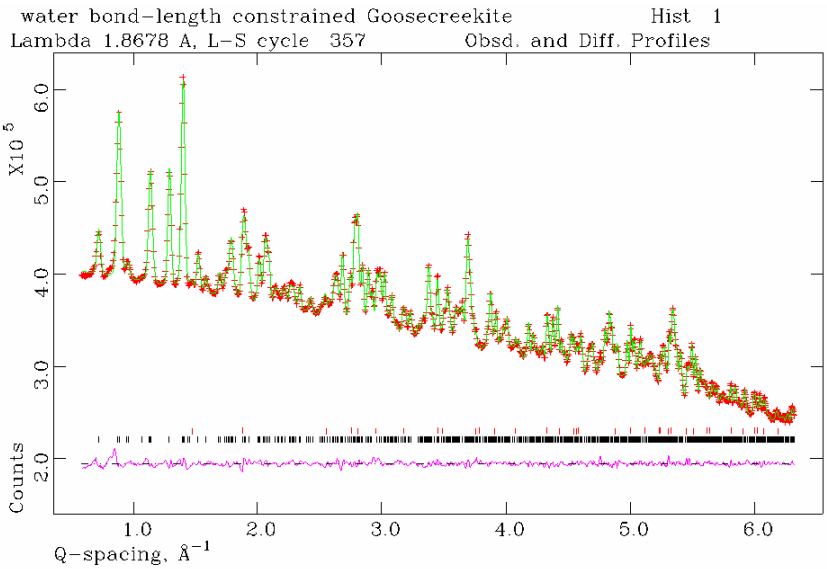
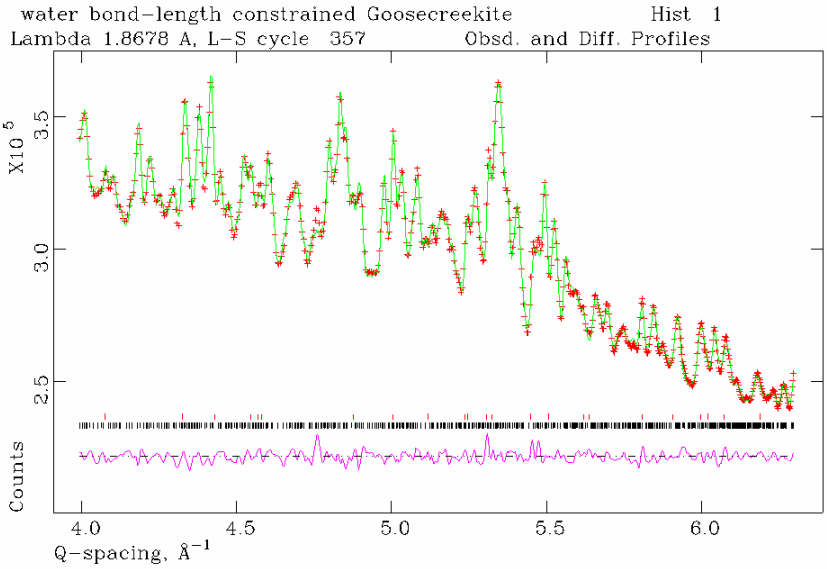
Electronic Supplementary Information

1. Constrained bond-length model Rietveld



Supplementary Figure 1 (Manuscript Figure 1) Final Rietveld refinement profile for goosecreekite for the bond length constrained model. The observed data are crosses, the calculated pattern a solid line, the tick marks show the allowed reflections (upper quartz, lower goosecreekite) and the lower solid line is the difference plot; R_{wp} = 6.6%, R_p = 5.0% and $R_{F^{**}2}$ = 3.01% for 1089 observations.

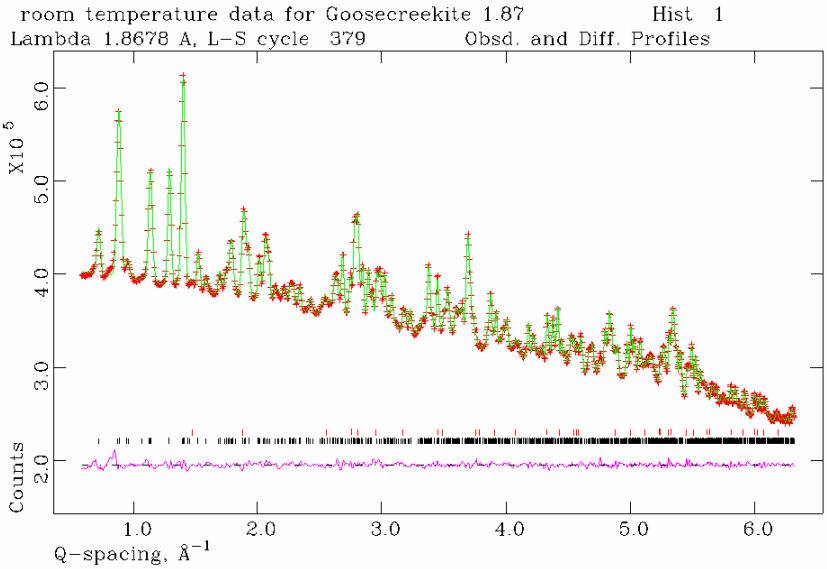


Supplementary Figure 2 High Q region of the Rietveld fit shown in SF1.

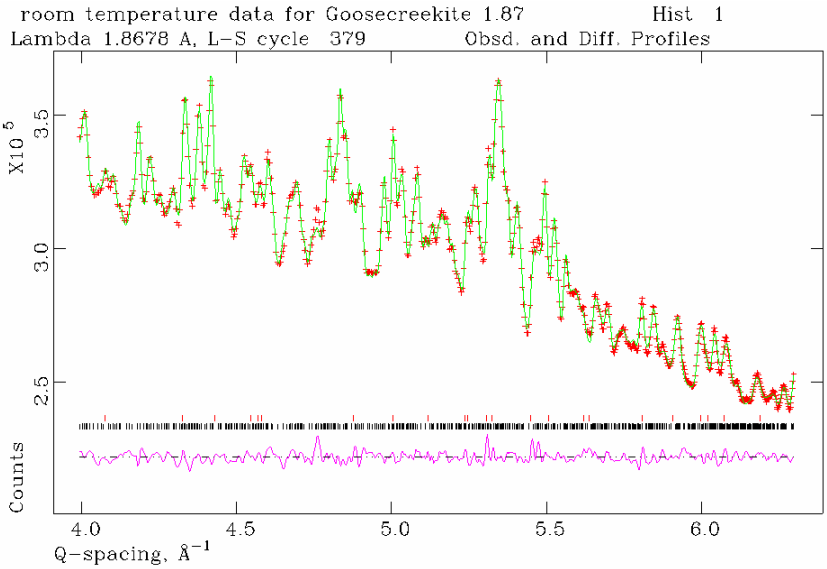
Supplementary Table 1 Atomic coordinates for the constrained water bond-length model for goosecreekite. Space group P 2_1. a = 7.43605(22)Å, b = 17.3988(5)Å, c = 7.29555(21)Å, β = 105.594(3)°

Atom	X	Y	Z	Ui/Ue*100 / Å^2	Occupancy
SI1	0.3328(9)	0.4735(4)	0.6296(10)	1.27(13)	1.0
SI2	0.3167(10)	0.1309(4)	0.6046(11)	1.27(13)	1.0
SI3	0.0227(9)	0.2616(4)	0.5187(10)	1.27(13)	1.0
SI4	0.1027(9)	0.0575(4)	0.8613(9)	1.27(13)	1.0
SI5	0.0947(10)	0.3835(4)	0.8348(10)	1.27(13)	1.0
SI6	0.2401(10)	0.0021(4)	0.2777(10)	1.27(13)	1.0
AL7	0.7485(10)	0.1302(4)	0.5913(10)	2.38(30)	1.0
AL8	0.0675(10)	0.3989(4)	0.2627(10)	2.38(30)	1.0
O9	0.7185(14)	0.0639(4)	0.4099(12)	1.03(4)	1.0
O10	0.5377(10)	0.1491(5)	0.6384(14)	1.03(4)	1.0
O11	0.8379(11)	0.2097(5)	0.5051(13)	1.03(4)	1.0
O12	0.9021(10)	0.1014(5)	0.8015(12)	1.03(4)	1.0
O13	0.2574(12)	0.4402(5)	0.8035(12)	1.03(4)	1.0
O14	0.2644(11)	0.4186(4)	0.4434(12)	1.03(4)	1.0
O15	0.5580(9)	0.4626(5)	0.7056(12)	1.03(4)	1.0
O16	0.1957(10)	0.2057(5)	0.5091(13)	1.03(4)	1.0
O17	0.2667(13)	0.1037(5)	0.7983(13)	1.03(4)	1.0
O18	0.2563(12)	0.0589(5)	0.4583(11)	1.03(4)	1.0
O19	0.0814(15)	0.3007(5)	0.7290(11)	1.03(4)	1.0
O20	0.9692(12)	0.3187(5)	0.3350(14)	1.03(4)	1.0
O21	0.9009(12)	0.4703(4)	0.2210(13)	1.03(4)	1.0
O22	0.1730(12)	0.0580(5)	0.0929(9)	1.03(4)	1.0
O23	0.1237(13)	0.3681(5)	0.0606(9)	1.03(4)	1.0
O24	0.9043(11)	0.4307(5)	0.7398(14)	1.03(4)	1.0
CA25	0.6593(18)	0.2717(9)	0.1951(18)	2.49(33)	1.0
OW1	0.3486(17)	0.2214(7)	0.0894(17)	5.06(29)	1.0
H1W1	0.2344(22)	0.2542(11)	0.0645(29)	5.06(29)	1.0
H2W1	0.3340(36)	0.1815(9)	-0.0117(23)	5.06(29)	1.0
OW2	0.6344(20)	0.2977(6)	0.8788(15)	5.29(33)	1.0
H1W2	0.3640(33)	0.8428(9)	0.2056(24)	5.29(33)	1.0
H2W2	0.6202(29)	0.2630(11)	0.7667(23)	5.29(33)	1.0
OW3	0.7582(17)	0.1517(6)	0.1028(14)	2.69(22)	1.0
H1W3	0.7977(26)	0.1303(9)	-0.0071(20)	2.69(22)	1.0
H2W3	0.7628(24)	0.1073(8)	0.1908(20)	2.69(22)	1.0
OW4	0.5685(16)	0.4013(6)	0.1625(19)	5.17(28)	1.0
H1W4	0.6767(24)	0.4376(10)	0.1989(34)	5.17(28)	1.0
H2W4	0.4580(22)	0.4241(11)	0.0689(24)	5.17(28)	1.0
OW5	0.5157(15)	0.3012(6)	0.4597(19)	4.72(27)	1.0
H1W5	0.4534(27)	0.3527(8)	0.4432(28)	4.72(27)	1.0
H2W5	0.4300(23)	0.2605(9)	0.4839(29)	4.72(27)	1.0

2. Unconstrained bond-length model Rietveld



Supplementary Figure 3 Final Rietveld refinement profile for goosecreekite for the unconstrained bond length model. The observed data are crosses, the calculated pattern a solid line, the tick marks show the allowed reflections (upper quartz, lower goosecreekite) and the lower solid line is the difference plot; R_{wp} = 6.4%, R_p = 4.8% and $R_{F^{**}2}$ = 2.93% for 1091 observations.



Supplementary Figure 4 High Q region of the Rietveld fit.

Supplementary Table 2 Atomic coordinates for the unconstrained water bond-length model for goosecreekite. Space group P 2_1. a = 7.43680(31) Å, b = 17.3957(7) Å, c = 7.29571(30) Å, β = 105.591(3) °.

Atom	x	y	z	Ui/Ue*100 / Å^2	Occupancy
SI1	0.3384(13)	0.4753(5)	0.6319(13)	1.49(20)	1.0
SI2	0.3167(14)	0.1315(5)	0.6126(13)	1.49(20)	1.0
SI3	0.0239(13)	0.2627(6)	0.5155(14)	1.49(20)	1.0
SI4	0.1011(13)	0.0591(6)	0.8721(13)	1.49(20)	1.0
SI5	0.1048(14)	0.3818(5)	0.8327(13)	1.49(20)	1.0
SI6	0.2350(13)	0.0017(5)	0.2842(14)	1.49(20)	1.0
AL7	0.7450(15)	0.1322(6)	0.5908(14)	2.6(4)	1.0
AL8	0.0657(13)	0.4014(5)	0.2610(14)	2.6(4)	1.0
O9	0.7136(18)	0.0657(6)	0.4094(16)	0.84(6)	1.0
O10	0.5366(14)	0.1525(7)	0.6424(19)	0.84(6)	1.0
O11	0.8414(16)	0.2094(6)	0.5032(17)	0.84(6)	1.0
O12	0.8980(14)	0.1007(7)	0.7987(17)	0.84(6)	1.0
O13	0.2517(16)	0.4469(7)	0.8032(16)	0.84(6)	1.0
O14	0.2614(16)	0.4225(6)	0.4425(15)	0.84(6)	1.0
O15	0.5637(12)	0.4613(8)	0.7006(16)	0.84(6)	1.0
O16	0.1946(13)	0.2049(7)	0.5093(18)	0.84(6)	1.0
O17	0.2707(16)	0.1002(6)	0.8057(16)	0.84(6)	1.0
O18	0.2564(15)	0.0606(7)	0.4624(14)	0.84(6)	1.0
O19	0.0719(21)	0.2983(6)	0.7293(15)	0.84(6)	1.0
O20	0.9711(16)	0.3199(6)	0.3322(20)	0.84(6)	1.0
O21	0.8977(16)	0.4724(5)	0.2133(18)	0.84(6)	1.0
O22	0.1726(16)	0.0605(7)	0.1041(13)	0.84(6)	1.0
O23	0.1238(18)	0.3707(7)	0.0592(12)	0.84(6)	1.0
O24	0.9178(16)	0.4326(7)	0.7456(18)	0.84(6)	1.0
CA25	0.6648(32)	0.2691(15)	0.1867(32)	5.2(6)	1.0
OW1	0.3414(29)	0.2250(11)	0.0797(26)	3.45(34)	1.0
H1W1	0.2465(42)	0.2481(15)	0.0724(38)	3.45(34)	1.0
H2W1	0.3147(43)	0.1807(17)	0.0091(41)	3.45(34)	1.0
OW2	0.6281(26)	0.2983(13)	0.8635(26)	4.15(41)	1.0
H1W2	0.3737(48)	0.8445(19)	0.1863(41)	4.15(41)	1.0
H2W2	0.6296(37)	0.2577(17)	0.7723(44)	4.15(41)	1.0
OW3	0.7551(23)	0.1517(9)	0.0996(24)	2.09(29)	1.0
H1W3	0.7768(35)	0.1312(14)	0.0114(39)	2.09(29)	1.0
H2W3	0.7769(31)	0.1036(15)	0.1887(34)	2.09(29)	1.0
OW4	0.5599(30)	0.4033(10)	0.1568(28)	4.10(36)	1.0
H1W4	0.6762(38)	0.4419(15)	0.2028(42)	4.10(36)	1.0
H2W4	0.4714(41)	0.4112(16)	0.0687(39)	4.10(36)	1.0
OW5	0.5151(31)	0.2990(12)	0.4526(29)	6.06(46)	1.0
H1W5	0.4497(47)	0.3538(17)	0.4532(44)	6.06(46)	1.0
H2W5	0.4126(40)	0.2619(17)	0.4921(44)	6.06(46)	1.0

Supplementary Table 3 Relevant bond distances in unconstrained goosecreekite illustrating the calcium coordination sphere and the H-bonding to the framework.

Bond	Distance / Å	Bond	Distance / Å
Ca-O11	2.544(14)	O17-H2W1	2.015(26)
Ca-O20	2.401(14)	O15-H1W2	2.141(27)
Ca-OW1	2.474(20)	O10-H2W2	2.296(26)
Ca-OW2	2.307(18)	H2W2-OW5	-#
Ca-OW3	2.451(17)	O12-H1W3	1.967(23)
Ca-OW4	2.327(20)	O9-H2W3	1.878(21)
Ca-OW5	2.497(18)	O21-H1W4	1.679(25)
Ave Si-O*	1.625(4)	O13-H2W4	2.146(23)
Ave Al-O*	1.725(3)	O14-H1W5	1.796(26)
Ave O-H*	See Table 4	O16-H2W5	2.077(26)
O23-H1W1	2.248(26)	O10-H2W5	2.271(25)

*constrained bond lengths.
#No value is recorded for this bond length as it is now greater than 2.3 Å.

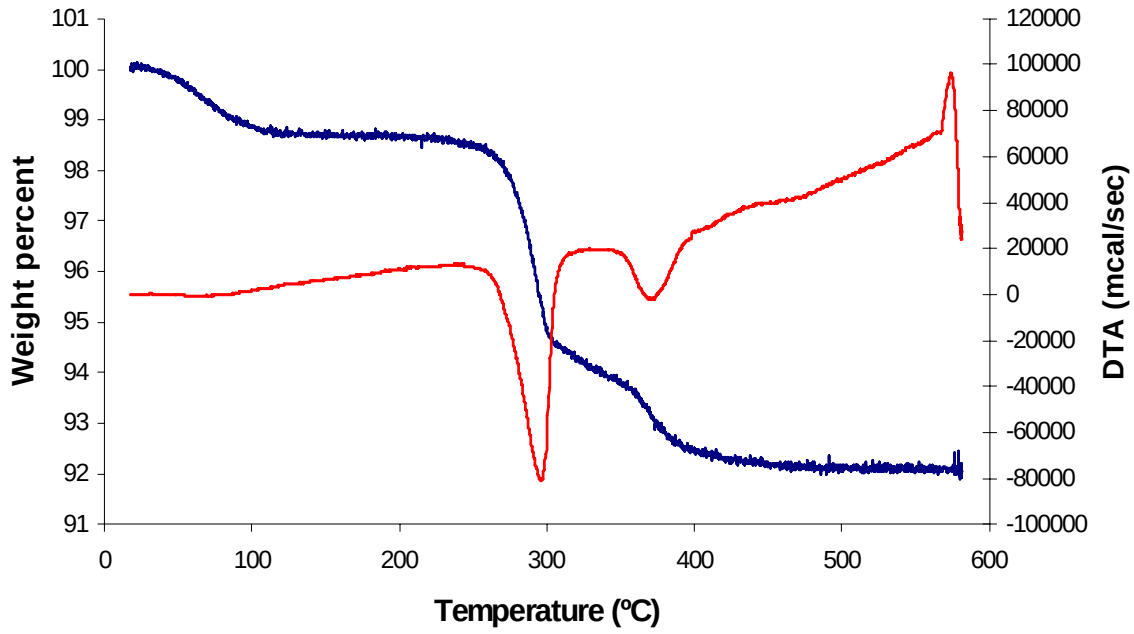
Supplementary Table 4 Relevant bond angles in the water molecules in the unconstrained model of goosecreekite.

Bond	Angle / °	Bond	Angle / °
H1W1-OW1-H2W1	117.3(20)	H1W4-OW4-H2W4	114.3(21)
H1W2-OW2-H2W2	90.7(18)	H1W5-OW5-H2W5	112.0(21)
H1W3-OW3-H2W3	109.4(16)		

Supplementary Table 5 Water bond lengths in the unconstrained model of goosecreekite.

Bond	Distance / Å	Bond	Distance / Å
H1W1-OW1	0.854(33)	H2W1-OW1	0.819(33)
H1W2-OW2	1.105(32)	H2W2-OW2	0.879(33)
H1W3-OW3	0.834(25)	H2W3-OW3	0.981(25)
H1W4-OW4	1.089(30)	H2W4-OW4	0.917(33)
H1W5-OW5	1.000(32)	H2W5-OW5	0.967(32)

In order to model the background for the refinements, a manual background was chosen and left fixed, fitted with 6 terms of a type 1 (shifted Chebyschev) background profile until the final cycles of the refinement when it was allowed to vary normally. The reason for this was to minimise the correlation between the ADPs and the background at higher values of Q where peak overlap becomes more significant.



Supplementary Figure 5 TGA/DTA plot of the dehydration of Goosecreekite, performed on a 15mg sample, referenced to an alumina standard. The heating rate used was 10 °C / min.