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Multifaceted orogenic fluid dynamics unraveled by hydrothermal epidote

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1 U–Pb data of allanite secondary reference materials and epidote unknowns

Sample P6b was dated during the same session as sample Gr0-e, in which AVC allanite gave a Tera–Wasserburg age of 281.3 ± 4.3 Ma (anchored to the $^{207}Pb/^{206}Pb$ value of 0.854 ± 0.015 at 275 Ma of Stacey and Kramers, 1975; see the supplementary material of Peverelli et al., 2022a). Sample GE4 was dated along with sample Heyuan-1 of Peverelli et al. (2021; see their Table 3), with CAP allanite returning a Tera–Wasserburg age of 288.5 ± 2.9 Ma and AVC allanite one of 292.4 ± 3.7 Ma, both anchored to a $^{207}Pb/^{206}Pb$ value of 0.854 ± 0.015 (at 275 Ma; Stacey and Kramers, 1975). Samples P8, OBA19-03B and GAS19-01A were dated on 22 February 2021, with CAP allanite returning an anchored Tera–Wasserburg age of 283.8 ± 4.2 Ma (Fig. A1a) and AVC allanite one of 283.0 ± 2.5 Ma (Fig. A1b; both anchored to the $^{207}Pb/^{206}Pb$ value of 0.854 ± 0.015 at 275 Ma of Stacey and Kramers, 1975). Sample Planggenstock was dated on 23 February 2021, when CAP allanite yielded a Tera–Wasserburg age of 285.6 ± 3.2 Ma (Fig. A1c) and AVC allanite one of 284.1 ± 4.1 Ma (Fig. A1d; both anchored to the $^{207}Pb/^{206}Pb$ value of 0.854 ± 0.015 at 275 Ma of Stacey and Kramers, 1975). All AVC ages are within uncertainty of the reference U–Pb age of 289.6 ± 5.6 Ma (Gregory et al., 2007). The age obtained from CAP allanite on 22 February 2021 is within the uncertainty of the reference age of 275.0 ± 4.7 Ma (Gregory et al., 2007), while that calculated on 23 February 2021 is outside uncertainty of it by ca. 1 % (2.7 myr). Such a difference is negligible if compared to the uncertainties in epidote U–Pb ages of no less than 10 %.

Table S1

Measurement conditions for U–Pb isotope data by LA-ICP-MS.

Laboratory & Sample Preparation	
Laboratory name	Institute of Geological Sciences, University of Bern (Switzerland)
Sample type/mineral	Epidote (unknowns), allanite (reference materials)
Sample preparation	Thin sections
Imaging	Backscattered electron imaging: 1 nA beam current, 20 kV acceleration voltage, working distances of 8.5–9.5 mm
Laser ablation system	
Make, Model & type	Resonetics RESOlutionSE 193 nm excimer laser
Ablation cell & volume	S-155 large volume constant geometry cell (Laurin Technic, Australia)
Laser wavelength	193 nm
Pulse width	5 nsec
On-sample fluence	3 J cm^{-2}
Repetition rate	5 Hz
Ablation duration	40 sec
Spot diameter	50 µm
Sampling mode	Static spot ablation
Carrier gas	Pure He (0.4 l min^{-1}) and N_2 (0.003 l min^{-1}) mixed with Ar (0.86 l min^{-1}) immediately after the ablation cell
Cell carrier gas flow	0.86 l min^{-1}
ICP-MS Instrument	
Make, Model & type	Agilent 7900 single quadrupole
Sample introduction	Ablation aerosol with squid device
RF power	1350 W (22 and 23 February 2021)
Sampler, skimmer cones	Ni
Detection system	Single collector secondary electron multiplier
Data acquisition protocol	Time-resolved analysis
Scanning mode	Peak-hopping one point per peak
Detector mode	Pulse counting, dead time correction applied and analog mode when signal intensity > 10^6 cps
Masses measured	206, 207, 208, 232, 238
Dwell times	40 msec (all masses)
Sweep time	ca. 0.21 sec
Th/U (232/238) ratios and oxide production (248/232)	> 98 % and 0.2 %; measured on NIST 612 using a beam size 50 µm, fluence 2.5 J cm^{-2}, repetition rate 5 Hz, scan rate 5 µm sec^{-1}
Sensitivity on mass 232	6585 cps ppm^{-1} (22 February 2021) and 6640 cps ppm^{-1} (23 February 2021); measured on NIST 612 using a beam size 50 µm, fluence 2.5 J cm^{-2}, repetition rate 5 Hz, scan rate 5 µm sec^{-1}
IC Dead time	38 nsec
Data Processing	
Gas blank	30 sec on-peak zero subtracted
Calibration strategy	Tara allanite used as primary reference material; CAP and AVC allanite used for quality control
Primary reference material	Tara allanite (Gregory et al., 2007; Smye et al., 2014; ^{207}Pb-correction using an initial ^{207}Pb/^{206}Pb ratio of 0.866 ± 0.079)
Data processing package	Iolite 7.08 and the VizualAge_UcomPbine Data Reduction Scheme (Chew et al., 2014)
Initial Pb correction	No initial Pb correction applied to epidote unknown and allanite secondary reference materials
Uncertainty level & propagation	Age uncertainties are given at 95 % confidence level, and do not include propagation of long-term reproducibility of secondary standards
Quality control / Validation	CAP and AVC allanite (Barth et al., 1994; Gregory et al., 2007);
Other information	The suitability of allanite as a sufficiently matrix-matched reference material for epidote U–Pb geochronology is outlined in Peverelli et al. (2021)

Table S2

U–Pb isotope data of allanite secondary reference materials. Uncertainties are given as 2 standard errors (2 SE). $f206$ indicates the fraction of initial Pb in each measurement and it is calculated as:

$$\left(\left({}^{207}Pb/{}^{206}Pb\right)_{meas} - \left({}^{207}Pb/{}^{206}Pb\right)_{rad}\right)/\left(\left({}^{207}Pb/{}^{206}Pb\right)_{in} - \left({}^{207}Pb/{}^{206}Pb\right)_{rad}\right).$$

Error correlation (rho) is calculated as:

$$\left(\left({}^{206}Pb/{}^{238}U\right)/2SE_{206/238}\right)/\left(\left(\left({}^{207}Pb/{}^{235}U\right)/2SE_{207/235}\right)\right).$$

in = initial; meas = measured; rad = radiogenic. Mass 235 is not measured, and therefore ^{235}U is calculated from measurements of mass 238 using the $^{238}U/^{235}U$ ratios of 137.818 according to Hiess et al. (2012).

22 February 2021

CAP allanite

Spot	$f206$	$^{238}U/^{206}Pb$	2 SE	$^{207}Pb/^{206}Pb$	2 SE	$^{207}Pb/^{235}U$	2 SE	$^{206}Pb/^{238}U$	2 SE	rho	$(^{207}Pb/^{206}Pb)_{in}$	$(^{207}Pb/^{206}Pb)_{rad}$
1	0.17	18.678	0.303	0.1845	0.0029	1.368	0.021	0.053	0.001	0.95	0.866	0.0518
2	0.21	17.422	0.282	0.2213	0.0035	1.747	0.028	0.057	0.001	0.99		
3	0.14	18.939	0.294	0.1642	0.0025	1.192	0.018	0.053	0.001	0.97		
4	0.12	19.701	0.307	0.1514	0.0022	1.06	0.01	0.051	0.001	0.85		
5	0.19	17.699	0.288	0.2067	0.0035	1.604	0.027	0.056	0.001	1.0		
6	0.13	19.120	0.303	0.154	0.002	1.109	0.016	0.052	0.001	0.91		
7	0.18	18.008	0.289	0.1981	0.0031	1.513	0.024	0.055	0.001	0.99		
8	0.23	16.858	0.276	0.2387	0.0043	1.949	0.037	0.059	0.001	1.2		
9	0.15	19.279	0.301	0.1704	0.0025	1.219	0.017	0.052	0.001	0.89		
10	0.18	18.103	0.301	0.1985	0.0038	1.51	0.03	0.055	0.001	1.2		
11	0.21	17.328	0.285	0.2237	0.0039	1.775	0.029	0.058	0.001	0.99		
12	0.24	16.537	0.273	0.2416	0.0045	2.015	0.036	0.060	0.001	1.1		

AVC allanite

Spot	$f206$	$^{238}U/^{206}Pb$	2 SE	$^{207}Pb/^{206}Pb$	2 SE	$^{207}Pb/^{235}U$	2 SE	$^{206}Pb/^{238}U$	2 SE	rho	$(^{207}Pb/^{206}Pb)_{in}$	$(^{207}Pb/^{206}Pb)_{rad}$
1	0.25	16.989	0.283	0.2523	0.0041	2.043	0.034	0.059	0.001	1.0	0.854	0.0518
2	0.18	18.365	0.287	0.1972	0.0026	1.477	0.018	0.054	0.001	0.78		
3	0.14	19.106	0.296	0.1666	0.0022	1.208	0.014	0.052	0.001	0.75		
4	0.23	17.059	0.274	0.2329	0.0035	1.888	0.026	0.059	0.001	0.86		
5	0.24	16.694	0.279	0.2481	0.0042	2.06	0.03	0.060	0.001	0.93		
6	0.28	15.883	0.277	0.28	0.01	2.425	0.046	0.063	0.001	1.1		
7	0.26	16.434	0.270	0.2642	0.0056	2.219	0.043	0.061	0.001	1.2		
8	0.32	15.283	0.257	0.3059	0.0054	2.756	0.043	0.065	0.001	0.93		
9	0.29	15.783	0.274	0.2858	0.0051	2.498	0.039	0.063	0.001	0.90		
10	0.18	18.318	0.292	0.1932	0.0033	1.463	0.025	0.055	0.001	1.1		

11	0.19	18.073	0.278	0.2057	0.0024	1.564	0.017	0.055	0.001	0.71		
12	0.18	18.142	0.276	0.2001	0.0022	1.515	0.016	0.055	0.001	0.69		
13	0.22	17.495	0.285	0.2303	0.0036	1.81	0.03	0.057	0.001	0.88		
14	0.20	18.024	0.286	0.2083	0.0039	1.61	0.026	0.055	0.001	1.0		
15	0.20	18.106	0.279	0.2148	0.003	1.646	0.03	0.055	0.001	1.0		
16	0.21	17.658	0.306	0.2176	0.0041	1.716	0.028	0.057	0.001	0.94		
17	0.21	17.841	0.286	0.2177	0.0029	1.681	0.022	0.056	0.001	0.81		
18	0.16	18.598	0.301	0.1819	0.0025	1.36	0.02	0.054	0.001	0.91		
19	0.24	16.647	0.277	0.2463	0.0039	2.039	0.032	0.060	0.001	0.94		
20	0.20	17.464	0.281	0.2124	0.0035	1.692	0.027	0.057	0.001	0.99		
21	0.18	17.999	0.311	0.1951	0.0048	1.509	0.034	0.056	0.001	1.3		
22	0.29	15.790	0.274	0.2826	0.0064	2.484	0.047	0.063	0.001	1.1		

23 February 2021

CAP allanite

Spot	*f*206	$^{238}U/^{206}Pb$	2 SE	$^{207}Pb/^{206}Pb$	2 SE	$^{207}Pb/^{235}U$	2 SE	$^{206}Pb/^{238}U$	2 SE	rho	$(^{207}Pb/^{206}Pb)_{in}$	$(^{207}Pb/^{206}Pb)_{rad}$
1	0.16	18.748	0.633	0.1804	0.0033	1.337	0.023	0.053	0.002	0.51	0.854	0.0518
2	0.10	20.032	0.682	0.1285	0.0027	0.884	0.019	0.050	0.002	0.63		
3	0.12	18.986	0.649	0.1505	0.0018	1.097	0.012	0.053	0.002	0.32		
4	0.12	19.342	0.636	0.1473	0.0021	1.049	0.015	0.052	0.002	0.43		
5	0.16	17.781	0.601	0.1835	0.003	1.431	0.021	0.056	0.002	0.43		
6	0.12	19.550	0.650	0.1487	0.0022	1.048	0.014	0.051	0.002	0.40		
7	0.18	18.430	0.611	0.1946	0.003	1.46	0.02	0.054	0.002	0.41		
8	0.11	19.631	0.655	0.137	0.002	0.964	0.012	0.051	0.002	0.37		
9	0.20	18.457	0.613	0.2107	0.0035	1.575	0.023	0.054	0.002	0.44		
10	0.19	17.966	0.614	0.203	0.004	1.562	0.026	0.056	0.002	0.49		
11	0.16	18.549	0.619	0.1831	0.0033	1.363	0.023	0.054	0.002	0.50		
12	0.18	17.969	0.613	0.1936	0.0034	1.487	0.024	0.056	0.002	0.47		
13	0.17	18.149	0.626	0.1915	0.0036	1.454	0.026	0.055	0.002	0.52		

AVC allanite

Spot	*f*206	$^{238}U/^{206}Pb$	2 SE	$^{207}Pb/^{206}Pb$	2 SE	$^{207}Pb/^{235}U$	2 SE	$^{206}Pb/^{238}U$	2 SE	rho	$(^{207}Pb/^{206}Pb)_{in}$	$(^{207}Pb/^{206}Pb)_{rad}$
1		17.425	0.577	0.2298	0.0037	1.829	0.028	0.057	0.002	0.46	0.584	0.0518
2		13.004	0.473	0.3852	0.0065	4.05	0.09	0.077	0.003	0.62		
3		15.671	0.540	0.2967	0.0058	2.615	0.047	0.064	0.002	0.52		
4		17.649	0.592	0.229	0.004	1.798	0.034	0.057	0.002	0.56		
5		15.741	0.545	0.284	0.005	2.49	0.04	0.063	0.002	0.46		
6		16.226	0.553	0.2641	0.0048	2.257	0.039	0.062	0.002	0.51		
7		15.664	0.540	0.2949	0.0055	2.599	0.041	0.064	0.002	0.46		
8		16.889	0.570	0.2528	0.0055	2.076	0.044	0.059	0.002	0.63		
9		15.126	0.526	0.306	0.006	2.805	0.051	0.066	0.002	0.52		
10		14.932	0.513	0.3146	0.0066	2.908	0.052	0.067	0.002	0.52		
11		16.401	0.565	0.2676	0.0059	2.255	0.045	0.061	0.002	0.58		

12	15.906	0.557	0.2837	0.0054	2.47	0.04	0.063	0.002	0.49
13	15.456	0.525	0.288	0.006	2.59	0.05	0.065	0.002	0.56
14	17.206	0.592	0.2209	0.0049	1.762	0.034	0.058	0.002	0.56
15	19.066	0.654	0.1632	0.0024	1.185	0.015	0.052	0.002	0.37
16	16.439	0.567	0.2473	0.0037	2.078	0.028	0.061	0.002	0.39
17	17.188	0.591	0.2252	0.0045	1.805	0.034	0.058	0.002	0.55
18	15.456	0.525	0.295	0.007	2.638	0.058	0.065	0.002	0.65
19	15.627	0.537	0.2852	0.0065	2.515	0.048	0.064	0.002	0.55
20	15.723	0.569	0.3005	0.0078	2.622	0.065	0.064	0.002	0.68
21	15.122	0.526	0.2999	0.0074	2.73	0.07	0.066	0.002	0.74

Table S3

U–Pb isotope data of epidote unknowns. Uncertainties are given as 2 standard errors (2SE). $f206$ indicates the fraction of initial Pb in each measurement and it is calculated as:

$$\left(\left({}^{207}Pb/{}^{206}Pb\right)_{meas} - \left({}^{207}Pb/{}^{206}Pb\right)_{rad}\right)/\left(\left({}^{207}Pb/{}^{206}Pb\right)_{in} - \left({}^{207}Pb/{}^{206}Pb\right)_{rad}\right).$$

Error correlation (rho) is calculated as:

$$\left(\left({}^{206}Pb/{}^{238}U\right)/2SE_{206/238}\right)/\left(\left(\left({}^{207}Pb/{}^{235}U\right)/2SE_{207/235}\right)\right).$$

Initial $^{207}Pb/^{206}Pb$ ratio are obtained from Tera – Wasserburg diagrams excluding outliers (see Sect. 5.1). in = initial; meas = measured; rad = radiogenic. Mass 235 is not measured, and therefore ^{235}U is calculated from measurements of mass 238 using the $^{238}U/^{235}U$ ratios of 137.818 according to Hiess et al. (2012).

						P6b						
Spot	**$f206$**	**$^{238}U/^{206}Pb$**	**2 SE**	**$^{207}Pb/^{206}Pb$**	**2 SE**	**$^{207}Pb/^{235}U$**	**2 SE**	**$^{206}Pb/^{238}U$**	**2 SE**	**rho**	**$(^{207}Pb/^{206}Pb)_{in}$**	**$(^{207}Pb/^{206}Pb)_{rad}$**
1	1.00	1.240	0.021	0.7837	0.0039	87.27	0.66	0.806	0.014	0.44	0.7846	0.0466
2	0.99	1.082	0.021	0.778	0.005	99.1	1.1	0.924	0.018	0.57		
3	0.99	2.411	0.039	0.7744	0.0049	44.39	0.34	0.415	0.007	0.47		
4	0.94	14.577	0.319	0.7439	0.0062	7.1	0.1	0.069	0.001	0.65		
5	0.99	5.537	0.095	0.7799	0.0056	19.38	0.18	0.181	0.003	0.54		
6	0.97	5.890	0.090	0.7628	0.0056	17.94	0.11	0.170	0.003	0.40		
7	1.00	2.371	0.047	0.785	0.005	45.91	0.63	0.422	0.008	0.69		
8	0.91	22.311	0.473	0.715	0.005	4.424	0.076	0.045	0.001	0.81		
9	1.01	3.068	0.050	0.793	0.004	35.68	0.24	0.326	0.005	0.41		
10	0.96	8.241	0.129	0.7553	0.0048	12.662	0.072	0.121	0.002	0.36		
11	0.99	2.239	0.035	0.7769	0.0051	47.88	0.26	0.447	0.007	0.35		
12	0.99	7.032	0.124	0.7802	0.0071	15.42	0.15	0.142	0.002	0.55		
13	0.97	7.197	0.114	0.7591	0.0049	14.567	0.092	0.140	0.002	0.40		
14	0.97	3.044	0.049	0.7618	0.0061	34.55	0.24	0.328	0.005	0.43		
15	0.99	3.578	0.056	0.7792	0.0063	30.25	0.18	0.279	0.004	0.38		
16	0.99	2.447	0.042	0.779	0.004	44.01	0.49	0.409	0.007	0.64		
17	1.00	1.592	0.028	0.7845	0.0053	68.33	0.73	0.628	0.011	0.61		
18	0.99	2.353	0.039	0.7743	0.0055	45.41	0.44	0.425	0.007	0.58		
19	1.00	1.012	0.017	0.786	0.003	107.27	0.85	0.988	0.017	0.46		
20	0.99	0.184	0.004	0.7805	0.0036	585.5	8.6	5.43	0.11	0.73		
21	0.98	0.510	0.009	0.7732	0.0055	210.6	1.9	1.962	0.035	0.51		
22	0.96	7.199	0.420	0.7571	0.0041	14.6	0.86	0.139	0.0081	1.01		
23	0.98	0.692	0.011	0.7705	0.0049	154.6	1.2	1.445	0.024	0.47		
24	0.97	0.727	0.012	0.7595	0.0046	144.8	1.1	1.376	0.022	0.47		
25	0.99	1.364	0.035	0.776	0.006	78.6	1.8	0.733	0.019	0.88		

P8												
Spot	***f*206**	**$^{238}U/^{206}Pb$**	**2 SE**	**$^{207}Pb/^{206}Pb$**	**2 SE**	**$^{207}Pb/^{235}U$**	**2 SE**	**$^{206}Pb/^{238}U$**	**2 SE**	**rho**	**$(^{207}Pb/^{206}Pb)_{in}$**	**$(^{207}Pb/^{206}Pb)_{rad}$**
1	0.99	0.646	0.010	0.7717	0.0052	164.3	1.7	1.547	0.025	0.64	0.7811	0.0466
2	0.99	3.189	0.065	0.7729	0.0048	33.36	0.58	0.314	0.006	0.85		
3	0.97	5.741	0.089	0.7621	0.0055	18.24	0.12	0.174	0.003	0.42		
4	0.98	3.119	0.051	0.7691	0.0066	33.98	0.33	0.321	0.005	0.60		
5	0.99	0.765	0.014	0.7729	0.0075	140.5	1.6	1.307	0.024	0.62		
6	1.00	0.886	0.016	0.7776	0.0052	120.7	1.1	1.13	0.02	0.51		
7	1.00	0.638	0.013	0.7824	0.0083	170.9	2.3	1.568	0.031	0.68		
8	1.00	2.370	0.067	0.7791	0.0044	45.2	1.1	0.422	0.012	0.86		
9	1.00	1.299	0.027	0.7803	0.0048	82.6	1.1	0.77	0.02	0.64		
10	0.98	2.407	0.050	0.769	0.006	43.95	0.78	0.415	0.009	0.86		
11	0.96	8.826	0.179	0.7515	0.0078	11.8	0.2	0.113	0.002	0.79		
12	0.98	3.223	0.049	0.7684	0.0052	32.82	0.25	0.310	0.005	0.50		
13	0.98	5.069	0.077	0.7653	0.0047	20.81	0.11	0.197	0.003	0.35		
14	0.98	5.942	0.099	0.7678	0.0091	17.76	0.17	0.168	0.003	0.57		
15	1.00	2.980	0.052	0.7797	0.0068	36.4	0.4	0.336	0.006	0.66		
16	1.00	2.483	0.056	0.7795	0.0058	43.43	0.82	0.403	0.009	0.84		
17	0.98	2.362	0.051	0.7698	0.0053	44.77	0.61	0.423	0.009	0.63		
18	0.99	3.233	0.050	0.7733	0.0062	32.87	0.25	0.309	0.005	0.49		
19	1.00	0.692	0.013	0.7828	0.0052	155.5	1.7	1.446	0.028	0.56		
20	0.91	23.2126	0.409	0.7137	0.0065	4.26	0.05	0.043	0.001	0.69		

GE4												
Spot	***f*206**	**$^{238}U/^{206}Pb$**	**2 SE**	**$^{207}Pb/^{206}Pb$**	**2 SE**	**$^{207}Pb/^{235}U$**	**2 SE**	**$^{206}Pb/^{238}U$**	**2 SE**	**rho**	**$(^{207}Pb/^{206}Pb)_{in}$**	**$(^{207}Pb/^{206}Pb)_{rad}$**
1	1.02	1.733	0.075	0.7476	0.0085	59.9	2.3	0.577	0.025	0.89	0.7308	0.0464
2	1.03	0.823	0.024	0.752	0.007	120.6	1.6	1.208	0.035	0.46		
3	1.01	2.540	0.071	0.74	0.01	40	0.81	0.394	0.011	0.72		
4	1.01	1.370	0.045	0.7345	0.0069	74.7	2	0.73	0.02	0.81		
5	1.02	5.461	0.146	0.742	0.008	18.98	0.2	0.183	0.005	0.39		
6	0.94	25.615	0.656	0.6873	0.0064	3.712	0.046	0.039	0.001	0.48		
7	1.00	5.249	0.234	0.728	0.016	18.41	0.79	0.190	0.008	0.96		
8	0.98	6.485	0.206	0.719	0.008	15.26	0.3	0.154	0.005	0.62		
9	0.99	3.070	0.085	0.7245	0.0091	32.77	0.41	0.326	0.009	0.45		
10	1.01	2.641	0.077	0.74	0.01	37.93	0.92	0.379	0.011	0.83		
11	0.96	7.605	0.202	0.706	0.008	13.05	0.15	0.131	0.003	0.43		
12	0.98	6.702	0.180	0.716	0.008	14.93	0.21	0.149	0.004	0.52		
13	0.98	9.930	0.276	0.71	0.01	10.17	0.19	0.101	0.003	0.67		
14	0.97	9.9108	0.383	0.709	0.013	10.06	0.31	0.101	0.004	0.80		
15	0.97	9.766	0.267	0.7072	0.0074	10.22	0.12	0.102	0.003	0.43		
16	0.97	3.698	0.137	0.711	0.012	26.72	0.77	0.27	0.01	0.78		
17	0.95	10.699	0.275	0.6984	0.0064	9.11	0.1	0.093	0.002	0.43		

18	0.97	2.538	0.064	0.7103	0.0059	38.83	0.53	0.39	0.01	0.54		
19	0.95	19.646	0.618	0.697	0.011	4.79	0.13	0.051	0.002	0.86		
20	0.96	12.270	0.346	0.703	0.010	8.08	0.13	0.081	0.002	0.57		
21	0.97	6.752	0.242	0.7136	0.0099	14.14	0.3	0.148	0.005	0.59		
22	0.94	19.642	0.579	0.690	0.009	4.86	0.11	0.051	0.001	0.77		
23	0.99	2.115	0.058	0.7255	0.0065	47.76	0.62	0.473	0.013	0.47		
24	0.96	0.656	0.018	0.7008	0.0059	148.2	2.3	1.525	0.041	0.58		

OBA19-03B

Spot	*f*206	$^{238}U/^{206}Pb$	2 SE	$^{207}Pb/^{206}Pb$	2 SE	$^{207}Pb/^{235}U$	2 SE	$^{206}Pb/^{238}U$	2 SE	rho	$(^{207}Pb/^{206}Pb)_{in}$	$(^{207}Pb/^{206}Pb)_{rad}$
1	0.99	5.822077317	0.0881311	0.801	0.004	19.16	0.07	0.172	0.003	0.25	0.8118	0.0463
2	0.98	9.305	0.1385	0.7975	0.0032	11.79	0.05	0.107	0.002	0.315		
3	0.97	13.4975	0.219	0.7902	0.0068	7.88	0.05	0.075	0.001	0.384		
4	0.99	5.712	0.085	0.8015	0.0035	19.34	0.09	0.175	0.003	0.30		
5	0.95	23.116	0.353	0.7769	0.0045	4.70	0.02	0.043	0.001	0.32		
6	0.97	10.734	0.161	0.7886	0.0044	10.24	0.05	0.093	0.001	0.32		
7	0.96	19.220	0.307	0.7815	0.0033	5.63	0.03	0.052	0.001	0.37		
8	0.95	25.8937	0.389	0.7723	0.0035	4.11	0.02	0.039	0.001	0.28		
9	1.00	4.320	0.065	0.8085	0.0031	25.78	0.11	0.231	0.003	0.28		
10	0.97	17.437	0.277	0.7904	0.0036	6.31	0.04	0.057	0.001	0.43		
11	0.97	12.293	0.181	0.7919	0.0032	8.87	0.04	0.081	0.001	0.34		
12	0.98	9.359	0.140	0.7978	0.0039	11.57	0.09	0.107	0.002	0.54		
13	0.98	7.771	0.115	0.7986	0.0036	14.15	0.06	0.129	0.002	0.29		
14	0.98	8.331	0.125	0.7988	0.0034	13.19	0.11	0.120	0.002	0.56		
15	0.98	10.801	0.163	0.7932	0.0044	10.23	0.05	0.093	0.001	0.31		
16	0.98	6.848	0.098	0.7988	0.0025	16.07	0.07	0.146	0.002	0.32		
17	1.01	0.361	0.006	0.8165	0.0042	297.3	3.9	2.77	0.049	0.74		
18	0.99	5.023	0.076	0.8055	0.0039	22.02	0.13	0.199	0.003	0.39		
19	0.99	7.677	0.118	0.8013	0.0044	14.52	0.07	0.130	0.002	0.33		

GAS19-01A

Spot	*f*206	$^{238}U/^{206}Pb$	2 SE	$^{207}Pb/^{206}Pb$	2 SE	$^{207}Pb/^{235}U$	2 SE	$^{206}Pb/^{238}U$	2 SE	rho	$(^{207}Pb/^{206}Pb)_{in}$	$(^{207}Pb/^{206}Pb)_{rad}$
1	0.99	6.835	0.107	0.8306	0.0071	16.78	0.15	0.146	0.002	0.57	0.8381	0.0463
2	1.00	1.797	0.035	0.841	0.006	64.41	0.93	0.556	0.011	0.73		
3	1.00	2.823	0.076	0.8358	0.0058	41	1	0.354	0.009	0.91		
4	1.01	1.108	0.018	0.8427	0.0072	104	1	0.903	0.015	0.58		
5	1.00	1.938	0.032	0.8362	0.0065	59.3	0.4	0.516	0.008	0.41		
6	1.00	0.022	0.001	0.8391	0.0038	5340	120	45.9	1.2	0.86		
7	1.00	0.845	0.015	0.8403	0.0072	134.3	1.6	1.183	0.021	0.67		
8	1.00	1.214	0.022	0.84	0.01	96.5	1.2	0.824	0.015	0.68		
9	1.01	1.253	0.025	0.8437	0.0071	93.8	1.4	0.798	0.016	0.74		
10	1.00	1.536	0.026	0.8349	0.0055	74.7	0.5	0.651	0.011	0.40		
11	0.99	3.733	0.067	0.8339	0.0071	30.71	0.39	0.268	0.005	0.71		

12	1.00	1.570	0.024	0.837	0.003	73.35	0.49	0.637	0.010	0.44
13	0.99	1.785	0.028	0.8338	0.0041	64.4	0.5	0.560	0.009	0.50
14	1.00	0.469	0.007	0.8373	0.0039	245.4	1.9	2.132	0.034	0.49
15	1.00	0.928	0.015	0.834	0.004	123.47	0.73	1.077	0.017	0.37
16	1.00	1.805	0.029	0.835	0.003	63.75	0.59	0.554	0.009	0.57
17	0.99	3.273	0.049	0.8289	0.0038	35.22	0.19	0.305	0.005	0.36
18	0.99	1.615	0.039	0.8305	0.0049	71	1	0.619	0.015	0.87
19	1.00	1.404	0.039	0.836	0.006	83	2	0.71	0.02	0.86
20	0.99	2.494	0.039	0.8287	0.0068	45.71	0.29	0.401	0.006	0.40
21	0.99	2.100	0.034	0.832	0.006	54.5	0.5	0.476	0.008	0.58
22	1.00	0.623	0.010	0.8344	0.0044	184.5	1.7	1.606	0.025	0.59
23	1.00	2.337	0.047	0.8343	0.0046	49.1	0.6	0.428	0.009	0.57
24	1.00	1.485	0.026	0.8399	0.0064	77.8	1.1	0.673	0.012	0.79
25	1.00	1.896	0.031	0.8343	0.0066	60.48	0.39	0.527	0.008	0.40
26	1.00	0.739	0.011	0.8381	0.0046	155.9	1.1	1.353	0.021	0.45
27	0.99	1.261	0.024	0.8335	0.0053	90.92	0.94	0.793	0.015	0.55
28	0.99	1.511	0.023	0.831	0.005	75.69	0.48	0.66	0.01	0.42

Planggenstock

Spot	*f*206	$^{238}U/^{206}Pb$	2 SE	$^{207}Pb/^{206}Pb$	2 SE	$^{207}Pb/^{235}U$	2 SE	$^{206}Pb/^{238}U$	2 SE	rho	$(^{207}Pb/^{206}Pb)_{in}$	$(^{207}Pb/^{206}Pb)_{rad}$
1	0.99	2.488	0.099	0.8017	0.0077	45	1	0.402	0.016	0.56	0.8071	0.0463
2	1.00	0.945	0.032	0.8089	0.0045	118.6	1.2	1.058	0.036	0.30		
3	0.99	2.964	0.105	0.8025	0.0045	37.49	0.37	0.337	0.012	0.28		
4	1.00	1.618	0.055	0.8056	0.0045	68.9	0.9	0.618	0.021	0.38		
5	0.98	9.385	0.317	0.7924	0.0048	11.68	0.08	0.106	0.004	0.19		
6	1.00	1.010	0.035	0.807	0.005	110.6	1.3	0.99	0.03	0.34		
7	1.01	1.316	0.045	0.8126	0.0061	84.88	0.86	0.760	0.026	0.30		
8	1.00	1.831	0.060	0.8063	0.0044	61	1	0.546	0.018	0.23		
9	1.00	1.431	0.049	0.8062	0.0043	78.02	0.88	0.699	0.024	0.33		
10	0.99	3.275	0.107	0.802	0.004	33.92	0.26	0.30	0.01	0.23		
11	0.99	3.116	0.107	0.799	0.004	35.56	0.37	0.321	0.011	0.30		
12	0.99	1.434	0.049	0.8016	0.0044	77.4	0.8	0.697	0.024	0.29		
13	0.99	2.950	0.096	0.7995	0.0046	37.52	0.26	0.339	0.011	0.21		
14	0.98	3.774	0.127	0.794	0.005	29.12	0.16	0.265	0.009	0.16		
15	0.99	3.532	0.120	0.799	0.005	31.41	0.27	0.283	0.010	0.25		
16	1.00	3.382	0.114	0.804	0.005	33	1	0.30	0.01	0.30		
17	1.00	2.623	0.089	0.8061	0.0062	42.32	0.56	0.381	0.013	0.39		
18	0.99	3.940	0.132	0.801	0.004	28.12	0.12	0.254	0.008	0.13		
19	0.99	2.089	0.070	0.8018	0.0044	53.17	0.23	0.479	0.016	0.13		
20	1.00	1.635	0.056	0.8038	0.0044	68.08	0.66	0.612	0.021	0.28		
21	0.84	83.822	3.091	0.683	0.017	1.14	0.02	0.012	0.001	0.48		
22	0.99	8.171	0.274	0.8011	0.0081	13.58	0.12	0.122	0.004	0.26		

23	0.99	6.588	0.239	0.797	0.013	16.93	0.26	0.152	0.005	0.42
24	1.01	3.2	0.1	0.8131	0.0096	35.1	0.4	0.312	0.011	0.29
25	0.99	1.659	0.055	0.8024	0.0048	66.98	0.38	0.60	0.02	0.17
26	1.00	2.226	0.074	0.8088	0.0065	50.4	0.4	0.449	0.015	0.24
27	0.99	2.011	0.069	0.8003	0.0075	55.09	0.44	0.497	0.017	0.23
28	0.99	3.418	0.114	0.8007	0.0052	32.43	0.18	0.293	0.010	0.17
29	1.00	1.636	0.056	0.804	0.005	68.09	0.39	0.611	0.021	0.17
30	1.00	1.780	0.060	0.8064	0.0048	62.81	0.34	0.562	0.019	0.16
31	0.98	9.288	0.311	0.793	0.005	11.83	0.07	0.108	0.004	0.17
32	0.99	5.653	0.195	0.801	0.005	19.58	0.22	0.177	0.006	0.33
33	0.99	6.743	0.227	0.7968	0.0049	16.37	0.08	0.148	0.005	0.14
34	1.00	4.843	0.171	0.8065	0.0063	23.06	0.27	0.206	0.007	0.33
35	0.98	9.952	0.337	0.7953	0.0051	11.09	0.07	0.100	0.003	0.18
36	1.00	1.333	0.044	0.8047	0.0045	83.47	0.46	0.75	0.02	0.16

3 Cerium, Pb, Th, U and sum of REE (ΣREE) of the studied epidote samples

Mass fractions of Ce, Pb, Th, U and the sum of REE (ΣREE; Table S4) were determined by LA-ICP-MS at the University of Bern. Measurements in samples P1 of spots 13–38 were carried out on a Geolas Pro 193 nm ArF excimer laser (Coherent, USA) coupled with an ELAN DRCe quadrupole ICP-MS (QMS; Perkin Elmer, USA). Ablation rate was ca. 0.1 µm per laser pulse, repetition rate 10 Hz and beam sizes between 24 and 60 µm, with ablation in 1 L min^{-1} He and 0.008 L min^{-1} H_2. Details on the measurement procedures are outlined in Pettke et al. (2012). All other samples and spots in sample P1 were analyzed on a RESOlutionSE 193 nm excimer laser system (Applied Spectra, USA) equipped with an S-155 large-volume constant-geometry chamber (Laurin Technic, Australia) coupled with an Agilent 7900 ICP-QMS. Repetition rate was 5 Hz, spot sizes between 20 and 30 µm, with ablation in He which admixed with the Ar carrier gas for transport to the ICP-MS. The NIST SRM612 was used for optimization of analytical conditions of both systems. The basalt glass GSD-1G was used as external standard, and the NIST SRM612 for quality control. Data reduction was carried out using SILLS (Guillong et al., 2008), using the sum of total oxides (98.3 % for epidote and 100 % for the NIST SRM612) for internal standardization (see Halter et al., 2002). Calculation of limits of detection used the formulation of Pettke et al. (2012) for each element in each analysis.

Table S4: Cerium, Th, U and sum of REE (ΣREE) measured by LA-ICP-MS in the studied epidote samples. Analyses below limits of detection are indicated by the symbol < followed by limit of detection calculated for that specific analysis. N/A = all REE measurements were below individual limits of detection.

Analysis	Ce [μg g^{-1}]	Pb [μg g^{-1}]	Th [μg g^{-1}]	U [μg g^{-1}]	ΣREE [μg g^{-1}]
Epidote P1					
1	0.03	19	0.004	4.9	0.84
2	2.1	44	0.75	6.3	9.4
3	0.22	26	0.003	4.8	1.9
4	0.02	28	0.88	3.3	0.04
5	0.02	12	*< 0.01*	0.28	0.20
6	1.2	24	*< 0.02*	2.0	3.3
7	0.11	7.3	*< 0.01*	2.7	2.0
8	0.14	12	*< 0.01*	1.2	1.7
9	0.69	12	*< 0.01*	1.2	3.4
10	1.2	13	*< 0.01*	1.2	4.9
11	0.55	17	*< 0.01*	1.4	2.7
12	1.6	91	*0.11*	0.88	7.9
13	1.2	23	0.23	2.8	19
14	1.0	32	0.10	4.8	12
15	2.9	26	0.24	0.69	11
16	1.6	13	0.08	1.8	10
17	*< 0.10*	28	*< 0.04*	0.24	*N/A*
18	0.20	12	0.01	0.92	1.9
19	0.47	23	0.06	2.4	5.2
20	0.71	35	*< 0.07*	5.0	7.0
21	0.04	7.6	*< 0.03*	0.53	0.04
22	< 0.07	15	*< 0.02*	0.87	0.11
23	4.2	28	*< 0.02*	0.51	8.3
24	*< 0.09*	27	*< 0.03*	0.49	*N/A*
25	8.3	9.5	*< 0.03*	1.2	24

26	0.78	7.5	0.04	0.85	3.7
27	0.30	16	0.02	1.2	3.0
28	0.26	8.6	< *0.02*	1.7	2.4
29	0.70	11	< *0.01*	2.2	3.4
30	0.29	11	0.08	0.52	0.89
31	0.12	24	0.06	6.0	1.5
32	1.1	16	< *0.07*	14	23
33	0.17	6.3	< *0.03*	1.5	0.83
34	0.56	67	0.02	7.4	2.0
35	0.12	58	0.03	11	1.4
36	0.20	12	< *0.01*	1.6	2.3
37	0.21	13	< *0.01*	0.95	2.4
38	0.16	10	< *0.02*	1.4	1.7
Epidote P2					
1	0.71	6.3	< *0.06*	2.1	1.9
2	0.13	11	< *0.06*	1.4	0.24
2	34	42	< *0.08*	2.3	79
3	18	39	< *0.05*	1.4	39
3	1.1	33	< *0.06*	0.78	3.6
4	0.92	12	< *0.11*	0.49	2.2
4	0.77	29	9.8	2.1	2.6
5	0.26	48	< 0.07	0.63	1.8
5	7.8	36	3.4	8.2	27
6	0.51	15	< *0.04*	1.0	3.1
6	0.42	18	< *0.05*	1.2	2.8
7	1.3	20	< *0.03*	0.99	5.0
7	0.42	18	< *0.05*	0.78	2.4
8	1.3	27	< *0.04*	1.8	6.7
8	15	17	< *0.03*	2.7	38
9	0.49	25	0.04	1.1	2.6
9	5.9	24	0.52	15	21

10	1.6	44	0.36	2.6	5.1
10	0.03	20	4.5	0.72	0.14
11	0.20	47	< *0.03*	0.45	1.6
11	0.11	47	< *0.04*	0.69	0.92
12	0.88	50	< *0.02*	1.5	3.2
12	0.05	21	0.39	2.6	0.50
13	5.3	121	< *0.02*	18	14
13	0.10	11	< *0.02*	0.69	0.25
14	0.32	23	< *0.01*	6.1	1.2
Epidote P3					
1	200	54	1.8	6.5	469
2	50	34	2.3	6.7	128
2	132	40	1.3	14	313
3	1.9	60	20	2.0	8.6
3	5.8	56	< 0.04	1.1	17
Epidote Grimsel-1					
1	10	172	8.9	133	92
2	8.1	150	0.18	137	96
2	4.1	125	0.15	188	48
3	7.6	135	0.30	186	69
3	7.4	133	0.26	174	70
4	4.3	118	0.14	164	48
4	6.6	130	0.36	160	54
5	7.6	136	2.2	185	64
5	8.2	132	2.0	192	63
6	5.2	133	0.12	195	57
6	8.5	151	7.6	182	67
7	20	152	0.99	226	125
7	14	127	0.47	79	131
8	18	131	< 0.09	129	163
8	8.8	155	0.82	353	89

9	1.9	171	0.15	266	67
9	9.3	97	0.07	60	42
10	15	169	0.71	125	87
10	9.3	87	0.30	137	55
11	17	174	0.56	242	131
11	12	148	2.6	102	79
12	9.7	175	0.47	225	82
12	7.2	93	4.9	103	34
13	8.5	193	0.81	136	53
13	3.0	78	1.9	158	40
14	2.5	179	0.14	263	79
14	0.97	93	0.04	54	23
15	3.0	152	0.84	140	48
15	6.3	93	0.83	118	41
16	2.9	113	3.0	105	30
Epidote Grimsel-2					
1	10	75	0.07	217	144
2	6.8	69	0.27	159	110
3	7.1	75	0.20	151	100
4	5.2	51	*< 0.06*	121	100
5	4.4	62	*< 0.05*	117	87
6	6.8	74	0.07	161	97
7	14	97	0.11	273	180
8	18	95	0.13	266	125
9	17	65	0.21	284	115
10	6.1	88	*< 0.02*	177	131
11	9.5	93	*< 0.03*	109	117
12	29	75	0.53	535	188
13	10	62	0.36	311	97
Epidote P6b					
1	2.1	39	*< 0.04*	180	71

2	1.7	109	< *0.03*	29	30
3	1.6	75	< *0.04*	11	24
4	6.1	56	0.25	43	37
5	4.6	51	0.60	40	30
6	0.69	33	< *0.04*	42	13
7	1.6	34	< *0.04*	174	40
8	3.6	32	0.04	12	23
9	0.99	41	< *0.01*	40	15
10	9.3	115	0.02	24	46
11	11	54	< *0.06*	5.2	40
12	2.3	79	< *0.02*	8.1	14
13	6.4	84	< *0.07*	8.3	28
14	0.89	60	< *0.04*	4.0	12
15	2.5	98	< *0.01*	18	28
16	0.70	67	< *0.04*	5.6	13
17	2.8	176	< *0.06*	7.7	27
18	19	46	0.08	17	68
19	34	119	0.52	40	109
20	6.2	59	< *0.05*	9.1	40
Epidote P8					
1	0.78	26	< *0.04*	30	25
2	3.5	53	0.22	201	62
3	4.7	35	0.08	67	49
4	1.5	58	0.01	24	22
5	1.2	26	< *0.03*	15	15
6	2.4	58	< *0.03*	79	54
7	2.9	68	0.05	11	28
8	0.89	56	< *0.02*	27	20
9	6.9	52	< *0.04*	7.1	37
10	1.2	45	< *0.01*	71	37
11	0.72	34	< *0.02*	57	33

12	0.94	48	< *0.02*	18	19
13	0.94	30	0.06	24	18
			Epidote GE4		
1	28	38	0.004	80	196
2	2.1	30	< *0.006*	20	42
3	0.55	30	< *0.008*	11	21
4	0.84	31	< *0.007*	41	59
5	3.1	61	0.02	162	132
6	0.38	58	0.01	5.4	13
7	84	63	1.3	147	441
8	1.7	43	0.07	160	99
9	2.1	63	0.06	76	156
10	2.0	77	0.01	53	107
11	2.1	29	< *0.005*	72	116
12	1.2	62	0.03	48	77
13	5.6	43	0.56	20	80
14	0.11	18	0.00	15	214
15	1.3	47	0.11	182	142
16	1.3	66	0.01	107	168
			Epidote OBA19-03B		
1	1.7	189	< *0.01*	363	160
2	6.4	128	0.01	263	282
3	8.8	200	< *0.007*	475	366
4	3.8	168	< *0.01*	531	348
5	365	127	7.9	365	1437
6	81	114	1.7	604	636
7	1.3	175	< *0.007*	415	265
8	6.2	151	0.02	439	328
9	321	151	13	452	1364
10	0.26	116	0.04	313	50
11	1.0	148	0.04	124	24

12	331	118	1.5	330	1293
13	0.26	117	0.02	245	95
14	0.20	120	0.02	158	51
15	0.25	134	0.03	101	29
16	240	164	12	191	893
17	422	166	2.9	345	1499
18	25	146	0.43	224	250
19	15	197	0.01	456	395
20	17	186	0.13	466	431
21	0.19	137	< *0.007*	342	165
22	3.6	116	< *0.01*	249	247
23	0.08	142	< *0.01*	247	87
24	6.2	115	*0.02*	246	209
25	0.02	116	< *0.005*	241	149
26	0.04	191	< *0.01*	221	75
Epidote GAS19-01A					
1	1.1	37	0.12	56	21
2	0.97	39	0.09	22	13
3	1.0	48	2.5	27	14
4	0.46	67	0.13	13	6.6
5	0.90	45	< *0.01*	18	10
6	2.0	83	0.02	13	12
7	25	199	0.86	349	273
8	8.7	47	0.12	43	88
9	2.6	45	0.02	36	23
10	2.7	34	0.08	25	25
11	4.5	226	0.37	735	82
12	0.77	95	0.82	9.2	6.0
13	0.65	96	0.12	45	19
14	2.1	74	0.64	67	41
Epidote Planggenstock					

1	156	105	0.27	44	443
2	82	69	0.14	35	257
3	50	89	0.04	27	165
4	62	44	0.10	30	218
5	84	57	0.11	45	299
6	64	53	0.04	33	245
7	39	48	0.02	21	172
8	35	40	0.02	18	158
9	22	25	0.02	15	103
10	59	47	0.02	51	212
11	48	43	0.04	44	181
12	27	15	0.08	29	103
13	17	15	0.02	18	77
14	46	153	0.28	26	142
15	46	154	0.28	27	141
16	49	185	0.40	53	175
17	44	148	0.44	53	164
18	38	149	0.24	21	118
19	41	142	0.22	22	126
20	49	126	0.31	25	146
21	54	168	0.26	28	162
22	58	154	0.31	29	170

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