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1 **High-precision zircon age spectra record the dynamics and evolution of large** 2 **open-system silicic magma reservoirs**

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12 13 Highlights

- 14 • U-Pb CA-ID-TIMS dates of zircon from plumbing system of a Permian caldera
- 15 • Distribution of ages interpreted through a new zircon saturation model
- 16 • Relative decrease of zircon mass with time implies monotonic cooling
- 17 • Late peak in zircon crystallization indicates increase in heat supply
- 18 • Evidence for open-system processes preserved in zircon record of super-eruptions

19 **ABSTRACT**

20 The emplacement history and thermal evolution of subvolcanic magma reservoirs determine
21 their longevity, size, and ability to feed volcanic eruptions. As zircon saturation is dependent on
22 melt temperature and composition, quantitative analysis of zircon age distributions provides
23 insight into the timing and magnitude of intensive parameter variations during the lifetime of a
24 magma reservoir. Here we present chemical abrasion-isotope dilution-thermal ionization mass

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4 25 spectrometry (CA-ID-TIMS) U-Pb zircon crystallization ages and in-situ zircon trace-element
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6 26 geochemistry for a suite of caldera-related, coeval plutonic and volcanic units of the Permian
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9 27 Sesia Magmatic System (northern Italy). This dataset documents the protracted growth and
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11 28 evolution (~1 Myr) of a voluminous (>1000 km³), upper crustal (3.5-2.0 kbar) silicic magma
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14 29 reservoir. Systematic changes in zircon composition with time reveal episodic intrusions of new
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16 30 magma into a single, progressively differentiating reservoir which was dominantly kept at high
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19 31 crystallinity conditions (>60 vol.%). The volcanic and plutonic units both show dispersed and
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21 32 heterogeneous CA-ID-TIMS age distributions. A stochastic, thermodynamics-based zircon
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24 33 saturation model accounting for fractional crystallization, recharge and thermal rejuvenation
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26 34 effects on zircon growth and stability in a rhyolitic magma body reproduces the observed
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29 35 distributions of zircon U-Pb ages. Model results suggest that zircon compositional variability and
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31 36 crystallization age heterogeneity, characteristic of the Sesia and other large silicic systems,
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34 37 distinguish open- and closed-system magmatic processes. Specifically, an increase of the mass of
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36 38 crystallized zircons over time is the hallmark of thermally and chemically mature magma
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39 39 reservoirs, capable of producing voluminous eruptions upon changes in the thermal and
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41 40 mechanical state of the system. Conversely, a decrease in crystallized zircon mass over time
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44 41 characterizes systems where crystallization dominates over magma mobility, hindering
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46 42 substantial mass release during eruptive events. We suggest that quantitative analyses of
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48 43 dispersed zircon crystallization age distributions obtained with high-precision techniques can be
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51 44 applied to the plutonic and volcanic record to identify mature silicic magma reservoirs whose
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53 45 properties and storage conditions allowed for catastrophic caldera-forming eruptions. This work
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56 46 provides the petrologic community with a new tool to investigate the genesis and evolution of
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4 47 zircon-bearing intermediate to silicic intrusive magmatism and caldera-forming volcanism
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6 48 through time and across tectonic settings.
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10 49 **Keywords:** U-Pb geochronology, zircon saturation, Magma Chamber Simulator, caldera-
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12 50 forming eruptions, pluton construction, Ivrea-Verbano Zone.
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15 51 **1. Introduction**

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18 52 Time-resolved records of the thermal and compositional evolution of shallow (<3 kbar)
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20 53 crustal magma bodies provide insights into the processes that regulate tempo, volume, style of
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22 54 volcanic eruptions and the origin of silicic plutons. Zircon, a common accessory mineral in
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24 55 silicic igneous rocks, represents the prime tracer of physico-chemical processes unfolding during
25
26 56 the lifetime of a subvolcanic magma reservoir, owing to its unparalleled ability as a timekeeper
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28 57 (Schoene & Baxter, 2017) and the slow diffusivity of most elements through its crystal lattice
29
30 58 (Cherniak and Watson, 2003). Recent advances in zircon geochronology by chemical abrasion-
31
32 59 isotope dilution-thermal ionization mass spectrometry (CA-ID-TIMS) allow for internal age
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34 60 precision better than the 0.02 % level for single zircon $^{206}\text{Pb}/^{238}\text{U}$ dates (Wotzlaw et al., 2017;
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36 61 Szymanowski and Schoene, 2020). The integration of CA-ID-TIMS U-Pb geochronology, trace
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38 62 element and isotopic analyses in zircons (e.g. Schoene et al., 2012; Wotzlaw et al., 2013;
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40 63 Szymanowski et al., 2017) with advances in thermomechanical modeling (e.g. Annen et al. 2009;
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42 64 Gelman et al., 2013; Karakas et al., 2017; Huber et al., 2019) have provided invaluable insights
43
44 65 into the nature and duration of upper-crustal magmatism. These works have contributed to the
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46 66 paradigm of long-lived (10^5 - 10^6 yr), largely crystalline magma reservoirs that are periodically
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48 67 recharged by new magma inputs, as the dominant model of shallow, silicic magma storage in the
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50 68 Earth's crust (Sparks et al., 2019).
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4 69 In U-Pb geochronological studies of upper-crustal magmatic reservoirs, the internal age
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6 70 resolution afforded by CA-ID-TIMS can resolve heterogeneities in zircon crystallization dates,
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9 71 i.e. dispersed age populations (Keller et al., 2018). Such age dispersion is interpreted as either
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11 72 indicative of protracted saturation and growth of autocrystic zircons (Wotzlaw et al., 2013;
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14 73 Samperton et al., 2015; Ratschbacher et al., 2018; Szymanowski et al., 2019; Klein and Eddy,
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16 74 2023) or as a record of recycled and juxtaposed zircons crystallized in a polybaric storage system
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19 75 (Schoene et al., 2012; Barboni et al., 2015; Farina et al., 2018). In the case of protracted
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21 76 saturation, as growth and survival of autocrystic zircons is linked to the physical and chemical
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24 77 conditions of magma reservoirs (Watson, 1996; Boehnke et al., 2013; Bindeman and Melnik,
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26 78 2016), their distribution in time has been proposed to provide quantifiable constraints on the
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29 79 compositional and thermal states of crystallizing magma bodies (Caricchi et al., 2014; Tierney et
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31 80 al., 2016; Samperton et al., 2017; Kent and Cooper, 2018; Kirkland et al., 2021) and their
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34 81 eruptive potential (Weber et al., 2020). Specifically, the simple distribution of zircon mass
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36 82 crystallized as a function of time and temperature observed in monotonically cooled plutonic
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39 83 systems (Samperton et al., 2017; Kirkland et al., 2021) can be reproduced by experimentally
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41 84 derived zircon solubility models (Watson and Harrison, 1983; Boehnke et al., 2013) and
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44 85 thermodynamic assessment of phase equilibria (Ghiorso et al., 2002; Holland and Powell, 2011).
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46 86 However, no study has yet linked the complex, heterogeneously dispersed CA-ID-TIMS zircon
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49 87 age datasets to open-system, crystal-melt dynamics evident from petrological studies of silicic
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51 88 systems (Szymanowski et al., 2020) and suggested by volcanic and plutonic zircon age spectra
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53 89 displaying crystallization gaps and apparent late peaks of zircon crystallization (e.g. Wotzlaw et
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55 90 al., 2013; Rioux et al., 2016).
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4 91 To address this issue, we integrate CA-ID-TIMS U-Pb zircon geochronology and in-situ
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6 92 geochemistry on co-genetic, silicic intrusive and eruptive products of a Permian magmatic
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9 93 plumbing system (Sesia Magmatic System; Quick et al., 2009; Karakas et al., 2019) with a novel
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11 94 approach to zircon saturation modeling that combines mass- and energy-constrained
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14 95 thermodynamic tools with stochastic sampling (Keller et al., 2017; Bohrson et al., 2020,
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16 96 Tavazzani et al., **accepted**). We demonstrate that the compositional variability and heterogeneous
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18 97 age distribution of autocrystic zircons from an internally complex subvolcanic intrusion and
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21 98 coeval voluminous rhyolitic eruptions are faithful chroniclers of incremental growth,
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24 99 differentiation and episodic recharge in a large upper-crustal magma reservoir. Based on this
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26 100 successful application, quantitative evaluation of zircon age dispersion is proposed as a tool to
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29 101 push the analyses of open-system thermomechanical processes into deep time plutonic and
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31 102 volcanic records.

32 33 34 103 **2. Geology of the Sesia Magmatic System**

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37 104 The Sesia Magmatic System (SMS) is a transcrustal igneous system that developed in the
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39 105 pre-Alpine basement of the Southern Alps during early Permian extension. Due to Mesozoic-
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42 106 Cenozoic exhumation, the SMS is now exposed in tilted crustal blocks in the Italian Alps as part
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44 107 of the Ivrea-Verbano and Serie dei Laghi terranes (Fig. 1a; Fountain, 1976; Handy and Zingg,
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46 108 1991). During its four-million-year lifetime (~285 to ~281 Ma; Karakas et al., 2019) the SMS
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48
49 109 produced synchronous magmatism across a thick crustal column, ranging from a stratified
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52 110 gabbroic intrusion in the lower crust (Rivalenti et al., 1981) to explosive volcanism at the surface
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54 111 (Quick et al., 2009). At the apex of its activity, the SMS generated the Sesia Caldera (>13 km in
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56 112 diameter) that erupted at least 400 km³ of dominantly silicic materials (Quick et al., 2009).
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4 113 The main eruptive unit of the Sesia Caldera (SC) is a densely welded, crystal-rich (25-30
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6 114 vol.%) rhyolitic ignimbrite that includes patches of megabreccia (Fig. 1b). Textural and
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9 115 compositional features of quartz phenocrysts indicate that the eruption of voluminous crystal-
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11 116 rich material was triggered by thermal rejuvenation (i.e. reduction in crystallinity; Huber et al.,
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14 117 2011) of an evolved magma body, stored at near-solidus, high crystallinity conditions and
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16 118 shallow depths (2.0 to 2.5 kbar; Tavazzani et al., 2020). A layer of volcanoclastic sediments (~30
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19 119 m) separates the main eruptive unit of the SC from a ~400 m thick, welded rhyolitic ignimbrite
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21 120 unit (Sbisà, 2009), characterized by high-silica content (75-77 wt.% SiO₂), diffuse sub-parallel
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24 121 eutaxitic texture and low crystallinity (~10 vol%). This laterally continuous but volumetrically
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26 122 limited (1-10 km³) rhyolitic unit is interpreted as the product of a post-caldera eruption due to its
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29 123 stratigraphic position, and it is referred in this work as the upper ignimbrite unit.
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32 124 Directly underneath the SC deposits, the upper-crustal segment of the SMS consists of
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34 125 three granitoid plutons emplaced in the Serie dei Laghi metamorphic sequence (Handy and
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36 126 Zingg, 1991; Brack et al., 2010). The southernmost of these bodies is the Valle Mosso Pluton
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39 127 (VMP), a composite silicic intrusion (~260 km³; Fig. 1b) ranging from quartz-monzonite to high-
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41 128 silica leucogranite (65-77 wt% SiO₂). The bulk of the intrusion consists of variably porphyritic
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44 129 monzogranite which contains petrological evidence of recharge by less evolved melts and
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46 130 fluctuating thermal conditions (e.g., disequilibrium textures in low temperature phases;
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49 131 Tavazzani et al., 2017, 2020). Overall, the architecture, textural and bulk-rock compositional
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51 132 features of the VMP (Fig. 1b) reflect the evolution of a silicic magma reservoir undergoing
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54 133 protracted differentiation, driven by crystal-melt separation, and episodically recharged by
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56 134 granitic and, to a lesser extent, mafic magma (Sinigoi et al., 2016; Tavazzani et al., 2020).
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4 135 The spatial proximity, overlapping compositional range and shared thermal history
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6 136 support a genetic link between the VMP intrusion and the eruptive products of the SC. Recent
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9 137 high-precision U-Pb zircon age determinations on VMP and SC samples (Karakas et al., 2019)
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11 138 further reinforce this relationship, providing an ideal opportunity to quantify timescales of
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14 139 assembly, storage and differentiation in an interconnected, upper crustal magmatic reservoir in
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16 140 relation to a voluminous caldera-forming eruption.

141 3. Materials and methods

142 Informed by recent, detailed mapping and petrologic investigation (Tavazzani et al.,
143 2017, 2020), we selected six samples for high-precision CA-ID-TIMS U-Pb zircon dating (Fig.
144 1b) that cover the full extent of textural and geochemical variability of the upper-crustal intrusive
145 and eruptive products of the SMS. We compile 39 new dates with recently published zircon ages
146 of Karakas et al. (2019) into a dataset of 96 single CA-ID-TIMS zircon U-Pb dates for upper-
147 crustal **lithologies of the SMS**.

148 3.1 LA-ICP-MS U-Pb zircon geochronology and trace element analyses

149 Zircon grains were separated from **thirteen** crushed rock specimens using standard heavy
150 mineral separation techniques. Selected zircons were hand-picked and annealed by heating to 900
151 °C for 48 hours in a muffle furnace before mounting in epoxy mounts and polishing to expose
152 central cross sections. Zircons were then texturally characterized employing cathodoluminescence
153 (CL) imaging (Supplementary Figures S2-S4). Following imaging, zircon grains were analyzed
154 by laser ablation-inductively coupled plasma mass spectrometry (LA-ICP-MS), to determine trace
155 element abundances and U-Pb ages from the same ablation crater, employing a 193 nm Resonetics
156 Resolution S155 laser ablation system coupled to a Thermo Element XR, sector-field single
157 collector ICP-MS (Guillong et al., 2014). Detailed procedures and analytical parameters are

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4 158 reported in Supplementary Text and Supplementary Data Table S2, following community-derived
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6 159 guidelines (Horstwood et al., 2016).
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9 10 160 **3.2 CA-ID-TIMS zircon U-Pb geochronology**

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12 161 Based on CL-imaging and LA-ICPMS results, fifty-six magmatic zircons from six
13 162 samples, showing no evidence of inheritance or contamination by inclusions were selected for CA-
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15 162 ID-TIMS analyses. Individual zircons were extracted from the epoxy mounts using stainless steel
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17 163 tools and chemically abraded for 15 hours at 190°C (Mattinson, 2005), cleaned in 6 M HCl and
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19 164 3.5 M HNO₃, loaded into 200 µl Savillex microcapsules with concentrated HF and spiked with ~5-
20 165 8 mg of the EARTHTIME ²⁰²Pb-²⁰⁵Pb-²³³U-²³⁵U solution (Condon et al., 2015; McLean et al.,
21
22 166 2015). Samples were dissolved in Parr bombs at 220°C for ~60 hours. After dissolution, samples
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24 167 were dried down and redissolved in 6 M HCl at 190°C for several hours and then dried down again
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26 168 before redissolving them in 3 M HCl for ion exchange chemistry. U and Pb were separated using
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28 169 a single-column HCl-based ion exchange chemistry (Krogh, 1973). The U-Pb fractions were
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30 170 loaded on outgassed Re filaments with ~1 µl of Si-gel emitter (modified from Gerstenberger and
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32 171 Haase, 1997). U and Pb isotopes were measured using a Thermo TRITON Plus thermal ionization
33
34 172 mass spectrometer at the Institute of Geochemistry and Petrology of ETH Zurich (Wotzlaw et al.,
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36 173 2017). Data reduction, age calculation and uncertainty propagation were performed using the
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38 174 Tripoli and ET_redux software package (Bowring et al., 2011; McLean et al., 2011). Detailed
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40 175 analytical procedures are reported in Supplementary Text.
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52 177 **3.3 Zircon saturation model**

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55 178 In parallel with the CA-ID-TIMS analytical effort, we developed a model to quantify and
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57 179 track changes in saturated zircon mass during protracted crystallization, differentiation and
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59 180 repeated intrusions of magmas in an upper crustal rhyolitic reservoir. In contrast to previous
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4 181 studies (e.g. Tierney et al., 2016; Kent and Cooper, 2018; Weber et al., 2020), this model does
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6 182 not rely on a parameterized relationship between temperature and fraction of zircon crystallizing
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9 183 (Harrison et al., 2007) but integrates temperature, crystallinity, melt major and trace element
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11 184 evolution to assess zircon saturation (Fig. 2). The trajectory of intensive variables during
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14 185 crystallization is derived using the software Magma Chamber Simulator (Bohrson et al., 2014,
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16 186 2020), a modeling tool combining thermodynamic assessment of phase equilibria (based on the
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18
19 187 MELTS family of algorithms of Ghiorso et al., 2002; Gualda et al., 2012) with energy and mass
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21 188 conservation.

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24 189 The mass of zirconium (Zr) available for crystallization (m_{Zr} , Fig. 2a) is calculated at
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26 190 every step of a crystallization simulation applying the workflow developed by Keller et al.
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29 191 (2017):

$$31 \quad 192 \quad m_{Zr} = \frac{m_{liq}}{100 ([Zr]_{calc}^{melt} - [Zr]_{sat}^{melt})} \cdot (1)$$

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33
34 193 where m_{liq} is the mass of liquid phase, $[Zr]_{calc}^{melt}$ is the Zr concentration in the liquid phase and
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37 194 $[Zr]_{sat}^{melt}$ is the Zr concentration required to saturate the melt in zircon using the calibration of
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39 195 Boehnke et al. (2013). At each step (n) of the simulation, relative change in zircon mass
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42 196 ($dm_{Zr_n} = m_{Zr_n} - m_{Zr_{(n-1)}}$) is used to derive zircon crystallization probability (p_{Zr_n} , Fig. 2b)

$$43 \quad 44 \quad 45 \quad 197 \quad p_{Zr_n} = \frac{dm_{Zr_n}}{\sum_{i=0}^N dm_{Zr_i}} \cdot (2)$$

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48 198 which is normalized to the overall variation in crystallized zircon mass ($\sum_{n=0}^N dm_{Zr_n}$, N represents
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50
51 199 the total number of steps in a crystallization simulation). A zircon crystallization probability
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53 200 density function $f_{p_{Zr}}(n)$ is defined for each simulation, with non-uniform distribution:

$$54 \quad 55 \quad 56 \quad 57 \quad 201 \quad p_{Zr_n} \geq 0 \quad \text{and} \quad \sum_{n=0}^N p_{Zr_n} = 1. (3)$$

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4 202 In order to simulate the stochastic process of zircon selection during geochronological analysis, a
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6 203 variable number of zircons (N_{Zr} , user defined) is randomly sampled from the cumulative
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9 204 distribution function $F_{p_{Zr}}(n)$ of the target probability density function $f_{p_{Zr}}(n)$ through a Monte
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11
12 205 Carlo based model (Fig. 2c). The Monte Carlo algorithm undergoes 10^3 iterations for each
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14 206 simulation to ensure robustness of the random selection process. Bootstrapped zircon *age*
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16 207 spectra, where *age* is expressed as relative zircon crystallization density functions $f_{x_{tal}}(t_r)$ in the
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19 208 time interval (t_r) from crystallization of the first zircon (t_{sat}), are generated from outputs of the
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22 209 Monte Carlo simulation and visualized through a truncated kernel density estimate of the scaled
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24 210 crystallization *ages* (Fig. 2d).

25
26 211 Taking advantage of this flexible setup, we simulated contrasting thermal and physical
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29 212 trajectories for a single rhyolitic magma batch stored at upper crustal depth, namely: (1)
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31 213 equilibrium crystallization, (2) fractional crystallization, or (3) intrusion during continuous
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34 214 differentiation (via fractional crystallization) by one to five recharges of variable volume and
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36 215 composition (i.e. rhyolitic or basaltic). Simulations are run until the magma body reaches solidus
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39 216 temperature or alternatively interrupted at 750°C (c. 50 vol.% melt), to simulate the abrupt
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41 217 termination of crystallization caused by an eruptive event. The whole-rock composition of a
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43 218 monzogranite from the VMP is used as the initial liquid composition, initial magmatic water
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45
46 219 content is set to 2.5 wt% H₂O and fO_2 to the QFM (quartz-fayalite-magnetite) buffer (Tavazzani
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48 220 et al., 2020). The complete list of input parameters, explored crystallization simulations, model
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51 221 limitations and applicability range are discussed in Tavazzani et al. (accepted). The full set of
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53 222 open-source MATLAB[®] scripts necessary to run zircon saturation simulations is available at
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56 223 <https://github.com/TavazzaniL/OpenSystemZrSat>.

57 58 224 4. Results

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4.1 CA-ID-TIMS geochronology

Out of fifty-six analyses, thirty-nine Th-disequilibrium corrected $^{206}\text{Pb}/^{238}\text{U}$ dates are interpreted as magmatic crystallization ages whereas the remaining dates are affected either by the presence of xenocrystic components or post-crystallization Pb-loss (discussion of rejected fractions is available in the Supplementary Text). This dataset is augmented with published zircon data of a Sesia Ignimbrite sample from Karakas et al. (2019). All samples display resolvable age dispersion (Fig. 3a), aside from a small microgranitic porphyry body (Gp) in the VMP in which textural and petrologic evidence suggest rapid cooling from near-liquidus conditions (Tavazzani et al., 2020). The range of individual zircon dates overlaps in an interval between 283.984 ± 0.057 and 282.97 ± 0.17 Ma (2σ uncertainty, Fig. 3a) with the exception of an older (up to 284.8 Ma) component represented by several analyses from the crystal-rich rhyolite (Sei) and in two zircons from the VMP homogeneous monzogranite samples (Hwg). The zircon fraction younger than 284 Ma shows dispersion in single crystal $^{206}\text{Pb}/^{238}\text{U}$ dates of 1.012 ± 0.178 Myr for the VMP, comparable to observations by Karakas et al. (2019) on other granitoid bodies of the SMS (Fig. 3a). In the volcanic samples, excluding the older component (>284 Ma), U-Pb dates record 0.309 ± 0.080 Myr of zircon crystallization in the crystal-poor rhyolite unit and 0.477 ± 0.188 Myr of crystallization in the crystal-rich Sesia Ignimbrite sample of Karakas et al. (2019), with crystallization intervals for each sample calculated with the iterative approach of Eddy et al. (2016; see Supplementary Text for details). The Sesia and Upper Ignimbrite eruption ages can be obtained as either the Th-corrected $^{206}\text{Pb}/^{238}\text{U}$ ages of the single youngest zircon of each eruptive unit (Sesia Ignimbrite: $283.46 \pm 0.14/0.16/0.35$ Ma; Upper Ignimbrite: $283.22 \pm 0.07/0.11/0.33$ Ma) or as the Bayesian estimate based on the bootstrapped zircon distribution (Keller et al., 2018) that yields $283.49 \pm 0.10/0.13/0.34$ Ma

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4 248 (Sei) and $283.21 \pm 0.10/0.13/0.34$ Ma (Ui, 2σ uncertainty given as internal only/with tracer
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6 249 calibration/with tracer and ^{238}U decay constant; Fig. 3a). The estimates obtained by these two
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9 250 approaches are indistinguishable and resolve a ~ 300 kyr temporal gap between the two eruptive
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11 251 events, consistent with field observations (erosional surface and sedimentation interval; Sbisà,
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14 252 2009).

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16 253 The two kernel density estimates obtained by bootstrapping zircon CA-ID-TIMS age
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19 254 distributions of all samples from volcanic (Fig. 3b) and plutonic (Fig. 3c) units between zircon
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21 255 saturation (t_{sat}) and full plutonic solidification (t_{sol}) or eruption (t_{er}) are similar and show an increase
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23
24 256 in relative number of crystallized zircons with time. The distributions are asymmetrical, with
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26 257 zircon crystallization peaks shortly preceding eruption in the volcanic domain and full plutonic
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28
29 258 crystallization in the intrusive units. The peaks are more evident when including the age fraction
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31 259 >284 Ma (defined as antecrysts in Fig. 3b, c) into the kernel density estimate.

32 33 34 260 **4.2 Zircon trace element composition**

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37 261 The dated zircons display a geochemical variability that coincides with the compositional
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39 262 range observed by Karakas et al. (2019) for zircons from upper crustal rocks of the SMS (Fig. 4).
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41
42 263 Overlapping zircon compositions from volcanic and plutonic units align along a trend consistent
43
44 264 with feldspar fractionation (decreasing Eu anomaly, Eu/Eu^* and increasing U concentration; Fig.
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47 265 4a, c) and are strongly correlated with the temperature-modulated, Ti content (Fig. 4b; Ferry and
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49 266 Watson, 2007). The investigated zircons also display a clear negative correlation between Ti,
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52 267 REE contents and heavy to middle REE ratios (i.e., Yb/Dy ; Fig. 4d, e). This relationship
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54 268 suggests that heavy to middle REE budgets in SMS melts are controlled by zircon and apatite
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57 269 crystallization without a significant role played by titanite (cf. Wotzlaw et al., 2013; Samperton
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59 270 et al., 2015; Szymanowski et al., 2017; Fig. 4d), consistent with the observed mineral assemblage
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4 271 (Tavazzani et al., 2020). Few zircons display resolvable intra-grain compositional variability (i.e.
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6 272 core to rim; Supplementary Figure S17).

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10 273 All zircons dated by bulk-grain CA-ID-TIMS reveal a systematic evolution of trace
11
12 274 element compositions over the ~1.5 Myr long range of U-Pb zircon crystallization ages (Fig. 5;
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14 275 for a complete discussion of the biases associated with matching in-situ compositions with bulk-
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16 276 grain U-Pb ages see Supplementary Text). The zircon fraction with U-Pb ages >284 Ma shows
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18 277 the least evolved compositions (e.g., Hf<10⁴ ppm, Eu/Eu* of 0.2-0.4, Ti between 5 and 15 ppm;
19
20 278 Fig. 5a, d, e) observed in the VMP and SC zircon record. Starting at ~284 Ma, zircons display an
21
22 279 increase in U, Hf and MREE concentrations paired with decreasing Ti and Eu/Eu*. During the
23
24 279 remaining lifetime of the reservoir (~1 Myr), zircons are characterized by scatter around low Ti
25
26 280 (<5 ppm; Fig. 5e), evolved compositions (e.g., Hf>10⁴ ppm, Dy ~200 ppm and Eu/Eu* of ~0.1;
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28 281 Fig. 5a, c, d).

282 283 **4.3 Zircon saturation modeling**

284 Three end-member scenarios explore systematic variations in saturated zircon mass
285 during the crystallization of a rhyolitic magma batch, as modulated by the dominant
286 crystallization processes (i.e. equilibrium or fractional crystallization), magma recharge
287 frequency, volume and composition. Simulation results are compared to SMS volcanic and
288 plutonic zircon age distributions (Fig. 6).

289 The first scenario, shown in Fig. 6a and 6d, simulates the temporal distribution of zircons
290 crystallized in a single magma batch during a fractional crystallization process. In this scenario,
291 after saturation is reached at ~775°C, zircons precipitate from progressively cooler residual melt
292 until the residual melt fraction is erupted at ~750°C (Fig. 6a) or until the solidus temperature is
293 reached (Fig. 6d). In saturation calculations truncated at eruption temperatures, early uniform,

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4 294 moderate crystallization rates persist after saturation and decrease sharply just before the eruptive
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7 295 event (Fig. 6a). At plutonic conditions, outputs display high crystallization rates close to zircon
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9 296 saturation temperature, followed by a steady decline approaching the solidus (Fig. 6d).
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12 297 Generally, in volcanic simulations, zircon distributions lack the pronounced asymmetry observed
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14 298 in plutonic simulations.

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17 299 The changes in temporal distribution of zircon mass induced by new magma injection in
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19 300 a crystallizing magma body are explored in two distinct simulations. In the first recharge
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21
22 301 scenario, three basaltic andesite magma batches are injected in a rhyolitic magma body at near-
23
24 302 solidus, high-crystallinity conditions (i.e. 730°C and *c.* 75 vol.% crystals) during monotonic
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26
27 303 cooling and differentiation. Each recharge has the same size (i.e. 1/10 of the initial magma mass)
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29 304 and it is added to the crystallizing rhyolite body when residual melt fraction drops below 25
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31
32 305 vol.% (Fig. 6b and 6e). In this scenario, the negative inflection in rate of zircon crystallization is
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34 306 an expression of the transient zircon undersaturation (*sensu* Bindeman and Melnik, 2016)
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36
37 307 triggered by stark compositional contrast between host (i.e. rhyolitic) and recharging (i.e. basaltic
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39 308 andesite) magmas. Increase in zircon growth rates is observed only after the last mafic recharge
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42 309 enters the system (Fig. 6b and 6e). As a result, zircon distributions resulting from these
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44 310 simulations are asymmetrical, with the majority of zircons precipitated from residual melt shortly
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46 311 before solidus is reached or the residual melt fraction is erupted.

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49 312 The last model setup (Fig. 6c and 6f) simulates zircon crystallization in a rhyolitic
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52 313 magma body, replenished at near-solidus, high-crystallinity conditions (i.e. 730°C and *c.* 75
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54 314 vol.% crystals) by magma batches compositionally identical to the starting material. In this
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57 315 scenario, three large (i.e. 1/2 of the initial magma mass), equally sized rhyolitic recharges are
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59 316 introduced in a fractionating magma batch to simulate the incremental growth of a granitic,

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4 317 upper-crustal body. The impact that a sequence of large, rhyolitic recharges has on the temporal
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6 318 distribution of precipitated zircon mass is overall comparable to the effect caused by small mafic
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9 319 injections. However, due to the limited thermal and compositional contrast between newly
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11 320 injected and crystallizing magma, zircon-saturated conditions are maintained throughout the
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14 321 whole simulation. Zircon crystallization rates increase steadily until they reach the highest values
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16 322 near eruption (Fig. 6c) or full plutonic crystallization (Fig. 6f).
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19 323 **5. Discussion**

22 324 **5.1 Time-resolved record of inception, maturation and eruption(s) of an upper-crustal** 23 24 25 325 **magma reservoir**

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27 326 The overlapping crystallization ages (Fig. 3a), matching temporal distributions (Fig. 3b,
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30 327 c) and coherent geochemical trends (Fig. 4 and Fig. 5) of zircons sampled across volcanic and
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33 328 intrusive lithologies of the SMS suggest zircon saturation and precipitation from melts stored in a
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35 329 common upper crustal reservoir over ~1 Myr. In this context, we rely on precise U-Pb ages (2σ
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38 330 uncertainties on individual dates of 40-190 kyr; Fig. 3) and paired in-situ geochemical analyses
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40 331 to track temporal variations in melt temperature, composition and degree of crystallization over
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43 332 the lifespan of the reservoir (e.g. Schoene et al., 2012; Samperton et al., 2015; Melnik and
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45 333 Bindeman, 2018; Large et al., 2021).
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48 334 Early crystallized zircons (i.e. >284.0 Ma; Fig. 3a) display low incompatible element
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50 335 concentrations (i.e. Hf, Dy), high Ti and Eu/Eu*. The composition of these zircons is comparable
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52
53 336 to that of zircons crystallized in lower to middle crustal dioritic and gabbroic magmas of the
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55 337 SMS (Karakas et al., 2019; Fig. 5). We interpret these zircons as antecrysts, crystallized during
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57
58 338 the initial stage of magmatism in the SMS at middle crustal depths and from melts of mafic to
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60 339 intermediate compositions. Ascent, progressive cooling and feldspar dominated fractionation of
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4 340 these parental melts, together with variable incorporation of crustal melting-derived melts, is
5
6 341 likely the source of upper crustal granitoid- and rhyolite-hosted zircons of the SMS (Fig. 4; see
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8
9 342 also Sinigoi et al., 2016; Storck et al., 2021). During a prolonged interval (*ca.* 283.9 to 283.2 Ma)
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11 343 of continuous or punctuated zircon crystallization, pronounced zircon Eu anomaly and high
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14 344 HREE (e.g. Dy) contents indicate the existence of a highly fractionated magma body, likely
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16 345 stored at high crystallinity conditions (estimated >60 vol.% crystals based on simple
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19 346 geochemical modeling; Fig. 4c, d) and invariant, nearly eutectic temperatures (Ti: 5 ± 1 ppm
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21 347 corresponding to 728 ± 20 °C at $a_{\text{SiO}_2} = 1.0$ and $a_{\text{TiO}_2} = 0.6$; Ferry and Watson, 2007).
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23
24 348 Acknowledging the limits of fractional crystallization modeling and the possibility of higher melt
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26 349 fractions at lower temperatures (Jackson et al., 2018), a similar record might be produced by a
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29 350 less crystallized reservoir whereby the compositional homogeneity is caused by frequent
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31 351 overturns or stirring of the magma body (e.g. Trubač et al., 2017). Regardless of the details of
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34 352 physical state of the upper crustal reservoir, the homogeneous, differentiated melts that
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36 353 characterize the system after 284 Ma (Fig. 5) indicate the establishment of a large, unified
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39 354 magma reservoir or an interconnected network of reservoirs in the upper crust underneath the
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41 355 SC, similar to that inferred for plumbing systems at a number of young calderas (e.g.
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43 356 Yellowstone, Huang et al., 2015; Wotzlaw et al., 2015; Toba, Jaxybulatov et al., 2014). In the
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46 357 case of a high crystallinity reservoir, deviations from the trend, in the form of individual zircon
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48 358 analyses or groups of grains showing increased Eu/Eu* (Fig. 5d), temperature-dependent Ti (Fig.
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51 359 5e) and reduced HREE contents (e.g. Dy; Fig. 5c) can be interpreted as signals of new magma
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53 360 influxes in the reservoir, adding heat and mass to the system with direct effect on melt
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56 361 temperature and mineral phase stability (e.g. Szymanowski et al., 2019). Alternatively, in a
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58 362 scenario where higher melt fraction can be sustained at lower temperatures and the reservoir is
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4 363 well stirred, the variability in composition could reflect the sampling of peripheral part of the
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6 364 system during overturning or rejuvenation events.
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10 365 Contrasting compositions and age distributions of zircon grains extracted from the two
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12 366 SC ignimbrite units analyzed in Karakas et al. (2019) and in this work (i.e. Sesia Ignimbrite and
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14 367 Upper Ignimbrite) suggest different processes involved in melt mobilization and eruption. The
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16
17 368 long crystallization interval (~500 kyr, excluding antecrystic components; Fig. 3a) and broad
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19 369 geochemical variability of zircons from the crystal-rich Sesia Ignimbrite (Fig. 4 and Fig. 5),
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21
22 370 which includes compositions corresponding to both highly differentiated and relatively
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24 371 unevolved melts (Fig. 5d, e), can be explained by thermal rejuvenation of an internally
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26 372 heterogeneous magmatic reservoir (Huber et al., 2011) in the build-up to the SC climactic
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29 373 eruption (>400 km³; Fig. 1b). This process was potentially accompanied by mechanical mixing
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32 374 of distinct zircon populations entrained by peripheral, variably crystallized portions of the system
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34 375 (e.g. Szymanowski et al., 2019; Curry et al., 2021). The narrow crystallization interval and the
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36 376 geochemical homogeneity of zircons from the volumetrically limited, high-silica rhyolite unit
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39 377 (Upper Ignimbrite: 1 to 10 km³; Fig. 1b) suggest a different eruption priming mechanism. In this
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41 378 unit, zircon crystallization ages span <300 kyr and display compositions compatible with
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44 379 crystallization from a highly differentiated melt (Fig. 4 and Fig. 5). Evolved melt domains,
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46 380 isolated in a crystalline framework, are likely buffered at the haplogranitic minimum (Schaen et
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49 381 al., 2018), thus explaining the limited compositional variability of zircons from this unit (at least
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51 382 in compatible elements; Fig. 5d, e). Moreover, the extraction and ascent of these highly
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53 383 differentiated, residual melts through a viscous crystal mush (Bachmann and Huber, 2019),
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56 384 rather than destabilization and homogenization of the entire mush column, provide a
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4 385 mobilization mechanism that accounts for the limited temporal variability of zircons sampled in
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6 386 the Upper Ignimbrite unit.

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10 387 This set of observations combines with existing textural (e.g. cryptic contacts) and bulk-
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12 388 rock geochemical data (Tavazzani et al., 2020) to convey the image of a large, mostly crystalline
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14 389 and periodically recharged upper crustal reservoir, containing isolated and/or interconnected melt
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17 390 domains capable of zircon precipitation, which persisted in the upper-crust for ~1 Myr and
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19 391 produced at least one catastrophic caldera forming eruption. This picture supports models
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22 392 emerging from a number of subvolcanic systems tapped by voluminous rhyolitic eruptions (e.g.
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24 393 Wotzlaw et al., 2013; Rioux et al., 2016; Szymanowski et al., 2017) or porphyry intrusions (e.g.
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26 394 Chelle-Michou et al. 2014; Buret et al., 2016) and agrees with timescales derived from thermal
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29 395 models (e.g. Gelman et al., 2013; Karakas et al., 2017), which imply storage at dominantly high-
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31 396 crystallinity conditions of episodically replenished silicic magma reservoirs for 10^5 to 10^6 years
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34 397 in the upper crust.

35 36 37 398 **5.2 Petrologic significance of CA-ID-TIMS zircon U-Pb age dispersion**

38 39 40 399 **5.2.1 Zircon age dispersion in the SMS**

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43 400 The theoretical trajectory of zircon growth during monotonic cooling and crystallization
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45 401 of an isolated magma batch has been codified in a seminal work by Watson (1996) and recently
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48 402 reproduced integrating thermodynamically derived phase proportions, residual melt
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50 403 compositional evolution and empirical zircon saturation equations (Keller et al., 2017; Kirkland
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52 404 et al., 2021; Klein and Eddy, 2023; Tavazzani et al., **accepted**). Results of these studies indicate
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55 405 that in a monotonically cooled magma batch both equilibrium and fractional crystallization
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58 406 pathways produce non-linear zircon growth. Specifically, a rapid increase in precipitated zircon
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60 407 mass upon saturation in Zr is followed by a steady decline in zircon crystallization rates until
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4 408 solidus is reached (Fig. 2d; Fig. 6d), with zircon crystallizing from progressively cooler residual
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6 409 melt. These predictions are backed by evidence from silicic intrusions characterized by
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9 410 comprehensive high-precision U-Pb zircon datasets, which display decreasing mass of
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11 411 crystallized zircon with time and linear decrease in temperature (e.g. Samperton et al., 2017;
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14 412 Large et al., 2021)

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17 413 In intrusive and volcanic units of the SMS, the relative distribution of zircon
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19 414 crystallization ages (i.e. increase in relative number of crystallized zircons with time; Fig. 3b and
20
21 415 3c) is incompatible with zircon growth from a monotonically cooled, isolated magma batch (Fig.
22
23 416 6). Additionally, the SMS zircon geochemical record shows non-linear variations in temperature-
24
25 417 modulated Ti contents (Fig. 5e), not consistent with crystallization in a monotonically cooled
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27 418 magma system. A good match between the shape of zircon age spectra from VMP and SC units
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29 419 and modeled zircon growth is obtained by simulating repeated recharges during fractionation
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32 420 (RFC; Fig. 6c, f), either by small basaltic andesite melt batches (Fig. 6b, e) or large rhyolite
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34 421 injections (Fig. 6 c, f). The equilibration of newly injected magma with the resident crystallizing
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36 422 magma causes fluctuations in zircon saturation parameters (e.g. temperature, Zr concentration,
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38 423 degree of melt polymerization) and is reflected in non-linear zircon crystallization probabilities
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41 424 (p_{Zr_n} ; Fig. 2b). Moreover, synthetic zircon populations from RFC simulations display a broad
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44 425 spread and systematic changes in crystallization temperatures that match Ti-in-zircon
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47 426 temperature evolution observed across VMP and SC zircons (see Tavazzani et al., [accepted](#)),
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49 427 when applying activity parameters appropriate for the SMS (i.e. $a_{TiO_2}=0.6$; $a_{SiO_2}=1.0$ from
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51 428 Tavazzani et al., 2020; Fig. 5e). Overall, the two RFC simulations produce comparable results,
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54 429 with the rhyolitic recharge scenario (Fig. 6c and 6f) favored by quantitative comparison of
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4 430 synthetic and empirical zircon age spectra via Kolmogorov-Smirnov statistical test (Press et al.,
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6 431 1988; see Tavazzani et al., [accepted](#), for a statistical match example).
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9 432 **5.2.2 Implications for the interpretation of zircon age dispersion in large silicic systems**

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12 433 Heterogeneous age spectra with late peaks in zircon crystallization are a distinctive
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15 434 feature of rhyolitic ignimbrites generated in caldera-forming eruptions (Fig. 7a; e.g. Huckleberry
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17 435 Ridge Tuff, Singer et al., 2014; Wotzlav et al., 2015; Kneeling Nun Tuff, Szymanowski et al.,
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19
20 436 2017, 2019). The same pattern of age dispersion is observed in upper-crustal granitoid intrusions
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22 437 that are contemporary and genetically related to centers of rhyolitic volcanism (inset in Fig. 7a;
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24
25 438 e.g. Organ Mountains, Rioux et al., 2016; SMS, Karakas et al., 2019, this work). Here, based on
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27 439 results from zircon saturation modeling, we present two non-mutually exclusive pathways of late
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29
30 440 zircon crystallization peak generation in plutonic and volcanic products of silicic magmatic
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32 441 systems.
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35 442 The first pathway is through zircon growth in an interconnected, continuously recharged
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37 443 reservoir, wherein a significant fraction of melt (>25 vol.%) remains in the system between
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40 444 successive magmatic pulses (Fig. 7b). In this scenario, each new magma injection contributes
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42 445 heat to the system and dilutes Zr concentration in the residual melt (i.e. assuming new injections
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45 446 are undersaturated in zircon) causing delay in zircon mass precipitation. A sharp increase in
46
47 447 zircon growth rates is attained only at the end of the recharge cycle. This pathway agrees with
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49
50 448 the prediction that sizeable eruptions (>0.1 km³) are preceded by incremental assembly and
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52 449 chemical evolution of shallow magma reservoirs (Forni et al., 2018) in systems characterized by
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54 450 intermediate magmatic fluxes (e.g. recharge rates of ~0.001 km³ yr⁻¹), which allow survival of
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57 451 melt between emplacement of successive batches (Gelman et al., 2013; Tierney et al., 2016;
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59 452 Townsend and Huber, 2020). A second pathway, whose applicability is limited to the
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4 453 interpretation of zircon age spectra in rhyolitic ignimbrites, is related to mechanical mixing of
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6 454 zircon populations crystallized in sequentially emplaced and monotonically cooled magma
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9 455 bodies shortly before (i.e. homogenization; Huber et al., 2011, 2012) or during a large eruptive
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11 456 event (simultaneous or sequential tapping; e.g. Pearce et al., 2020). If the majority of magma
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14 457 bodies are emplaced close to eruption, pre- or syn-eruptive mixing can generate volcanic age
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16 458 spectra with an apparent late peak in zircon crystallization (Fig. 7c; Tavazzani et al., **accepted**).
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19 459 The reported increase in magma flux approaching the climactic eruption stage of caldera-forming
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21 460 magmatic centers (Bouvet de Maisonneuve et al., 2021) make this pathway a viable alternative
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24 461 for the generation of late zircon crystallization peaks in rhyolitic ignimbrites generated in
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26 462 caldera-forming eruptions.
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29 463 In summary, late zircon crystallization peaks may reflect either thermochemically
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31 464 modulated zircon saturation during open-system crystallization of an interconnected magma
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34 465 reservoir, or the pre/syneruptive mechanical mixing of zircon populations crystallized in
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36 466 monotonically cooled, isolated magma batches. Overall, the increase in crystallized zircon mass
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39 467 with time, when observed in the plutonic or volcanic record of silicic systems (Fig. 7a), reflects
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41 468 long-term magma storage conditions, magma fluxes and short-term perturbations, which would
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44 469 allow the discharge of large volumes of mobile melt in single, catastrophic eruptions.
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47 470 By contrast, age spectra of silicic intrusive suites not known to be directly associated with
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49 471 large ignimbritic eruptions display a relative decrease in zircon crystallization with time (Fig.
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51 472 8a). This characteristic shape of plutonic zircon age spectra is matched by synthetic zircon
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54 473 distributions generated in equilibrium and fractional crystallization simulations of monotonically
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56 474 cooled, isolated magma batches (Fig. 8b, c; Samperton et al., 2017; Kirkland et al., 2021). As
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59 475 true closed-system crystallization is unlikely in natural magmatic systems (Cashman et al.,
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4 476 2017), an alternative process capable of producing these zircon age distributions is incremental
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6 477 growth characterized by high instantaneous flux (recharge rates $>0.01 \text{ km}^3 \text{ yr}^{-1}$; e.g. Eddy et al.,
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8
9 478 2016) in a long-term averaged low flux environment (i.e. $<0.001 \text{ km}^3 \text{ yr}^{-1}$; Caricchi et al., 2016)
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11 479 where no melt remains in the system between successive batches. The absence of melt hinders
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14 480 substantial thermal and compositional interaction between pulses (Barboni et al., 2015) and leads
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16 481 to the monotonic cooling of each separate magma batch, approximating a closed-system
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19 482 crystallization scenario.
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22 483 It follows that those plutonic systems that are characterized by zircon age spectra
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24 484 showing relative decrease in saturated zircon mass with time (Fig. 8a) are unlikely to have
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26 485 experienced a developmental stage corresponding to a thermochemically interconnected magma
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29 486 reservoir (i.e. mature reservoir, Bouvet de Maisonneuve et al., 2021). The absence of late zircon
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31 487 crystallization peaks also eliminates the possibility of a homogenization event, related to
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34 488 increasing magma flux into the system (Fig. 7c). Overall, the absence of reservoir maturation or
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36 489 late-stage homogenization signals in the plutonic record of many silicic systems implies that
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39 490 long-term and/or transient conditions favorable for the evacuation of large volumes of magma in
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41 491 a single, catastrophic event were never established. If present, volcanic activity associated with
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44 492 these plutons would have accounted for volumetrically limited, distributed eruptions fed by
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46 493 individual magma batches intruded and differentiated in the upper crust.
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49 494 **5.3 Implications for eruption age estimation of volcanic units**

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52 495 High-precision geochronology of eruptive products in sedimentary successions is a
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54 496 fundamental tool to calibrate the Earth's geologic history. However, the use of zircon U-Pb dates
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57 497 to define the eruption (or emplacement) age is invariably biased by pre-eruption zircon
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59 498 crystallization, Pb loss and methodological biases (e.g. undersampling of the zircon population,
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4 499 over-representation of late-stage crystallization events; Klein and Eddy, 2023; this work).
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7 500 Therefore, increasingly sophisticated models that rely on statistical likelihood of a group of
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9 501 zircons representing an eruptive event have been proposed and tested on a number of geological
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11 502 objects (Schoene et al., 2019; Keller et al., 2018). These models rely on assumptions about the
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13
14 503 distribution of zircon mass during crystallization of the source magma body (Keller et al., 2018).
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16 504 A key observation from this study, although it does not specifically treat the kinetics of zircon
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19 505 crystallization from a multi-phase magma (e.g. not-constant growth rate, armoring effects;
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21 506 Bindeman and Melnik, 2016; Klein and Eddy, 2023), is that the mass of zircon crystallized in
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24 507 upper crustal magmatic bodies feeding large eruptions is not normally distributed, with relatively
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26 508 more zircon mass generated towards the end of the lifetime of the system (i.e. eruption or
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29 509 complete crystallization). In light of this observation, we suggest that models such as that of
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31 510 Keller et al. (2018), which allows the selection of a-priori distribution of zircon mass in the melt,
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34 511 should be used with a maximally informed prior distribution reflecting this observation. For
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36 512 large-n samples (e.g. LA-ICP-MS datasets) a prior distribution generated by bootstrapping the
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39 513 data may be the most appropriate; however, in cases where few zircon dates are available to
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41 514 estimate eruption/deposition age (e.g. CA-ID-TIMS datasets), significant biases may be
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43 515 introduced via bootstrapping, or by choosing an inappropriate prior. Our forward modelling (Fig.
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46 516 6) and data compilation (Fig. 7, 8) suggest that in absence of other information, the best choice to
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48 517 estimate eruption age of large volcanic explosive events might be approximating the time
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51 518 evolution of mass of zircon crystallized with a truncated normal distribution (Keller et al., 2018),
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53 519 as this distribution reproduces the late peak of zircon crystallization characteristic of long-lived,
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55 520 interconnected magmatic reservoirs (Fig. 7a).

521 **5.4 Quantification of undersampling effect on zircon age distribution**

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4 522 The success of quantitative analyses of zircon age spectra relies on the acquisition of a
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6 523 large enough number of zircons per sample, such that the entire history of zircon saturation is
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9 524 represented (Klein and Eddy, 2023). In this context, the stochastic aspect of our modeling
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11 525 template can quantify the effect that the limited number of zircons per sample routinely analyzed
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13
14 526 with the CA-ID-TIMS technique have on the representativeness of zircon age distributions in
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16 527 natural systems (Samperton et al., 2015). A compilation of zircon age spectra generated by CA-
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19 528 ID-TIMS displays the influence that undersampling has on apparent relative distribution of
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21 529 zircon in time (Fig. 7 and Fig. 8). Assuming equivalent crystallization histories, well
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24 530 characterized systems (i.e. >20 zircons per sample) display accentuated asymmetries in zircon
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26 531 crystallization distribution, whereas systems with a low number of zircons analyzed (i.e. <10
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29 532 zircons per sample) show smoother age spectra. Our workflow can reproduce the “smoothing”
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31 533 effect by controlling the number of zircons randomly sampled from a synthetic probability
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34 534 distribution (Fig. 7b, c and Fig. 8b, c). Such observation confirms the stochastic nature of
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36 535 undersampling in high-precision U-Pb geochronology and roughly indicates a minimum number
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39 536 of 10 zircon dates per igneous sample in order to assign petrological significance to any age
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41 537 dispersion.

42 43 538 **6. Conclusion**

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45 539 Comparison of high-precision (CA-ID-TIMS) zircon age spectra from volcanic and
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48 540 plutonic products of large silicic systems with results of a thermodynamic-based zircon
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51 541 saturation model highlights the link between magmatic fluxes, zircon crystallization histories and
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53 542 eruptive potential of shallow silicic magmatic reservoirs:

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55 543 (1) The dominant crystallization process (i.e. equilibrium or fractional crystallization)
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58 544 controls the shape of zircon age spectra during monotonic cooling and crystallization of isolated
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4 545 magma batches. In systems characterized by extensive, supra-solidus interaction between
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6 546 successive magma recharges the frequency, volume and composition of new magma injections
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9 547 affect zircon growth curves.

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11 548 (2) It follows that in evolved, subalkaline systems, an increase in relative number of
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14 549 crystallized zircons with time denotes ‘active’, thermochemically connected magma reservoirs,
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16 550 where increase in heat supply can produce system-wide crystallinity variations conducive to
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19 551 evacuation of large volumes of magma in a single, voluminous eruption. Conversely, a decrease
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21 552 in crystallized zircon mass with time identifies ‘passive’ systems, which are incrementally
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24 553 assembled by rapid emplacement of individual magma batches, but never experience system-
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26 554 wide crystallinity variations allowing for the segregation and release of large mobile magma
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29 555 volumes.

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31 556 (3) Finally, saturation modeling results inform on the stochastic undersampling effect on
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34 557 zircon age distribution that is associated with the limited number of U-Pb zircon dates typical of
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36 558 CA-ID-TIMS studies and the potential biases for determination of eruption age in long-lived
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38 559 rhyolitic caldera systems. A compilation of TIMS results suggest a minimum number of 10
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41 560 individual zircon dates per sample to be acquired in order to assign petrological significance to
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43 561 the shape of any dispersed zircon age spectra.

44 45 46 562 **CRedit authorship contribution statement**

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49 563 **Lorenzo Tavazzani:** Conceptualization, Investigation, Methodology, Software, Visualization,
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51 564 Data Curation, Writing – Original Draft. **Jörn-Frederik Wotzlaw:** Investigation, Validation,
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11 571 **Declaration of competing interest**

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15 572 The authors declare that they have no known competing financial interests or personal
16
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54 863 **Figure captions**

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56 864 **Figure 1.** Location of the study area and sampling sites. (a) Schematic geologic map of the Sesia
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59 865 Magmatic System (SMS) and its location in the Ivrea-Verbano and Serie dei Laghi units of
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4 866 southern Alpine basement (northern Italy), redrawn after Handy and Zingg (1991). **(b)** Geologic
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7 867 map of the upper crustal segment of the SMS to the South of the Cremosina Line with zircon
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9 868 sample localities, after Tavazzani et al. (2020).

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12 869 **Figure 2.** Zircon saturation modeling workflow, exemplified for closed-system (equilibrium
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14 870 crystallization – EQ, fractional crystallization – FC) and open-system (recharge fractional
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17 871 crystallization – RFC; Bohrson et al., 2014) crystallization scenarios. **(a)** Normalized zircon
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19 872 saturated mass distribution $m_{Zr}(n^*)$ derived for the three crystallization scenarios. **(b)**
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21 873 Cumulative zircon crystallization probability distribution functions $F_{pZr}(n^*)$ derived from
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24 874 integration of $m_{Zr}(n^*)$ over the zircon saturation interval. **(c)** Three synthetic zircon age datasets
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27 875 (number of zircons, $N_{Zr} = 10$) extracted from $F_{pZr}(n^*)$ curves of EQ, FC and RFC simulations.
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30 876 In these datasets the ratio of true crystallization timescale (ΔT) to analytical uncertainty (σ) is
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32 877 equal to 20 to simulate results of CA-ID-TIMS analyses (e.g. $\Delta T = 1$ Myr and $\sigma = 50$ kyr,
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35 878 corresponding to the typical precision of our analytical data). **(d)** Relative zircon crystallization
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37 879 distributions $f_{xtal}(t_r)$ (i.e. *age spectra*) scaled from initiation to termination of zircon
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40 880 crystallization, shown as kernel density estimates of EQ, FC and RFC simulations at $N_{Zr} = 30$.
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42 881 The black line in **(d)** corresponds to the kinetic zircon crystallization model of Watson (1996),
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45 882 based on zirconium diffusion constraints. (For interpretation of the colors in the figures, the
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47 883 reader is referred to the web version of the article).

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50 884 **Figure 3.** Geochronology of the Valle Mosso Pluton and related silicic volcanic rocks. **(a)** Rank-
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52 885 order plot of CA-ID-TIMS zircon dates from the SMS. Colored bars represent Th-corrected CA-
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55 886 ID-TIMS $^{206}\text{Pb}/^{238}\text{U}$ dates of individual zircon crystals with their 2σ uncertainty. Analyses
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57 887 interpreted as antecrysts are semi-transparent. Δt_h is the difference between the oldest and
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60 888 youngest zircon dates in a sample; Δt_a is the difference between oldest and youngest autocrystic
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4 889 zircon dates. Eruption ages are defined for each volcanic sample as the Bayesian estimate (Keller
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6 890 et al., 2018) using the bootstrapped distribution of Th-corrected $^{206}\text{Pb}/^{238}\text{U}$ dates as prior, with 2σ
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9 891 uncertainty given as internal only/with tracer calibration/ with tracer and ^{238}U decay constant.
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11 892 Individual zircon dates for crystal-rich rhyolite sample MBR and range of zircon dates from 4
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13 893 granitoids of the Serie dei Laghi are reproduced from Karakas et al. (2019). **(b, c)** Temporal
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15 894 distribution of individual zircon ages reported as kernel estimates of relative zircon
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17 895 crystallization density functions $f_{xtal}(t_r)$, where t_r is relative crystallization time, normalized
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19 896 between $x = 1$, denoting zircon saturation (t_{sat}) and $x = 0$, corresponding to full plutonic
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21 897 solidification (t_{sol}) or eruption (t_{er}). The two kernel density estimates represent zircon
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23 898 crystallization distributions bootstrapped over all dated samples in the eruptive **(b)** and intrusive
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25 899 **(c)** domains of the SMS.
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32 900 **Figure 4.** Zircon trace element co-variation diagrams. LA-ICPMS trace element data for zircons
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34 901 also analyzed by CA-ID-TIMS **(a)**, **(d)** overlap with SMS upper-crustal zircon compositions
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36 902 from Karakas et al. (2019). Data are color-coded for Ti-concentration in zircon in **(b)**, **(e)**, zircon
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38 903 crystallization temperatures are calculated based on Ferry and Watson (2007) using an a_{TiO_2} of
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40 904 0.6 and a_{SiO_2} of 1.0 (see Tavazzani et al., 2020 for details on activity coefficients determination).
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42 905 In **(c)**, **(f)** fractional crystallization models are superimposed on trace elements co-variation
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44 906 diagrams. The median composition of zircons with the highest Ti-in-zircon crystallization
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46 907 temperature is selected as initial composition. Percent crystallinity (20%, 50%, etc.) is indicated
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48 908 along the fractionation trajectories. Partition coefficients used and details of each crystallization
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50 909 models are provided in the Supplementary Material. The two models in **(c)** use either constant
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52 910 partition coefficients (Constant D) or partition coefficients that vary with temperature (Variable
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54 911 D) during the course of crystallization. Crystallizing mineral assemblage constitutes of sanidine,
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4 912 plagioclase and quartz in near-eutectic proportions (Luth et al., 1964). In (f), REE evolution
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6 913 scenarios are evaluated as a function of the amount of zircon (zrn), apatite (ap) and titanite (ttn)
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9 914 in the crystallizing mineral assemblage.

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11 **Figure 5.** Temporal variations of trace elements in the upper crustal part of the SMS zircons,
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14 916 with mid- and lower-crustal data from Karakas et al. (2019) shown for comparison. Each point
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17 917 indicates one in-situ LA-ICPMS analyses acquired prior to bulk-crystal CA-ID-TIMS U-Pb
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19 918 dating, plotted against individual zircon crystallization age. Rim analyses are plotted as circles
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22 919 with dark lines extending to composition of the respective cores. Box plots show median,
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24 920 interquartile ranges (IR) and extreme values for bins of 100 kyr (7 youngest bins) or longer (200
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27 921 kyr for dates >283.80 Ma) calculated on rim and core compositions. Vertical lines indicate
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29 922 eruption ages defined for each volcanic sample as the Bayesian estimate of the zircon Th-
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32 923 corrected $^{206}\text{Pb}/^{238}\text{U}$ dates bootstrapped distribution (Keller et al., 2018) with 2σ uncertainty
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34 924 given as internal only/with tracer calibration/with tracer and ^{238}U decay constant. Ti-in-zircon
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36
37 925 crystallization temperatures estimates use the model of Ferry and Watson (2007) and activity
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39 926 values from Tavazzani et al. (2020).

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42 927 **Figure 6.** Comparison of SMS zircon CA-ID-TIMS crystallization age distributions and synthetic
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44 928 zircon distributions obtained through zircon saturation modeling from three different magma
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47 929 crystallization scenarios. (a, d) Result of fractional crystallization (FC) simulations. (b, e)
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49 930 Recharge and fractional crystallization (RFC) models, employing a sequence of three basaltic-
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52 931 andesite recharges. (c, f) Recharge and fractional crystallization (RFC) simulation using a rhyolitic
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54 932 recharge magma in three consecutive injection episodes. Zircon age distributions, expressed as
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57 933 relative zircon crystallization density functions $f_{x\text{tal}}(t_r)$, where t_r is relative time, scaled from zircon
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59 934 saturation (t_{sat}) to full plutonic solidification (t_{sol}) or eruptive truncation (t_{er}), are shown as kernel
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4 935 density estimate truncated at ± 0.5 kernel bandwidth (Keller et al., 2018). Each synthetic zircon
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6 936 distribution (i.e. bold black lines) is the product of random sampling of 30 zircons from synthetic
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9 937 FC and RFC distributions averaged over 10^3 stochastic iterations. Temperature/crystallinity
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11 938 evolution and increments of magma mass over scaled crystallization time are shown in the top
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14 939 panels of each figure. Crystallinity and solidus temperature are based on rhyolite-MELTS (Gualda
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16 940 et al., 2012) derived temperature crystal-fraction curve (i.e. solidus-liquidus relation) for starting
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19 941 granitic composition used in the simulation (PST214; Tavazzani et al., [accepted](#)).

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22 942 **Figure 7.** Conceptualization of the effect of thermal and mechanical processes on the shape of
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24 943 CA-ID-TIMS zircon age spectra in subvolcanic magma reservoirs. **(a)** Empirical kernel density
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26 944 estimates of zircon crystallization distributions from four rhyolitic ignimbrites generated in
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29 945 catastrophic caldera-forming eruptions (N_{Zr} : range of number of zircons analyzed per sample for
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31 946 each eruption). In the inset panel, zircon crystallization distribution form two upper-crustal
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34 947 intrusions contemporary and genetically related to caldera-forming silicic volcanism. **(b)**
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36 948 Synthetic zircon age spectra output from fractional crystallization (RFC) simulations employing
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38
39 949 a sequence of three rhyolitic recharges in a cooling and differentiating magma reservoir. **(c)** Age
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41 950 spectra resulting from pre- or syn-eruptive mixing of zircon populations from sequentially
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44 951 emplaced, distinct magma batches during a caldera-forming event. The kernel density
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46 952 distributions are generated through the randomized sampling of zircon populations from four
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49 953 individual rhyolitic magma batches emplaced, monotonically cooled and differentiated in the
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51 954 upper crust (FC). Zircon crystallization probability (p_{Zr}) evolution is shown in the top panels of
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53 955 each figure, each roman numeral represents a new episode of recharge during the evolution of
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56 956 the system. The number of zircons (N_{Zr}) randomly sampled from RFC and composite
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58 957 FC+homogenization probability density functions $f(p_{Zr})$ is variable between 5 and 30.
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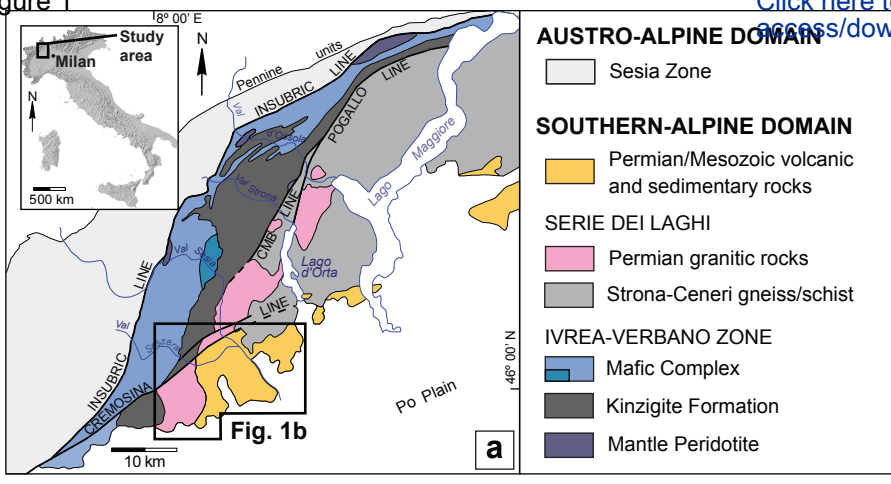
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4 958 Conceptual sketches are adapted from Szymanowski et al. (2017) and Forni et al. (2018).

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6 959 References: 1. Kneeling Nun Tuff (KNT, Szymanowski et al., 2019); 2. Huckleberry Ridge Tuff
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9 960 (HRT, Singer et al., 2014; Wotzlaw et al., 2015); 3. Sesia Caldera (SC, Karakas et al., 2019, this
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12 961 work); 4. Organ Needle caldera (ONC, Rioux et al., 2016); 5. Valle Mosso pluton (VMP, this
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14 962 work); 6. Organ Needle pluton (ONP, Rioux et al., 2016).

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17 963 **Figure 8.** Summary of petrologic implications of the shape of zircon age spectra in silicic

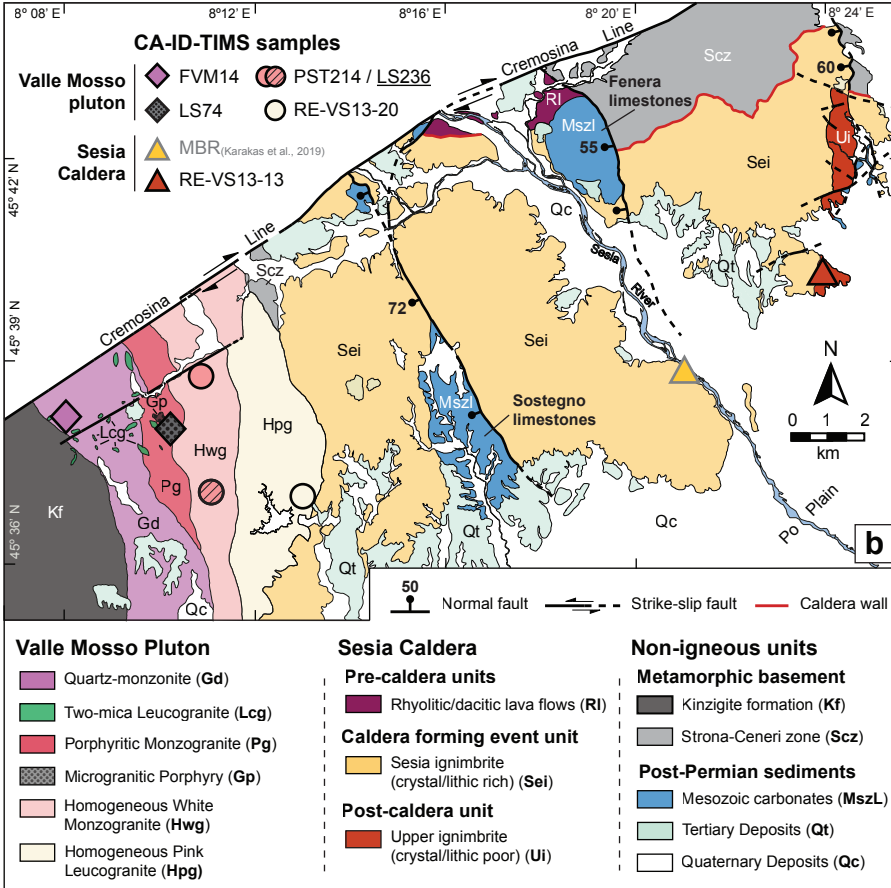
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19 964 intrusive suites. **(a)** Bootstrapped zircon crystallization distributions from four silicic intrusive
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22 965 suites (N_{Zr} : range of number of zircons analyzed per sample for each intrusion). **(b, c)** Synthetic
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24 966 zircon age spectra from fractional crystallization (FC) and equilibrium crystallization (EQ)
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27 967 simulations. Zircon crystallization probability (p_{Zr}) evolution is shown in the top panels of each
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29 968 figure. The number of zircons (N_{Zr}) randomly sampled from synthetic FC and EQ probability
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31
32 969 density functions $f(p_{Zr})$ is variable between 5 and 30. Conceptual sketches are adapted from
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34 970 Szymanowski et al. (2017) and Forni et al. (2018). References: 1. Bergell pluton (Samperton et
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37 971 al., 2015); 2. Mt. Capanne pluton (Barboni et al., 2015); 3. La Gloria pluton (Gutiérrez et al.,
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39 972 2018); 4. Golden Horn Batholith (Eddy et al., 2016).
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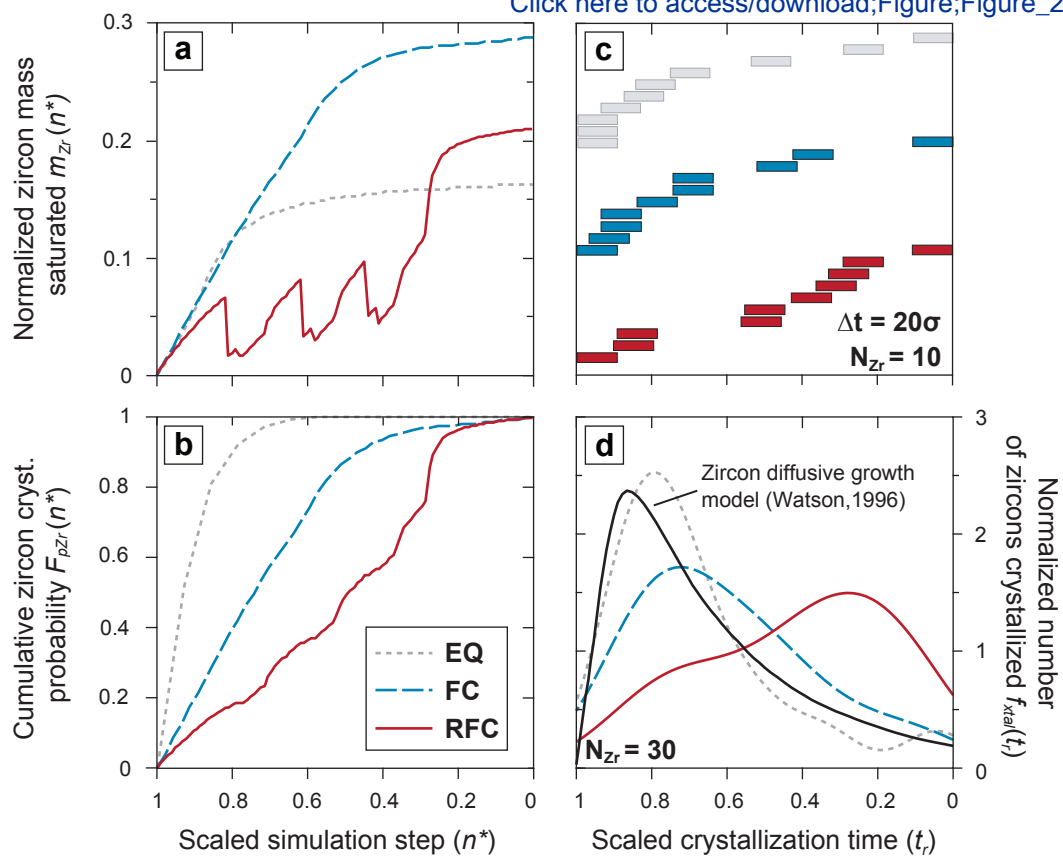
Figure 1



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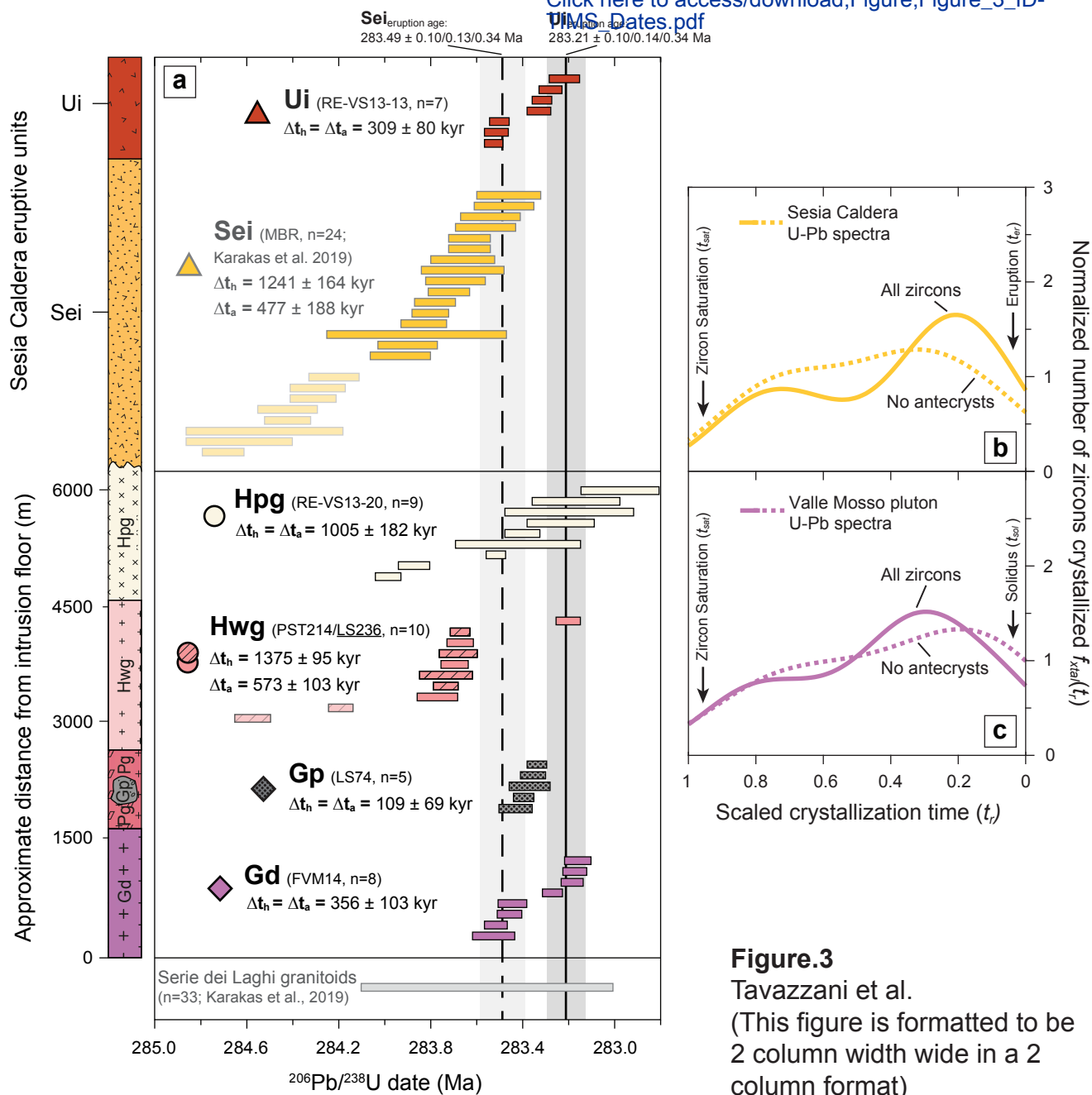
Figure 1
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**Figure. 2**

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**Figure 3**

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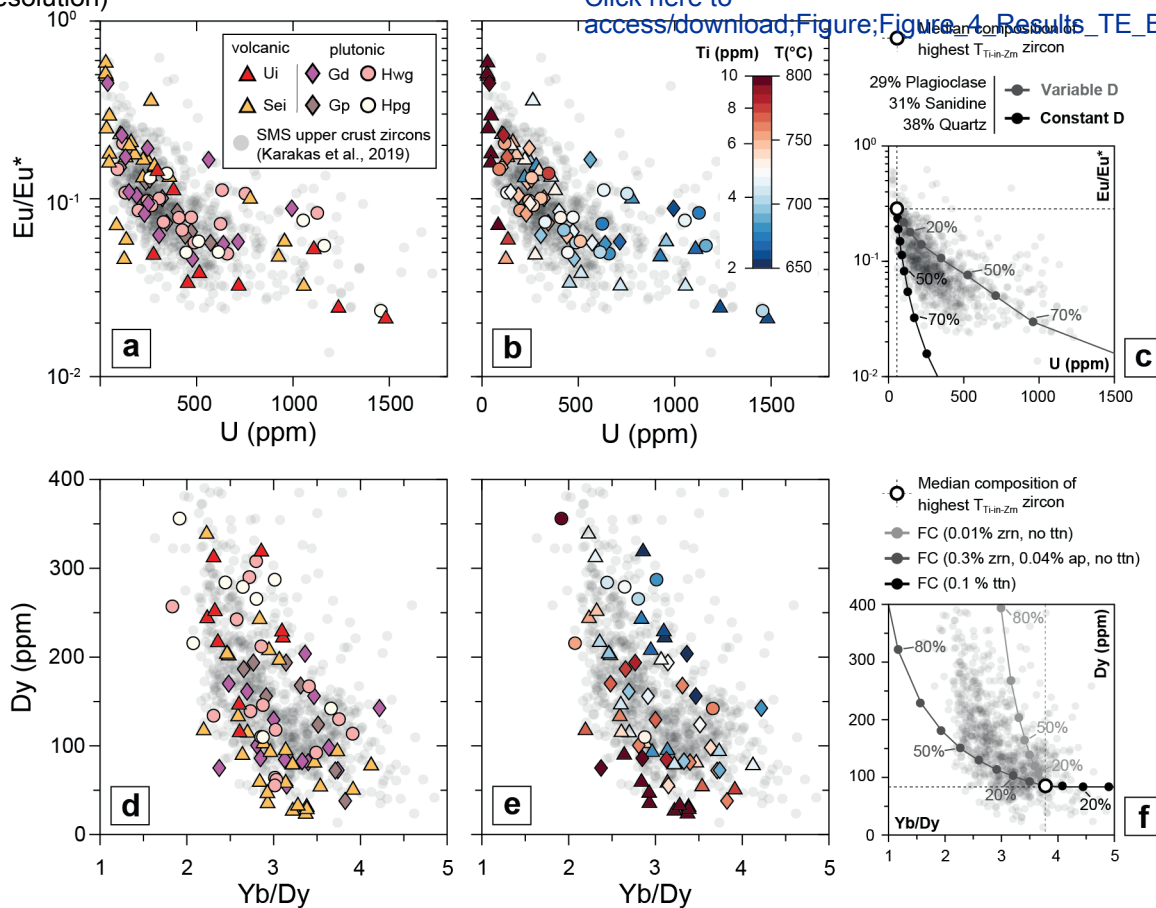
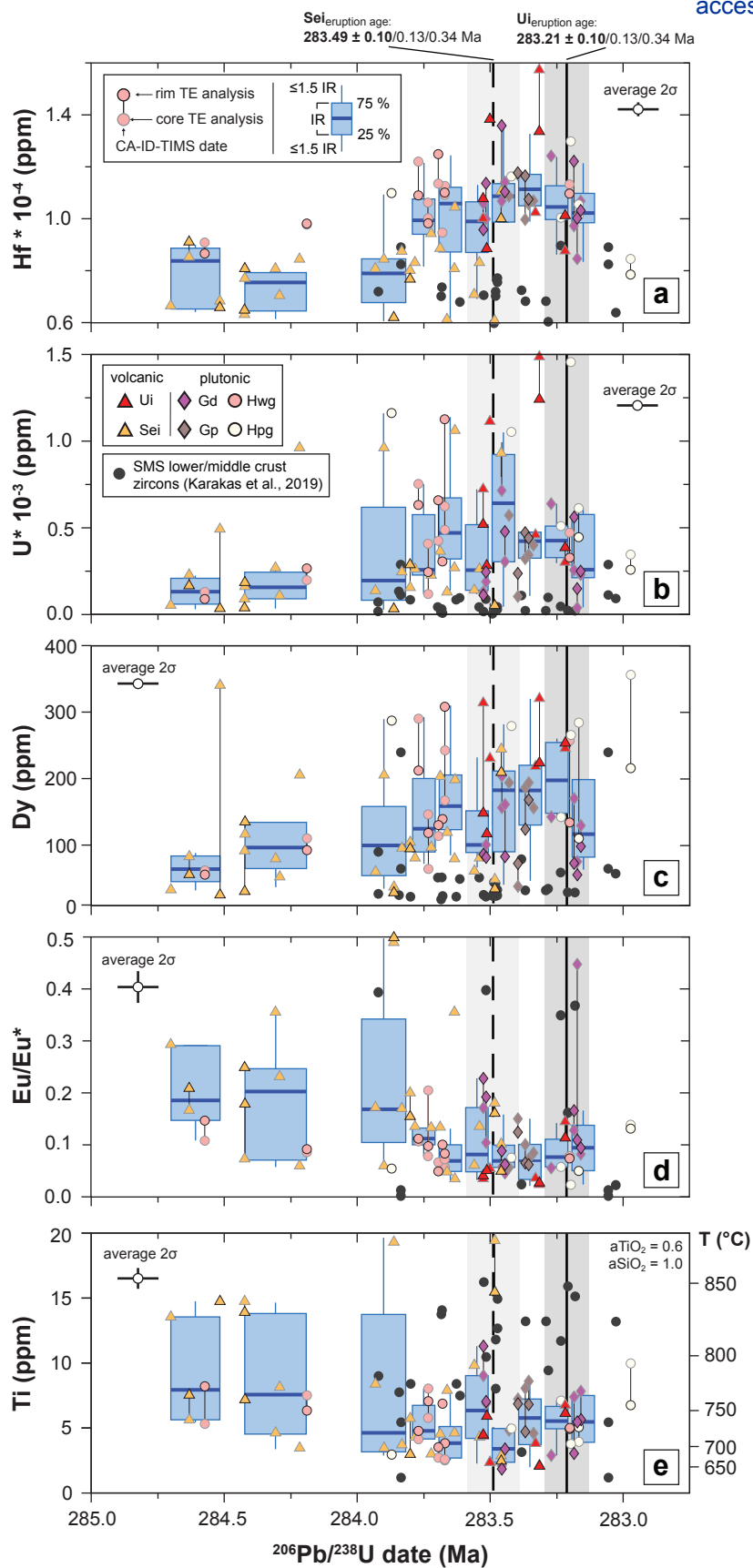


Figure. 4

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**Figure. 5**

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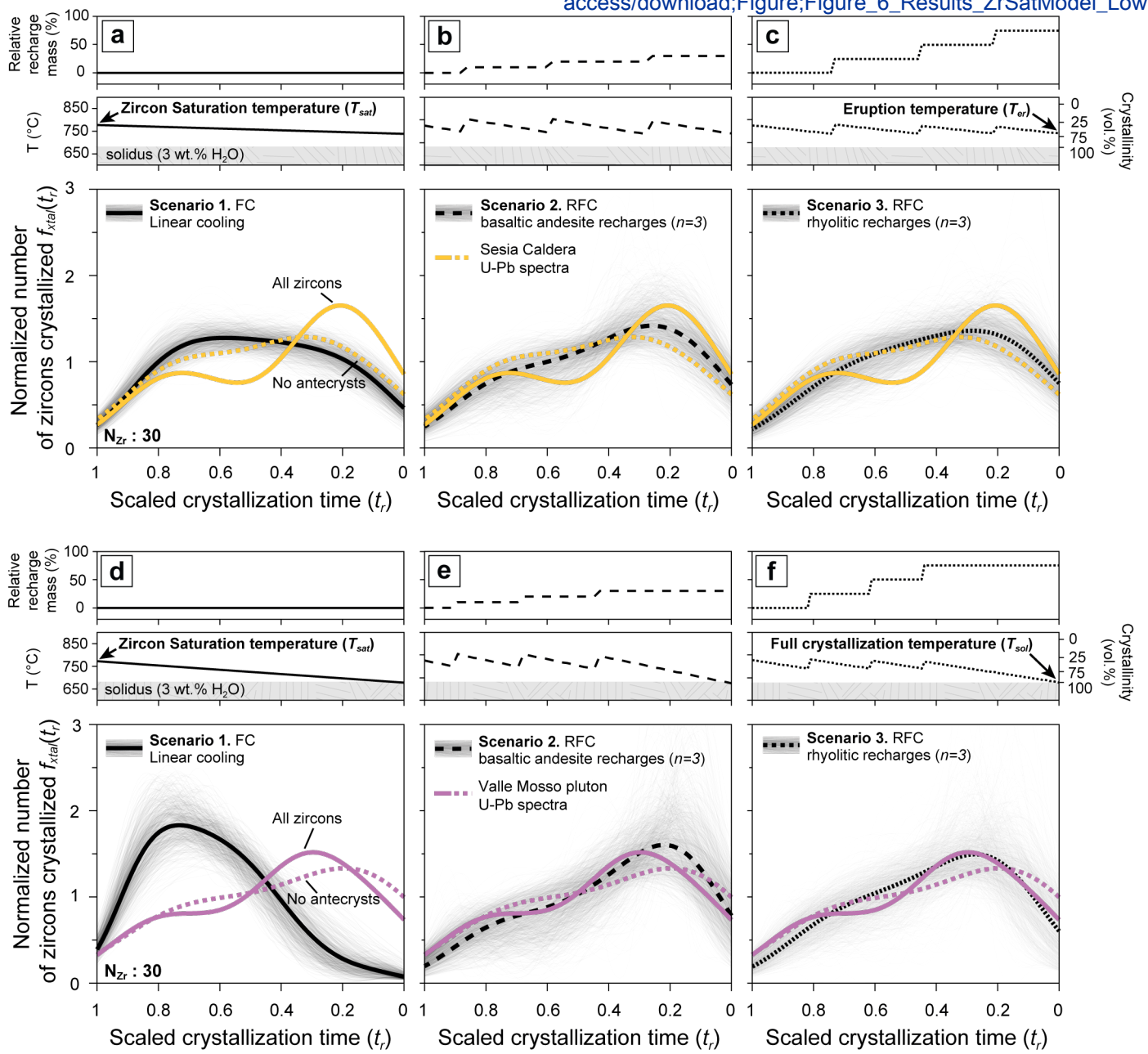
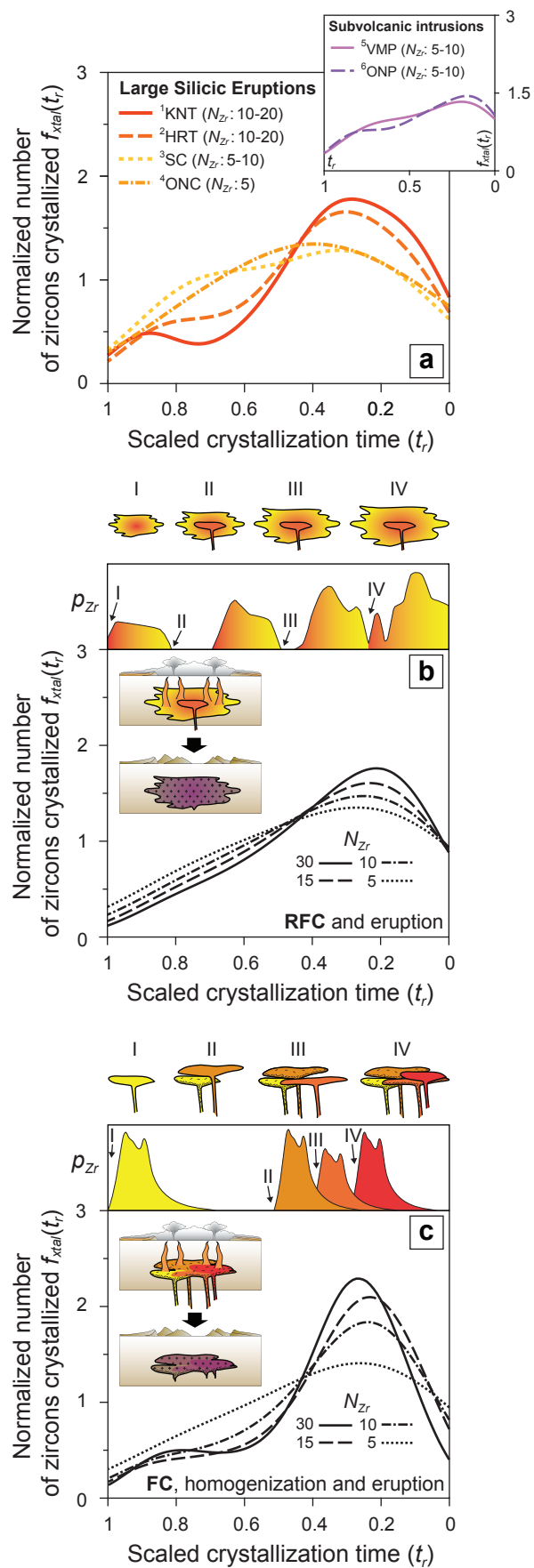


Figure.6

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**Figure. 7**

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Figure 8

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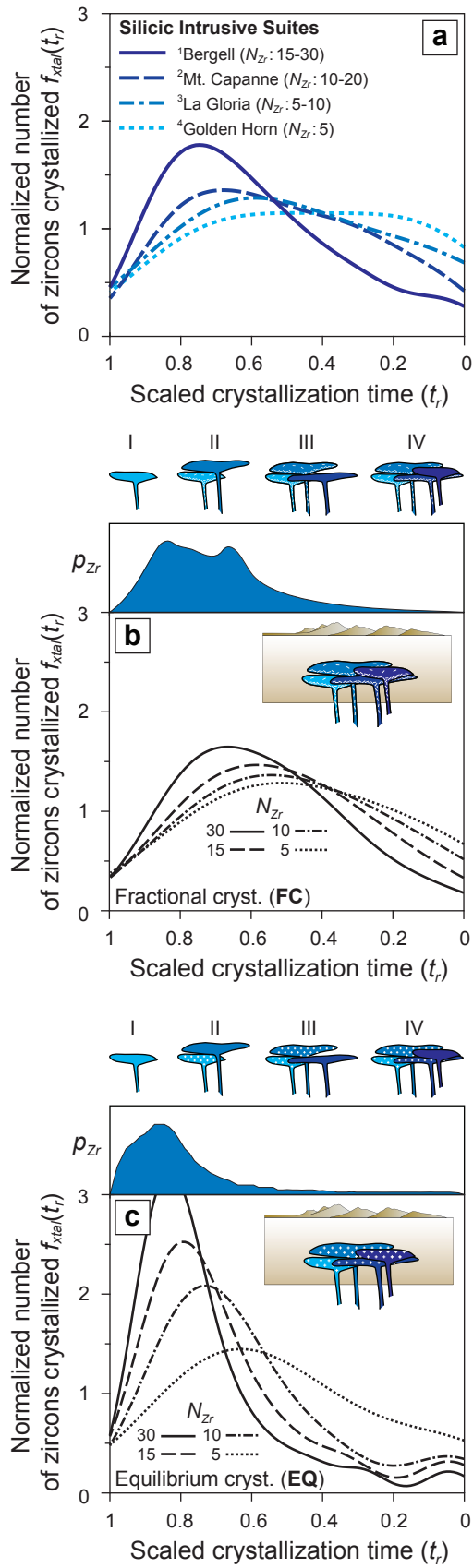


Figure 8

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