Supplementary Information for the manuscript

Natrolite is not a "soda-stone" anymore: Structural study of alkali (Li^{+}), alkaline-earth (Ca^{2+}, Sr^{2+}, Ba^{2+}) and heavy metal (Cd^{2+}, Pb^{2+}, Ag^{+}) cation-exchanged natrolites

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Supplementary Table1. EDS chemical composition of the fully cation-exchanged natrolites.[a]

	Elements	Atomic percent (%)[c]					Composition
		1	2	3	4	5	
Ca-NAT	Ca	5.63	5.93	4.45	5.69	6.64	$Ca_{7.7(1)}Al_{16}Si_{24}O_{80}\cdot xH_2O$
	K	0.00	0.00	0.00	0.00	0.00	
	Al	11.94	12.01	11.13	12.43	12.43	
Sr-NAT	Sr	5.93	5.62	6.36	5.96	5.76	$Sr_{7.6(1)}Al_{16}Si_{24}O_{80}\cdot xH_2O$
	K	0.00	0.00	0.00	0.00	0.00	
	Al	12.41	12.49	12.95	11.96	11.37	
Ba-NAT	Ba	5.74	6.26	6.17	6.48	6.74	$Ba_{7.9(1)}Al_{16}Si_{24}O_{80}\cdot xH_2O$
	K	0.00	0.00	0.00	0.00	0.00	
	Al	12.59	13.08	13.3	12.87	12.07	
Ag-NAT	Ag	12.78	12.31	12.32	12.32	13.05	$Ag_{16.4(1)}Al_{16}Si_{24}O_{80}\cdot xH_2O$
	K	0.00	0.00	0.00	0.00	0.00	
	Al	12.20	12.24	12.3	12.64	11.7	
Cd-NAT	Cd	6.39	6.19	6.38	6.3	6.52	$Cd_{8.2(1)}Al_{16}Si_{24}O_{80}\cdot xH_2O$
	K	0.00	0.00	0.00	0.08	0.14	
	Al	12.07	12.9	12.42	11.99	12.72	
Pb-NAT	Pb	6.63	6.11	6.58	6.28	6.24	$Pb_{8.2(1)}Al_{16}Si_{24}O_{80}\cdot xH_2O$
	K	0.00	0.00	0.00	0.00	0.01	
	Al	12.74	11.47	12.54	12.85	12.24	
Li-NAT	K	0.00	0.07	0.00	0.00	0.07	$Li_{15.97(1)}K_{0.03(1)}Al_{16}Si_{24}O_{80}\cdot xH_2O$[b]
	Al	13.57	14.39	13.41	12.97	12.54	

[a]Values are normalized based on 16 aluminum atoms per unit cell.
[b]Lithium contents are estimated based on the measured potassium contents.
[c]Instrumental detection limit down to the third decimal point.

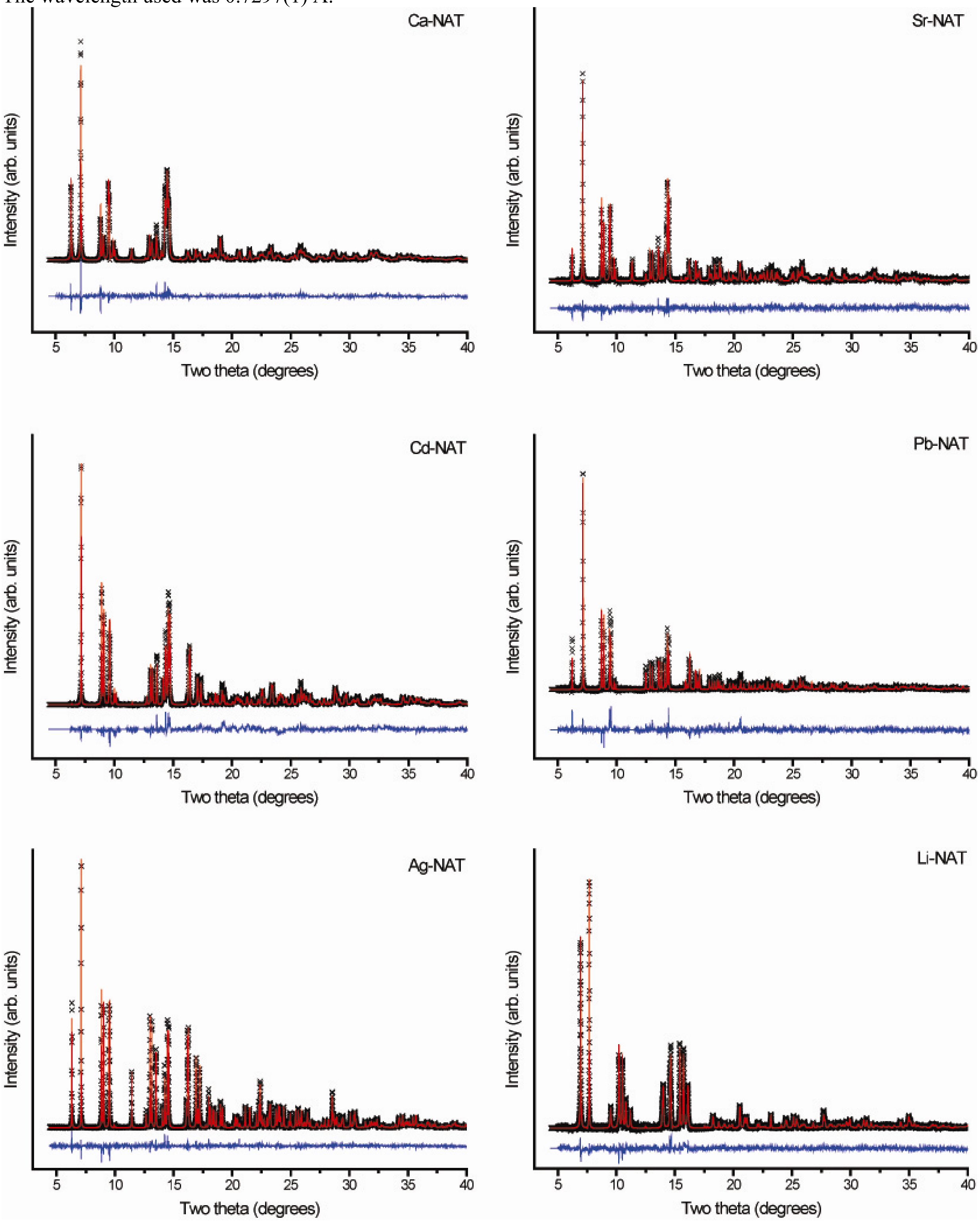
Supplementary Table2. Strcutural comparison between the Ca-NAT model and that of natural scolecite by Comodi et al. (2002).

Scolecite			
a	6.533(2)		
b	19.030(3)		
c	9.830(3)		
alpha	90		
beta	109.95(3)		
gamma	90		
volume	1148.76(4)		
	x	y	z
Ca	0.1613(1)	0.14323(2)	0.0521(1)
Si1	0.5	0.37057(3)	0
Si2	0.2298(1)	0.33184(3)	0.2001(1)
Si3	0.5400(1)	0.08257(3)	0.3312(1)
Al1	0.9344(1)	0.46209(3)	0.0998(1)
Al2	0.3555(1)	0.21662(3)	0.4338(1)
O1	0.5420(3)	0.0315(1)	0.4608(2)
O2	0.4472(3)	0.0460(1)	0.1744(2)
O3	0.3836(3)	0.1513(1)	0.3156(2)
O4	0.1154(3)	0.1998(1)	0.4681(2)
O5	0.3535(3)	0.2994(1)	0.3852(2)
O6	0.0868(3)	0.2712(1)	0.0905(2)
O7	0.4143(3)	0.3587(1)	0.1345(2)
O8	0.0767(3)	0.3956(1)	0.2148(2)
O9	0.7894(3)	0.1101(1)	0.3565(2)
O10	0.6602(3)	0.4369(1)	0.0342(2)
Wat1	0.8908(4)	0.0803(1)	0.1083(3)
Wat2	0.9085(5)	0.326(1)	0.4404(4)
Wat3	0.5782(4)	0.4454(1)	0.3742(2)

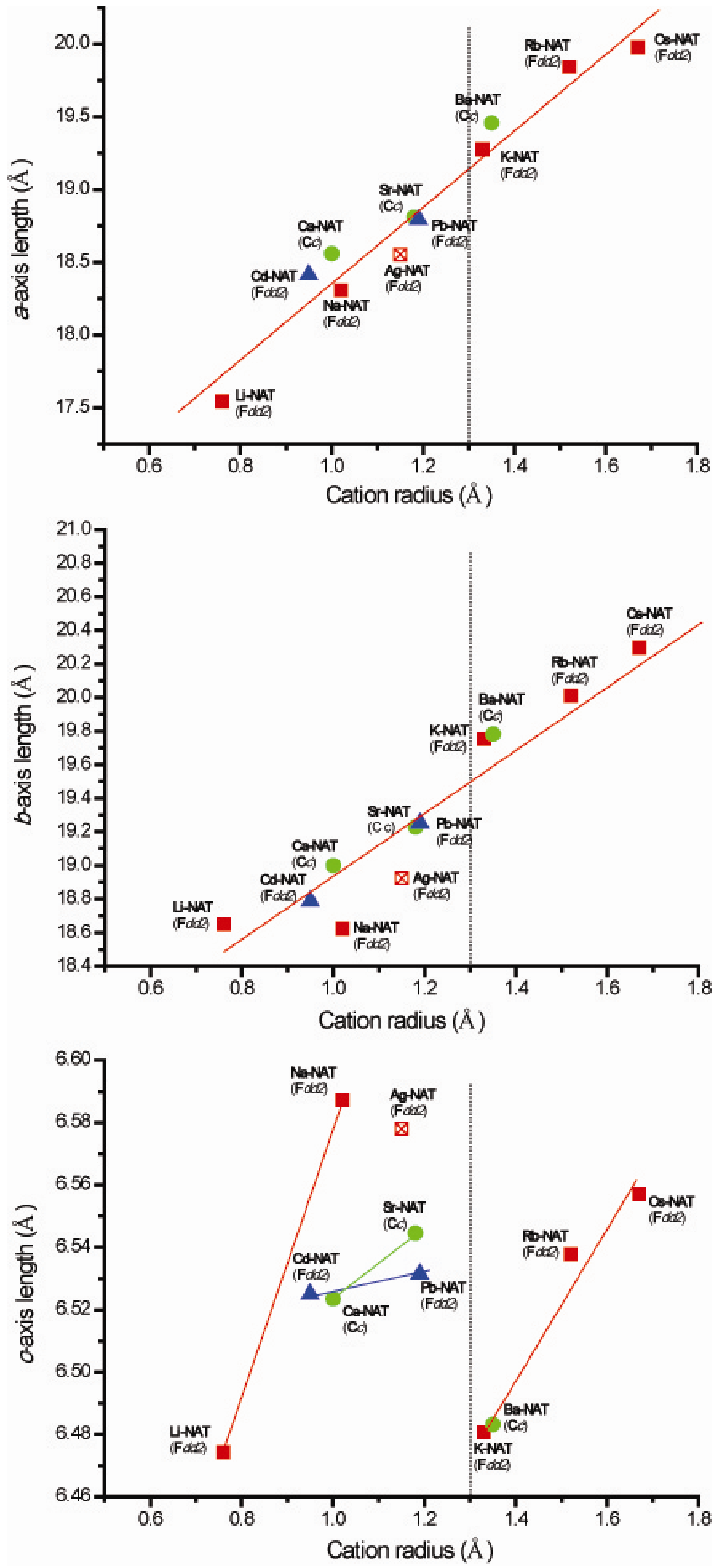
Ca-NAT			
a	6.5242(3)		
b	18.9885(7)		
c	9.8466(5)		
alpha	90		
beta	109.70(1)		
gamma	90		
volume	1148.4(1)		
	x	y	z
Ca	0.157(1)	0.1412(3)	0.0562(7)
Si1	0.503(1)	0.3690(4)	0.0001(8)
Si2	0.229(1)	0.3335(3)	0.1997(7)
Si3	0.549(1)	0.0826(4)	0.3337(7)
Al1	0.933(1)	0.4628(4)	0.0989(7)
Al2	0.356(1)	0.2186(4)	0.4391(7)
O1	0.544(2)	0.0276(5)	0.459(1)
O2	0.447(2)	0.0453(5)	0.1776(9)
O3	0.401(2)	0.1520(5)	0.330(1)
O4	0.116(2)	0.2038(5)	0.477(1)
O5	0.342(2)	0.3008(5)	0.3595(9)
O6	0.085(1)	0.2730(5)	0.0937(9)
O7	0.425(2)	0.3573(6)	0.139(1)
O8	0.086(2)	0.3995(5)	0.221(1)
O9	0.797(1)	0.1104(6)	0.357(1)
O10	0.658(1)	0.4385(5)	0.037(1)
Wat1	0.874(3)	0.0755(8)	0.086(2)
Wat2	0.903(3)	0.3256(8)	0.429(2)
Wat3	0.568(3)	0.4537(7)	0.354(2)

Difference			
a	0.0088(10)		
b	0.0415(15)		
c	-0.0166(15)		
alpha	0		
beta	0.25(2)		
gamma	0		
volume	0.36(5)		
	x	y	z
Ca	0.0043(1)	0.00203(2)	-0.0041(4)
Si1	-0.0032(5)	0.00157(20)	-0.0001(4)
Si2	0.0008(5)	-0.00166(15)	0.0004(4)
Si3	-0.009(5)	0.00003(2)	-0.0025(4)
Al1	0.0014(5)	-0.00071(2)	0.0009(4)
Al2	-0.0005(5)	-0.002(1)	-0.0053(4)
O1	-0.0020(10)	0.0039(3)	0.0018(5)
O2	0.0002(10)	0.0007(3)	-0.0032(5)
O3	-0.0174(10)	-0.0007(3)	-0.0144(5)
O4	-0.0006(10)	-0.0040(3)	-0.0089(5)
O5	0.0115(10)	-0.0014(3)	0.0257(5)
O6	0.0018(5)	-0.0018(3)	-0.0032(5)
O7	-0.0107(10)	0.0014(3)	-0.0045(5)
O8	-0.0093(10)	-0.0039(3)	-0.0062(5)
O9	-0.0076(5)	-0.0003(3)	-0.0005(5)
O10	0.0022(2)	-0.0016(3)	-0.0028(5)
Wat1	0.0168(15)	0.0048(4)	0.0223(10)
Wat2	0.0055(15)	0.0004(4)	0.0114(10)
Wat3	0.0102(15)	-0.0083(4)	0.0202(10)

Supplementary Figure 1. The Rietveld refinement fits of the structural models of Ca-NAT, Sr-NAT, Cd-NAT, Pb-NAT, Ag-NAT, and Li-NAT to the synchrotron X-ray powder diffraction data measured at room temperature. Backgrounds were subtracted from the data. Points represent observed data. The continuous lines are the calculated profiles. The lower curves represent the differences between the observed and calculated profiles (I_{obs} - I_{calc}) plotted on the same scale as the observed data. The wavelength used was 0.7297(1) Å.



Supplementary Figure 2. The refined unit cell lengths of the caion-exchanged natrolites, normalized to the orthorhombic unit cell. Data for Na-NAT are from the work of Baur et al. (1990), and K-, Rb-, and Cs-NAT from Lee et al. (2010). The lines are guides to the eyes. The dotted line indicates a possible threshold for the order-disorder transition.



Supplementary Figure 3. Changes in the degree of orthorhombicity, defined as 2(b-a)/(b+a), of the caion-exchanged natrolites plotted as a function of the cation radius.