# Supplementary information



(**a**)

(**b**)
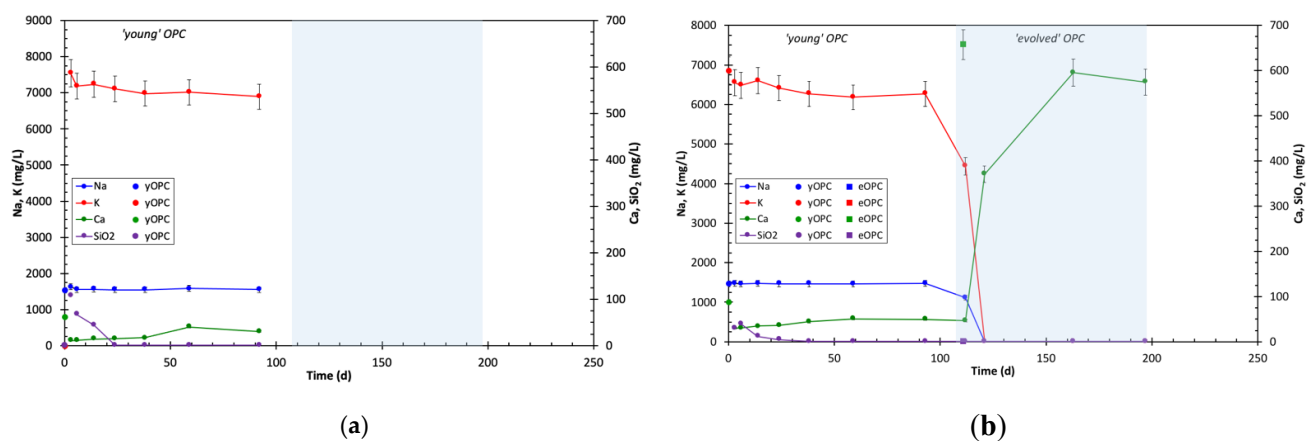
Figure S1 Major changes in fluid chemistry with time. (**a**) Toki Granite with 'young' OPC leachate (Col-1); (**b**) Toki Granite with 'young' OPC, then 'evolved OPC leachate (Col-2). Legend text: yOPC – 'young' OPC leachate; eOPC–'evolved' OPC leachate. Lines indicate concentrations in reacted samples; Single points the original concentration in the reacting fluids. X-axis at same scale as Figure 4 for direct comparison of data.
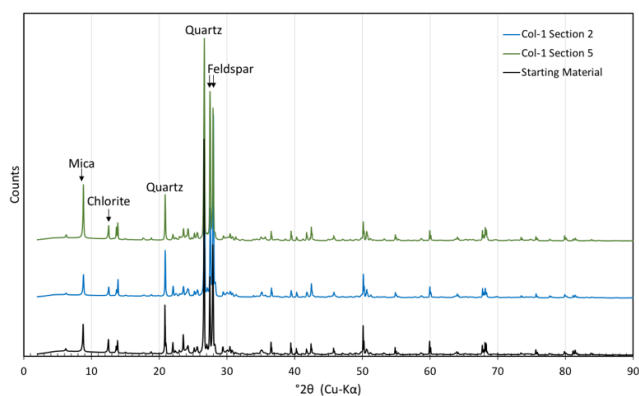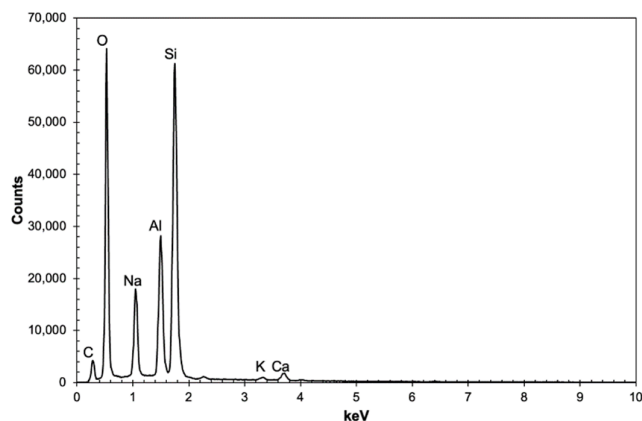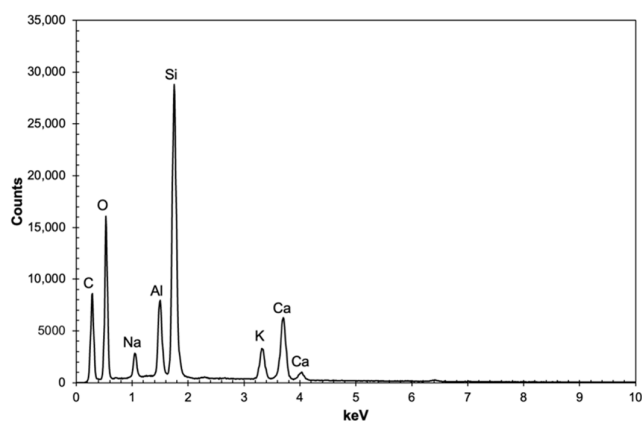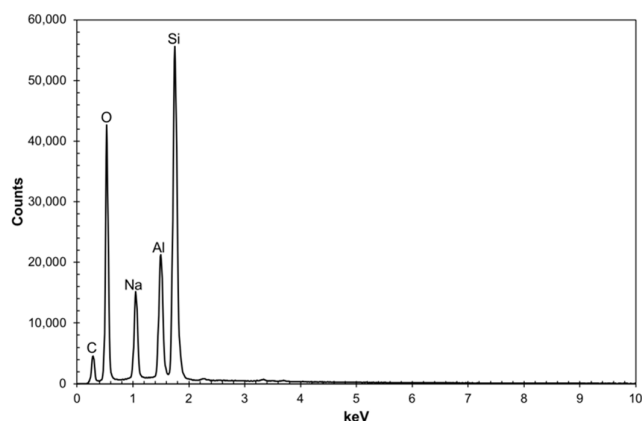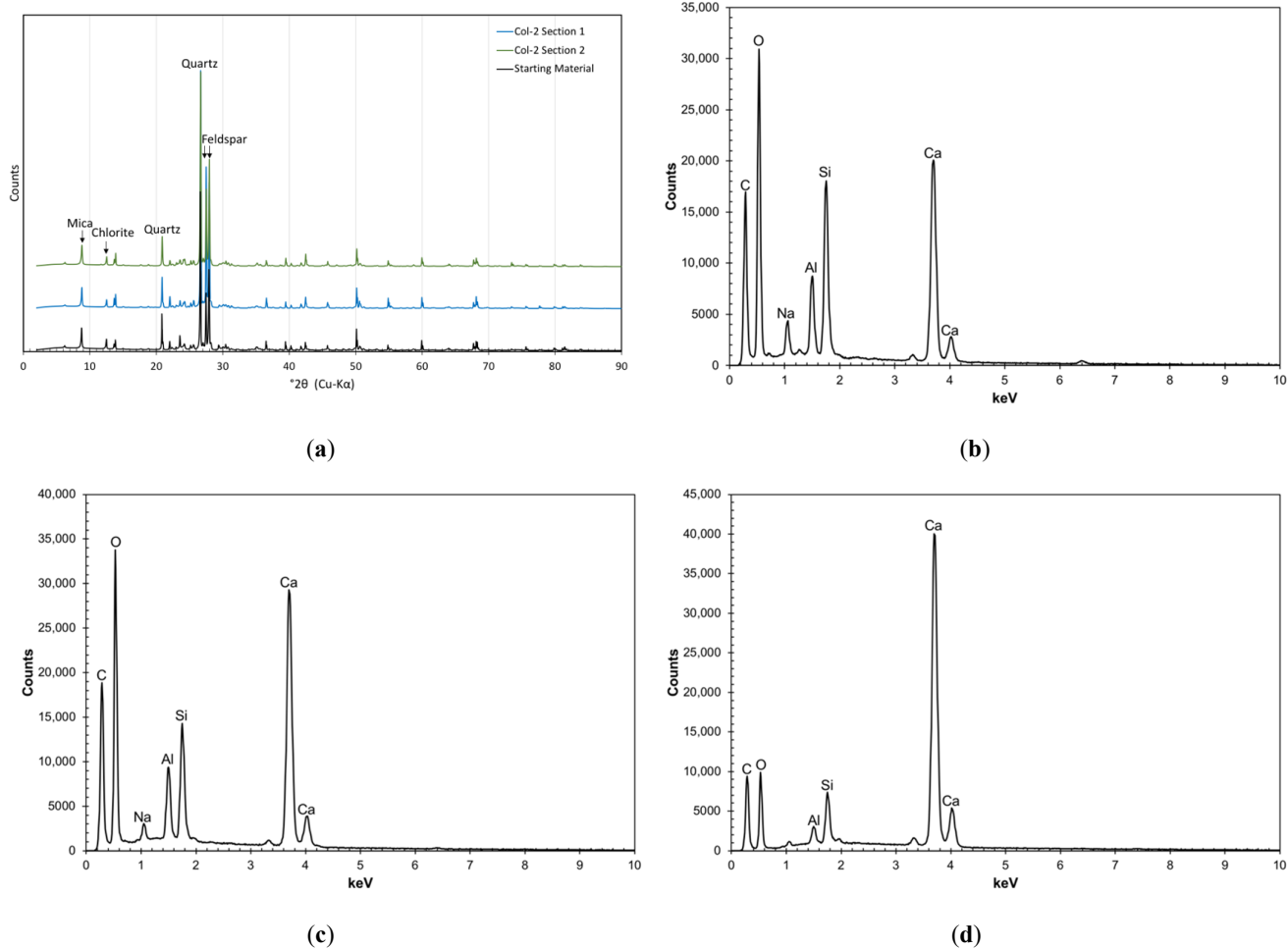
Figure S2 (**a**) X-ray diffractograms (main diffraction lines of key minerals labelled) of unreacted and reacted granite samples experiment with 'young' OPC, (Col-1, Sections 2,5). Showing little difference between unreacted and reacted solids identified by XRD. (**b-d**) SEM-EDS spectra, see Figure 5 for analysis locations (**b**) Section 1; (**c**) Section 5; (**d**) Section 10.
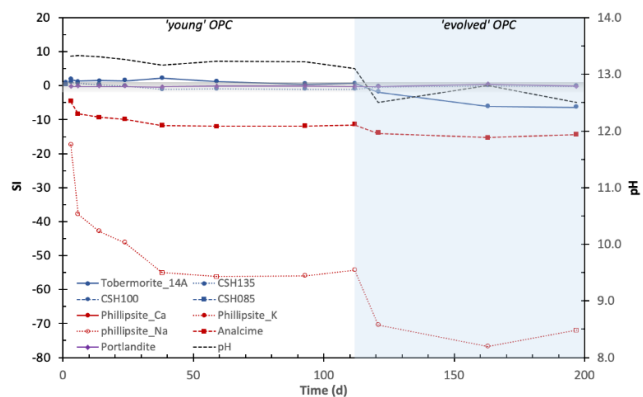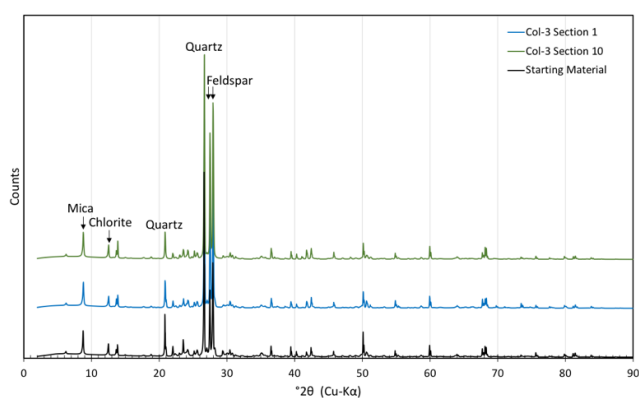
Figure S3 (**a**) X-ray diffractograms (main diffraction lines of key minerals labelled) of unreacted and reacted granite samples exper-
iment with 'young' OPC, then the 'evolved OPC leachate (Col-2, Sections 1,2). Showing little difference between unreacted
and reacted solids identified by XRD. (**b-d**) SEM-EDS spectra, see Figure 7 for analysis locations (**b**) Section 1; (**c**) Section
5; (**d**) Section 10.

Figure S4 Selected primary mineral and C-S-H phase saturation states in reacted fluids, experiment with 'young' OPC, then the
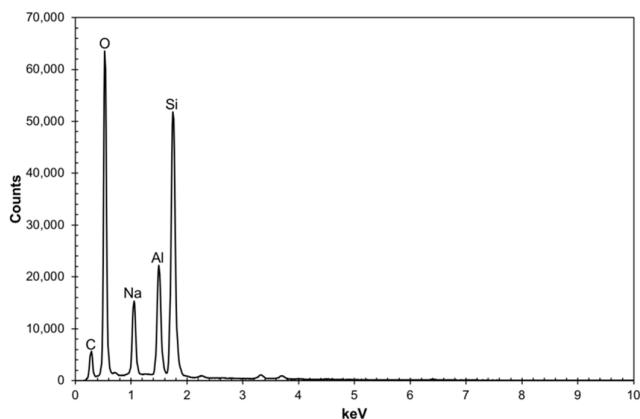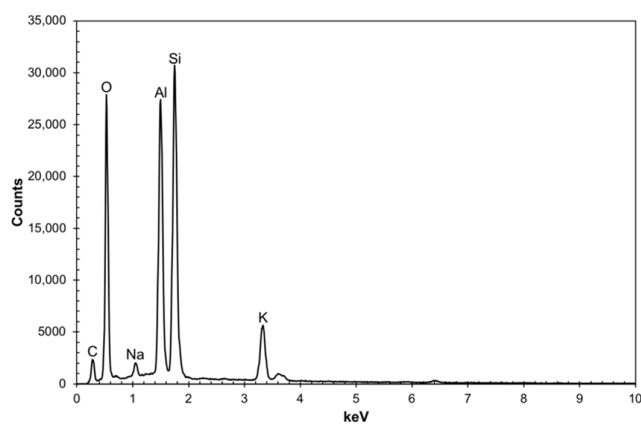'evolved OPC leachate (Col-2).

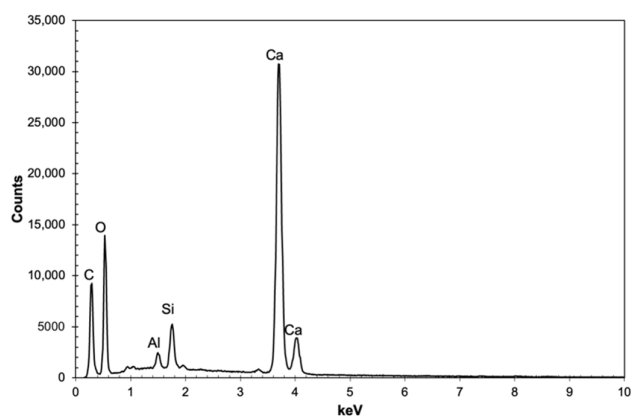**Figure S5** (**a**) X-ray diffractograms (main diffraction lines of key minerals labelled) of unreacted and reacted granite samples experiment with 'young' OPC, then the 'evolved OPC leachate followed by MGW (Col-3, Sections 1,10). Showing little difference between unreacted and reacted solids identified by XRD. (**b-d**) SEM-EDS spectra, see Figure 8 for analysis locations (**b**, **c**) Section 5; (**d**) Section 18.

CABARET model parameters.

In order to create the reacting fluids in situ within the CABARET model three hypothetical mineral assemblages were defined. These were then reacted at appropriate timesteps (0, ~40, and ~80 days) with 'pure' water to produce fluids with aqueous chemistry matching that of the OPC leachates and the Horonobe Groundwater.

Mineral name	Formula	Dissolution reaction	Calculated Log K
A_YoungOPC_calcium	$Ca(OH)_2$	$[Ca(OH)_2] + 2[H^+] \Leftrightarrow [Ca^{++}] + 2[H_2O]$	22.8
A_YoungOPC_chlorine	HCl	$[HCl] \Leftrightarrow [H^+] + [Cl^-]$	-33.4
A_YoungOPC_alminium	$Al(OH)_3$	$[Al(OH)_3] + [H_2O] \Leftrightarrow [Al(OH)_4^-] + [H^+]$	-10.4
A_YoungOPC_carbon	H_2CO_3	$[H_2CO_3] \Leftrightarrow 2[H^+] + [CO_3^-]$	-36.9
A_YoungOPC_potassium	KOH	$[KOH] + [H^+] \Leftrightarrow [K^+] + [H_2O]$	12.4
A_YoungOPC_magnesium	$Mg(OH)_2$	$[Mg(OH)_2] + 2[H^+] \Leftrightarrow [Mg^{++}] + 2[H_2O]$	4.86
A_YoungOPC_sodium	NaOH	$[NaOH] + [H^+] \Leftrightarrow [Na^+] + [H_2O]$	11.9
A_YoungOPC_silicon	$Si(OH)_4$	$[Si(OH)_4] \Leftrightarrow [Si(OH)_{4(aq)}]$	-24.3
A_YoungOPC_water	H_2O	$[H_2O] \Leftrightarrow [H_2O]$	-0.002
A_YoungOPC_sulfur	H_2SO_4	$[H_2SO_4] \Leftrightarrow 2[H^+] + [HSO_4^-]$	-47.3

Table S1 Calculated Log $K_{(297.15 K)}$ for the hypothetical minerals; 'young' OPC leachate.

Mineral name	Formula	Dissolution reaction	Calculated Log K
A_OldOPC_calcium	$Ca(OH)_2$	$[Ca(OH)_2] + 2[H^+] \Leftrightarrow [Ca^{++}] + 2[H_2O]$	22.9
A_OldOPC_chlorine	HCl	$[HCl] \Leftrightarrow [H^+] + [Cl^-]$	-32.7
A_OldOPC_alminium	$Al(OH)_3$	$[Al(OH)_3] + [H_2O] \Leftrightarrow [Al(OH)_4^-] + [H^+]$	-9.74
A_OldOPC_carbon	H_2CO_3	$[H_2CO_3] \Leftrightarrow 2[H^+] + [CO_3^-]$	-35.9
A_OldOPC_potassium	KOH	$[KOH] + [H^+] \Leftrightarrow [K^+] + [H_2O]$	-7.60
A_OldOPC_magnesium	$Mg(OH)_2$	$[Mg(OH)_2] + 2[H^+] \Leftrightarrow [Mg^{++}] + 2[H_2O]$	4.05
A_OldOPC_sodium	NaOH	$[NaOH] + [H^+] \Leftrightarrow [Na^+] + [H_2O]$	-7.60
A_OldOPC_silicon	$Si(OH)_4$	$[Si(OH)_4] \Leftrightarrow [Si(OH)_{4(aq)}]$	-23.7
A_OldOPC_water	H_2O	$[H_2O] \Leftrightarrow [H_2O]$	-0.00068
A_OldOPC_sulfur	H_2SO_4	$[H_2SO_4] \Leftrightarrow 2[H^+] + [HSO_4^-]$	-45.6

Table S2 Calculated Log $K_{(297.15 K)}$ for the hypothetical minerals; 'evolved' OPC leachate.

Mineral name	Formula	Dissolution reaction	Calculated Log K
A_Mizu_calcium	$Ca(OH)_2$	$[Ca(OH)_2] + 2[H^+] \Leftrightarrow [Ca^{++}] + 2[H_2O]$	13.4
A_Mizu_chlorine	HCl	$[HCl] \Leftrightarrow [H^+] + [Cl^-]$	-11.3
A_Mizu_alminium	$Al(OH)_3$	$[Al(OH)_3] + [H_2O] \Leftrightarrow [Al(OH)_4^-] + [H^+]$	7.90
A_Mizu_carbon	H_2CO_3	$[H_2CO_3] \Leftrightarrow 2[H^+] + [CO_3^-]$	-11.7
A_Mizu_potassium	KOH	$[KOH] + [H^+] \Leftrightarrow [K^+] + [H_2O]$	3.58
A_Mizu_magnesium	$Mg(OH)_2$	$[Mg(OH)_2] + 2[H^+] \Leftrightarrow [Mg^{++}] + 2[H_2O]$	11.7
A_Mizu_sodium	NaOH	$[NaOH] + [H^+] \Leftrightarrow [Na^+] + [H_2O]$	6.11
A_Mizu_silicon	$Si(OH)_4$	$[Si(OH)_4] \Leftrightarrow [Si(OH)_{4(aq)}]$	-3.30
A_Mizu_water	H_2O	$[H_2O] \Leftrightarrow [H_2O]$	-0.0000869
A_Mizu_sulfur	H_2SO_4	$[H_2SO_4] \Leftrightarrow 2[H^+] + [HSO_4^-]$	-21.2

Table S3 Calculated Log $K_{(297.15 K)}$ for the hypothetical minerals; Mizunami groundwater.

Aqueous Basis species	Aqueous Complex species		
H_2O	$Al(OH)_2^+$	CO_3^{--}	$MgSiO(OH)_3^+$
	$Al(OH)_{3(aq)}$	$KCl(aq)$	$NaAl(OH)_{4(aq)}$
Al^{+++}	$Al(OH)_4^-$	KCO_3^-	$NaCl_{(aq)}$
Ca^{++}	$CaAl(OH)_4^+$	$KHCO_3(aq)$	$NaCO_3^-$
Cl^-	$CaCl^+$	$KOH(aq)$	$NaHCO_{3(aq)}$
H^+	$CaCl_{2(aq)}$	$KSiO_2(OH)_2^-$	$NaOH_{(aq)}$
HCO_3^-	$CaCO_{3(aq)}$	$KSiO(OH)_{3(aq)}$	$NaSiO_2(OH)_2^-$
K^+	$CaHCO_3^+$	$MgAl(OH)_4^-$	$NaSiO(OH)_{3(aq)}$
Mg^{++}	$CaOH^+$	$MgCO_{3(aq)}$	OH^-
Na^+	$CaSiO_2(OH)_{2(aq)}$	$MgHCO_3^+$	$SiO(OH)_3^-$
$Si(OH)_{4(aq)}$	$CaSiO(OH)_3^+$	$MgOH^+$	$SiO_2(OH)_2^{--}$
SO_4^{--}	$CO_{2(aq)}$	$MgSiO_2(OH)_{2(aq)}$	

Table S4 Details of the dissolved chemical species included in the CABARET reactive transport model.