we  
621 obtain a volume of  $24 \pm 4 \text{ km}^3$  of emitted products, giving an eruptive rate of  $0.9 \pm 0.2 \text{ km}^3/\text{ka}$ ,  
622 similar to Tungurahua I. Finally, the magmatic productivity rate calculated for Tungurahua III

623  $(2.5 \pm 1.0 \text{ km}^3/\text{ka}$ ; Table 3) is consistent with the eruptive rate of  $\sim 1.5 \text{ km}^3/\text{ka}$  estimated by  
624 Hall et al. (1999) over the last 2300 years, for a similar volume to ours. The relatively high  
625 production rate calculated for Tungurahua III may be explained by its recent construction and  
626 rather continuous activity, compared to the two older edifices, which are averaged over a  
627 larger time scale, certainly including periods of quiescence.

628

#### 629 *5.4.2. Comparison between eruptive rates of Tungurahua volcano and other studies*

630 Many phenomena may impact on the magmatic productivity, such as the geodynamic  
631 context, the nature and thickness of the crust, or the period of time considered. In a  
632 compilation of the magmatic productivity calculated over the world, White et al. (2006)  
633 obtained an average rate of  $4.4 \pm 0.8 \text{ km}^3/\text{ka}$  for continental crust volcanoes and  
634  $2.3 \pm 0.8 \text{ km}^3/\text{ka}$  for andesitic edifices. Our eruptive rate of  $2.5 \text{ km}^3/\text{ka}$ , corresponding to  
635 Tungurahua III activity (Table 3 and 4), falls within this latter range. Note that the rates of 0.6  
636 and  $0.9 \text{ km}^3/\text{ka}$ , obtained here for Tungurahua I and II, respectively (Table 3 and 4), are  
637 comparable to the ones obtained on Chimborazo ( $0.1$  to  $1 \text{ km}^3/\text{ka}$ ; Samaniego et al., 2012) and  
638 Pichincha ( $\sim 0.6 \text{ km}^3/\text{ka}$  for both Rucu and Guagua Pichincha; Robin et al., 2010) volcanoes,  
639 both also from Ecuador (Fig. 1). Tungurahua eruptive rates are also similar to those from  
640 other arc volcanoes, for instance in Chile at Parinacota ( $\sim 0.3$  and  $1 \text{ km}^3/\text{ka}$  for the whole  
641 edifice and the young cone, respectively; Hora et al., 2007), Puyehue-Cordón Caulle  
642 ( $0.42 \text{ km}^3/\text{ka}$ ; Singer et al., 2008) and El Misti ( $0.63$  and  $2.1 \text{ km}^3/\text{ka}$  for the whole edifice and  
643 peak eruptive rates, respectively; Thouret et al., 2001) volcanoes, which were active since less  
644 than 320 ka, or with Ceboruco volcano in Mexico ( $\sim 0.5 \text{ km}^3/\text{ka}$ ; Frey et al., 2004). However,  
645 Tungurahua eruptive rates are significantly higher than at Aucanquilcha volcano in Chile  
646 ( $0.04$  and  $0.16 \text{ km}^3/\text{ka}$  for the whole edifice and the highest eruptive rate stage, respectively;  
647 Klemetti and Grunder, 2008) and at Tequila volcano in Mexico ( $0.13$  and  $> 0.24 \text{ km}^3/\text{ka}$  for

648 the whole field constructed during the last million years and the youngest cone, respectively;  
649 Lewis-Kenedi et al., 2005). These data show that eruptive rates may vary significantly in the  
650 same geodynamic context. Moreover, volcanoes which experienced a long period of activity  
651 (in the order of one million years) seem to have lower eruptive rates than younger edifices,  
652 averaged over shorter times (e.g., Klemetti and Grunder, 2008). The magmatic productivity  
653 rates also vary during construction stages of a single edifice, where a few thousand years  
654 period of high activity will display a higher eruptive rate than the one averaged on the whole  
655 eruptive history of the volcano. Indeed, stratovolcanoes grow in spurts (e.g., Hildreth and  
656 Lanphere, 1994; Lewis-Kenedi et al., 2005), with sporadic eruptive pulses, separated by long-  
657 time periods of quiescence, producing most of the volcano volume. The magmatic  
658 productivity rates averaged over the whole period of construction are therefore systematically  
659 lower, such observed for Tungurahua with  $\sim 0.4 \text{ km}^3/\text{ka}$  for the whole volcano (Table 3), while  
660 the eruptive rate of the three individual edifices is higher than  $0.6 \text{ km}^3/\text{ka}$ . Magmatic  
661 productivity rates of Tungurahua I, II and the whole edifice are long-term averaged eruptive  
662 rates, which include quiescence periods, whereas Tungurahua III, younger than 3 ka,  
663 represents a « peak » of activity with a significantly higher eruptive rate (Table 3). A similar  
664 picture seems to occur in Peru at Ampato-Sabancaya volcanic complex. Ampato, the older  
665 edifice, has an eruptive rate of  $0.08\text{-}0.09 \text{ km}^3/\text{ka}$ , whereas it is of  $0.6\text{-}1.7 \text{ km}^3/\text{ka}$  for the  
666 younger edifice Sabancaya, constructed since 6-10 ka (Samaniego et al., 2016).

667 As expected, Tungurahua magmatic productivity rates are lower than in a subduction  
668 context where the velocity of the plate is faster ( $5 \text{ km}^3/\text{ka}$  for Mount Fuji in Japan; Crisp,  
669 1984), and higher than in a subduction context where the convergence rate is slower, as for  
670 instance in the Lesser Antilles. Indeed, eruptive rates there vary between  $0.13 \text{ km}^3/\text{ka}$  in  
671 Martinique (Mount Pelée; Germa et al., 2015),  $0.15 \text{ km}^3/\text{ka}$  at Montserrat (Harford et al.,  
672 2002), and  $0.04$  to  $0.7 \text{ km}^3/\text{ka}$  in Guadeloupe (Lahitte et al., 2012; Ricci et al., 2015a; 2015b;

673 Table 4). Finally, Tungurahua eruptive rates are lower than those obtained in Argentina for  
674 Payún Matrú volcano ( $4 \text{ km}^3/\text{ka}$ , averaged over the last 300 ka; Germa et al., 2010), where  
675 back-arc extension may have favoured the magma ascent.

676

#### 677 5.4.3. Comparison between erosion rates of Tungurahua volcano and other studies

678 Since only a few studies have investigated erosion rates, their relationship with the  
679 geodynamic context or the climate variability is still poorly understood. Our erosion rate for  
680 Tungurahua I volcano of  $0.2 \pm 0.1 \text{ km}^3/\text{ka}$ , equivalent to  $6.5 \pm 1.7 \text{ t/km}^2/\text{ka}$  and  
681  $2.4 \pm 0.6 \text{ mm/a}$ , calculated for the last 80 ka, is significantly higher than rates obtained in the  
682 arid central Andes and in Peru (between 0.01 and 0.11 mm/a; Karátson et al., 2012) or in  
683 tropical environments, such as in the Lesser Antilles with  $0.05 \pm 0.02 \text{ km}^3/\text{ka}$  in Martinique  
684 (Germa et al., 2015), and between  $0.2 \pm 0.2$  and  $1.4 \pm 0.6 \text{ t/km}^2/\text{ka}$  in Guadeloupe (Ricci et al.,  
685 2015a; 2015b) islands. This difference could be explained by the succession of glacial and  
686 interglacial periods experienced by Tungurahua volcano, whereas the Antilles islands were  
687 only subject to rainfalls. However, morphology and extreme rainfalls experienced by La  
688 Réunion island, as well as the faster weathering of basaltic than andesitic lavas (e.g., Gislason  
689 et al., 1996; Louvat and Allègre, 1997), may explain its high erosion rate of  $1.2 \pm 0.4 \text{ km}^3/\text{ka}$   
690 (Salvany et al., 2012), compared to the one of Tungurahua volcano.

691

## 692 6. Conclusions

693 This study allows us to refine the eruptive history of Tungurahua volcano, which was  
694 built in three stages since the last 300 ka. The overall volume calculated for the remaining  
695 edifice is  $\sim 110 \text{ km}^3$ , which corresponds to a mean eruptive rate of  $\sim 0.4 \text{ km}^3/\text{ka}$ . Tungurahua I  
696 was a  $\sim 125 \text{ km}^3$  andesitic to dacitic stratovolcano, built between  $293 \pm 10$  and  $79 \pm 3 \text{ ka}$  with  
697 a magmatic productivity rate of  $0.6 \pm 0.2 \text{ km}^3/\text{ka}$ . Pondoá, Runtún, Ulba, Minsas and

698 Patacocha plateaux are present-day remnants of this first edifice. After a ~50 ka long period  
699 of quiescence, the first western sector collapse occurred at ~33 ka (Le Pennec et al., 2013).  
700 Tungurahua II was built between  $29 \pm 2$  and ~3 ka (Hall et al., 1999; Le Pennec et al., 2013).  
701 Around  $25 \text{ km}^3$  of volcanic products filled the collapse amphitheatre, with an eruptive rate of  
702  $0.9 \pm 0.2 \text{ km}^3/\text{ka}$ , similar to those of Tungurahua I. They mainly crop out in the southwestern  
703 flank and around Juive Grande area, in the northwestern flank (Fig. 2). Collapse of  
704 Tungurahua II western flank was followed by rapid construction of Tungurahua III during the  
705 last 3 ka. This third and still active edifice constitutes the western flank of the present-day  
706 volcano, and is being built with a relatively high productivity rate of  $2.5 \pm 1.0 \text{ km}^3/\text{ka}$ . We  
707 also estimated that around  $16 \text{ km}^3$  of Tungurahua I volcanic material was eroded since the end  
708 of its construction, corresponding to an erosion rate of  $0.2 \pm 0.1 \text{ km}^3/\text{ka}$ . The composition of  
709 Tungurahua lavas is rather homogeneous, and the magmatic signature of the three edifices do  
710 not seem to have been affected by the two sector collapses experienced by the volcano.  
711 Moreover, the geochemistry do not show changes in the deep magmatic source as observed  
712 for northern Ecuador volcanoes.

713

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728

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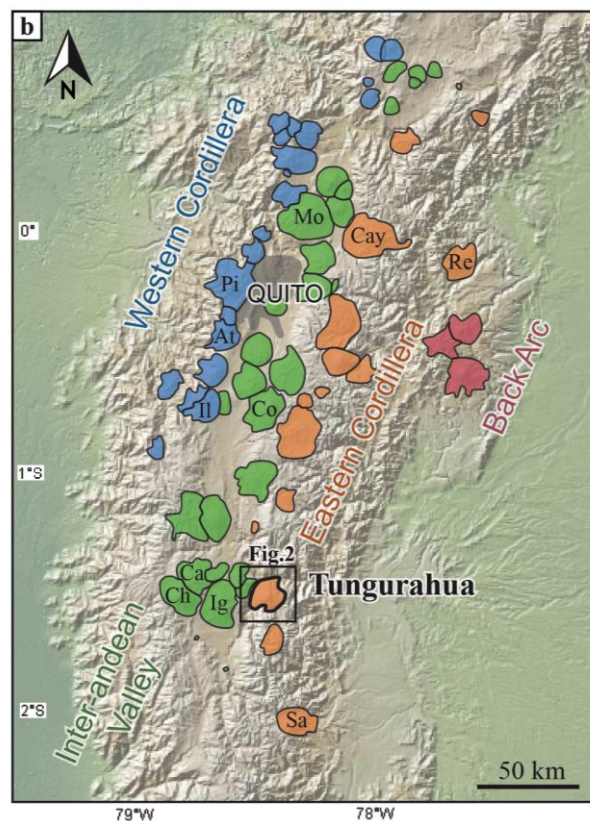
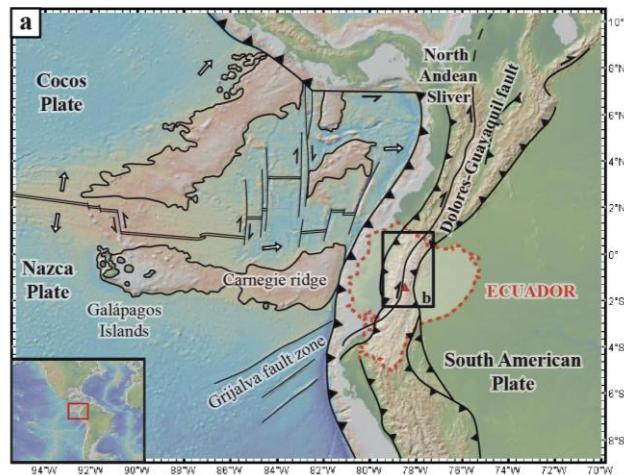
1016 **Figure captions**

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1018 **Fig. 1:**

1019 a) Regional setting of Ecuador, showing main tectonic structures and geodynamic context of the Northern  
1020 Volcanic Zone of the Andes. Arrows indicate the direction of plate motion (DeMets et al., 1990). Bathymetry  
1021 image is from GeoMapApp, using data of Smith and Sandwell (1997). Red triangle represents location of  
1022 Tungurahua volcano. b) Map of Ecuadorian volcanic arc, modified from Hall et al. (2008). Volcanoes are  
1023 distributed along the Western Cordillera (volcanic front, represented in blue), the Inter-andean valley (green), the  
1024 Eastern Cordillera (orange) and the back arc (red). Quito, the capital city, is represented in grey. Tungurahua  
1025 volcano is surrounded by heavy black borders. At: Atacazo; Ca: Carihuairazo; Cay: Cayambe; Ch: Chimborazo;  
1026 Co: Cotopaxi; Il: Iliniza; Mo: Mojanda; Pi: Pichincha; Re: Reventador; Sa: Sangay.

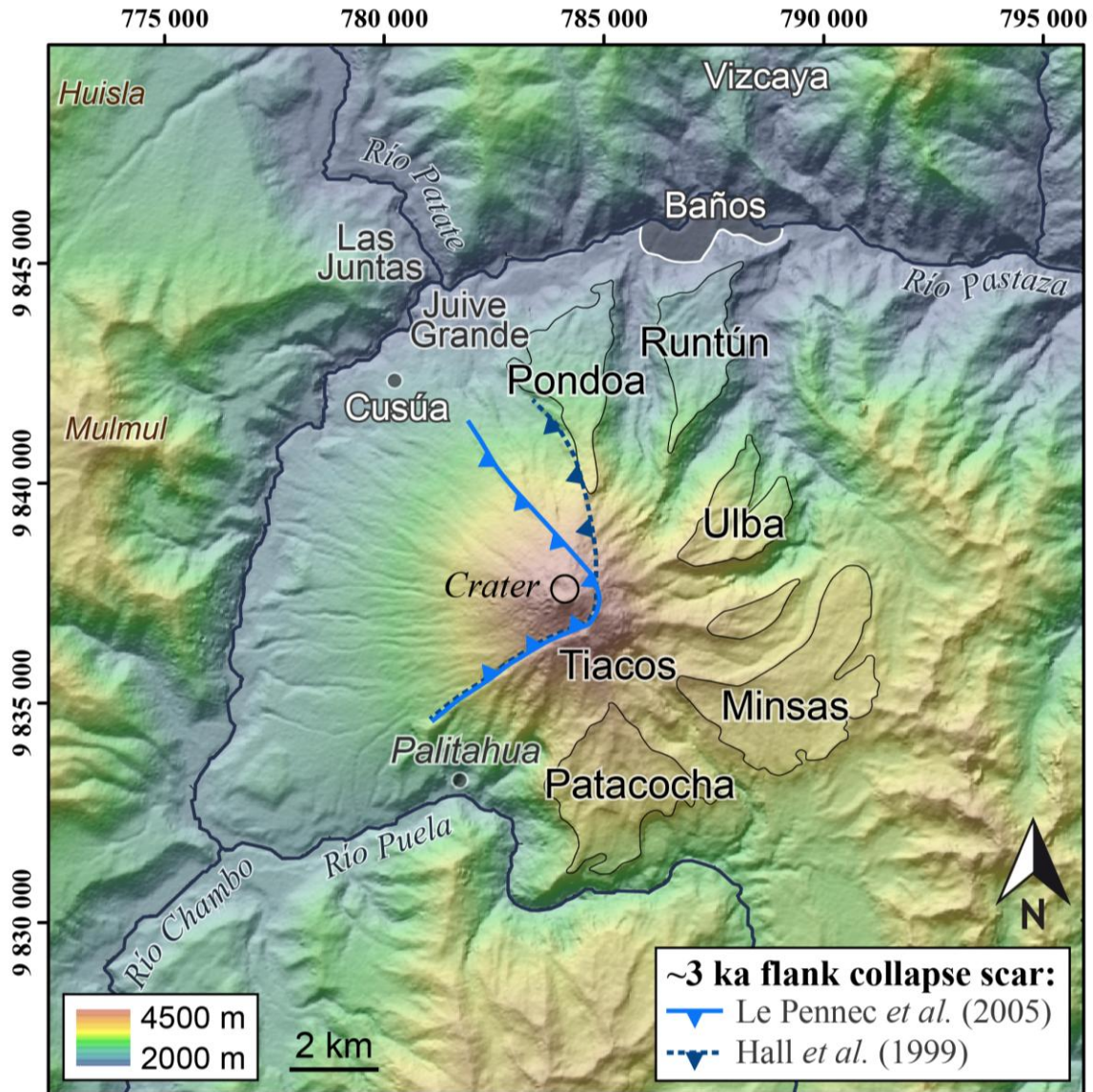
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1029 **Fig. 2:**

1030 Shaded view of the Shuttle Radar Topography Mission (SRTM) Digital Elevation Model (DEM; 30-m  
1031 resolution) of Tungurahua volcano and places mentioned in the text (location is indicated on Fig.1b).  
1032 Coordinates are in Universal Transverse Mercator (UTM; zone 17). Mulmul and Huisla are neighbour volcanoes  
1033 located in the Inter-andean valley. Blue lines represent main rivers and thin black lines the boundary of plateaux  
1034 upper surfaces. Tiacos unit corresponds to a 80-100 m-thick sequence of Tungurahua II lava flows that cover  
1035 older lavas of Tungurahua I (Hall et al., 1999). Vizcaya lava flow extends over at least 5 km along the valley  
1036 north of Tungurahua, from an unknown source.  
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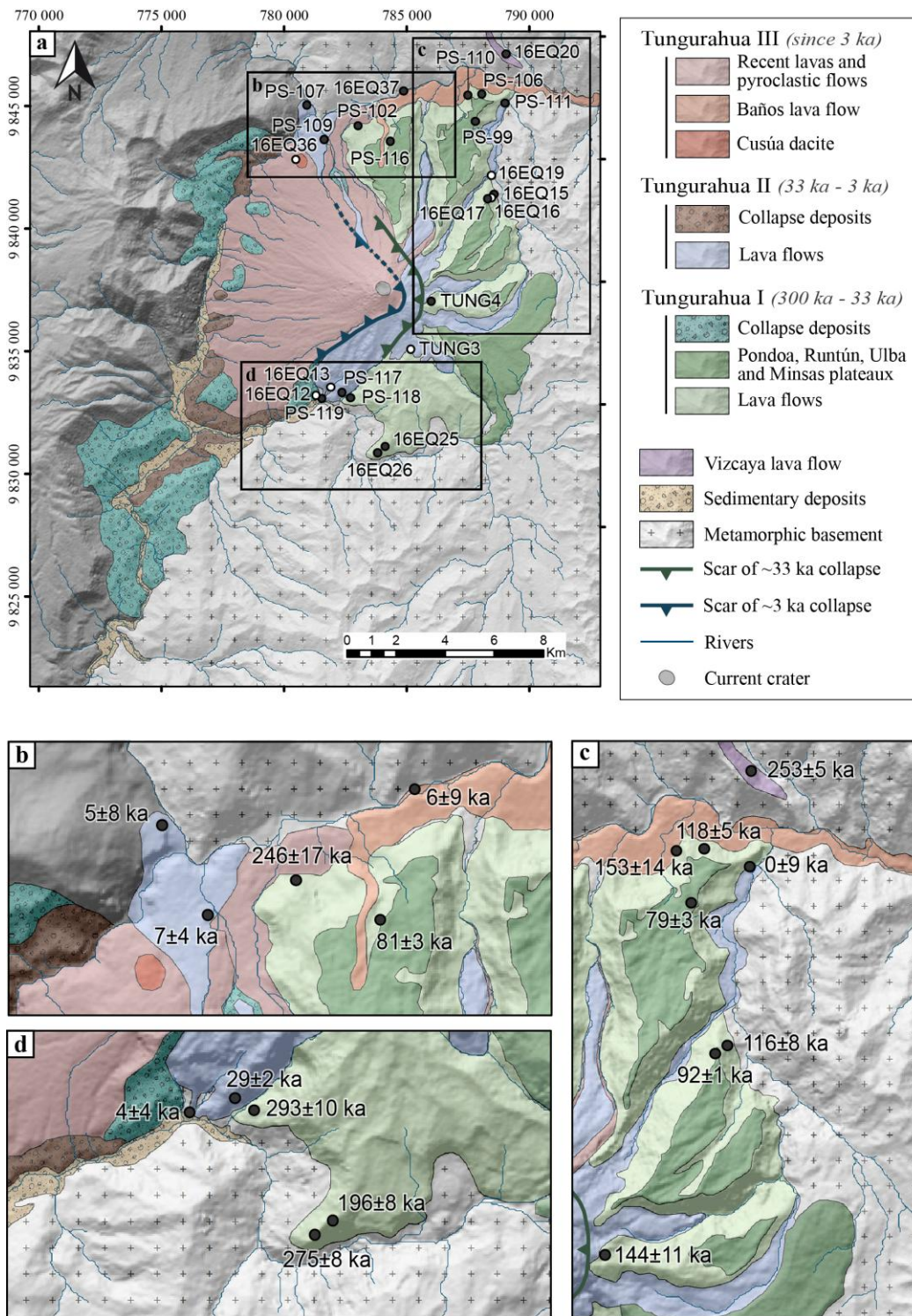
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1043 **Fig. 3:**

1044 a) Revised geological map of Tungurahua volcano reported on shaded SRTM DEM after considering our new  
 1045 ages and field observations. Sample locations presented in this study are indicated by a black (K-Ar dated  
 1046 samples) or a white (non-dated samples) symbol. All samples have been analysed for major and trace elements.  
 1047 Geological units are updated from Hall et al. (1999), and the collapse deposits cartography is presented in  
 1048 Bustillos et al. (2011). b), c), and d) are detailed views of the northern, southern, and eastern flanks respectively,  
 1049 showing location of the new K-Ar ages.  
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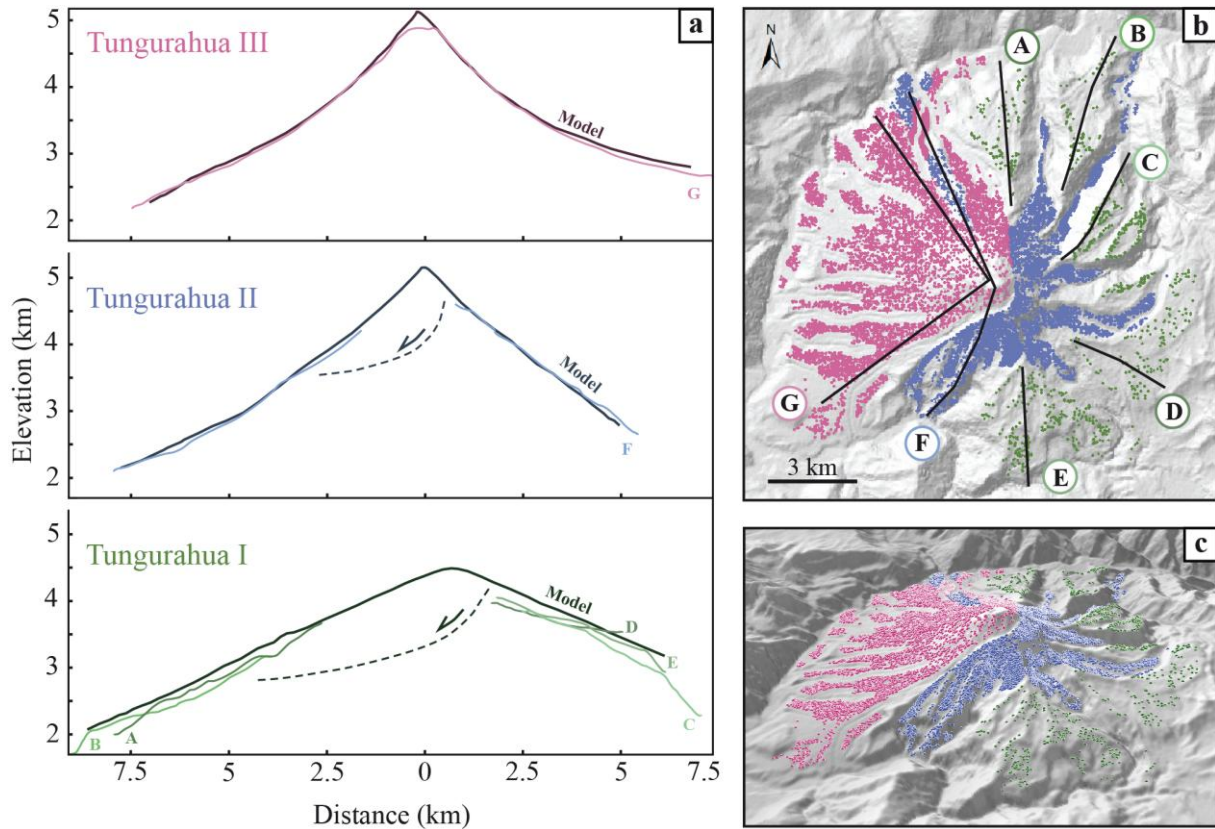


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1052 **Fig. 4:**

1053 a) Current elevation profiles of the three edifices, with their respective topographic models before erosion. The  
1054 location of the profiles is indicated in Fig. 4b. Asymmetrical altitude is due to the basement located higher to the  
1055 west. b) Selected points used for reconstructions reported on shaded DEM of Tungurahua volcano. c) 3D view  
1056 with the same selected point.  
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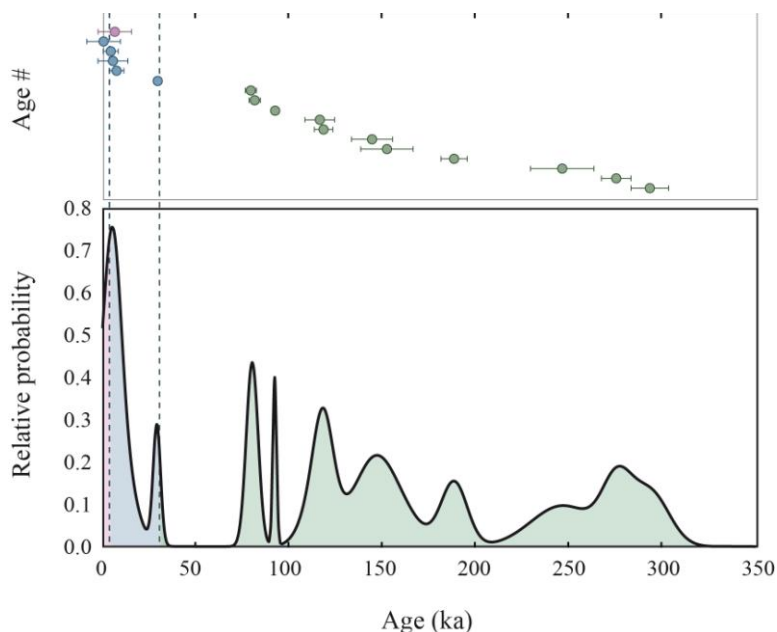
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1069 **Fig. 5:**

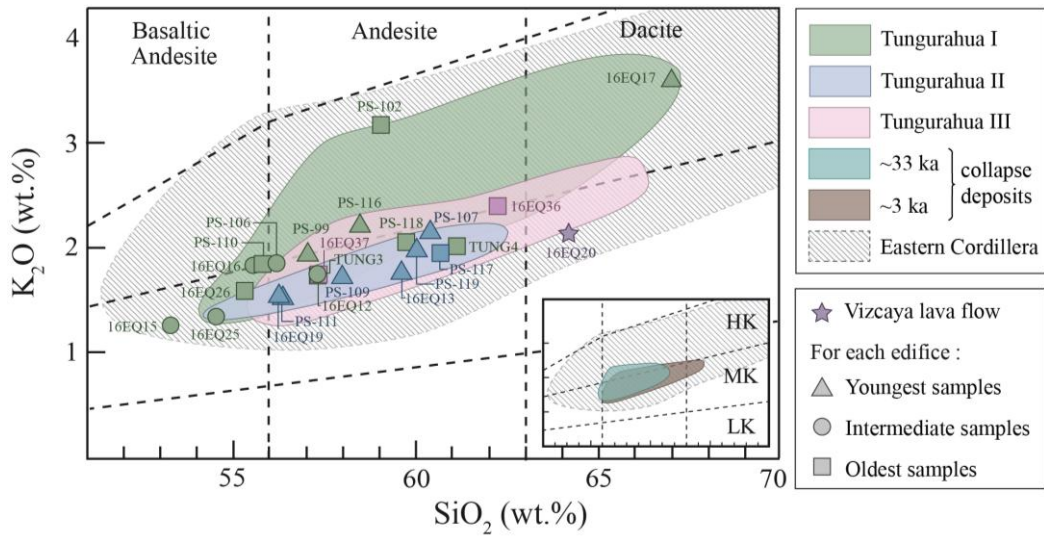
1070 Age-probability spectrum, outlining potential gaps of activity between 78 and 33 ka for Tungurahua I, and  
1071 between 27 and 15 ka for Tungurahua II. The method used to calculate the relative probability is detailed in  
1072 Deino and Potts (1992). The ages are plotted above with their  $1\sigma$  uncertainty error bars. Green dotted line: ~33 ka  
1073 collapse; blue dotted line: ~3 ka collapse.  
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1076 **Fig. 6:**

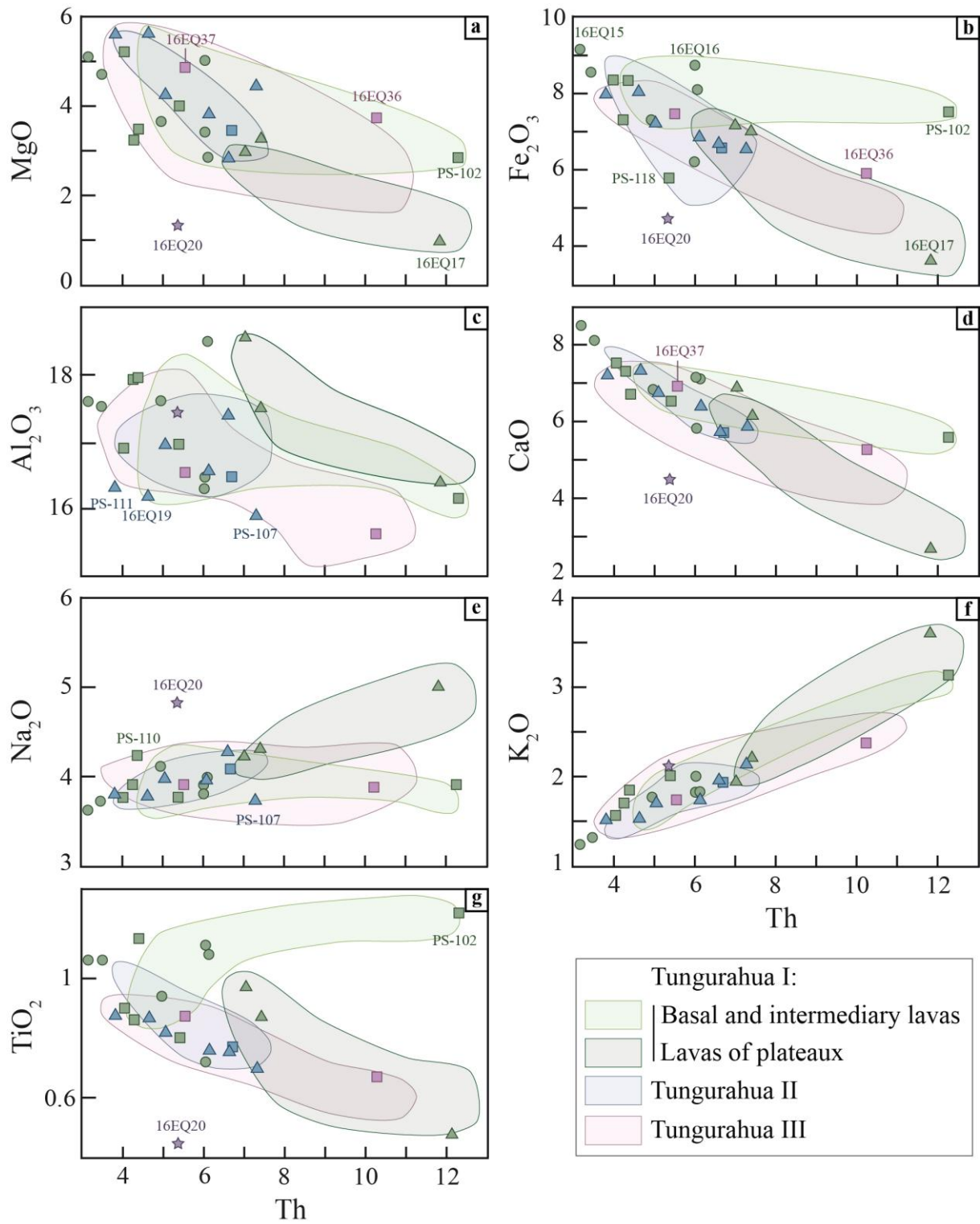
1077  $K_2O$  vs.  $SiO_2$  diagram (Peccerillo and Taylor, 1976). Hatched gray domain represent whole rock data of Eastern  
1078 Cordillera volcanoes, from the GEOROC database (<http://georoc.mpch-mainz.gwdg.de/> ; using Samaniego et al.  
1079 (2005), Bryant (2006), Hidalgo et al. (2012) and Chiaradia et al. (2014) studies). Data for Tungurahua I (green  
1080 field), Tungurahua II (blue) and Tungurahua III (pink) originate from Schiano et al. (2010); Samaniego et al.  
1081 (2011) and Le Pennec et al. (2013), and new data presented in Appendix B. Due to small amount of  
1082 Tungurahua II outcrops, the blue domain was drawn from a smaller number of measurements. The inset  
1083 represents the compositional fields of blocks belonging to Tungurahua I (turquoise) and II (brown) sector  
1084 collapse deposits. HK: high-K calc-alkaline serie, MK: medium-K calc-alkaline serie, LK: low-K calc-alkaline  
1085 serie. .  
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1088 **Fig. 7:**

1089 Variation diagrams showing the evolution of major elements (in wt. %) as a function of Th (in ppm). Symbols  
 1090 are the same used in Fig. 6. Coloured fields have been obtained from data published in Schiano et al. (2010) and  
 1091 analyses given in Appendix B. Samples from the latter have been associated to a unit considering their location  
 1092 and stratigraphical position.  
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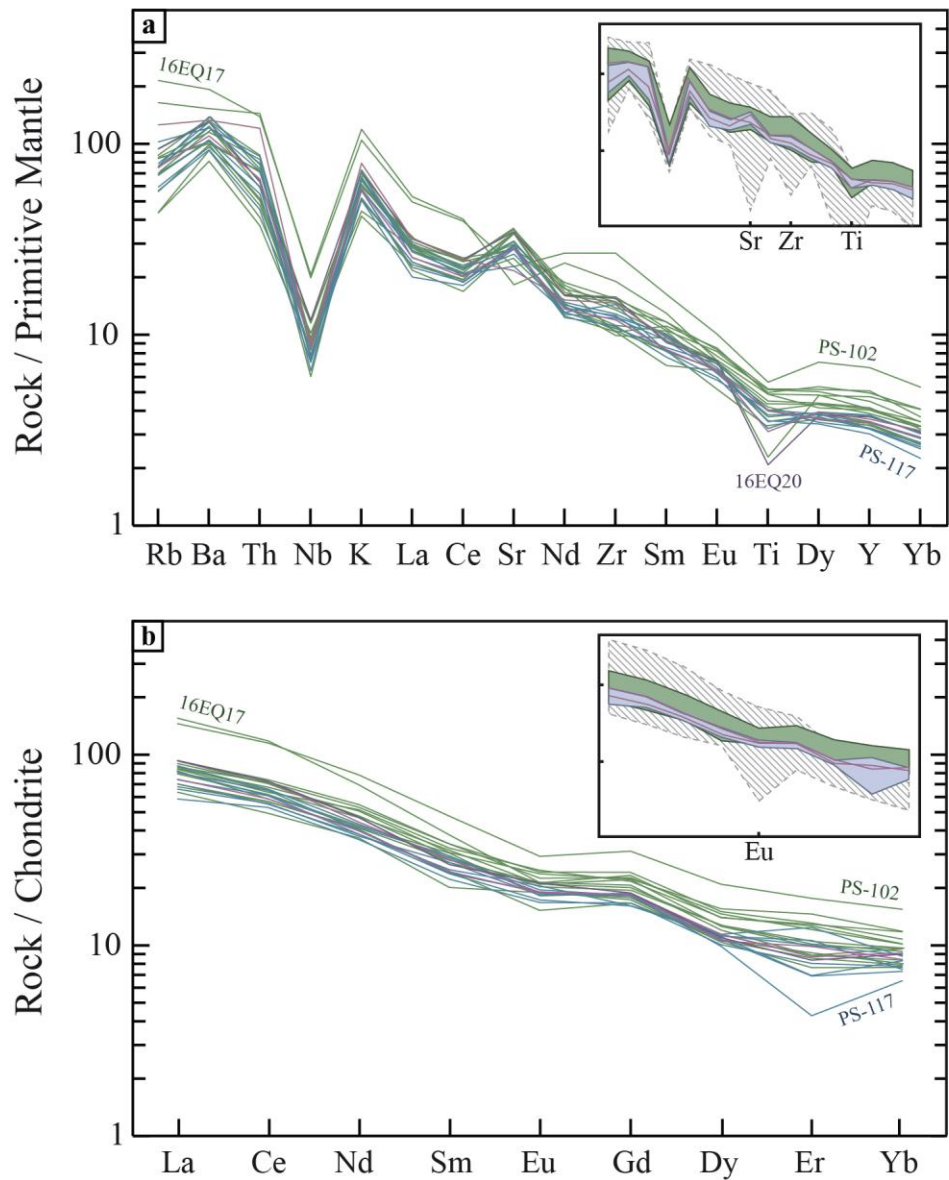
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1096 **Fig. 8:**

1097 a) Incompatible elements normalized to primitive mantle spider diagram (Sun and McDonough, 1989). Hatched  
 1098 grey domain represent Eastern Cordillera data. Analysed samples are represented in green (Tungurahua I), blue  
 1099 (Tungurahua II), pink (Tungurahua III) and purple (Vizcaya lava flow). b) Rare Earth Elements normalized to  
 1100 chondrites diagram (Sun and McDonough, 1989). Symbols are the same used in Fig. 8a.

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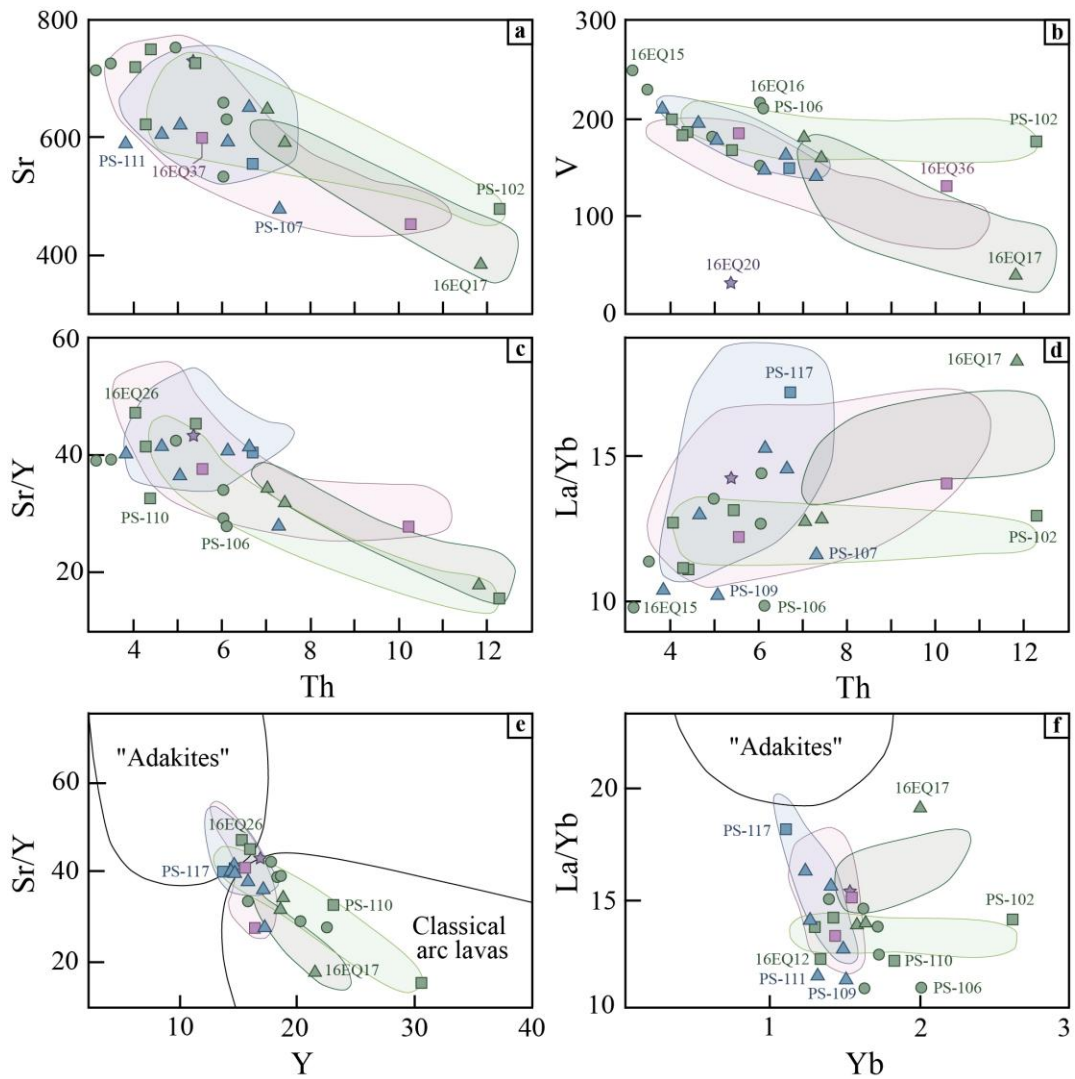
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1108 **Fig. 9:**

1109 Variation diagrams of trace elements (in ppm). a-b) Sr and V vs. Th. c-d) (Sr/Y) and (La/Yb) vs. Th. e-f) (Sr/Y)  
 1110 vs. Y and (La/Yb) vs. Yb. "Adakite" and classical arc lavas fields from Castillo et al. (1999). Symbols, caption  
 1111 and references are the same used in Fig. 7.  
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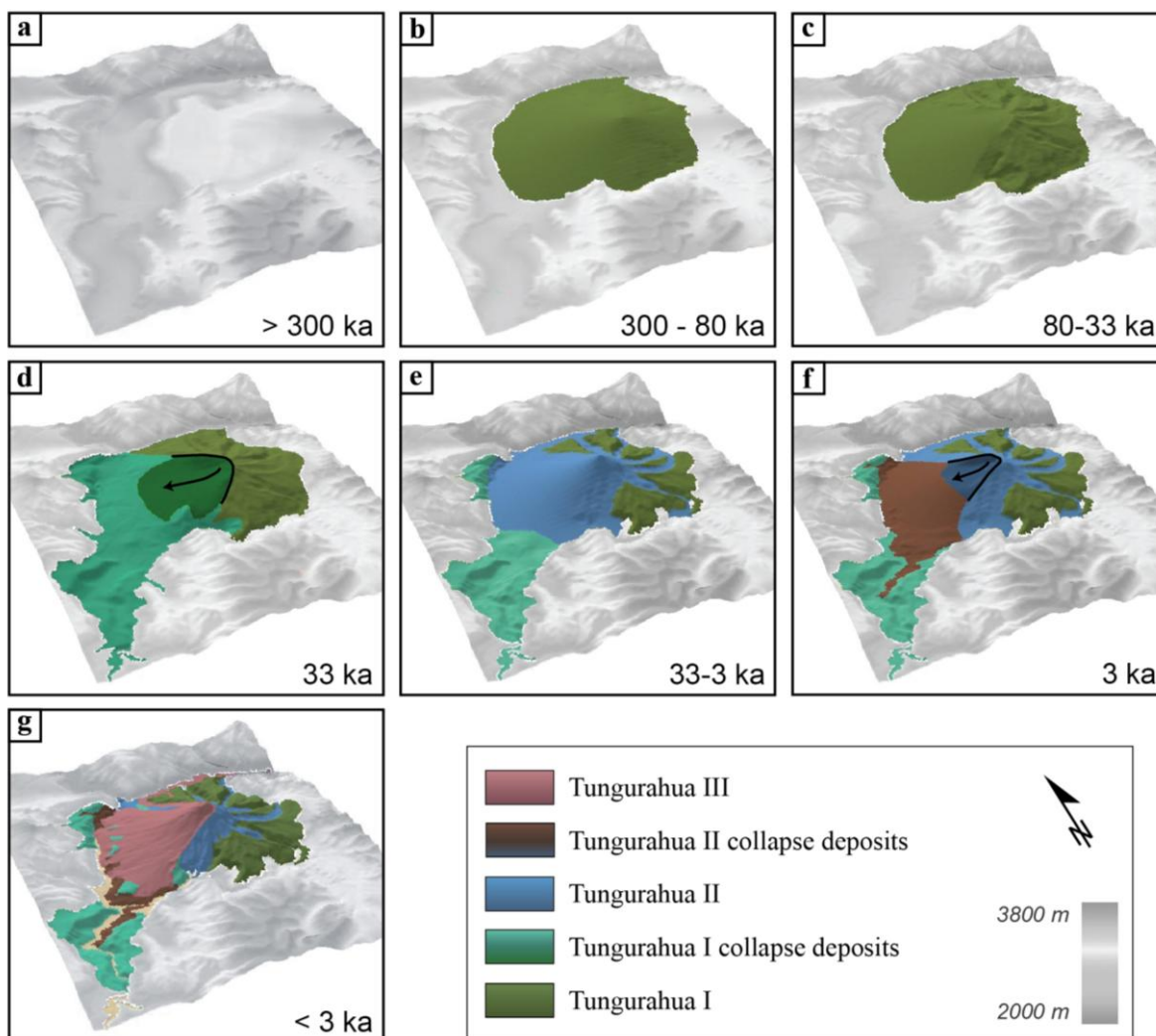
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1121 **Fig. 10:**

1122 Morphological reconstruction of Tungurahua volcano: a) before the onset of activity, b) at the end of  
 1123 Tungurahua I construction, c) after the quiescent period preceding the first flank collapse (note that the western  
 1124 flank no longer crops out today, this is therefore a modelling based on the volumes of the deposits, and does not  
 1125 consider incised valleys), d) after the first western sector collapse, e) at the end of Tungurahua II construction, f),

1126 after the collapse of the summit of Tungurahua II, and g) the current morphology, with the construction of  
 1127 Tungurahua III.  
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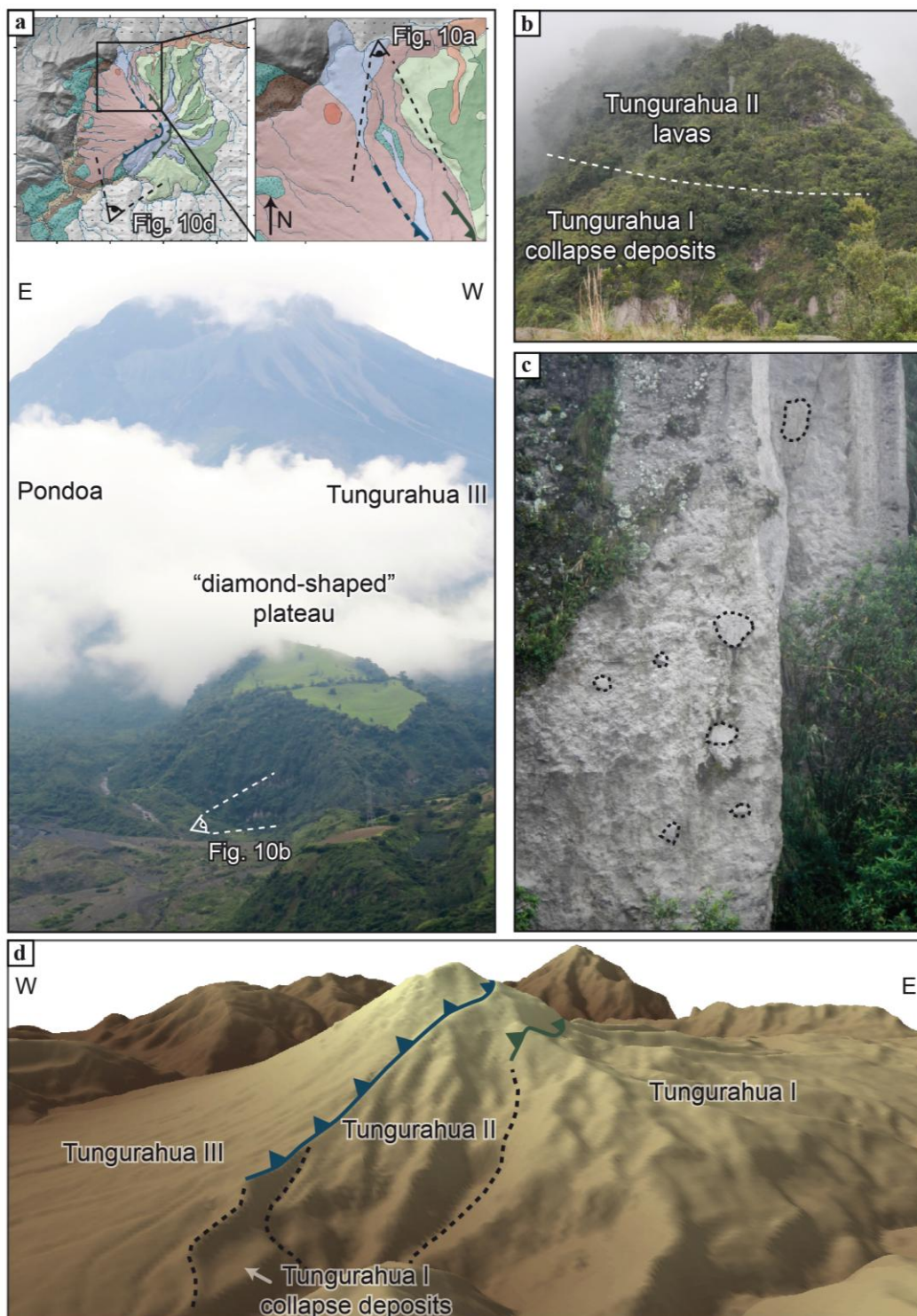


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1136 **Fig. 11:**

1137 a) Overview of the “diamond-shape” plateau, located between Pondoia and Tungurahua III deposits. b)  
 1138 Cross-sectional view of the plateau, composed of Tungurahua I collapse deposits overlaid by Tungurahua II lava  
 1139 flows. c) Detailed view of Tungurahua I collapse deposits, showing blocks in a light breccia matrix. Most

1140 massive blocks are highlighted by black dotted lines. d) 3-D view of the southwestern flank. Green line: ~33 ka  
 1141 collapse scar; blue line: ~3 ka collapse scar. Dotted lines delimit the three edifices.  
 1142



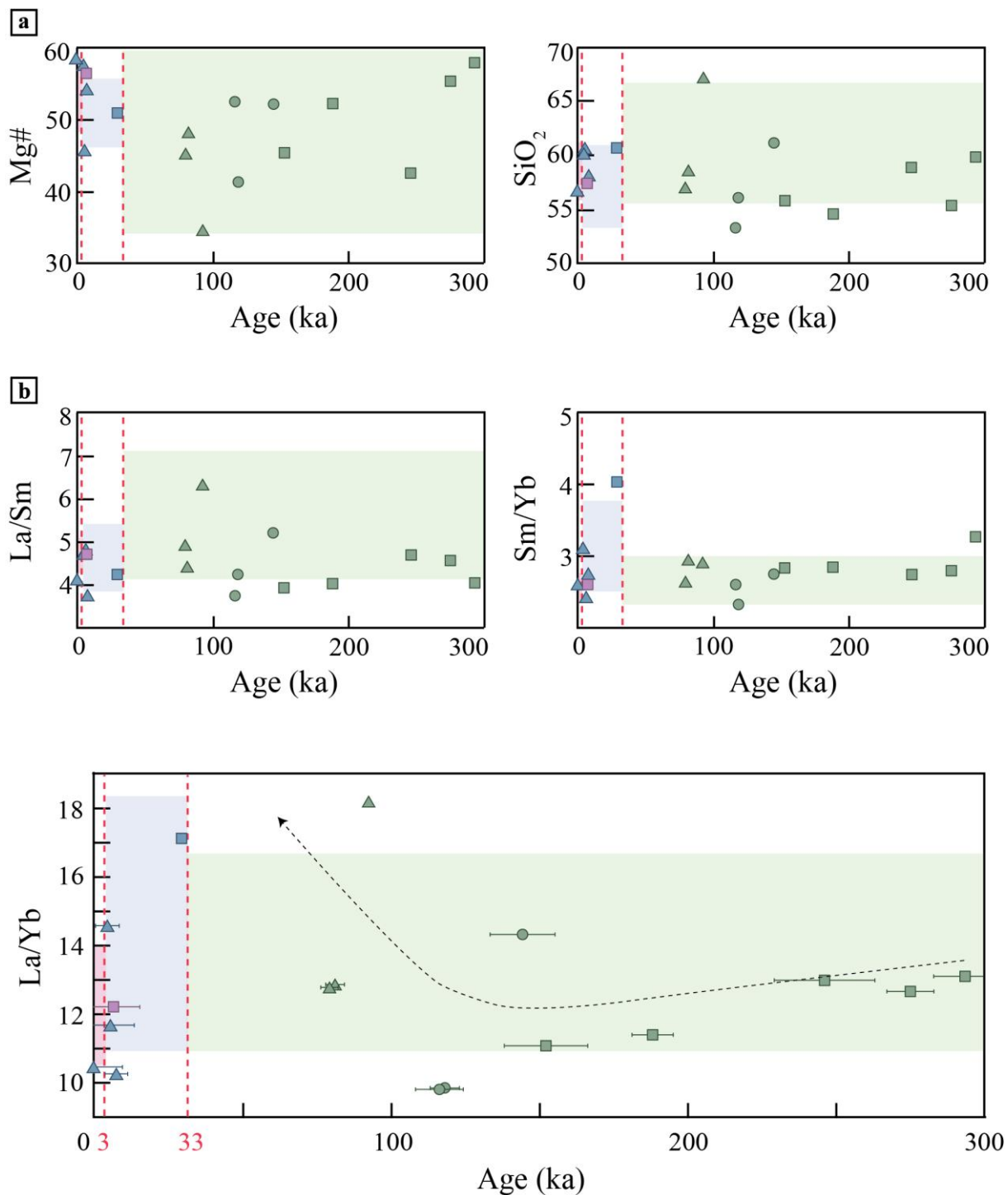
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1144 **Fig. 12:**

1145 a) Mg# (=100\*Mg/(Mg+Fe<sup>2+</sup>)) and SiO<sub>2</sub> (in wt. %) content as a function of chronology. b) La/Yb, La/Sm and  
 1146 Sm/Yb ratios as a function of chronology. Red dotted lines indicate the two sector collapses. Symbols are the



1147 same used in Fig. 6. Coloured fields represent the range of Mg#, SiO<sub>2</sub> and REE ratios obtained from our non-  
 1148 dated samples and analyses given in Appendix B.  
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**Table captions**

1155 **Table 1:**

1156 New K-Ar ages obtained on groundmass in this study. Results are listed from the oldest to the youngest. Column  
1157 headings indicate sample name, detail of the outcrop location, sample coordinates projected using the Universal  
1158 Transverse Mercator (UTM) coordinate system (Zone 17), potassium concentration in percent, radiogenic argon  
1159 content in percent and  $10^{11}$  atoms per gram, ages and weighted mean age in ka, with 1-sigma accuracy. Ages  
1160 marked with an asterisk (\*) were calculated using unweighted average.  
1161

1162 **Table 2:**

1163 Major and trace element composition of whole-rock samples. All major data were brought down to a total of  
1164 100%. Samples marked with an asterisk (\*) have not been dated.  
1165

1166 **Table 3:**

1167 Results of volumes, magmatic productivity and erosion rates calculations, given at 1-sigma accuracy, obtained  
1168 from the palaeosurfaces modelling. Raw volume corresponds to the volume calculated from the surface model  
1169 without taking into account erosion that occurred during the construction. Total volume corresponds to the sum  
1170 of the raw volume and the volume eroded during the construction, based on the erosion rate. Min-max values  
1171 were calculated by dividing the minimum volume by the maximum time period, and the maximum volume by  
1172 the maximum time period, respectively. Erosion rate in T/km<sup>2</sup>/ka was calculated considering a rock mean  
1173 density of 2.7. Ages marked with an asterisk (\*) are previous <sup>14</sup>C published ages (Le Pennec et al., 2013).  
1174

1175 **Table 4:**

1176 Summary of Tungurahua eruptive and erosion rates, compared to rates from other published studies.  
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1181 **Appendix captions**

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1183 **Appendix A:**

1184 Thin sections of all samples (in alphabetical order), observed under an optical microscope. Photographs in  
1185 left-hand column were taken with a polarized light, and in right-hand column with a natural light.  
1186

1187 **Appendix B:**

1188 Major and trace element composition of Tungurahua whole-rock samples, presented in Hall et al. (1999) and in  
1189 Bustillos (2008). All major data were brought down to a total of 100%. Column headings indicate edifice,  
1190 sample name, outcrop coordinates projected using the Universal Transverse Mercator (UTM) coordinate system  
1191 (Zone 17) and given within 100 meters, and volcanic unit. Results are listed in two parts: lavas, from potentially  
1192 oldest to youngest, then blocks belonging to sector collapse deposits. pl : plateau.  
1193

1194 **Appendix C:**

1195 KMZ file containing the location of the samples described in this article.  
1196