Hermann, J., and Lakey, S., 2021, Water transfer to the deep mantle through hydrous, Al-rich silicates in subduction zones: Geology, v. 49, https://doi.org/10.1130/G48658.1

Supplementary Material

Starting Materials

The starting materials consist of a blend of natural minerals. Chlorite is from a chlorite-rich vein in Cierro di Almirez (Sample Al08-16 from Padron-Navarta et al., 2011) and contains 1.55 wt% Cr_2O_3, 12.94 wt% Al_2O_3 and a Mg# of 94. About 1% of antigorite is present as impurity. Antigorite with 0.31 % wt% Cr_2O_3, 3.04 wt% Al_2O_3 and a Mg# of 92 is from the same locality. Tremolite derives from Val Malenco ultramafic rocks and has 0.72 wt% Al_2O_3 and a Mg# of 90. Seed garnet is a pyrope from Dora-Maira with Mg# of 92. The first starting material used for the peridotite phase relations consists of a blend of 80 wt% chlorite, 8 wt% antigorite, 11.5 wt% tremolite and 0.5 wt% pyrope. The composition is enriched in Al_2O_3 with respect to a peridotite. It was designed to best document reactions 1-3 relevant for peridotites in a single starting composition and to have abundant Al-phases in the reaction products for identification of hydrous Al phases. The second starting material is a chlorite-rich metasomatic rock consisting of 98.5 wt% chlorite, 1 wt.% antigorite and 0.5 wt% pyrope. The composition of the minerals and the starting materials are given below. The mineral blends were ground for 45 minutes in an agate mortar with acetone. The resulting paste was placed in a drying oven over night at 110°C before loading into capsules.

STARTING MIXES

(a) CHLORITE SCHIST

	Oxide	CHL (n=11)		ATG (n=2)		GRT (n=9)	
	wt%	Mean	sd	Mean	sd	Mean	sd
Wt%	Na2O	0.00	0.00	0.00	0.00	0.08	0.06
	MgO	34.02	0.44	37.77	1.40	26.75	0.20
	SiO2	33.78	0.41	42.78	0.19	43.38	0.29
	Al2O3	12.94	0.29	0.16	0.01	24.42	0.22
	NiO	0.19	0.15	0.16	0.16	0.00	0.00
	FeO	3.63	0.14	5.25	0.01	4.08	0.40
	MnO	0.00	0.00	0.08	0.08	0.00	0.00
	Cr2O3	1.55	0.39	0.00	0.00	0.00	0.00
	CaO	0.00	0.00	0.00	0.00	0.43	0.02
	TiO2	0.00	0.00	0.00	0.00	0.00	0.00
	H2O (calc)	12.53	0.13	12.57	0.19	0.00	0.00
	Total	98.64	0.94	98.77	1.54	99.15	0.69
Cations (a.p.f.u.)	Na	0.00	0.00	0.00	0.00	0.00	0.00
	Mg	4.86	0.03	2.68	0.06	2.75	0.03
	Si	3.24	0.01	2.04	0.02	3.00	0.01
	Al	1.46	0.03	0.01	0.00	1.99	0.01
	Ni	0.01	0.01	0.01	0.01	0.00	0.00
	Fe	0.29	0.01	0.21	0.00	0.24	0.02
	Mn	0.00	0.00	0.00	0.00	0.00	0.00
	Cr	0.12	0.03	0.00	0.00	0.00	0.00
	Ca	0.00	0.00	0.00	0.00	0.03	0.00
	Ti	0.00	0.00	0.00	0.00	0.00	0.00
	Total	9.98	0.01	4.95	0.02	8.01	0.00
	Mg#	0.94	0.00	0.93	0.00	0.92	0.01
	Cr#	0.07	0.02	0.00	0.00	0.00	0.00

(b)

Mineral	Proportion in starting material
Chlorite	98.50
Antigorite	1.00
Garnet (seeds)	0.50
Total	100.00

(c)

Oxide	wt%
H2O(calc.)	12.64
MgO	34.49
SiO2	34.38
Al2O3	13.05
NiO	0.19
FeO	3.70
Cr2O3	1.55
Total	100.00

CHLORITE PERIDOTITE

(d)

	Oxide	TRE (n=8)		ATG (n=11)		GRT (n=9)		CHL (n=9)	
		Mean	sd	Mean	sd	Mean	sd	Mean	sd
Wt%	H2O (calc)	2.13	0.02	12.63	0.07	0.00	0.00	12.47	0.10
	Na2O	0.80	0.29	0.00	0.00	0.08	0.06	0.00	0.00
	MgO	21.27	0.35	36.67	0.29	26.75	0.20	33.82	0.30
	SiO2	56.55	0.61	41.85	0.33	43.38	0.29	33.66	0.35
	Al2O3	0.72	0.28	3.04	0.20	24.42	0.22	12.78	0.31
	NiO	0.00	0.00	0.02	0.07	0.00	0.00	0.21	0.16
	FeO	4.39	0.31	3.83	0.10	4.08	0.40	3.72	0.39
	MnO	0.15	0.09	0.02	0.05	0.00	0.00	0.00	0.00
	Cr2O3	0.09	0.12	0.31	0.06	0.00	0.00	1.65	0.38
	CaO	11.69	0.51	0.00	0.00	0.43	0.02	0.00	0.00
	TiO2	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	Total	97.79	0.87	98.37	0.48	99.15	0.69	98.31	0.72
Cations (a.p.f.u.)	Na	0.05	0.02	0.00	0.00	0.00	0.00	0.00	0.00
	Mg	4.47	0.06	2.60	0.01	2.75	0.03	4.85	0.02
	Si	7.97	0.03	1.99	0.01	3.00	0.01	3.24	0.02
	Al	0.12	0.05	0.17	0.01	1.99	0.01	1.45	0.03
	Ni	0.00	0.00	0.00	0.00	0.00	0.00	0.02	0.01
	Fe	0.52	0.04	0.15	0.00	0.24	0.02	0.30	0.03
	Mn	0.02	0.01	0.00	0.00	0.00	0.00	0.00	0.00
	Cr	0.01	0.01	0.01	0.00	0.00	0.00	0.13	0.03
	Ca	1.76	0.08	0.00	0.00	0.03	0.00	0.00	0.00
	Ti	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00
	Total	14.91	0.02	4.92	0.00	8.01	0.00	9.98	0.01
	Mg#	0.90	0.01	0.94	0.00	0.92	0.01	0.94	0.01
	Cr#	0.06	0.07	0.07	0.01	0.00	0.00	0.08	0.02

(e)

Mineral	Proportion in starting material
Chlorite	79.87
Tremolite	11.67
Antigorite	7.96
Garnet (seeds)	0.50
Total	100.00

(f)

Oxide	wt%
Na2O	0.10
MgO	33.12
SiO2	37.70
Al2O3	10.84
NiO	0.17
FeO	3.87
MnO	0.02
Cr2O3	1.38
CaO	1.40
TiO2	0.00
H2O (calc)	11.41
Total	100.00

Experimental techniques

The starting material was loaded in gold capsules of 2.3 mm outer diameter and placed into MgO inserts. These parts were put into a graphite heater, hosted in a low friction assembly consisting of NaCl lined by Teflon foil. The assembly was

then loaded in a pre-stressed Strecon vessel and mounted on a 500T end-loaded press (called ultra-high pressure (UHP) piston cylinder) housed at the Research School of Earth Sciences, The Australian National University. 28.06 mm long pistons of K8 grade tungsten carbide were used. The piston travel and ram pressure in all experiments was continuously monitored for the whole run duration and the ram pressure is adjusted automatically when needed (Hermann et al., 2016). Generally after 24 hours, the piston travel reached a plateau, indicating that friction has dissipated and that the experiment reached steady state. All experiments were left at steady state conditions for 44 to 144 h. Temperature was controlled using type B thermocouples ($Pt_{94}Rh_6/Pt_{70}Rh_{30}$) and is accurate to ±10°C.

Analytical techniques

At the termination of the experiments, the capsules were pierced and fluid loss was determined by weighing after drying the capsule overnight at 110°C. The sample was then exposed and impregnated with epoxy in a bench vacuum chamber. The sample was ground with 1200 dry emery paper and consecutively polished with 6, 3, 1 and ¼ µm grit diamond paste on a lapping machine for a total time between 15-30 minutes. The surface was regularly checked under a reflected light microscope. The sample was then coated with a 12 nm carbon coat for chemical analyses.

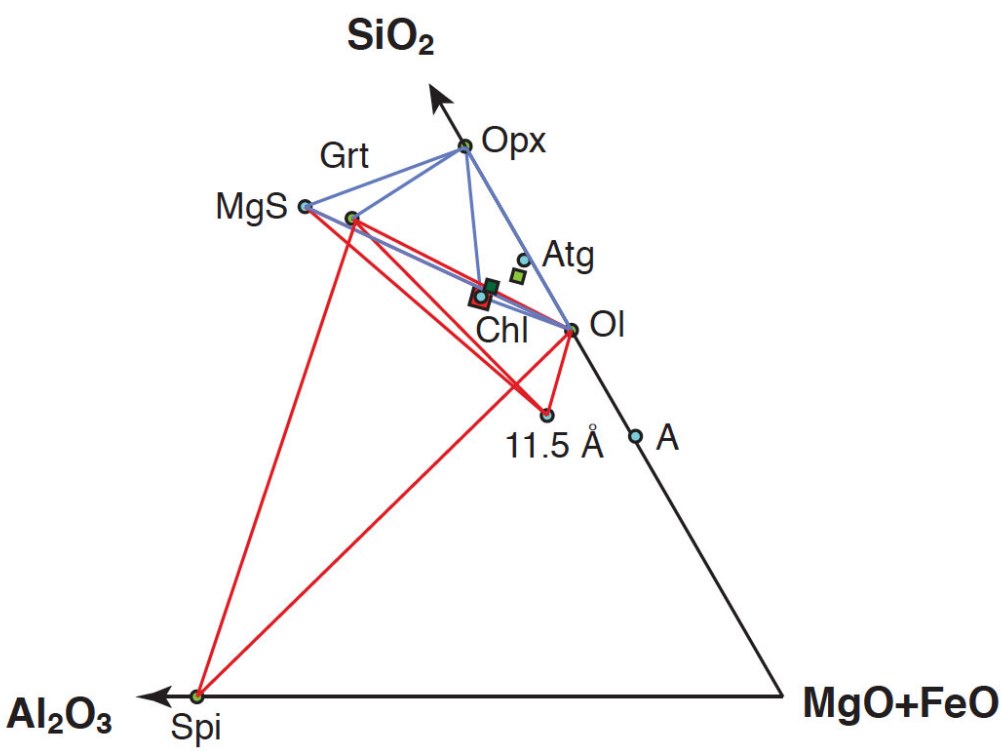
Textural analyses in BSE and SE mode as well as compositional analyses were performed on a Hitachi 4300 SE/N Shottky field emission SEM housed at the Microscopy Unit, The Australian National University. Mineral compositions were determined in EDS mode using an acceleration voltage of 15 kV, a beam current of 0.6 nA and a count time of 90s. A suite of natural minerals and synthetic materials were used for standardisation. Typical detection limits are at 0.2 wt.%. To ascertain the results, some large mineral grains were also measured in WDS mode with a Cameca SX-100 electron microprobe. There was excellent agreement between the two methods and thus the reported analyses (Table S2) of the generally fine-grained run products were done with the higher resolution FE-SEM.

Chemography

Main phases, abbreviations and the simplified stoichiometry in the MgO-Al_2O_3-SiO_2-H_2O (MASH) system relevant for the experiments:

Phase	**Abbreviation**	**Formula**
Antigorite	Atg	$Mg_{5.95}Al_{0.1}Si_{3.95}O_{10}(OH)_8$
Chlorite	Chl	$Mg_{5.25}Al_{1.5}Si_{3.25}O_{10}(OH)_8$
11.5 Å phase	11.5 Å	$Mg_{11}Al_2Si_4O_{16}(OH)_{12}$
Mg-Sursassite	MgS	$Mg_5Al_5Si_6O_{21}(OH)_7$
Phase A	A	$Mg_7Si_2O_8(OH)_6$
Olivine	Ol	Mg_2SiO_4
Orthopyroxene	Opx	$Mg_2Si_2O_6$
Garnet	Grt	$Mg_3Al_2Si_3O_{12}$
Spinel	Spi	$Mg_2Si_2O_4$
Clinopyroxene	Cpx	$CaMgSi_2O_6$

Chemography of these minerals in the MgO-Al_2O_3-SiO_2 composition space, projected from H_2O, together with the bulk rock composition of the starting materials (red diamond for chlorite-rich metasomatic ultramafic rocks, dark green diamond for chlorite peridotite) and a model harzburgite (light green diamond).



References

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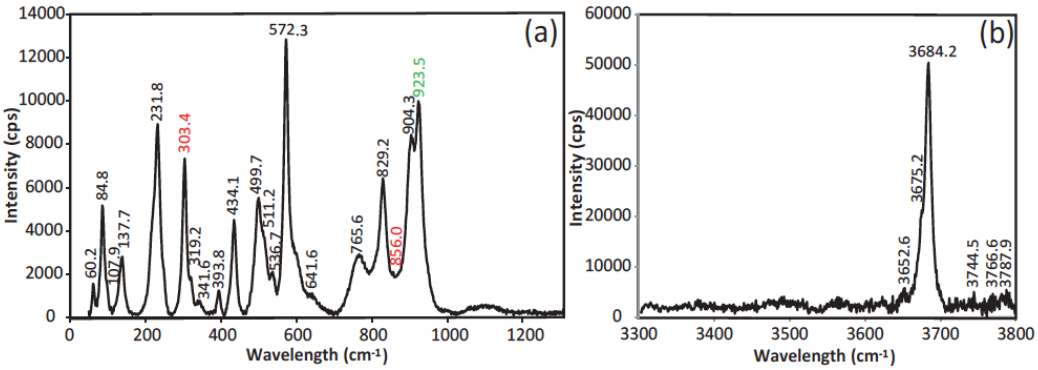
Figure S1:

Raman spectrum of 11.5 Å phase for the main cation-oxygen vibrations (a) and for the O-H vibrations (b). Bands from minor olivine (red) and garnet (green) are present.