SUPPLEMENTARY INFORMATION

doi:10.1038/nature09948

This Addendum provides material additional to our published paper[1] in response to a concern that we have not published our large data sets of phase compositions as functions of water, pressure (P) and temperature (T). Similarly, although our paper includes important information on water storage capacity within the upper mantle, we have not published the numerical data of water contents in nominally anhydrous minerals (NAMs), but only the methods by which our data can be replicated. We wish to clarify the context and purposes of our article and our intentions for the large data sets on which our article is based. We provide, in Table 1, two examples of phase analyses from below and above the solidus of vapour-saturated lherzolite, and in Table 2 are six examples of quantification of water contents in NAMs. Two water-in-olivine analyses are from the experiments in Table 1 and two other experiments provide data on co-existing olivine + orthopyroxene, and orthopyroxene + clinopyroxene at the same P, T, and bulk water contents. Tables 1 & 2 provide examples of our full data, which are close to submission in three papers which detail (a) the 2.5 ± 0.1 GPa experiments with variable water content and variable T; (b) the 1.5 to 6 GPa experiments with variable T and several water contents; (c) the FTIR study of monomineralic layers adjacent to the HZ1 lherzolite layers.

The context for our 'Letter to Nature'[1] is current debate on roles of water in planetary interiors. The P, T dependence of the water-saturated solidus of peridotitic compositions is recognised as a major constraint on Earth's upper mantle properties and processes. Publication[2] of an experimental study of the water-saturated peridotite solidus which discounted prior experimental studies including the demonstration of the importance of pargasite, created an unresolved conflict in the published literature. Furthermore, this experimental determination[2] of the water-saturated solidus was adopted in models of hydrous upper mantle melting[3,4], particularly in models of mantle wedge melting[2,5,6]. For these reasons, our new experimental study[1] clarifying the apparently conflicting interpretations by demonstrating the effects of different water contents, was published in a summary article[1]. In Fig. 1 we illustrate the differences between the two recent experimental water-saturated

solidus determinations under discussion[1,2], and the solidi predicted for hydrous upper mantle with different bulk water contents using a modeling approach[3]. In the modeling approach[3] the positions of the solidi for different water contents are derived from the water-saturated solidus and using mineral/melt water partitioning values and inferred high water contents in NAMs. The data on phase assemblages and bulk water contents published in our 'Letter to Nature'[1] are sufficient to correct the position of the water-saturated solidus, the role of pargasite and the positions of the solidi for bulk water contents greater than 0.05 wt% H_2O. Resolution of the controversy illustrated in Fig. 1 required a novel experimental approach. Our 'Letter to Nature'[1] is thus a notice of this study introducing a new experimental methodology; an application of our results to a major issue in upper mantle geodynamics; a summary of our results; and presentation of information enabling our experiments to be repeated.

In Table 1 we illustrate examples of mineral compositions in a subsolidus assemblage in which pargasite is present, and an above-solidus assemblage containing quenched water-rich silicate melt segregated into the olivine layer. The spot analysis of Fe-rich quench amphibole, the mean composition of 15 area-scan analyses including both olivine and quench aggregates, and the calculated melt composition are also included in Table 1. The melt was calculated[7] from plots of oxides vs. Mg# (Mg# = 100Mg/(Mg+Fe)) assuming a value of Mg# = 75 for the melt. These data are illustrative of those to be submitted for publication, for all experiments listed in Tables S3 & S4[1].

In our article[1] we emphasized the importance of pargasite stability for water storage capacity (Fig. 1)[1] in the uppermost mantle and also reported a water content retained in NAMs at the water saturated solidus of ~180 ppm. As the water content in near-solidus, basaltic melt is 25-30 wt% H_2O at 2.5 to 3 GPa, the partitioning of water between residual lherzolite and melt, $D_{mineral/melt}$ ~ 0.0007. This summary of water partitioning at the vapour-saturated solidus is derived from our full data set but was not supported by publication of the full data. We now provide examples of infra-red spectra (Fig. 2) and of measured water contents in olivine, orthopyroxene and clinopyroxene (Table 2) as illustrations of the material on which we have based our summary article[1]. Knowing the bulk composition of Lherzolite HZ1 and having analyses of all phases (except excess aqueous vapour in O85 at 1000°C) we

can calculate modal proportions (Column 2 in Table 1). Using the measured 40 ppm H_2O in olivine and values of 10 and 2.5 for orthoyroxene/olivine and clinopyroxene/orthopyroxene partitioning respectively, we calculate ~200 ppm H_2O in NAMs in O85 at 1000°C at 2.5 GPa. This experiment is subsolidus, with ~0.14 wt% H_2O held in pargasite and ~1.3 wt% H_2O in water-rich vapour. In O-77 at 1050°C at 2.5 GPa, the modal abundances of phases yield an estimate of ~220 ppm H_2O in NAMs coexisting with 4 wt% melt containing 30 wt% H_2O i.e. $D_{mineral/melt}$ = 0.0007. Our full data set provides data over a range of temperature, pressure and variable activity (concentration) of water in the vapour phase. An example of the effects of variable water activity is in the nominally 'dry' experiment P-30 at 1000°C at 2.5 GPa (Table 2). The experimental methods do not completely exclude hydrogen and activities of H_2O, Na_2O, K_2O, TiO_2 etc. are appropriate to stabilise pargasite. However, there is only about half the water contents in olivine and orthopyroxene as in the 'water-saturated' at the same P, T experiments. In our paper[1] we applied our new values for water contents in minerals at the water-saturated solidus to a model mantle mineralogy of Hirth and Kohlstedt[11] (rather than the mineral modes in our own experiments) to facilitate comparison with earlier literature, and we assumed that there is equal amount of water present in olivine and garnet[12] to derive an approximate water content of 180 ppm in mantle residue at the vapour-saturated solidus. The uncertainty in the estimation of residual water concentration is approximately 30 %.

The article[1] is not a geochemical/mineralogical paper but an integration of the new data obtained by our novel technique and from published papers. The evidence for and consequences of solidus temperatures, pargasite stability at the solidus, and conditions for vapour phase leaching rather than hydrous melting, are demonstrated by the data originally provided[1]. We have a large data set of phase compositions, including water contents in NAMs, which are relevant to many other issues such as mantle geothermometry and geobarometry. These data could not be incorporated in a 'Letter' format but are close to submission to specialist journals.

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Table 1. Compositions of O85 and O77 ('HZ1' compositions) experimental run products (wt%) at 2.5 ± 0.1 GPa. Modal proportions of phases calculated by mass balance. Analyses have been normalised to 100 wt% for OL, OPX, CPX, GA, and Q, and to 98.5 wt% for PAR before averages have been calculated. Mg# = 100Mg/(Mg+Fe) in atomic units; n, number of analyses; n.d., not detected; OL, olivine; OPX, orthopyroxene; CPX, clinopyroxene; GA, garnet; PAR, pargasite; AMP, amphibole; Q, quenched liquid or quench from vapour phase; Calc. Melt, calculated melt.

Phase	modal%	n	SiO_2 ± 1σ	TiO_2 ± 1σ	Al_2O_3 ± 1σ	Cr_2O_3 ± 1σ	NiO ± 1σ	FeO ± 1σ	MgO ± 1σ	CaO ± 1σ	Na_2O ± 1σ	K_2O ± 1σ	Mg#
HZ1 Peridotite at 2.5 GPa													
Run O-85 (1000°C, 1.45% H_2O)													
OL	52	3	40.83±0.32	0.01±0.10	n.d.	0.11±0.03	0.49±0.32	9.64±0.15	48.85±0.24	0.07±0.03	n.d.	n.d.	90.0
OPX	21	6	55.79±1.10	0.14±0.09	2.44±0.20	0.18±0.12	0.13±0.16	6.91±1.30	33.43±0.55	0.91±0.16	0.07 ± 0.08	n.d.	89.6
CPX	10	10	53.36±0.31	0.34±0.17	3.73±0.34	0.82±0.33	0.12±0.11	3.27±0.61	17.03±0.27	20.28±0.67	1.03±0.15	0.01±0.05	90.3
GA	10	8	42.16±0.25	0.49±0.22	21.73±0.36	1.59±0.70	0.07±0.12	8.12±0.28	19.12±0.48	6.68±0.71	0.03±0.02	0.01±0.03	80.8
PAR	7	7	45.61±0.47	0.95±0.17	13.47±0.59	0.60±0.15	0.17±0.15	4.16±0.25	19.82±0.36	10.00±0.54	3.27±0.12	0.45±0.04	89.5
Run O-77 (1050°C, 1.45% H_2O)													
OL	52	5	41.01±0.11	0.01±0.09	0.01±0.02	0.10±0.03	0.38±0.14	9.59±0.13	48.79±0.14	0.10±0.03	n.d.	n.d.	90.1
OPX	24	5	56.12±0.54	0.11±0.06	2.78±0.49	0.46±0.13	0.08±0.20	5.88±0.04	33.69±0.42	0.85±0.07	0.03±0.04	0.01±0.04	91.1
CPX	10	5	53.15±0.39	0.30±0.04	3.78±0.52	0.95±0.26	0.19±0.12	2.94±0.20	17.56±0.34	20.31±0.39	0.82±0.07	0.01±0.05	91.4
GA	10	5	42.20±0.13	0.22±0.10	22.59±0.32	1.43±0.70	n.d.	7.79±0.72	19.84±0.60	5.93±0.36	n.d.	0.00±0.01	81.9
Q-AMP		1	45.50	0.88	14.56	n.d.	0.27	7.20	25.25	0.43	3.80	0.84	86.2
Q+OL		15	44.70±2.58	0.62±0.21	9.29±4.10	0.00±0.09	0.27±0.19	7.63±1.01	28.42± 9.68	5.99±3.39	2.58±1.06	0.50±0.31	85.9
Calc. Melt	4		47.50	1.40	19.60	—	—	4.60	9.0	12.80	5.50	1.40	75.0

Table 2. Examples of quantification of water contents in NAMs. Abbreviations similar to Table 1; OL(SC), San Carlos olivine; low-Al, low alumina.

Sample	Mineral	P (GPa)	T (°C)	Mix	Bulk H_2O wt.%	Hydrous phase	Non-solid phase in equilibrium	Mineral layers	n	H_2O (ppm) ±30 %
P-30	OL	2.5	1000	HZ1	'dry'	PAR	-	OL(SC) & low-Al OPX	12	24
P-30	OPX	2.5	1000	HZ1	'dry'	PAR	-	OL(SC) & low-Al OPX	10	224
O-85	OL	2.5	1000	HZ1	1.45	PAR	vapour	OL(SC)	9	41
O-77	OL	2.5	1050	HZ1	1.45	-	melt	OL(SC)	10	42
P-7	CPX	2.5	1000	HZ1	1.45	-	vapour	low-Al CPX & low-Al OPX	15	978
P-7	OPX	2.5	1000	HZ1	1.45	-	vapour	low-Al CPX & low-Al OPX	10	365

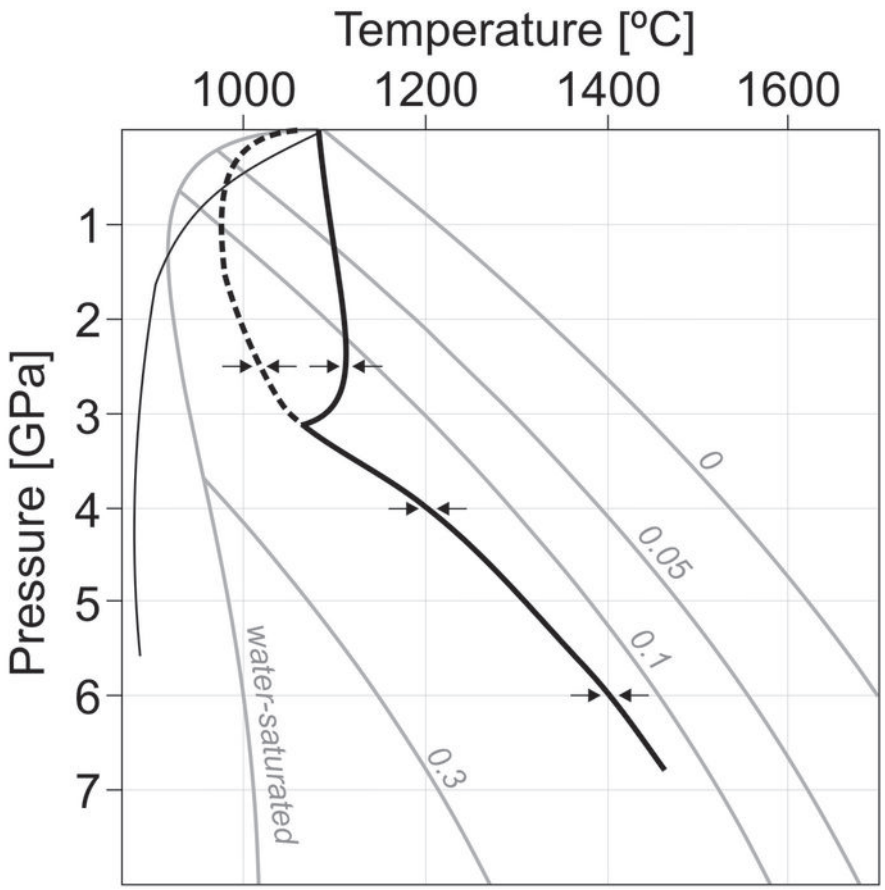

Figure 1 | The experimentally determined vapour-saturated solidus + H_2O (ref. 1) is shown as a heavy black line at P > 3 GPa and a dashed line at P < 3 GPa. The solidus for pargasite lherzolite without excess vapour (dehydration solidus)[1] is shown as a heavy black line below 3 GPa. Arrows indicate where the solidi have been fixed by experiments[1]. The finer black line is the interpreted vapour-saturated solidus of Grove et al.[2,5] and Till et al.[6] and the preliminary results of these studies were used to fix the 'water-saturated' solidus in a model for mantle melting[3] from which solidi for bulk water contents of 0.05, 0.1 and 0.3 wt% have been plotted (grey lines). The published experimental results[1] show that at >3 GPa solidi for 0.05, 0.1 and 0.3 wt% water are coincident as all are vapour-saturated (only the melt fraction immediately above the solidus varies). Below 3 GPa solidi are on or below the dehydration solidus and above or on the vapour-saturated solidus, depending on the bulk water content and the modal abundance of pargasite possible at the particular P, T condition[1].

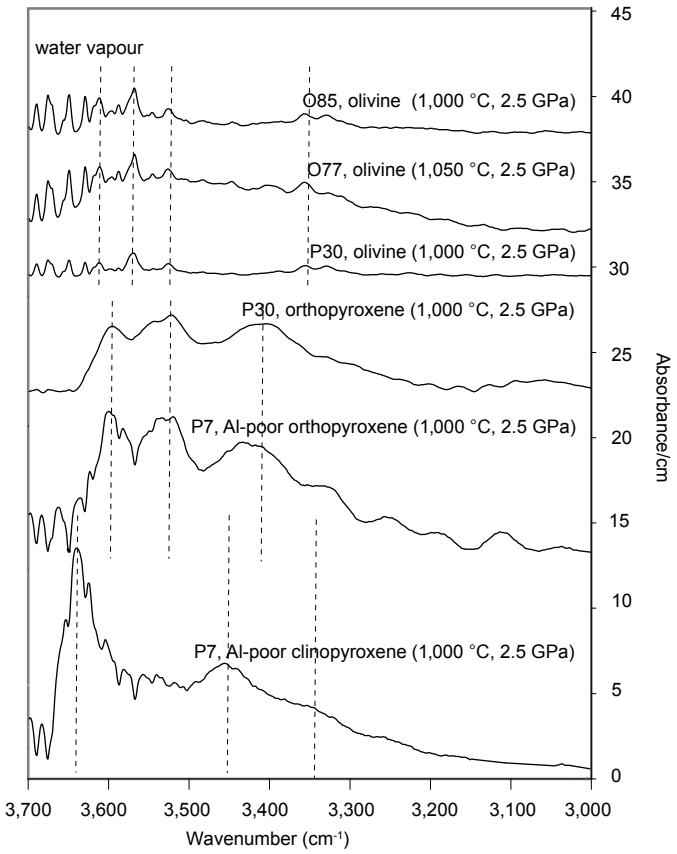

Figure 2 | Examples of FTIR spectra from monomineralic olivine and pyroxene layers. The average unpolarised infrared spectra are normalized to 1 cm of thickness. The spectra from monomineralic layers were used for quantitative estimation of their water content. Calibrations of spectra to derive water content are based on Kovács et al.[8] and Bell et al.[9,10] for total absorbance and appropriate calibration factors respectively. Water vapour bands occurring at above ~3600 cm^{-1} are excluded from quantification. Experiment O-85, HZ1 lherzolite composition with San Carlos olivine layers at 2.5 GPa, 1,000 °C, 1.45 wt.% H_2O; experiment O-77, HZ1 lherzolite composition with San Carlos olivine layers at 2.5 GPa, olivine and low-Al orthopyroxene layers at 2.5 GPa, 1,000 °C, dry (that is, no H_2O added); experiment P-7, HZ1 lherzolite composition with low-Al orthopyroxene and clinopyroxene layers at 2.5 GPa, 1,000 °C, 1.45 wt.% H_2O.