

Electronic Supplementary Information (ESI):

Tables S1 – S3 (ESI) for:

**Accuracy of Laser-Ablation (LA)-MC-ICPMS Sr Isotope Analysis of
(Bio)Apatite – a Problem Reassessed**

Wolfgang Müller and Robert Anczkiewicz

Table S1 (ESI): LA-MC-ICPMS results of $^{87}\text{Sr}/^{86}\text{Sr}$, $^{84}\text{Sr}/^{86}\text{Sr}$ and $^{85}\text{Rb}/^{86}\text{Sr}$ ratios of two modern shark enameloid in-house reference samples analyzed interchangeably over four years (Aug. 2010 – Sep. 2014; n=88). The resultant long-term averages of $^{87}\text{Sr}/^{86}\text{Sr}$ and $^{84}\text{Sr}/^{86}\text{Sr}$ ratios are 0.709171 ± 0.000053 (2SD) and 0.056500 ± 0.000040 (2 SD), respectively (n=84; two high and low outliers each (**)) have been excluded). WA...shark live-caught offshore W Australia (smaller), SA...South African shark (larger). Plot-Numbers ('plot-no.') are arbitrary consecutive numbers; missing numbers only serve to highlight the different analysis periods and no data are omitted. (See text)

analysis period	plot-no.	sample ID	$^{87}\text{Sr}/^{86}\text{Sr}$	2 S.E.	$^{84}\text{Sr}/^{86}\text{Sr}$	2 S.E.	$^{85}\text{Rb}/^{86}\text{Sr}$	2 S.E.
Aug-10	1	WAs shark1	0.709210	0.000015	0.056529	0.000008	0.000127	0.000017
	2	WAs shark2	0.709194	0.000014	0.056503	0.000006	0.000271	0.000021
	3	WAs shark3	0.709191	0.000034	0.056481	0.000013	0.000465	0.000055
		WAs shark4**	0.709222	0.000014	0.056458	0.000010	0.001669	0.000016
	4	WAs shark5	0.709181	0.000020	0.056455	0.000018	0.001518	0.000031
	5	WAs shark6	0.709159	0.000033	0.056458	0.000025	0.000227	0.000024
Jan-11	10	WAs shark1	0.709182	0.000016	0.056500	0.000008	0.000305	0.000029
	11	WAs shark2	0.709180	0.000031	0.056532	0.000019	0.000264	0.000036
	12	WAs shark3	0.709156	0.000030	0.056516	0.000021	0.000200	0.000019
	13	WAs shark4	0.709148	0.000032	0.056543	0.000026	0.000273	0.000040
	14	WAs shark5	0.709202	0.000024	0.056513	0.000014	0.000194	0.000028
	15	WAs shark6	0.709161	0.000026	0.056536	0.000014	0.000204	0.000018
	16	WAs shark7	0.709179	0.000038	0.056571	0.000020	0.000283	0.000032
	17	WAs shark8	0.709163	0.000029	0.056498	0.000016	0.000204	0.000016
	18	WAs shark9	0.709178	0.000030	0.056509	0.000017	0.000170	0.000022
Apr-12	25	WAs shark1	0.709181	0.000026	0.056524	0.000010	0.000088	0.000009
	26	WAs shark2	0.709209	0.000019	0.056509	0.000008	0.000249	0.000028
	27	WAs shark3	0.709180	0.000021	0.056520	0.000012	0.000250	0.000037
	28	WAs shark4	0.709216	0.000018	0.056503	0.000008	0.000240	0.000043
Dec-12	35	WAs shark1	0.709147	0.000056	0.056464	0.000026	0.000375	0.000040
	36	WAs shark2	0.709105	0.000031	0.056515	0.000018	0.000672	0.000090
	37	SA shark1	0.709163	0.000024	0.056500	0.000017	0.000107	0.000013
	38	SA shark2	0.709154	0.000019	0.056495	0.000009	0.000175	0.000010
	39	SA shark3	0.709176	0.000020	0.056507	0.000011	0.000227	0.000016
Apr-13	45	SA shark1	0.709176	0.000021	0.056488	0.000010	0.000065	0.000015
	46	SA shark2	0.709231	0.000025	0.056490	0.000017	0.000165	0.000016
	47	SA shark3	0.709188	0.000019	0.056517	0.000013	0.000239	0.000014
	48	SA shark4	0.709225	0.000018	0.056486	0.000010	0.000093	0.000008
	49	SA shark5	0.709156	0.000019	0.056517	0.000010	0.000241	0.000011
	50	SA shark6	0.709178	0.000021	0.056484	0.000012	0.000099	0.000014
	51	SA shark7	0.709190	0.000021	0.056499	0.000012	0.000139	0.000012
	52	SA shark8	0.709190	0.000019	0.056507	0.000011	0.000089	0.000010
	53	SA shark9	0.709128	0.000027	0.056510	0.000016	0.000234	0.000038
	54	SA shark10	0.709131	0.000016	0.056499	0.000009	0.000241	0.000010
	55	SA shark11	0.709198	0.000020	0.056493	0.000009	0.000252	0.000013
	56	SA shark12	0.709167	0.000023	0.056480	0.000010	0.000180	0.000011
	57	SA shark13	0.709187	0.000034	0.056490	0.000018	0.000081	0.000020
	58	WAs shark1	0.709134	0.000016	0.056492	0.000009	0.001654	0.000034
	WAs shark2**	0.709099	0.000020	0.056496	0.000010	0.001473	0.000039	
59	WAs shark3	0.709145	0.000022	0.056487	0.000013	0.000958	0.000021	
60	WAs shark4	0.709166	0.000025	0.056515	0.000016	0.000201	0.000014	
61	WAs shark5	0.709151	0.000024	0.056504	0.000015	0.000219	0.000011	
62	WAs shark6	0.709196	0.000018	0.056506	0.000011	0.000638	0.000114	

Jul-13	70	SA shark1	0.709175	0.000039	0.056510	0.000018	0.000163	0.000029
	71	SA shark2	0.709176	0.000027	0.056496	0.000015	0.000215	0.000022
	72	SA shark3	0.709179	0.000022	0.056477	0.000011	0.000147	0.000017
	73	SA shark4	0.709221	0.000016	0.056461	0.000010	0.000118	0.000014
	74	SA shark5	0.709208	0.000019	0.056465	0.000011	0.000117	0.000012
	75	SA shark6	0.709160	0.000025	0.056492	0.000013	0.000198	0.000025
	76	SA shark7	0.709165	0.000061	0.056497	0.000033	0.000016	0.000033
	77	SA shark8	0.709141	0.000024	0.056502	0.000016	0.000142	0.000027
	78	SA shark9	0.709177	0.000021	0.056477	0.000009	0.000128	0.000012
	79	SA shark10	0.709170	0.000019	0.056488	0.000011	0.000114	0.000015
	80	SA shark11	0.709182	0.000021	0.056482	0.000009	0.000106	0.000012
	81	SA shark12	0.709184	0.000018	0.056488	0.000010	0.000092	0.000010
	82	SA shark13	0.709174	0.000019	0.056477	0.000009	0.000015	0.000009
	83	SA shark14	0.709167	0.000018	0.056488	0.000009	0.000086	0.000008
	84	SA shark15	0.709208	0.000020	0.056497	0.000011	0.000108	0.000009
	85	SA shark16	0.709192	0.000019	0.056495	0.000011	0.000141	0.000011
Apr-14	90	SA shark1	0.709178	0.000029	0.056506	0.000017	0.000064	0.000042
	91	SA shark2	0.709145	0.000059	0.056512	0.000017	0.000266	0.000085
	92	SA shark3	0.709140	0.000029	0.056528	0.000014	0.000315	0.000015
	93	SA shark4	0.709156	0.000028	0.056507	0.000016	0.000189	0.000028
	94	SA shark5	0.709128	0.000046	0.056527	0.000022	0.000141	0.000027
	95	SA shark6	0.709139	0.000023	0.056508	0.000016	0.000215	0.000027
	96	SA shark7	0.709156	0.000029	0.056492	0.000017	0.000179	0.000030
	97	WAs shark1	0.709128	0.000022	0.056484	0.000013	0.000317	0.000021
	98	WAs shark2	0.709130	0.000020	0.056507	0.000013	0.000256	0.000016
	99	WAs shark3	0.709118	0.000030	0.056523	0.000015	0.001725	0.000037
	100	SA shark8	0.709130	0.000022	0.056527	0.000016	0.000210	0.000014
	101	SA shark9	0.709173	0.000029	0.056511	0.000014	0.000136	0.000014
	102	SA shark10	0.709189	0.000023	0.056507	0.000013	0.000124	0.000014
		SA shark11**	0.709098	0.000030	0.056488	0.000018	0.000203	0.000025
	103	SA shark12	0.709165	0.000032	0.056518	0.000023	0.000116	0.000014
	104	SA shark13	0.709144	0.000037	0.056521	0.000022	0.000119	0.000019
	105	SA shark14	0.709133	0.000030	0.056504	0.000015	0.000223	0.000032
	106	SA shark15	0.709179	0.000044	0.056523	0.000022	0.000167	0.000020
	107	SA shark16	0.709148	0.000031	0.056503	0.000027	0.000150	0.000029
Sep-14	115	SA shark1	0.709161	0.000024	0.056482	0.000015	0.000129	0.000014
	116	SA shark2	0.709160	0.000024	0.056496	0.000012	0.000220	0.000020
	117	SA shark3	0.709177	0.000021	0.056467	0.000012	0.000155	0.000011
	118	SA shark4	0.709186	0.000021	0.056488	0.000011	0.000077	0.000010
	119	SA shark5	0.709211	0.000024	0.056488	0.000016	0.000088	0.000014
	120	SA shark6	0.709195	0.000026	0.056485	0.000012	0.000097	0.000018
	121	SA shark7	0.709153	0.000016	0.056505	0.000011	0.000214	0.000019
		SA shark8**	0.709239	0.000020	0.056473	0.000011	0.000100	0.000015
	122	SA shark9	0.709210	0.000019	0.056479	0.000012	0.000082	0.000009
	123	SA shark10	0.709208	0.000022	0.056495	0.000013	0.000129	0.000017
		average +/- 2 SD	0.709171	0.000053	0.056500	0.000040	0.000277	0.000711

Table S2 (ESI): Results of Rb-Sr isotopic analyses of aliquots of a virtually Rb-Sr-free, concentrated Ca-P solution (50 ppm Ca – 22 ppm P) to which varying amounts of SRM984 (Rb) and SRM987 (Sr) were added. This serves to emulate LA-MC-ICPMS apatite analysis and constrains the effective $^{85}\text{Rb}/^{87}\text{Rb}$ ratio used to correct for ^{87}Rb interference (cf. Fig. 5).

Aliquot number	$^{85}\text{Rb}/^{86}\text{Sr}_{\text{measured}}$	2 S.E.	$^{87}(\text{Sr}+\text{Rb})/^{86}\text{Sr}^1$	2 S.E.
1	0.002246	0.000008	0.711169	0.000022
2	0.002254	0.000008	0.711164	0.000022
3	0.000759	0.000002	0.710567	0.000016
4	0.000758	0.000003	0.710557	0.000016
5	0.000129	0.000001	0.710301	0.000015
6	0.000126	0.000001	0.710298	0.000015
7	0.000091	0.000001	0.710283	0.000014
8	0.011578	0.000003	0.714839	0.000016
9	0.005853	0.000003	0.712561	0.000016
10	0.061821	0.000004	0.734639	0.000016
11	0.129484	0.000021	0.761346	0.000018

¹...corrected for Sr mass bias ($^{86}\text{Sr}/^{88}\text{Sr}=0.1194^{21}$)

Table S3 (ESI): Sr(-Rb)-isotopic results ($^{87}\text{Sr}/^{86}\text{Sr}$, $^{84}\text{Sr}/^{86}\text{Sr}$ and $^{85}\text{Rb}/^{86}\text{Sr}$) from both LA-MC-ICPMS and comparative microsampling-TIMS of two archaeological teeth from the same individual excavated at the medieval mining community Sulzburg (Black Forest, SW Germany)^{37, 38}.

a) LA-MC-ICPMS:

SG85 36 (M1): LA-MC-ICPMS profile			
distance [mm]	$^{87}\text{Sr}/^{86}\text{Sr}$	$^{84}\text{Sr}/^{86}\text{Sr}$	$^{85}\text{Rb}/^{86}\text{Sr}$
0.042	0.706910	0.056417	0.009769
0.084	0.707127	0.056420	0.009076
0.126	0.706884	0.056431	0.008327
0.168	0.707138	0.056453	0.008377
0.210	0.706977	0.056564	0.007349
0.252	0.706858	0.056585	0.007855
0.295	0.707212	0.056395	0.007698
0.337	0.707143	0.056472	0.008369
0.379	0.707353	0.056394	0.009351
0.421	0.707183	0.056502	0.009738
0.463	0.707097	0.056427	0.010406
0.505	0.707503	0.056479	0.008613
0.547	0.707539	0.056448	0.007473
0.589	0.707717	0.056551	0.007847
0.631	0.707575	0.056661	0.007283
0.673	crack, no data		
0.715	crack, no data		
0.757	crack, no data		
0.799	crack, no data		
0.841	crack, no data		
0.884	crack, no data		
0.926	crack, no data		
0.968	crack, no data		
1.010	0.707263	0.056507	0.007097
1.052	0.707029	0.056440	0.006199
1.094	0.706977	0.056455	0.006328
1.136	0.706912	0.056565	0.006366
1.178	0.707378	0.056503	0.006231
1.220	0.707113	0.056534	0.007208
1.262	0.707643	0.056594	0.006455
1.304	0.708203	0.056362	0.006784
1.346	0.707931	0.056530	0.006516
1.388	0.707870	0.056491	0.006314
1.430	0.707658	0.056600	0.006268
1.473	0.707920	0.056474	0.006153
1.515	0.707872	0.056533	0.006492
1.557	0.707926	0.056493	0.006355
1.599	0.707766	0.056587	0.006885
1.641	0.707908	0.056538	0.007750
1.683	0.708049	0.056460	0.007158
1.725	0.707045	0.056561	0.006642
1.767	0.707308	0.056569	0.006340
1.809	0.707233	0.056474	0.006115
1.851	0.707189	0.056449	0.006193
1.893	0.706961	0.056447	0.005928
1.935	0.707218	0.056493	0.005880
1.977	0.707139	0.056441	0.005452
2.019	0.707219	0.056600	0.005630
2.062	0.707074	0.056429	0.005582
2.104	0.707345	0.056645	0.005474
2.146	0.707375	0.056562	0.005850
2.188	0.707813	0.056391	0.005348
2.230	0.707420	0.056367	0.005304
2.272	0.707115	0.056602	0.006114
2.314	0.707180	0.056553	0.005994
2.356	0.707093	0.056493	0.005698
2.398	0.707026	0.056656	0.006535
2.440	0.707277	0.056509	0.005973
2.482	0.707063	0.056604	0.005727
2.524	0.707119	0.056499	0.005612
2.566	0.707024	0.056341	0.005100

distance [mm]	$^{87}\text{Sr}/^{86}\text{Sr}$	$^{84}\text{Sr}/^{86}\text{Sr}$	$^{85}\text{Rb}/^{86}\text{Sr}$
2.608	0.707075	0.056503	0.005291
2.651	0.707091	0.056539	0.005107
2.693	0.707138	0.056510	0.004979
2.735	0.707467	0.056522	0.004758
2.777	0.707388	0.056406	0.004394
2.819	0.707355	0.056410	0.004794
2.861	0.707451	0.056405	0.004440

SG85 48 (M3): LA-MC-ICPMS profile			
distance [mm]	⁸⁷ Sr/ ⁸⁶ Sr	⁸⁴ Sr/ ⁸⁶ Sr	⁸⁵ Rb/ ⁸⁶ Sr
0.042	0.713300	0.056600	0.007245
0.084	0.713185	0.056662	0.007277
0.126	0.713518	0.056571	0.007308
0.168	0.713272	0.056470	0.006996
0.210	0.713413	0.056473	0.006602
0.252	0.713849	0.056736	0.006594
0.295	0.714518	0.056512	0.006489
0.337	0.713717	0.056611	0.006105
0.379	0.713820	0.056414	0.005942
0.421	0.713583	0.056578	0.006105
0.463	0.714864	0.056447	0.005953
0.505	0.714707	0.056523	0.006151
0.547	0.715466	0.056186	0.006201
0.589	0.715096	0.056561	0.005972
0.631	0.715014	0.056622	0.005902
0.673	0.715269	0.056452	0.005873
0.715	0.715052	0.056671	0.005962
0.757	0.714683	0.056542	0.005683
0.799	0.714906	0.056435	0.005712
0.841	0.714767	0.056566	0.005886
0.884	0.714952	0.056428	0.005735
0.926	0.714668	0.056310	0.005533
0.968	0.714733	0.056469	0.005600
1.010	0.714858	0.056521	0.005549
1.052	0.714947	0.056657	0.005484
1.094	0.714802	0.056387	0.005291
1.136	0.714535	0.056600	0.005631
1.178	0.715018	0.056595	0.005404
1.220	0.714920	0.056508	0.005508
1.262	0.715312	0.056440	0.005349
1.304	0.715053	0.056501	0.005458
1.346	0.715245	0.056534	0.005547
1.388	0.714823	0.056709	0.005679
1.430	0.714944	0.056496	0.005511
1.473	0.714836	0.056721	0.005499
1.515	0.714787	0.056377	0.005368
1.557	0.715231	0.056340	0.005103
1.599	0.714475	0.056462	0.005221
1.641	0.714810	0.056619	0.005128
1.683	0.714926	0.056402	0.005259
1.725	0.714772	0.056676	0.005333
1.767	0.715486	0.056615	0.005424
1.809	0.715453	0.056667	0.005248
1.851	0.715252	0.056501	0.005051
1.893	0.714773	0.056428	0.005012
1.935	0.714933	0.056495	0.005099
1.977	0.715412	0.056517	0.005116
2.019	0.715444	0.056404	0.005220
2.062	0.715285	0.056505	0.005204
2.104	0.714794	0.056446	0.005199
2.146	0.715420	0.056647	0.004921
2.188	0.715286	0.056544	0.005241
2.230	0.714845	0.056711	0.005139
2.272	0.715030	0.056509	0.005041
2.314	0.715164	0.056759	0.005104
2.356	0.715204	0.056646	0.005025
2.398	0.714850	0.056487	0.004809
2.440	0.714998	0.056565	0.004734
2.482	0.714914	0.056429	0.004721
2.524	0.715127	0.056730	0.004868
2.566	0.715799	0.056644	0.005132
2.608	0.716225	0.056377	0.005413
2.651	0.716394	0.056642	0.005610
2.693	0.716334	0.056684	0.005545
2.735	0.716478	0.056505	0.005527
2.777	0.716675	0.056557	0.005813

distance [mm]	$^{87}\text{Sr}/^{86}\text{Sr}$	$^{84}\text{Sr}/^{86}\text{Sr}$	$^{85}\text{Rb}/^{86}\text{Sr}$
2.819	0.715416	0.056642	0.005381
2.861	0.715022	0.056499	0.005112
2.903	0.714561	0.056566	0.004784
2.945	0.714719	0.056590	0.004825
2.987	0.714818	0.056348	0.004846
3.029	0.714343	0.056625	0.004732
3.071	0.714515	0.056510	0.004454
3.113	0.714533	0.056529	0.004742
3.155	0.714951	0.056595	0.004926
3.198	0.714968	0.056550	0.005241
3.240	0.714978	0.056579	0.005464
3.282	0.714823	0.056390	0.005943
3.324	0.714672	0.056741	0.005748
3.366	0.714790	0.056400	0.005646
3.408	0.714749	0.056564	0.006019
3.450	0.714978	0.056542	0.005506
3.492	0.714717	0.056632	0.005372
3.534	0.714689	0.056531	0.005453
3.576	0.714917	0.056408	0.005431
3.618	0.714433	0.056393	0.005351
3.660	0.714384	0.056312	0.005171
3.702	0.714442	0.056644	0.005338
3.744	0.714899	0.056512	0.005388
3.787	0.714696	0.056516	0.005910
3.829	0.714985	0.056746	0.006992
3.871	0.714698	0.056556	0.007115
3.913	0.715089	0.056612	0.006827
3.955	0.715168	0.056456	0.007003
3.997	0.715156	0.056487	0.006885
4.039	0.714993	0.056629	0.006637
4.081	0.714544	0.056680	0.006588
4.123	0.714270	0.056342	0.006378
4.165	0.713748	0.056474	0.005939
4.207	0.714212	0.056651	0.006235
4.249	0.714573	0.056617	0.006728
4.291	0.714675	0.056761	0.006838
4.333	0.714796	0.056246	0.006925
4.376	0.714449	0.056447	0.007532
4.418	0.713945	0.056771	0.007588
4.460	0.713460	0.056456	0.007547
4.502	0.713505	0.056504	0.006891
4.544	0.713322	0.056503	0.007051
4.586	0.713401	0.056475	0.007709
4.628	0.713766	0.056479	0.008354
4.670	0.714332	0.056463	0.008312
4.712	0.714252	0.056561	0.008497
4.754	0.714332	0.056506	0.008589
4.796	0.714323	0.056344	0.008244
4.838	0.713960	0.056333	0.008525
4.880	0.714523	0.056380	0.008888
4.922	0.714013	0.056866	0.008660
4.965	0.713212	0.056487	0.007940
5.007	0.712575	0.056405	0.007164
5.049	0.712624	0.056603	0.007265
5.091	0.713513	0.056628	0.007536
5.133	0.713564	0.056667	0.007586
5.175	0.713594	0.056486	0.007351
5.217	0.713688	0.056468	0.006422
5.259	0.713341	0.056457	0.006160
5.301	0.713084	0.056589	0.006218
5.343	0.713144	0.056510	0.005963
5.385	0.713204	0.056537	0.006042
5.427	0.712920	0.056436	0.006123
5.469	0.712884	0.056519	0.005566
5.511	0.712268	0.056597	0.004957
5.554	0.712489	0.056453	0.005188
5.596	0.712803	0.056547	0.005604

distance [mm]	$^{87}\text{Sr}/^{86}\text{Sr}$	$^{84}\text{Sr}/^{86}\text{Sr}$	$^{85}\text{Rb}/^{86}\text{Sr}$
5.638	0.712797	0.056583	0.005738
5.680	0.712973	0.056389	0.005442
5.722	0.712669	0.056374	0.005470

b) Microsampling-TIMS:

Sample ID	mass [µg]	$^{87}\text{Sr}/^{86}\text{Sr}$	2 SE
SG85 36	720	0.707311	0.000018
SG85 48a	350	0.713718	0.000034
SG85 48c	160	0.712894	0.000033