1	Weekly to	monthly	time scale	of melt in	clusion	entrapment
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- 2 prior to eruption [[indi@ed?]] by phosphorus distribution
- 3 in olivine from mid-ocean ridges
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14 ABSTRACT

15 Melt inclusions (MIs) hosted in euhedral olivine have been proposed to represent 16 droplets of primary melt, protected from processes occurring near Earth's surface during 17 eruption. The complex zoning of phosphorus (P) in some olivines and the presence of a 18 P-depleted zone around MIs indicate a complex history for the host-MI system. We 19 analyzed P in olivine and MIs from two mid-oceanic ridge basalt (MORB) samples from 20 the Mid-Atlantic Ridge (MAR) by electron probe microanalyzer, secondary ion mass 21 spectrometry (SIMS), and NanoSIMS. Phosphorus dendrites in olivine suggest an initial 22 fast olivine growth followed by a stage of slower growth. Dissolution textures around

23	some MIs were identified and were probably caused by adiabatic decompression melting.
24	Based on diffusion modeling of P in olivine, we infer that olivine beneath the MAR
25	remains in the system (1) for days to weeks after crystallization of P-rich lamellae, and
26	(2) for a few hours after recrystallization of dissolved olivine. Dissolution and
27	reprecipitation of olivine containing boundary layers suggests that most MIs might be
28	affected by late post-entrapment processes.

29 INTRODUCTION

30 Olivine-hosted melt inclusions (MIs) are widely used to constrain primary melt 31 compositions because they are isolated from the magma and assumed to preserve the 32 composition of the melt from which their host grew (e.g., Kent, 2008; Métrich and 33 Wallace, 2008; Schiano, 2003). Despite agreement that olivine-hosted MIs are entrapped 34 at relatively shallow levels (<10 km), the origin of melt entrapped in MIs is debated. 35 Some studies indicate that MIs represent the melt composition of the last reequilibration 36 in the mantle (e.g., Hart, 1993; Sobolev, 1996), while others argue that MIs record 37 interaction with the magmatic plumbing system (e.g., Kent et al., 2002; Saal et al., 2005; 38 Kamenetsky and Gurenko, 2007), or localized, grain-scale dissolution-reaction-mixing 39 processes (Danyushevsky et al., 2004). Finally, they can lose their initial water content 40 and change oxygen fugacity (f_{O_2}) (Gaetani and Watson, 2000), calling into question the 41 interpretation that the large compositional variations observed in MIs directly represent 42 the heterogeneity of the mantle source.

The idea that euhedral olivine grows in concentric layers from core to rim has
been challenged due to the discovery of irregular, rhythmic zonation in incompatible,
slow-diffusing elements such as phosphorus (P) (e.g., Milman-Barris et al., 2008; Welsch

46	et al., 2013, 2014). It has been suggested that euhedral olivine forms by infilling the
47	spaces of this skeletal and/or dendritic framework (Welsch et al., 2013), formed during
48	fast diffusion-controlled growth (> 10^{-6} to 10^{-7} cm/s; Jambon et al., 1992), when growth
49	rate slows down (Welsch et al., 2014). It has been demonstrated that MIs formed during
50	diffusion-controlled, rapid skeletal or dendritic growth of olivine may trap surface
51	boundary layers that modify their composition (Faure and Schiano, 2005). It is thus
52	important to understand the timing of MI entrapment. Indeed, P-depleted olivine halos
53	around MIs located close to P-zoned olivine have been described (e.g., Milman-Barris et
54	al., 2008, Shea et al., 2015'' matches multiple references. Please add
55	letters (e.g. "Smith, 2000a").]], Bouvet de Maisonneuve et al., 2016), suggesting
56	interaction (diffusion or dissolution) between the dendritic olivine and MIs.
57	Phosphorous diffusion is slow compared to that of other elements in olivine [log
58	$(D_0, \text{m}^2/\text{s}) = -10.06 \pm 0.8$, $E_a = 229 \pm 16 \text{ kJ}$, where D_0 is the pre-exponential factor, and
59	E_{a} is activation energy [Watson et al., 2015], two to four orders of magnitude slower than
60	Fe-Mg interdiffusion in olivine]. Acquisition of P profiles allows the residence time of
61	the olivine in the system to be determined, as well as the relative timing of the different
62	processes occurring in the olivine during mineral growth, melt inclusion entrapment, and
63	eruption. Estimated residence times of olivine within the mid-oceanic ridge basalt
64	(MORB) magmatic system based on diffusion modeling of different elements (Ni, Al,
65	Ca, Fe-Mg) has been proposed to be between <1 day and 4 yr, with shorter times being
66	prevalent (e.g., Nabelek and Langmuir, 1986; Zellmer et al., 2011; Colin et al., 2012). P
67	diffusion profiles are therefore expected to be very short, and require analysis by
68	nanoscale secondary ion mass spectrometry (NanoSIMS).

Publisher: GSA Journal: GEOL: Geology DOI:10.1130/G39463.1 We combined P X-ray maps and high-spatial-resolution NanoSIMS profiles of P,

- Al, Mg, and Si with MELTS software (http://melts.ofm-research.org) calculations and
- 71 quantitative measurements of P and major element concentrations in olivine and in their
- 72 MIs in order to better understand entrapment mechanisms. NanoSIMS profiles of P in

olivine allow the residence time of olivine in the system after MI entrapment and before

ruption to be estimated. We show that decompression can cause olivine dissolution and

75 MI formation upon reprecipitation on very short time scales.

76 SAMPLES AND ANALYTICAL METHODS

The selected olivines contain 50–350 µm glassy MIs and are from two different

78 locations on the Mid-Atlantic Ridge (MAR). The first sample is a dredged olivine-rich

picrite (sample ARP73-10-03, 36°8372'N, 33°2482'W) from the FAMOUS zone

80 (Bougault and Hekinian, 1974). This sample contains >20% olivine phenocrysts, rare

81 clinopyroxene, and plagioclase xenocrysts (Laubier et al., 2007). The geological context,

82 geochemistry of lavas, and MIs were described in several studies (e.g., Langmuir et al.,

83 1977; Laubier et al., 2007; Gale et al., 2013). The second sample (CH77-DR6-203) is a

84 dredged fresh basalt containing a high volatile proportion from the 14° MAR triple

s5 junction (14.12°N, 45°W) (Bougault et al., 1988; Javoy and Pineau, 1991).

86 Major element composition and X-ray element maps of olivine and MIs (Fig. 1;

Fig. DR2 in the GSA Data Repository¹) were acquired by electron probe microanalyzer

- 88 (EPMA; University of Lausanne, Switzerland). The P content of MIs and olivine was
- 89 determined by measuring ³¹P and ²⁸Si in monocollection using the CAMECA IMS 1280-
- 90 HR at the SwissSIMS laboratory (University of Lausanne). High-resolution ³¹P, ²⁸Si, Al
- 91 (²⁷Al¹⁶O), and Mg (²⁴Mg¹⁶O) profiles were acquired with a CAMECA NanoSIMS 50L

92	(École Polytechnique Fédérale de Lausanne, Switzerland) (Fig. 1; Fig. DR3). ³¹ P
93	measurements were normalized to ²⁸ Si, measured at the same time, in order to monitor
94	possible instrumental drift. Analytical solutions to the one-dimensional diffusion equation
95	were used to obtain least-square fits to the normalized profiles and determine the
96	residence time of olivine in the system both (1) after the crystallization of P-rich lamellae
97	and (2) after the dissolution event (Fig. 2). Uncertainties reported for calculated diffusion
98	time scales are from the fitting procedure and do not include the uncertainty of the
99	diffusion coefficient for P in olivine (Watson et al., 2015). Analytical details for all
100	methods are provided in the Data Repository.
101	RESULTS
102	Within each MI, the major element composition is homogeneous, except for a
103	slight Mg and Fe depletion along the $5-8-\mu$ m-wide MI wall (Fig. DR2). The studied parts
104	of olivine crystals do not show any significant chemical zoning except for P, which
105	outlines a dendritic pattern (Fig. 1; Fig. DR2). Nine of the 15 mapped olivine crystals
106	from both samples show P zoning. MIs were trapped in both P-zoned (containing P-rich
107	olivine lamellae) and unzoned areas of olivine. All MIs located in P-zoned domains have
108	a P-depleted halo, which has a variable and irregular shape truncating the P-rich lamellae.
109	Based on two-dimensional estimates on X-ray maps, the volume of the surrounding P-
110	depleted halo is at least as much as that of the MI. P concentrations in olivine are 30-
111	1200 ppm, and in MI, 300–500 ppm (Fig. 1).
112	At the olivine-MI boundary, the difference in composition between melt and
113	olivine on NanoSIMS profiles is reflected by a gradual decrease in ²⁴ Mg/ ²⁸ Si and increase
114	in ²⁷ Al/ ²⁸ Si and ³¹ P/ ²⁸ Si ratios (Fig. DR3), likely due to the geometry of the inclusion

115	walls. The ${}^{31}P/{}^{28}Si$ transition (Fig. 1; Fig. DR3) is too wide to be the result of this
116	geometry only and is likely related to growth kinetics, primarily affecting P, as it diffuses
117	more slowly than Mg or Al in the melt (Spandler et al., 2007; Chakraborty, 2010; Watson
118	et al., 2015). The small P-enriched zone (Fig. 1, profile 4) around the MI corresponds to
119	post-entrapment growth of olivine. The gradational variation in P concentration between
120	lamellae in the P-zoned olivine most likely represents diffusion profiles, whereas the
121	transition between P-depleted halo and P-zoned or unzoned olivine domains is sharp (Fig.
122	1; Fig. DR3).
123	DISCUSSION
124	A key observation is the systematic finding of a P-depleted halo around MIs
125	hosted in direct contact with P-rich olivine. This halo truncates the P-rich zoning (e.g.,
126	Fig. 1; Fig. DR2). Different processes could form P-depleted halos (Milman-Barris et al.,
127	2008): (1) slow olivine crystallization on the wall of a melt pool; (2) dissolution and
128	reprecipitation of olivine; or (3) diffusion of P between olivine and MI. The sharp
129	truncations of the P-zoned domains (Fig. 1) by the halo do not support the first and third
130	hypotheses. Also, the preservation of P-rich lamellae on the micrometer scale does not
131	support the formation of diffusion halos over tens of micrometers, because the bands
132	should also broaden at the same rate. Based on the irregular shape of the P-depleted halo
133	and the clear truncation of the P zonation (Fig. 1), a coupled dissolution and
134	reprecipitation mechanism is more likely (e.g., Milman-Barris et al., 2008). Dissolution
135	of olivine is favored along fractures and defect zones, and around high-energy surfaces
136	like MIs and embayments. Dissolution can be triggered by increase in temperature,
137	mixing of melts with different composition, decompression, increase of volatile content

138	in the melt, or a combination of these processes. H_2O input may induce dissolution by
139	lowering the melting temperature. In MORB settings, H ₂ O input is limited, and the
140	similar composition of MIs and olivine domains suggests neither fluid input nor mixing
141	of significantly different melts. As olivine is compositionally homogeneous (Fig. DR2)
142	and the different MIs hosted in a single olivine grain have similar composition (Table
143	DR2 in the Data Repository), temperature increase is an unlikely cause for olivine
144	dissolution. MI migration by dissolution of the olivine in front of the MI and
145	reprecipitation of olivine behind the MI requires temperature gradients in the mineral
146	(Schiano et al., 2006). Such a migration process could also create P-depleted halos
147	around MIs. Even if the presence of P-depleted halos around melt embayments has been
148	observed in olivine crystallization experiments by cooling of a single melt at constant
149	pressure (Milman-Barris et al., 2008), we do not favor dissolution triggered by increase in
150	temperature, as it seems unlikely that the necessary temperature gradient is present on the
151	crystal scale. We would expect to see temperature-induced zoning, inconsistent with the
152	homogeneity of the studied olivines. The third possibility for dissolution of olivine is
153	adiabatic decompression. Simple MELTS calculations (Ghiorso and Sack, 1995; Ghiorso
154	et al., 2002) demonstrate that moderate decompression (<2 kbar) can dissolve ~15%–
155	30% of the olivine (Table DR3; Fig. DR4), while affecting the forsterite content by
156	<0.7%. As the P-depleted halos represent only a small volume of the total olivine, the
157	required decompression is <2 kbar.
158	The MIs shown in Figure 1 were all entrapped during a slow crystal growth
159	period, as they have similar compositions. Indeed, if MIs "a" and "b" (Fig. 1) were

160 trapped during a period of fast growth, they would be enriched in elements incompatible

161	in olivine such as CaO or Al_2O_3 (Faure and Schiano, 2005). They can be trapped either
162	during initial olivine growth (called case 1 hereafter) or during dissolution-precipitation
163	along defects (case 2). For case 1, it is unlikely that pressure changes would be
164	transferred into the fully enclosed MI. Even if it was the case, local disequilibrium
165	between MI and the P-rich olivine would facilitate dissolution of the P-rich lamellae only
166	during decompression, a feature not observed here. In case 2, an olivine melt pocket is
167	connected with the surrounding melt in an open system (embayment). Decompression
168	induces dissolution of P-zoned and unzoned olivine. The dissolution of P-zoned olivine
169	will locally enrich the melt in P. As P diffusion in melt is slow and P is incompatible, the
170	new melt will locally have a higher P content. Subsequent temperature drop causes
171	olivine crystallization and entrapment of the MI. Our data favor the open-system
172	hypothesis.
173	NanoSIMS profiles across the P-rich dendrites highlight P diffusion in olivine.
174	Based on these diffusion profiles and on the diffusion coefficient from Watson et al.
175	(2015), we estimate that at 1270 °C, olivine can remain in the system for about ~1 week
176	$(7.5 \pm 0.3 \text{ days}, based on profile 9 [P9] from ARP73-10-03_16; Fig. 2)$ prior to eruption,
177	in agreement with previously estimated short residence times for MORB samples (e.g.,
178	Nabelek and Langmuir, 1986; Danyushevsky et al., 2002; Pan and Batiza, 2002; Zellmer
179	et al., 2011; Colin et al., 2012). A temperature of 1270 °C was chosen based on results
180	from sample ARP73-10-03[[Cor]] MI rehomogenization, which give temperatures
181	between 1270 °C and 1300 °C (Laubier et al., 2007). These temperatures match estimates
182	using MELTS (Ghiorso and Sack, 1995; Ghiorso et al., 2002) and Petrolog software
183	(Danyushevsky et al., 2000; Danyushevsky et al., 2002). Even at lower temperature, the

184	residence time of olivine remains short (~2 weeks at 1200 °C and ~2 months at 1100 °C).
185	The sharp boundary between P-zoned olivine and the P-depleted halo located around MIs
186	(profiles 4 and 7; Fig. 1) suggests olivine residence times of a few hours during MI
187	inclusion.
188	In summary, our data suggest that growth of some olivine crystals is initially
189	diffusion controlled, as reflected by P zoning (Fig. 3, stage 1), followed by interface-
190	controlled growth (Fig. 3, stage 2) (Welsch et al., 2014). During its ascent to the surface,
191	olivine starts dissolving, preferentially around embayment or along fracture/defect zones
192	(Fig. 3, stage 3), probably due to adiabatic decompression. Decreasing temperature
193	and/or degassing of the melt during ascent increases the temperature difference from the
194	liquidus (Applegarth et al., 2013), leading to reprecipitation of a P-depleted olivine and
195	the preservation of MIs with elevated P contents (Fig. 3, stage 4) at an interface-
196	controlled slow growth rate in the last hours before the olivine reaches the surface.
197	Olivine crystallizing at the wall of the MI, seen in almost all MIs (e.g., Kent, 2008), is
198	highly enriched in P, as shown in the NanoSIMS profiles, suggesting fast growth for this
199	last process (Fig. 3, stage 5).
200	CONCLUSIONS
201	Olivine dissolution-precipitation during[[Missing part of sentence here
202	(including verb)]], as a P-poor halo around MIs has been observed in olivine from two
203	MORB samples studied here, as well as in olivine from different settings (e.g., Milman-
204	Barris et al., 2008; Welsch et al., 2014). Phosphorous concentration maps of olivine
205	combined with NanoSIMS P profiles strongly suggest that olivine, after skeletal growth,
206	remains for a few days in the system, and that MIs are enclosed only shortly (hours)

- 207 before eruption. MI composition can be affected by olivine dissolution and
- 208 recrystallization processes, at least for P, and cannot be simply taken as representative of
- 209 the mantle source. Care should thus be taken when interpreting MI P and other
- 210 incompatible element compositional variability.

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- 349 **FIGURE CAPTIONS**
- 350 Figure 1. Electron probe microanalyzer (EPMA) phosphorus (P) elemental X-ray map of
- 351 olivine ARP73-10-03_16. P concentrations were determined quantitatively by secondary
- 352 ion mass spectrometery (SIMS). Note higher P concentration in melt inclusions (MIs)
- 353 situated near P-zoned olivine compared to ones located in P-unzoned olivine. Pink arrows
- 354 represent nanoscale SIMS (NanoSIMS) profiles; arrow indicates direction of profile, and
- number is profile number. NanoSIMS profiles used ~150-nm-diameter beam. ${}^{31}P/{}^{28}Si$
- profiles 4, 7, and 9 are located at boundary between MI and olivine (profiles 4 and 7) and
- in olivine (profile 9). Interface between MI and olivine was determined based on Al and
- 358 Mg NanoSIMS profiles (Fig. DR3 [see footnote 1]). Peak in rectangle in profile 9 is peak
- fitted in Figure 2. Peak situated at 47 μ m in profile 9 is originally composed of three
- 360 lamellae (indicated by gray rectangles). Because their diffusion profiles overlap, they
- 361 were not fitted.

362

363	Figure 2. Comparison of measured ${}^{31}P/{}^{28}Si$ profile in olivine ARP73-10-03_16 (see Fig. 1
364	for profile location) with calculated curve derived from point-source diffusion modeling.
365	Note excellent fit. Diffusion was ~8 days $a \square 70 \degree C[[Define "t_{1270°C}"]]$, using diffusion
366	coefficient from Watson et al. (2015). This is a maximum estimate for temperature,
367	because an infinitesimally small point source was assumed.
368	
369	Figure 3. Schematic evolution of olivine crystal and melt inclusion (MI) entrapment in
370	(100) section. Phosphorus content during different steps of olivine growth and MI
371	entrapment is shown schematically on the profiles. Stages 1 and 2 correspond to growth
372	of dendritic olivine followed by overgrowth of dendrites, leading to formation of
373	euhedral olivine. Melt may be entrapped at this growth stage. Stage 3 illustrates partial
374	olivine dissolution due to adiabatic decompression melting. P-zoned olivine domain
375	might be dissolved at this stage. P-depleted halo will then recrystallize at slow growth
376	rate (stage 4), passively enriching MIs in P. Once olivine rapidly cools (stage 5),
377	remaining melt will be quenched and will form glassy MI. Submicron layer of olivine
378	might crystallize at MI wall and will be enriched in P. De the figure, correct the
379	spelling of "crystallization"; check the location of the right-hand arrow in the "stage
380	3" panel – should it point to the orange area?]]
381	
382	¹ GSA Data Repository item 2017xxx, additional information about the sample
383	preparation and EPMA, SIMS, and NanoSIMS measurements, as well as information

384 about fitting parameters, is available online at

Publisher: GSA Journal: GEOL: Geology DOI:10.1130/G39463.1 385 http://www.geosociety.org/datarepository/2017/ or on request from

386 editing@geosociety.org.

1 FIGURE CAPTIONS









- 8 Fig.3

GSA Data Repository 2017359

Manzini et al., 2017, Weekly to monthly time scale of melt inclusion entrapment prior to eruption recorded by phosphorus distribution in olivine from mid-ocean ridges: Geology, doi:10.1130/G39463.1.

1 Supplementary data

2

3 Methodology

4 Sample preparation

Polished olivine grains with melt inclusions exposed at the surface were pressed into 1-inch
indium mounts. Different glasses and San Carlos olivine were added to the sample mount to
be used as reference material for P content determination by SIMS (see below).

8 To ensure that the melt inclusions were totally glassy, we acquired SEM images with a Tescan

9 Mira LMU scanning electron microscope at the University of Lausanne.

10

11 Electron microprobe

12 All samples were analyzed by a JEOL 8200 Superprobe at the University of Lausanne. 13 Analytical conditions for major element measurements in olivine were 15kV acceleration 14 voltage, 20nA beam current and 5 µm beam size. Counting time was 30 sec for Si, Mg, Mn, 15 Al and Fe and 40 sec for Ca, Ni and Cr. Natural minerals and synthetic oxides were used as 16 standards. Analytical conditions for major element analyses in melt inclusions were 15kV, 17 10nA and 10 µm. Counting time was 30 sec on all elements, except for K (20 sec) and Na (16 18 sec). A glass standard (KL2-G) was used for SiO₂, Al₂O₃. Other elements were calibrated on 19 specific minerals (e.g., a fayalite for Fe). ML3B-G was used as internal standard to check the 20 calibration. Post-entrapment modification due to olivine crystallization (PEC% (% olivine

added), Table DR1) at the wall of melt inclusion was corrected for using the Petrolog software
(Danyushevsky et al., 2000, 2002; Danyushevsky and Plechov, 2011).

23 X-rays distribution maps of melt inclusions and olivines were acquired by electron 24 microprobe using a JEOL 8200 Superprobe at University of Lausanne. Analytical conditions 25 were 15 kV, 10 nA, 0-1 µm probe diameter and 100 ms dwell-time with a 1-2 µm pixel size 26 for major elements and 15kV, 200-250nA, 0-1 µm probe diameter and 100-230 ms dwell-time 27 with a 1 µm pixel size for P, Al, Ni and Ca. X-ray elemental maps display higher count rate 28 for P in MI compared to P-zoned olivine, as a result of the overlapping of the Ca (K β) second 29 order peak with the P (K α) peak. As olivine contains much less Ca than glass, the interference 30 is negligible in olivine but not in MI. In any case, as olivine is homogeneous in Ca (Figure 31 DR2), the structure on the phosphorus maps and profiles are only caused by variable P 32 concentrations.

33

34

35 Secondary ion mass spectrometer

36 P concentrations in olivines and melt inclusions were analysed using a Cameca 1280-HR SIMS at the SwissSIMS laboratory (Lausanne, Switzerland). San Carlos (P = 40 ppm; Welsch 37 38 et al., 2014) was used as standard for the olivine and BHVO-2G, BCR-2G, BIR-1G (P 39 concentrations are from the certified values furnished by USGS) for the melt inclusions. Samples were sputtered with a 10 kV ¹³³Cs⁺ primary beam of 1 nA. Electron gun was used to 40 41 compensate sample charge. A 15µm raster was used during pre-sputtering (90 sec) and analyses. Counting time was 0.48 on 29.9 (background), 1.04 s on ³⁰Si and 2.96 sec on ³¹P. 42 43 Waiting time was 0.8 s between each mass. Total time for each analysis was <6 min, including automatic centering of the secondary beam.³⁰Si and ³¹P were detected on an axial 44 45 electron multiplier. Entrance slit was set to 60 µm and exit slit to 200 µm, resulting in ~6000 46 mass resolving power. Field aperture was closed to 4000 µm. Energy slit was 50 eV.

47 Uncertainty on each point measurement (internal error) was < 5% (2σ %, average uncertainty 48 of 0.95% 2σ) for P in olivine and <0.8% (2σ %, average uncertainty of 0.2% 2σ) in melt 49 inclusion. The difference in reproducibility is due to counting statistic, melt inclusions having 50 100 times more counts on P. Reproducibility on the standard was < 4.5% (2SD) on the San 51 Carlos and <1% (2SD) on BCR-2G.

52

53 Nanoscale secondary ion mass spectrometer

Profiles in olivines were obtained using the NanoSIMS at the Center of Advanced Surface Analysis (CASA, Lausanne, Switzerland). A Cs⁺ primary beam was used to sputter olivine and the ion species ¹⁶O⁻, ²⁸Si⁺, ³¹P⁻, ²⁴Mg¹⁶O⁻, ²⁷Al¹⁶O⁻ and ³¹P¹⁶O⁻ were extracted and measured using electron multipliers. In order to prevent the build-up of a charge on the sample's surface, the surfaces were coated with a 10-15nm gold layer. MRP was > 8000 and no peak overlap was observed.

60 After identification of the area of interest using the CCD camera of the NanoSIMS, a 25 x 25 61 µm image of the area of interest was first acquired (with a 256 x 256 pixel resolution 62 corresponding to a pixel size ~ 100 nm). Using the NanoSIMS software, the locations of the 63 linescan were precisely selected to be perpendicular to the transition identified by EMPA 64 analyses. The line scans were acquired using a two-step approach. First a pre-sputtering phase 65 was performed for 2 cycles with a dwell time of 2sec/pixel, using slightly larger beam size to 66 clean up the surface (diaphragm D1-2, and 3.8pA for the Cs⁺ primary beam). The acquisition 67 of the high resolution line scans were performed using a finer beam (D1-3, ~150nm, and 68 1.7pA for the Cs+ beam) over 5 cycles with 2sec/pixel. ²⁴Mg¹⁶O⁻ data were used to monitor 69 possible local variation in the ionization and extraction processes as it is expected to be homogeneous in the olivine measured. 16O, 18O, 24Mg, 27Al, 28Si, 31P and 31P16O were 70 71 acquired at the same time. In order to minimize the instrumental drift, we normalized the profiles to ²⁸Si, which is, based on EMPA X-ray maps, homogeneous within the olivine and 72

the MI. The NanoSIMS session was not standardized, resulting in a ³¹P count rate higher in
the MI than in P-zoned olivine, due to matrix effects.

75

76 Diffusion Chronometry and Fit of Diffusion Profiles

The 2D profiles were measured perpendicular to the zonation. The following parameters were assumed for the diffusion calculations and fitting of the profiles: 1- each P-enriched band was homogeneous at the beginning or represents a line source; 2- the contact between the P-rich and P-poor zones was a step function. Thin, linear features with no plateau were modelled as a line source, while P-enriched zones displaying a plateau were modeled as step functions.
Specifically, a P-peak of a few µm width (e.g. Fig. 2) was modelled as line source, while b) P-

83 enriched zones larger than 10 µm were modelled as individual step functions (e.g. Fig. DR5).

We used the diffusion coefficient for P diffusion in olivine reported by Watson et al. (2015) based on their diffusion experiments in olivine which are $log(D_0,m^2/s) = -10.06 \pm 0.80$ and E_a $= 229 \pm 16$ kJ/mol. Error functions were fit to each of the measured profiles using a weighted Marquardt fitting routine (Press et al., 1992).

The uncertainty of the fit was estimated using the χ^2 per point. The uncertainty, expressed as 2 σ , for each point was estimated from the variation in the part of the profiles considered to be flat. Initial conditions of the model are the bigger source of uncertainty. Since the line source diffusion modeling assumes an infinite band width of P-enrichment, the calculated values are maximum time estimates.

93

94 **MELTS calculations**

MELTs version 5.0 was used. The principles of the program are summarised in Ghiorso and
Sack (1995). We used the average composition of the individual melt inclusions and 0.15 wt%
H₂O. We then determined the liquidus temperature, followed by decompression calculations
slightly below the liquidus temperature. Isentropic, isenthalpic and isothermal decompression

99 calculations were done between 2 kbar and 0.1 kbar in steps of 0.1 kbar, with a fixed fO_2

- 100 (QFM -2).
- 101
- 102

103 **Figures**



Figure DR1: Plot of ${}^{31}P/{}^{28}Si$ ratios obtained during NanoSIMS measurements versus (a) ${}^{48}Si$ counts and (b) ${}^{31}P$ counts. The absence of correlation in (a) and the good correlation in (b) indicates that ${}^{31}P/{}^{28}Si$ ratio depends on ${}^{31}P$ concentration only.





110 Figure DR2: X-ray maps for A- Mg concentration in ARP73-10-03 16; B- Al concentration 111 in ARP73-10-03 16; C- Ca concentration in ARP73-10-03 16; D- P concentration in CH77-112 DR6-203 52; E- P concentration in CH77-DR6-203 4; F- P concentration in CH77-DR6-113 203 8. Note the homogeneity of the olivine in major elements (A, B and C), whereas nice P 114 zonations are present in the same olivine (Figure 1). MI are homogeneous in P, except in Fig. 115 DR2-E. Apparent heterogeneity of this specific MI is explained by the presence of a previous 116 SIMS analysis spot, creating a small topography, able to slightly modify the P signal during P 117 imaging by EMPA.



120 Figure DR3:

³¹P/²⁸Si profiles realized in the ARP73-10-03_16 olivine and melt inclusions for all the
 measured elements. The contact between MI and olivine can be determined based on the Mg
 and Al profiles. The location of the profiles is presented in Figure 1.

124

125



Figure DR4: Results of the MELTS decompression calculations at fixed fO_2 (QFM) between 2 to 0.1 kbar for isothermal, isenthalpic and isentropic decompression. Ol is for olivine. Isentropic and isenthalpic decompression causes weak to moderate initial resorption, while isothermal decompression only causes crystallization.

127



Figure DR5: Comparison of the measured ³¹P/²⁸Si profile in olivine with the calculated curve derived from semi-infinite source diffusion modeling for profile 9, at the contact between Prich olivine and P-depleted halo. Diffusion was 0.8 days at 1270°C, using D from Watson et al. (2015). This estimated time scale is a maximum, since the profile is near vertical, and its width is constraint by one point which could represent an analytical artefact. In fact, the profile width could be significantly overestimated.



- 142
- 143 Figure DR6: P distribution in ARP73-10-03_16 olivine.
- 144
- 145 **Tables**
- 146 Table DR1
- 147 NanoSIMS measurements of the profile 9 realized in the ARP73-10-13_16 olivine.

Distance (um)	31P (cps)	28Si (cps)
18.0858400	19	97678
18.3477400	26	121670
18.5226800	11	111065
18.7845900	18	126832
18.9595300	18	108121
19.2214400	24	125457
19.3963800	23	105542
19.6582900	34	142189
19.8332200	41	147602
20.0951400	37	145394
20.2700700	37	111402
20.5319900	46	127164
20.7069200	44	102883
20.9688400	72	126160
21.1437600	82	100907
21.4056900	136	117867
21.5806100	92	101973
21.8425400	159	124376
22.0174600	148	106587
22.2793900	149	122282
22.4543000	133	101183
22.7162500	182	119846
22.8911500	157	109615
23.1531000	194	127241
23.3280000	149	108946
23.5899500	145	125116
23.7648500	147	105254
24.0268000	164	124435
24.2017000	125	104253
24.4636500	143	126520
24.6385400	107	108187
24.9005000	93	127452
25.0753900	70	112467
25.3373500	42	129259
25.5122400	33	109530
25.7742000	42	122799
25.9490900	32	109173
26.2110500	42	126676
26.3859400	20	111107

Distance (um)	31P (cps)	28Si (cps)
26.6479000	20	133279
26.8227900	21	112554
27.0847500	27	135998
27.2596300	24	119142
27.5216000	33	127031
27.6964800	32	133735
27.8716300	27	115507
28.1333300	23	123117
28.3084800	16	108431
28.5701800	26	124510
28.7453200	17	109618
29.0070300	20	133930
29.1821600	14	126906
29.4438800	23	134133
29.6190000	27	145317
29.8807300	25	147300
30.0558500	25	115786
30.3175800	23	136501
30.4926900	16	126328
30.7544300	24	129967
30.9295300	18	117576
31.1912700	15	135174
31.3663800	18	116562
31.6281200	18	136904
31.8032200	23	121893
32.0649700	22	130770
32.2400700	15	118591
32.5018200	17	142127
32.6769100	15	120532
32.9386700	25	137011
33.1137600	19	121271
33.3755200	32	134642
33.5506000	22	120888
33.8123700	27	136803
33.9874500	20	119072
34.2492200	21	125619
34.4242900	12	106236
34.6860700	14	126237
34.8611400	12	110990

- Table DR2
- 150 Major element compositions (determined by EPMA) and P concentration (determined by
- 151 SIMS) of the melt inclusions hosted in the ARP73-10-13_16 olivine. Values measured for
- 152 ML3B glass, used as external standard for EPMA glass analyses, are also reported. Major
- element concentrations are given in wt% and P concentrations in ppm. For ML3B FeO
- 154 represents FeO_{tot}.

Table DR2

Sample	SiO2	TiO2	Al2O3	Fe2O3	FeO	MnO	MgO	CaO	Na2O	К2О	% ol addec	P (ppm)	2sd%
ARP73_16a	49.52	0.59	14.42	0.91	7.83	0.12	12.07	12.96	1.51	0.07	10.9	374	0.07
ARP73_16b	49.81	0.53	14.44	0.88	7.58	0.09	11.68	13.41	1.51	0.08	12.7	495	0.06
ARP73_16c	49.71	0.55	14.53	0.87	7.63	0.14	11.80	13.28	1.43	0.06	17.0	297	0.09
ARP73_16d	48.97	0.59	14.85	0.90	7.77	0.12	11.83	13.33	1.57	0.07	17.3	311	0.08
ML3B	51.56	2.10	13.72	n.d.	11.59	0.17	6.59	10.33	2.52	0.39		n.d.	

- 156 Table DR3
- 157 Results of the MELTs calculations for the three different models tested.

Table DR3

iso	thermal o	decompress	sion	ise	ntropic d	ecompress	ion	isenthalpic decompression				
T (C)	P (kbars) mass (mg)	forsterite	т (С)	P (kbars) mass (mg)	forsterite	T (C)	P (kbars) mass (mg)	forsterite	
1240	2	5.194	84.27	1250	2	4.1818	84.70	1230	2	5.2288	83.59	
1240	1.9	5.1211	84.30	1248.7	1.9	4.242	84.67	1229.6	1.9	5.3998	83.65	
1240	1.8	5.0479	84.33	1248.3	1.8	4.2044	84.69	1229.1	1.8	5.5621	83.71	
1240	1.7	4.9744	84.36	1248	1.7	4.1666	84.71	1228.6	1.7	5.7184	83.76	
1240	1.6	4.9005	84.40	1247.6	1.6	4.1286	84.72	1228.1	1.6	5.8691	83.82	
1240	1.5	4.8262	84.43	1247.2	1.5	4.0904	84.74	1227.6	1.5	6.0147	83.87	
1240	1.4	4.7516	84.46	1246.9	1.4	4.0521	84.76	1227.6	1.4	5.9872	83.92	
1240	1.3	4.6767	84.50	1246.5	1.3	4.0135	84.77	1229.2	1.3	5.7603	84.02	
1240	1.2	4.6014	84.53	1246.1	1.2	3.9748	84.79	1230.7	1.2	5.5322	84.12	
1240	1.1	4.5258	84.56	1245.8	1.1	3.936	84.81	1232.3	1.1	5.303	84.23	
1240	1	4.4498	84.60	1245.4	1	3.8969	84.83	1233.8	1	5.0728	84.33	
1240	0.9	4.3735	84.63	1245	0.9	3.8577	84.84	1235.4	0.9	4.8414	84.43	
1240	0.8	4.2968	84.66	1244.6	0.8	3.8182	84.86	1236.9	0.8	4.6089	84.53	
1240	0.7	4.2197	84.70	1244.3	0.7	3.7787	84.88	1238.5	0.7	4.3753	84.63	
1240	0.6	4.1423	84.73	1243.9	0.6	3.7389	84.90	1240	0.6	4.1406	84.73	
1240	0.5	4.0645	84.77	1243.5	0.5	3.6989	84.92	1241.6	0.5	3.9048	84.83	
1240	0.4	3.9864	84.80	1243.2	0.4	3.6588	84.94	1243.1	0.4	3.6678	84.93	
1240	0.3	3.9079	84.83	1242.8	0.3	3.6185	84.95	1244.6	0.3	3.4297	85.03	
1240	0.2	3.829	84.87	1242.4	0.2	3.578	84.97	1246.1	0.2	3.1905	85.13	
1240	0.1	3.7497	84.90	1242	0.1	3.5373	84.99	1247.6	0.1	2.9501	85.23	

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