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Anthropogenic lead as a tracer of rock varnish growth: Implications for rates of formation

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SUPPLEMENTAL BACKGROUND

Rock varnish is an enigmatic feature, the origin of which has been argued for centuries. The first scientific descriptions of rock varnish and its possible biogenic origin date as far back in the literature as Alexander von Humboldt (1852), demonstrating that the deposition of Mn and Fe coatings on rocks has occupied the minds of many generations of scientists (cf. Krumbein and Jens, 1981). It is present on the surface of rocks in nearly all types of terrestrial environments (Dorn, 1991, 2007a, b). However, it is most common and thickest in arid regions, where it is also called desert varnish. In these areas, it occurs as a veneer of manganese and/or iron oxides interlaminated with clays and/or silica, the whole coating rarely exceeding 200 μm thick (Liu and Broecker, 2000. Although the exact mechanisms of rock varnish formation remain controversial, most workers agree on some combination of airborne dust to supply materials and microbial action to concentrate manganese, modified by considerable diagenesis (Potter and Rossman, 1977; Nagy et al., 1991; Krinsley, 1998; Liu et al., 2000; Lee and Bland, 2003; Perry et al., 2006; Dorn, 2007a, b; Dietzel et al., 2008; Garvie et al., 2008; Northup et al., 2010).

Rock varnishes have attracted considerable research interest as a potential Quaternary dating tool for rock surfaces (e.g. Dorn and Oberlander, 1982); as archeological features and artifacts (e.g. VandenDolder, 1992); as indicators of paleoclimatic change (e.g. Dorn, 1994; Liu et al., 2000; Lui and Broecker, 2000; 2007); as environmental monitors because of the great scavenging abilities of Mn-oxides for certain heavy metals such as lead (Dorn, 1991; Fleisher et al., 1999; Wayne et al., 2006); because of the likely role of microbes in their formation (e.g. Perry and Adams, 1978; Krumbein and Jens, 1981; Nagy et al., 1991; Gorbushina, 2007); as analogous environs for the search for life on other planets (DiGregorio, 2002; Gorbushina et al., 2002; Allen et al., 2004; Edwards, 2004); and to assist in interpretations of remote sensing studies of varnished rock surfaces on Earth and Mars (e.g. Israel et al., 1997;

Kraft and Greeley, 2000). Archeologists are interested in dating the age of varnishes in order to place petroglyphs carved into varnish by ancient cultures into their full historical context (Stasack et al., 1996; Dietzel et al., 2008). Geomorphologists have attempted to date landforms and surfaces using the age of desert varnish on rocks present on these land surfaces (Friend et al., 2000; French and Guglielmin, 2002). Early suggestions that the thickness of varnish indicated age have been thoroughly discredited (Liu and Broecker, 2000; Dorn, 2007). Attempts to directly date varnish include the cation-ratio method of Dorn (1983, 1989) and various radiogenic methods (Phillips et al. 1991; Watchman, 2000; Dorn, 1996, 1997). However, radiometric dating of varnish itself has not proven possible because varnish contains little carbon and some is recycled (Dorn 1996; Broecker and Liu, 2001), and its near-unity ratio of $^{232}Th/^{238}U$ activity precludes ^{230}Th dating (Broecker and Liu, 2001). Likewise, the cation-ratio method of Dorn (1983) has been shown to have significant problems (Reneau and Raymond 1991; Bierman and Gillespie, 1994).

The best constraints on the age of rock varnish have come from dating the rocks beneath the varnish such as lava flows (Liu, 2003, Phillips, 2003) or associated carbonates (Dragovich, 1988; Liu et al., 2000). Using these constraints, Liu et al. (2000) suggest that rock varnish may provide a record of paleoclimate. Variations in Ba and Mn concentrations, which produce color banding in ultrathin sections examined by transmitted light, are believed to reflect fluctuations in wetness of the environment and thus provide a proxy for climate change (Liu et al. 2000, Broecker and Liu, 2001; Liu and Dorn 1996). Manganese-rich layers are believed to have formed during wetter periods (Broecker and Liu, 2001), and the microstratigraphy has been correlated to climatological records in the southwestern U.S. (Liu et al., 2000; Friend et al., 2000; Liu and Broecker, 2007, 2008), Australia (Lee and Bland, 2003) and the Sahara (Zerboni, 2008).

However, although the microstratigraphy can be correlated on a regional scale (Liu and Broecker, 2000), on a single rock surface, varnish may exhibit variations in overall thickness and composition of microlayers, from hundreds of micrometers in thickness in microbasins and depressions to near nonexistence on neighboring surfaces, all on a scale of millimeters to centimeters laterally. Such extreme spatial change suggests variability in accretion rates, coupled with variable removal, temporally, by abrasion, spalling, compaction, and chemical leaching that may reduce the original thickness (Krinsley, 1998, Dragovich, 1988, 1994; Liu and Broecker, 2000).

Using careful dating of the underlying surface, Liu and Broecker (2000) document varnish growth rates of 1 to 40 μm/ky in the southwest U.S. The oldest samples have the slowest rates, with all samples ≥ 50 ka having rates ≤ 2 μm/ky, while the youngest sample (1.5 ka) has the fastest rate of 40 μm/ky (Liu and Broecker, 2000). Some workers have focused on the lower end of this range, citing a few microns/ky (Broecker and Liu, 2001; Dorn 2007a,b; Krinsley et al., 2009); while others cite the entire range (Kuhlman et al., 2006, 2008; Liu and Broecker, 2008). Dorn and Meek (1995) note varnish formation on iron slags only 40 years old. These rapid growth rates have important implications for the formation of rock varnish.

LEAD IN ROCK VARNISH

Elevated lead levels in rock varnish have been reported by a number of workers. Dorn (1998) first reported lead concentrations heavy-metal concentrations in rock varnish. Fleischer et al. (1999), confirmed by Hodge et al. (2005), report trace amounts of ^{210}Pb, a radiogenic isotope with a half-life of only 22 years, in Great Basin (U.S.A.) rock varnish. Thiagarajan and Lee (2004) report elevated Pb values of up to 1257 ppm (0.1257 wt% Pb). These studies attribute the elevated Pb values to atmospheric sources, either natural or anthropogenic. Wayne et al. (2007) found values of up to 0.296 wt% Pb in the surface layers of varnish from the Four Corners region (U.S.A.) and attributed the high values to proximity to several large coal-fired power plants. Broecker and Liu (2001) report up to 20000 ppm (2 wt%) Pb in the outermost 10-20 μm of a varnish from eastern California. They attribute the 20 μm depth as an artifact and instead consider the lead to be from leaded gasoline and contained in the outer 1 μm of the varnish. Fleischer et al. (1999) also report a 10-fold enrichment (no numbers given) in lead from eastern California which they attribute to anthropogenic input from unidentified smelters. In all of these studies, the lead is considered to be in the outermost layers and is attributed to anthropogenic sources.

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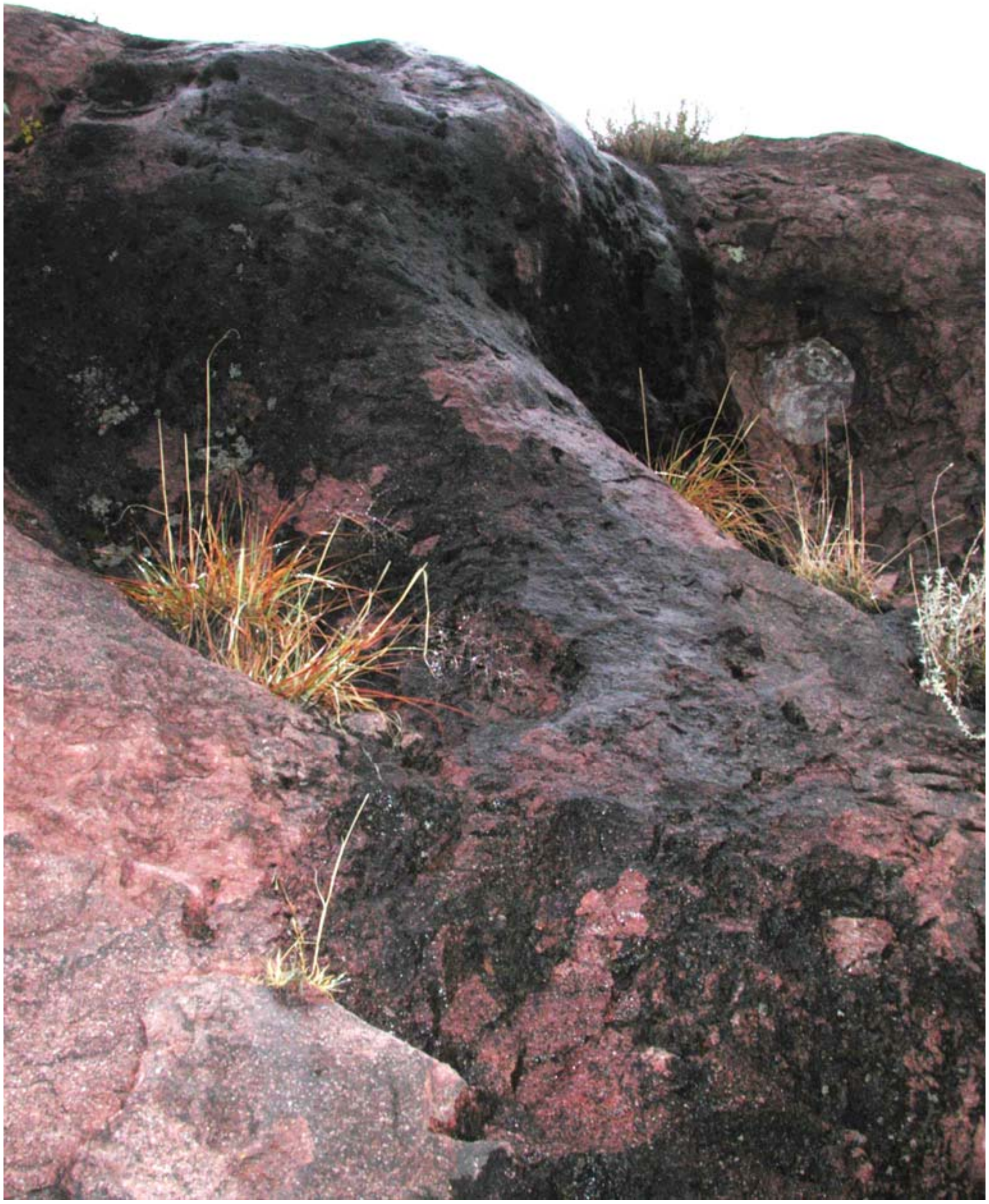

Figure DR1. Discontinuous rock varnish on an outcrop of rhyolite bedrock at the Socorro sampling site number 2.

Figure DR2. Fragments of reddish-colored rhyolite covered with rock varnish from Socorro Sites 1 and 2. The upper piece (about 3 cm in length) exhibits patchy varnish whereas the other rock fragments display heavy black coatings.

TABLE DR1. ELECTRON MICROPROBE TRAVERSE ACROSS BOTRYOIDAL VARNISH SAMPLE, FROM INNER EDGE TO OUTER RIM, SOCORRO SITE 2

No.	Distance (μm)	Na2O	MgO	Al2O3	SiO2	K2O	CaO	MnO2	Fe2O3	ZnO	BaO	PbO	Total
1	0.0	0.049	1.747	20.189	29.208	1.803	0.954	22.743	12.701	0.217	0.910	0.000	90.604
2	2.0	0.052	1.893	19.874	30.740	1.702	0.853	15.278	21.813	0.183	0.749	0.016	93.216
3	4.0	0.070	2.035	19.732	29.558	2.072	0.947	21.672	15.299	0.213	1.198	0.004	92.847
4	7.1	0.088	1.555	11.759	21.249	1.403	1.508	46.092	7.106	0.533	4.519	0.000	95.851
5	9.1	1.541	1.179	10.842	21.026	1.115	1.703	45.415	5.072	0.454	4.521	0.003	92.929
6	12.0	0.054	1.574	16.478	22.877	1.350	1.347	34.863	11.722	0.311	2.158	0.008	92.816
7	15.0	0.094	1.343	13.475	20.338	1.331	1.877	42.060	10.019	0.434	3.476	0.015	94.508
8	17.0	0.038	1.445	16.964	27.929	1.238	1.331	34.734	4.481	0.429	3.116	0.021	91.780
9	20.1	0.066	1.882	18.443	28.441	1.747	0.796	22.090	13.873	0.248	1.074	0.012	88.727
10	22.1	0.066	1.652	18.487	25.201	1.683	0.951	31.376	11.143	0.283	1.877	0.039	92.871
11	24.1	0.042	1.467	18.779	24.931	1.617	0.941	28.194	11.969	0.292	1.198	0.065	89.592
12	28.1	0.036	1.474	17.420	23.640	1.576	1.059	34.911	9.943	0.324	1.720	0.033	92.241
13	30.1	0.068	1.447	15.906	22.172	1.609	1.097	39.983	7.921	0.410	3.204	0.085	94.029
14	32.1	0.066	1.748	17.871	26.190	1.721	0.980	31.049	8.972	0.364	1.421	0.019	90.479
15	35.1	0.068	1.432	15.324	22.496	1.597	1.332	40.154	7.890	0.468	2.486	0.024	93.344
16	37.1	0.062	1.302	13.137	19.845	1.366	1.580	46.288	6.122	0.588	3.410	0.025	93.766
17	40.1	0.084	1.294	12.677	17.847	1.342	1.641	47.749	6.888	0.736	3.289	0.037	93.653
18	43.1	0.033	2.077	19.654	28.643	1.796	0.968	26.307	10.886	0.224	1.020	0.019	91.734
19	45.2	0.071	1.568	17.503	23.701	1.570	1.250	35.177	10.315	0.400	1.829	0.041	93.450
20	48.2	0.093	1.127	12.926	16.094	1.153	1.626	48.085	8.446	0.526	3.232	0.050	93.425
21	50.2	0.078	1.229	13.105	17.710	1.261	1.579	45.405	9.407	0.566	3.049	0.015	93.493
22	52.2	0.095	1.590	18.462	27.445	1.624	0.789	22.229	17.136	0.173	0.762	0.048	90.437
23	55.2	0.054	1.830	19.148	29.377	1.717	0.828	21.848	15.738	0.203	0.762	0.036	91.618
24	57.2	0.088	1.380	16.956	23.035	1.504	1.196	35.771	10.089	0.399	2.636	0.027	93.163
25	60.2	0.085	1.231	12.597	18.468	1.194	1.595	45.954	7.925	0.551	3.165	0.028	92.837
26	63.2	0.055	1.738	19.256	27.953	1.665	0.984	27.091	11.094	0.235	1.127	0.022	91.318
27	65.2	0.055	1.696	18.865	27.524	1.745	0.907	26.546	13.672	0.226	1.109	0.045	92.470
28	67.3	0.057	1.391	15.255	22.569	1.448	1.243	37.710	9.445	0.454	2.518	0.034	92.217
29	70.3	0.095	1.511	13.876	20.385	1.278	1.240	47.046	5.817	0.784	3.746	0.004	95.808
30	72.2	0.027	1.884	16.787	34.422	1.636	0.702	18.336	15.303	0.181	0.685	0.027	90.041
31	75.2	0.040	1.621	18.551	33.303	1.783	0.746	20.321	13.610	0.152	0.960	0.017	91.147
32	78.2	0.052	1.613	15.446	47.313	1.559	0.612	17.360	11.194	0.143	1.042	0.004	96.377
33	80.2	0.062	1.807	17.446	32.336	1.635	0.896	23.932	12.820	0.222	2.366	0.021	93.606
34	83.2	0.080	0.951	13.042	14.588	1.162	1.244	42.772	7.085	0.358	10.940	0.051	92.352
35	85.2	0.092	0.822	8.245	9.578	1.097	1.808	58.797	6.142	0.634	5.820	0.040	93.136
36	87.3	0.116	0.899	8.060	9.230	0.986	2.091	61.543	5.322	0.808	5.491	0.000	94.591
37	90.3	0.081	1.581	15.523	22.792	1.725	1.129	37.725	9.243	0.349	2.458	0.018	92.669
38	92.3	0.090	2.921	15.809	24.508	2.154	0.918	31.374	9.537	0.188	2.321	0.034	89.913
39	95.3	0.101	1.157	13.135	18.378	1.544	1.230	42.290	9.899	0.355	2.848	0.024	91.051
40	98.2	0.074	1.179	12.237	16.882	1.330	1.388	46.442	8.600	0.474	3.011	0.021	91.704
41	100.3	0.073	1.518	16.223	24.378	1.626	1.031	33.839	10.771	0.284	1.808	0.018	91.628
42	102.3	0.077	1.368	16.583	25.924	1.942	1.012	31.626	11.372	0.193	1.850	0.021	92.025
43	105.3	0.047	3.044	16.318	42.106	2.680	0.598	17.211	7.951	0.185	0.976	0.015	91.186
44	107.3	0.047	3.299	14.752	51.057	2.014	0.713	20.152	6.659	0.191	1.359	0.000	100.261
45	110.3	0.045	1.655	18.555	27.615	1.637	0.858	28.948	9.609	0.301	1.191	0.000	90.499
46	113.4	0.046	1.785	21.147	27.507	1.656	0.808	25.924	11.626	0.227	0.892	0.007	91.722
47	115.4	0.050	1.528	17.187	22.607	1.554	1.355	36.187	8.785	0.423	1.666	0.000	91.399
48	118.3	0.068	1.578	18.550	27.811	2.844	1.453	29.706	5.849	0.420	1.704	0.000	90.034
49	120.3	0.088	2.007	15.931	29.915	1.762	1.173	28.402	10.047	0.271	1.153	0.038	90.840
50	123.3	0.058	1.319	17.030	21.182	1.615	1.338	36.851	9.563	0.441	1.544	0.026	91.071
51	126.4	0.092	1.106	12.597	16.879	1.315	1.940	51.856	6.416	0.807	3.038	0.035	96.130

TABLE DR1. (CONT)

No.	Distance (um)	Na2O	MgO	Al2O3	SiO2	K2O	CaO	MnO2	Fe2O3	ZnO	BaO	PbO	Total
52	128.4	0.076	1.665	16.742	27.009	1.831	1.161	28.778	9.423	0.270	1.177	0.005	88.167
53	131.4	0.023	0.289	3.693	89.003	0.385	0.130	2.672	1.090	0.000	0.123	0.000	97.408
54	133.4	0.077	1.215	11.567	56.931	1.936	0.697	14.917	5.190	0.190	0.685	0.009	93.441
55	135.4	0.060	1.544	14.631	34.115	1.389	1.214	31.405	8.311	0.385	1.924	0.003	95.007
56	138.4	0.088	1.678	16.476	22.125	1.425	1.313	34.343	9.942	0.337	1.410	0.029	89.204
57	141.4	0.066	1.813	16.054	21.789	1.661	1.244	31.126	12.049	0.262	1.255	0.013	87.400
58	143.4	0.043	1.160	10.359	57.108	1.474	0.675	14.711	6.241	0.111	0.398	0.034	92.339
59	146.4	0.008	0.495	4.083	82.152	0.524	0.170	2.751	1.841	0.002	0.163	0.000	92.211
60	148.5	0.070	2.344	17.718	33.241	2.029	0.925	26.976	7.305	0.275	0.687	0.000	91.597
61	150.5	1.190	0.817	20.651	50.359	7.341	0.765	12.071	4.001	0.108	0.632	0.000	97.939
62	153.5	1.554	0.737	16.600	48.785	6.618	0.813	13.702	3.877	0.093	1.010	0.000	93.822
63	155.5	0.070	1.457	16.263	28.447	1.839	1.630	31.087	9.078	0.386	1.361	0.012	91.674
64	158.5	0.034	1.024	17.502	33.291	1.813	1.021	22.603	9.951	0.253	0.689	0.006	88.224
65	161.5	0.044	1.883	14.356	58.404	1.764	0.629	13.765	7.649	0.170	0.386	0.029	99.118
66	163.5	0.097	2.091	19.151	35.129	2.087	0.832	21.771	8.816	0.311	0.712	0.000	91.054
67	166.5	0.071	1.594	17.625	27.974	2.012	1.268	33.169	7.635	0.582	1.804	0.032	93.809
68	168.6	0.098	1.416	11.423	18.378	1.365	1.824	49.029	5.269	1.239	3.043	0.091	93.213
69	170.6	0.060	1.797	16.540	27.681	1.589	1.065	29.223	10.751	0.378	0.813	0.410	90.365
70	173.6	0.058	2.282	17.688	33.736	2.292	0.777	23.661	7.233	0.471	0.394	0.701	89.368
71	175.6	0.112	1.940	16.910	31.958	1.665	1.083	27.106	9.132	0.482	0.516	0.986	91.933
72	178.5	0.039	1.932	18.378	35.657	1.890	0.787	20.928	7.153	0.385	0.625	0.996	88.863
73	181.6	0.074	2.284	18.345	33.920	2.738	1.282	20.957	8.019	0.299	0.433	0.792	89.300
74	183.6	0.100	1.602	14.301	24.061	1.747	1.747	35.919	6.047	0.915	1.108	1.893	89.671
75	185.6	0.144	0.882	9.195	14.473	1.148	2.074	54.629	4.477	1.659	2.706	1.195	92.706
76	188.6	0.135	1.068	10.593	17.877	1.321	1.859	46.687	4.130	1.694	2.301	1.273	89.174
77	190.6	0.116	0.804	8.848	13.193	1.350	2.177	53.373	4.440	1.102	2.187	1.616	89.576
78	193.7	0.226	0.752	7.730	12.369	2.035	2.422	55.197	3.614	0.853	2.052	3.691	91.269
79	196.7	0.124	0.979	9.107	15.299	1.313	2.569	55.486	3.656	1.050	2.168	0.852	92.861

TABLE DR2. ELECTRON MICROPROBE TRAVERSE ACROSS LAYERED VARNISH SAMPLE, FROM INNER EDGE TO OUTER RIM, SOCORRO SITE 2

No.	Distance (μm)	Na2O	MgO	Al2O3	SiO2	K2O	CaO	MnO2	Fe2O3	ZnO	BaO	PbO	Total
1	0.0	0.09	1.69	16.70	24.97	1.65	1.00	26.88	12.40	0.18	1.00	0.03	86.65
2	1.0	0.06	1.56	16.19	24.03	1.33	1.21	32.04	12.88	0.16	1.19	0.01	90.70
3	2.0	0.08	1.73	15.61	23.10	1.55	1.12	32.30	13.22	0.16	1.41	0.06	90.38
4	3.2	0.09	1.76	18.62	25.65	1.92	1.03	27.70	12.29	0.23	1.18	0.05	90.53
5	4.1	0.08	1.68	19.53	27.07	1.58	1.03	31.01	9.57	0.30	1.61	0.00	93.49
6	5.4	0.07	1.89	20.98	30.26	1.35	0.92	27.40	8.01	0.26	1.34	0.04	92.53
7	6.3	0.07	1.91	17.92	26.64	1.78	0.95	28.44	13.23	0.23	1.26	0.01	92.53
8	7.3	0.09	1.71	16.07	23.17	2.10	1.02	30.32	12.99	0.24	1.84	0.03	89.64
9	8.2	0.14	1.13	13.12	17.42	1.71	1.24	40.78	11.83	0.41	1.95	0.02	89.80
10	9.2	0.23	1.50	15.45	26.66	2.48	1.18	33.80	9.29	0.41	1.65	0.00	92.70
11	10.4	0.19	1.41	14.61	21.85	2.20	1.27	39.88	9.47	0.38	2.91	0.01	94.20
12	11.4	0.19	1.29	13.70	17.58	1.99	1.33	43.51	10.42	0.36	2.31	0.00	92.74
13	12.4	0.14	1.44	14.84	19.88	2.18	1.21	39.19	10.71	0.26	2.08	0.06	92.05
14	13.3	0.10	1.52	15.74	21.94	2.27	1.13	34.90	11.66	0.20	1.85	0.05	91.45
15	14.6	0.12	1.54	16.53	23.22	2.14	1.05	32.20	11.87	0.20	1.84	0.06	90.85
16	15.5	0.08	1.59	17.16	23.57	2.09	1.05	34.18	12.09	0.16	1.61	0.06	93.72
17	16.5	0.08	1.39	15.90	22.53	2.12	1.11	37.40	11.00	0.26	1.66	0.03	93.56
18	17.5	0.09	1.08	13.73	19.51	1.80	1.20	44.77	8.07	0.42	2.21	0.05	93.01
19	18.4	0.10	1.03	12.27	17.64	1.81	1.33	50.08	6.74	0.51	2.65	0.03	94.22
20	18.4	0.13	1.02	12.48	17.49	1.83	1.42	49.24	6.05	0.53	2.70	0.02	92.95
21	19.4	0.12	1.06	12.03	17.51	1.96	1.56	49.54	6.03	0.57	2.98	0.03	93.43
22	20.6	0.15	1.22	12.12	17.81	2.02	1.49	47.65	6.44	0.56	2.87	0.01	92.38
23	22.6	0.14	0.95	10.60	14.86	1.91	1.41	47.74	6.04	0.62	2.64	0.03	87.02
24	23.5	0.13	1.16	11.46	17.14	1.99	1.71	51.10	5.86	0.72	3.27	0.00	94.57
25	24.5	0.12	1.22	13.07	18.87	2.11	1.50	48.02	6.26	0.53	2.52	0.02	94.30
26	25.7	0.09	1.34	15.51	21.42	2.01	1.20	42.03	7.03	0.43	1.91	0.00	93.00
27	25.7	0.09	1.40	15.45	21.15	2.00	1.09	40.62	8.00	0.39	1.62	0.00	91.87
28	26.7	0.09	1.01	13.40	17.81	1.85	1.08	40.73	7.82	0.44	1.69	0.04	86.00
29	27.7	0.13	1.25	12.63	18.15	1.95	1.55	48.27	5.79	0.66	2.65	0.01	93.09
30	29.6	0.11	1.31	13.18	19.72	2.11	1.49	45.50	6.24	0.69	2.34	0.02	92.74
31	30.8	0.12	1.22	13.92	20.23	1.88	1.28	44.11	6.57	0.68	2.24	0.26	92.52
32	31.8	0.06	1.49	16.13	25.38	1.76	1.00	34.67	8.07	0.45	1.53	1.80	92.41
33	32.8	0.05	1.48	14.78	25.91	1.69	0.93	31.63	9.94	0.25	0.90	2.32	90.00
34	32.8	0.06	1.24	12.00	19.10	1.56	1.29	41.03	8.74	0.28	1.04	3.44	90.00
35	34.0	0.09	1.01	7.84	13.24	1.24	1.27	29.37	5.18	0.23	0.73	2.12	62.49
36	34.9	0.05	0.59	4.02	6.98	0.76	0.74	16.93	2.60	0.15	0.44	1.37	34.71