7 Statistical Petrology Reveals a Link Between Supercontinents

8 Cycle and Mantle Global Climate

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10 Jérôme Ganne¹, Xiaojun Feng¹, Patrice Rey², Vincent De Andrade³

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12 1 - IRD, UR 234, GET, Université Toulouse III, 14 Avenue Edouard Belin, 31400 Toulouse, France

13 2 - Earthbyte Research Group, School of Geosciences, The University of Sydney, Sydney NSW 2006, Australia

14 3- Argonne National Laboratory, 9700 S. Cass Avenue, Argonne, IL 60439, Chicago, USA

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ABSTRACT - The breakup of supercontinents is accompanied by the emplacement of 16 continental flood basalts and dyke swarms, the origin of which is often attributed to 17 18 mantle plumes. However, convection modeling has showed that the formation of supercontinents result in the warming of the sub-continental asthenospheric mantle 19 (SCAM), which could also explain syn-breakup volcanism. Temperature variations 20 during the formation then breakup of supercontinents are therefore fundamental to 21 understand volcanism related to supercontinent cycles. Magmatic minerals record the 22 thermal state of their magmatic sources. Here we present a data mining analysis on the 23 first global compilation of chemical information on magmatic rocks and minerals 24 25 formed over the past 600 million years; a time period spanning the aggregation and breakup of Pangea, the last supercontinent. We show that following a period of 26 increasingly hotter Mg-rich magmatism with dominant tholeiitic affinity during the 27 aggregation of Pangea, lower-temperature minerals crystallized within Mg-poorer 28 29 magma with a dominant calc-alkaline affinity during Pangea disassembly. These trends 30 reflect temporal changes in global mantle climate and global plate tectonics in response to continental masses assembly and dispersal. We also show that the final amalgamation 31

of Pangea at ~300 Myr led to a long period of lithospheric collapse and cooling until the
major step of Pangea disassembly started at ~125 Myr. The geological control on the
geosphere magma budget has implications on the oxidation state and temperature of the
Earth's outer envelopes in the Phanerozoic and may have exerted indirect influence on
the evolution of climate and life on Earth.

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38 INTRODUCTION

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Mantle convection and plate tectonics are two coupled processes driving the cooling of the 40 Earth's interior (Labrosse and Jaupart, 2007). In the last decade, numerical studies have 41 shown that the distribution and size of continental plates at the Earth surface control the 42 mantle potential temperature (*Tp*) below continents (Gurnis, 1988; Yale and Carpenter, 1998; 43 Grigné et al., 2005; Phillips and Bunge, 2005). The concept of mantle warming below 44 45 supercontinents (Anderson, 1982; Coltice et al., 2009) has challenged the plume paradigm (Hill, 1991; Ernst and Buchan, 1997; Courtillot et al., 1999) to explain continental flood 46 47 basalts (CFBs) and their dyke swarms which are now group as Large Igneous Provinces 48 (LIPs) (Ernst, 2014). Two- and three-dimensional numerical studies (Gurnis, 1988; Grigné et 49 al., 2005; Coltice et al., 2007; Lénardic et al., 2011) show that the potential temperature of the convective mantle gradually increases up to ~ 150 °C as a response of the lengthening of the 50 convective wavelength as plates aggregate into a supercontinent. Locally, this seems to be 51 52 confirmed as primary magmas from the ~200 Myr old Central Atlantic Magmatic Province 53 (CAMP), emplaced at the onset of Pangea dispersal, point to a Tp 50 to 150 °C warmer than normal, but not warm enough for a plume origin according to some authors (Hole, 2015; 54 Rey, 2015). However, it remains quite uncertain how great an excess Tp might be derived 55 from a plume originating from the deep mantle (Bunge, 2005). Though, if the supercontinent 56

57 cycle modulates the temperature in the convective mantle then, on a global scale, magmatic 58 minerals like olivine, pyroxene, amphibole and plagioclase should have recorded over the past 59 ~600 Myr a measurable increase in crystallization temperature (T) in relation to the Pangea 60 assembly phase, and decrease during its dispersal.

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Despite uncertainties in measuring absolute values of T and Tp (Herzberg, 2011; Putirka, 62 63 2016), we hypothesize that their trends over several tens to hundreds Myrs are likely to be representative of changing thermal regimes in the sub-continental asthenospheric mantle 64 (SCAM). Figure 1a illustrates how the melting temperature at the solidus, described by the 65 potential temperature, impacts the chemical signature (MgO content) of magmas formed at 66 the solidus (i.e. primary magmas). During mantle upwelling, including that induced by 67 thinning of the continental lithosphere, the "exhumed " mantle follows an adiabat (blue or red 68 dashed lines) intersecting the solidus at a depth which depends on the potential temperature 69 70 (blue and red dots in **Fig.1a**). Primary melts are produced between the solidus where melting 71 begins (blue and red stars in **Fig.1a**) and the base of the lithosphere where melting ceases. 72 This is called decompression melting. Melting is fractional, and the primary melt is the average of the melt compositions that are generated between the pressures at which melting 73 74 begins and ends. Should these melts be extracted as soon as they are produced, they would follow their respective melt adiabat (black arrows). During fast ascent through a thin and/or 75 hot continental lithosphere, the composition of basaltic magmas reaching the Earth's surface 76 77 remains largely unchanged (Putirka, 2008). As the magmas approach the surface, olivine crystallizes within a few 10's of km from the surface (Ghiorsio & Sack, 1995). The potential 78 temperature of the mantle, from which these olivine-bearing tholeiitic basalts are extracted, 79 can be derived from the composition of the olivine (Putirka, 2016). In contrast, during slow 80 81 ascent through a thick and/or cooler continental lithosphere, sequential crystallization and

82 separation of minerals causes basaltic melts to evolve toward a progressively more calc-83 alkaline, Si-rich, composition (Grove and Baker, 1984), especially if water is present. If the 84 starting basalt was not sufficiently hydrous, fractional crystallization will likely lead to 85 alkaline, not calc-alkaline differentiation. Importantly, clinopyroxene and plagioclase start to crystallize earlier and at higher-pressure than olivine (Grove and Baker, 1984; Ghiorsio & 86 Sack, 1995; Villiger et al, 2007; Whitaker et al., 2007; Smith, 2014). As MgO is incorporated 87 88 into the clinopyroxene, the residual melt in calc-alkaline basalts tends to have a lower MgO content. In addition, as pressure (and water) promotes the incorporation of Ca in plagioclase, 89 90 basaltic melts become depleted in Ca as they approach the surface. Continued fractionation at lower pressure leads to magmatic rocks of dacitic and rhyolitic composition (Bachmann, 91 92 2016). These first order petro-geochemical rules suggest that magmatic systems characterized by low level of fractionation, in the near absence of water, can potentially capture changing 93 94 thermal regimes in the sub-continental asthenospheric mantle (SCAM), whereas in 95 fractionated and more hydous (i.e. calc-alkaline) magma systems the complementary compositional trends of pyroxene and feldspar can give insights into the thermal and hydrous 96 97 state and/or thickness of the lithosphere.

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99 METHOD

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Following the methods of Putirka (2008), built on a comprehensive review of thermobarometers for magmatic rocks, we compared different magmatic mineral compositions with bulk rock compositions using experimentally derived thermometers to obtain temperatures of crystallization. To unravel the thermal evolution of magmas we have used only the most robust thermometers as proposed in Putirka (2008) and Ridolfi and Renzulli (2012). While magmatic temperatures can be confidently calculated from many silicate minerals in

107 equilibrium with its hosting magma (Putirka, 2008), the calculation of pressure suffers larger 108 uncertainties and was not considered here. To estimate global magmatic temperature and map 109 its trend over the past 600 Myr, we compiled from GEOROC a database including over 16 110 million data points derived from geo-referenced bulk-rock composition (dominantly volcanic) and associated mineral analysis (see **Methods** in the supplementary material). The evolution 111 of global mean intensive (geochemical) and extensive (T, Tp) parameters is reported with 112 113 associated 1-standard-error of the sample mean at 50 Myr intervals. These means were generated by Monte Carlo analysis with bootstrap resampling techniques to mitigate sampling 114 bias (Keller and Schoene, 2012). A polynomial curve (N, R²) fitting the bootstrapped values 115 was reported on the graphs. Data mining on large geochemical datasets gives access to global 116 trends by integrating much of the high-frequency variations and complex details that can exist 117 in magmatic systems. However, because contrasting trends can reflect contrasting tectono-118 119 magmatic systems we have organized our data into three groups according to the inferred 120 tectonic environment proposed by the authors of data referenced in GEOROC. These three 121 groups are (1) continental margins, (2) intra-continental settings (CFBs, LIPs, rift-related 122 magmatism and intraplate volcanism, including syn- to post orogenic magmatism) and (3) oceanic domains. By considering the broad tectonic setting of all samples we can explore 123 124 their temporal relationship with respect to the timing of amalgamation and disassembly of Pangea. 125

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127 **RESULTS**

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The temporal distribution of magmatic rocks and minerals is documented in **Fig. 1 and 2** and in the supplementary material (**SI Fig. 1 to 18**). The oceanic record is missing prior to 250 Myr (**Fig. 1b**) due to seafloor recycling through subduction during Pangea amalgamation. It

132 progressively increases after ~250 Myr as predicted by seafloor spreading accompanying 133 Pangea dispersal. Conversely, arc-magmatism at continental-margins decreases from 600 to 134 ~250 Myr, before increasing again in the last ~200 Myr (Fig. 1b). This pattern is consistent 135 with the expected decrease in the number of subduction zones during Pangea assembly as continental blocks get sutured, from 460 to 275 Myr ago, along a global network of orogenic 136 belts (e.g. Appalachian, Caledonian, Alleghanian, Variscan, Mauritanides, Ural Mountains). 137 138 This global tectonic trend predicts and explains the observed global magmatic transition from dominant calc-alkaline signature (Fig. 1c and SI Fig. 11 & 12) characterizing arc magmatism 139 (Chiaradia, 2014), to dominant tholeiitic composition between ~300 Myr and ~200 Myr when 140 Pangea was stable. The increase in the continental record of calc-alkaline magmas after ~200 141 Myr can be linked to the initiation, during Pangea dispersal, of many subduction zones 142 promoting fractional crystallization in the deep crust of hydrous arc magmas (Chiaradia, 143 144 2014), and increasing contribution of shallow and more differentiated magma sources from 145 continental reworking (magma mixing, Grove and Baker, 1984). Moreover, one highly 146 efficient category of continental reworking, that is too often overlooked, is via sediment 147 subduction and subduction erosion - whereby subducted material of continental crustal origin is subducted and incorporated directly into new magmas. If these mechanisms are important 148 149 for crustal reworking, this would produce a strong positive correlation between calc-alkaline magmatism and crustal reworking. 150

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Continental reworking is recorded in the geochemical signature of magmatic biotite (Shabani et al., 2003) forming at supra-subduction or crustal levels, with a transition from metaluminous (mantle-derived I-type granitoids) to peraluminous (crustal-derived S-type granitoids) signature before ~300 Myr and after ~200 Myr (**Fig. 2d** and **SI Fig. 11d**). In addition, a recent global compilation of zircon isotopic data shows that continental crust reworking decreases from ~600 to ~225 Myr (**Fig. 1c**), during a period of gradually higher net crustal growth rate (Dhuime et al., 2012) . Subsequently, the crustal reworking rate increases, the shift correlating with the breakup of Pangea. The major tectono-magmatic trend captured by our dataset over the aggregation and dispersal of Pangea gives reasonable confidence that it is representative of exposed magmatic systems through time with minimum sampling bias.

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163 In the continental record, the Ti-content in clinoamphibole, the Ca-content of plagioclase, the Mg-content of pyroxene and the Mg-content of olivine (e.g. those equilibrated with liquid) 164 show an evolution from ~600 Myr ago involving an increase then a decrease to present (Fig. 165 **2a,b**). The compositional peaks are diachronous, being reached at 325 ± 25 Myr (Ti-content 166 in clinoamphibole), ~225 Myr (Mg-content of pyroxene and the Ca-content of plagioclase), 167 and ~125 Myr (Mg-content of olivine). Not surprisingly, crystallization temperatures shows a 168 169 similar trend for pyroxene, amphibole and plagioclase with increasing crystallization 170 temperatures of ~150 °C from ~600 to ~225 Myr, and decreasing temperature since ~225 Myr 171 in both the continental and oceanic records (Fig. 2c and SI Fig. 13). Interestingly, the thermal 172 peak of olivine is preceded by a high-temperature plateau evolution, over a duration of ~ 100 Myr, overlapping with the thermal peak of pyroxene and plagioclase (Fig. 2a and 2c). The 173 olivine- and glass-based thermometers (Putirka, 2008) yield information on mantle-derived 174 primary magma compositions and mantle potential temperatures (see Method, section 3 in 175 the supplementary material). We observe a progressive temperature decrease of ~ 100 °C since 176 177 ~125 Myr that correlates, in the continental record, with a decrease in MgO-content in olivine and their host rocks (Fig. 2a). Consistent Tp estimates have been obtained using the 178 PRIMELT3 MEGA software (Herzberg and Asimow, 2015) (Fig. 2c and SI Fig. 14 to 17). 179 180

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182 **DISCUSSION**

Figure 2 shows that the maxima in composition and the maxima in crystallization 184 185 temperatures for minerals in continental to oceanic rift magmatic systems are reached 186 between the final stages of Pangea assembly (~325 Myr), and the final stage of its dispersal (~125 Myr). On a plate scale, warming by up to ~150 °C of magmatic systems during Pangea 187 aggregation is predicted by convection numerical models (Gurnis, 1988; Phillips and Bunge, 188 189 2005; Coltice et al, 2007). Therefore, we interpret the peaks magmatic temperatures between ~325 and 125 Myr, including the maximum temperature of primary magmas (T_p) derived 190 191 from continental basalts, as the consequence of a mantle warming climax following Pangea assembling phase (Fig. 3a). During the same period, the rate of crustal reworking reached its 192 193 minima (Fig. 1c) as the internal orogens stitching Pangea's continental crust ceased to operate (Collins et al., 2011), and magmatic systems became dominantly tholeiitic and metaluminous 194 (Fig. 2d) as subduction zones decreased in numbers. During Pangea dispersal, from ~200 to 195 ~125 Myr ago, Pangea blocks moved away from each other. We propose that the associated 196 shortening of the flow wavelength (e.g. Grigné et al., 2005) and the lateral advection of cooler 197 198 oceanic asthenosphere underneath continental blocks (Farrington et al., 2010) explain the 199 cooler mantle magmatic systems observed in continental areas (Fig. 2c). This temperature 200 evolution correlates with the observed change in the geochemistry of magmas as shown by the switch from dominantly tholeiitic magmatic systems when Pangea was stable (~300 to 201 202 200 Myr, Fig. 3a) to dominantly calc-alkaline during its dispersal as subduction zones became more prominent (Fig. 3b). This is consistent with the switch towards increasing 203 crustal reworking (Fig. 1c) which has been emphasized in recent studies (e.g. Dhuime et al., 204 2012), as well as the reworking of the SCLM in orogenic systems (i.e the Circum-Pacific 205 accretionnary orogens, Collins et al., 2011). 206

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208 The evolution of magmatic temperature at crustal, mantle lithosphere and asthenosphere 209 levels follow similar trends with a ~200 Myr offset between the onset of crustal cooling (~325 Myr, purple curve in Fig. 3c), and the onset of the cooling of convective mantle (~125 Myr, 210 211 green curve in **Fig. 3c**). Aggregation of Pangea between 450 and 275 Myr (Veevers, 2004) 212 led to a network of orogenic belts (Collins et al., 2011). Towards the end of this orogenic cycle, crustal thickening, radiogenic heating and mafic magma underplating (Lyubetskaya 213 214 and Ague, 2010) led to a peak in magmatic temperature at crustal level, associated locally with crustal anataxis and the formation of migmatites (Augier et al., 2015) (Fig. 3c). From 215 216 ~300 Myr onwards, orogenic crusts recovered a normal thickness via gravitational collapse and the average crustal geotherm became progressively cooler. In the lithospheric mantle, the 217 warming preceding the peak magmatic temperature may be linked to warming in the orogenic 218 crusts above as well as warming of the convective mantle underneath. The peak magmatic 219 220 temperature in the lithospheric mantle (bracketed by the yellow trend for the upper 221 lithospheric mantle, and blue curve for the lithosphere-asthenosphere boundary) was likely 222 reached at 250±25 Myr. This delay can be explained by thermal inertia and the diffusion time 223 required for the hotter potential temperature of the convective mantle to propagate through the overlying lithospheric mantle. The peak magmatic temperature in the convective mantle 224 underneath Pangea lasted from ~225 until ~125 Myr when the major step of Pangea breakup 225 and dispersal occurred in the Albian times (Veevers, 2004) (i.e. opening of the South Atlantic 226 ocean (Fig. 3b). 227

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The global change of magma compositions and fluxes between the lithosphere and outer envelopes of the planet have a potentially fascinating connection with the evolution of the

²²⁹ IMPLICATIONS

oxidation and temperature state of the atmosphere and ocean through the Earth's history. Indeed, increasing magma temperature may first lead to an increased drawdown of atmospheric oxygen. Sulfur degassing has been shown to be more efficient at high magma temperatures (Scaillet et al., 1998), and sulfur is an important participant in oxygen drawdown (through formation of SO_4^{2-} compounds). Massive degassing of cooler calc-alkaline magmas is thus likely to reduce the sink for oxygen (Kump & Barley, 2007) and consequently drive or amplify a decrease in global temperatures at the surface of the planet (Royer et al., 2004).

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Other broader feedbacks between the chemical record of magmas and climate change 241 (Schmidt et al., 2015) in the last ~300 Myrs remain to be explored, acknowledging that the 242 more calc-alkaline composition of magmas, the more explosive the activity of these 243 volcanoes, placing more dust in the stratosphere, thereby cooling the Earth. The coupling 244 245 between the inner and outer envelops of the Earth through magma activity is particularly 246 relevant for better assessing the origins of Global Cooling of the planet since ~250 Myrs (Rover et al., 2004), following the "Great Dying" Permian Mass Extinction (Raup & 247 248 Sepkoski, 1982) caused by the Siberian Trap LIPs (Burgess and Bowring, 2015). Overall, our results have profound implications on the biosphere-hydrosphere-geosphere interactions in 249 250 the Earth history (McKenzie et al., 2016).

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262 Author contribution

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J.G. conceived the study and wrote the paper. X.F. contributed to the database building. P.R helped generate the research idea with J.G. and contributed to the writing and focusing of the paper. V.D.A. developed the Matlab scripts.

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269 Author information

270 Correspondence and requests for materials should be addressed to J.G. (jerome.ganne@ird.fr)

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420 FIGURES CAPTIONS

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422 Figure 1. Chemical record of magmas through times. (a) Phase diagram, contoured for MgO, of mantle melting (after Herzberg and Asimow, 2015; modified). Mantle melting at the 423 424 solidus (blue and red stars) for contrasting potential temperatures (blue and red circles) leads to contrasting MgO content in primary magmas. During their ascent to the surface (black 425 426 arrows) these primary magmas are modified by (often fluid-induced) fractional crystallization in the lithosphere and by crustal assimilation (Grove and Baker, 1984), forming calc-alkaline 427 series in which the mineralogical composition depends on the depth of crystallization. The 428 less modified magmas (tholeiitic affinity) keep the memory of the potential temperature. The 429 colored bars in (a), which are labeled by mineral type, are not precisely constrained by 430 temperature data. (b) Intracontinental, arc and oceanic magmatic systems through time. 431 Oceanic magmatic systems and magmatic systems at continental margins are increasing 432 during Pangea dispersal from ~175 Myr onwards. (c) Peak production of tholeiitic magmas 433 (TH index : more Fe₂O_{3 total}-enriched at MgO ~4-6 wt%; Chiaradia, 2014) and minimum crustal 434 reworking (yellow curve, from Dhuime et al., 2012) are reached during Pangea stability 435 phase. 436

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Figure 2. Chemical record of magmatic minerals. (a) Temperatures of crystallization through times based on magmatic olivines from continental settings (red points) and evolution of Mg-content of olivines and their host rocks. (b) Chemical evolution of pyroxenes, plagioclases and clinoamphiboles. (c) Crystallization temperature of magmatic minerals (open grey circles) obtained with different thermometers (Putirka, 2008; Ridolfi and Renzulli, 2012). Statistical assessment of their averaged evolution through time is given by the colored drawbars. From ~200 Myr onwards, the increasing occurrence of lower magmatic

temperatures for pyroxenes and plagioclases point toward increasing fractionation of magmas 446 at progressively deeper levels in a progressively cooler continental lithosphere. Mantle 447 potential temperature (T_p) is calculated with the Ol-Liq. thermometer (green points), using a 448 melt fraction of 0.15. (a) Samples of basalts have been re-analysed using PRIMELT3 MEGA 449 (Herzberg and Asimow, 2015) software to obtain complementary Tp estimates (red points; 450 451 see Method, section 3 in the supplementary material). (d) Correlation between the occurrence of Al-rich biotite and the Aluminium Saturation Index (ASI) of magmas defined as the molar 452 ratio Al_2O_3 (CaO + K₂O + Na₂O) (Zen, 1986). Numbers in the middle of the purple curve 453 correspond to the ASI values. 454

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Figure 3. Supercontinent cycle and thermal regime. (a, b) Sketches illustrating plate and 457 mantle dynamics and climate (Farrington et al., 2010) during amalgamation and breakup of 458 459 the Pangea supercontinent. Paleogeographic configurations are based on a Triassic and Cretaceous reconstruction (Veevers, 2004). The external (circum-Pacific) system comprises a 460 461 number of discrete orogens that, together, have probably existed for 550 Myr (Collins et al., 2011). (c) Thermal peaks for magmatic pyroxenes and plagioclases (~225 Myr), dominantly 462 463 tapping (or considered to reflect conditions in) the sub-continental lithospheric mantle, span a period of orogenic collapse for the belts suturing the Pangea supercontinent. Thermal peak for 464 olivines (~125 Myr), dominantly tapping the sub-continental asthenospheric mantle, 465 corresponds to a period of enhanced supercontinent breakup. The green, blue and yellow 466 curves come from **Fig. 2**; the shape of the purple curve "metamorphic record" is not 467 constrained by temperature data. 468

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Figure 1



Figure 2



Peak of temperature in the lithosphere, dominant tholeiitic record, minimum crustal reworking



Lithosphere and asthenosphere cooling , switch to more crustal reworking and more calc-alkaline affinity



Figure 3