

Electronic Supplementary Information

In-situ characterization of aggregates of nanoscale zero-valent iron (nZVI) in water: an engineering aspect

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Table of Contents:

1. Figure captions

2. Figures

3. Table

Figure captions

Figure S1 (A) The cone-shaped mixing tank; (B) a schematic of the tank dimension.

Figure S2 Principle diagram of the Turbiscan stability analyzer (A). Gravitational compression of the 50 g/L nZVI slurry in the Turbiscan vial: photo (B1) and the Turbiscan spectra in one hour (B2) and in four days (C). Change of the height of the interface during a 96-h gravitational compression: the three nZVI slurries have initial nZVI concentrations at 40, 50 and 60 g/L, respectively (D). The interface height was deduced from the Turbiscan spectra.

Figure S3 PSDs of nZVI aggregates in aqueous solution at different mixing conditions (nZVI concentrations range from 1 ppm to 500 ppm).

Figure S4 The raw optical images of the settling nZVI aggregates.

Figure S5 The d_{\max} -AR plot (A) and the concentration-AR average (AR_{ave}) plot (B) of the settling aggregates.

Figure S6 Deduction process of eqn (11).

Figure S1

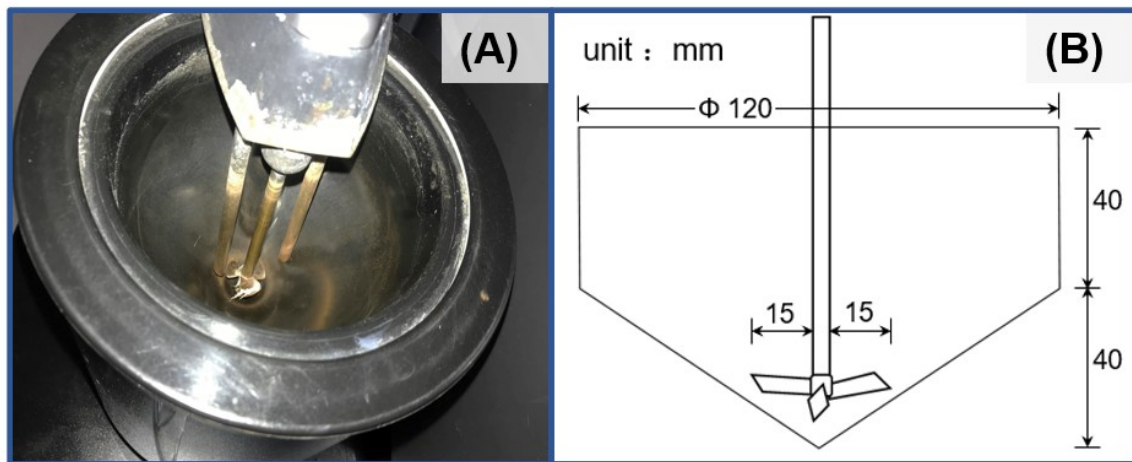
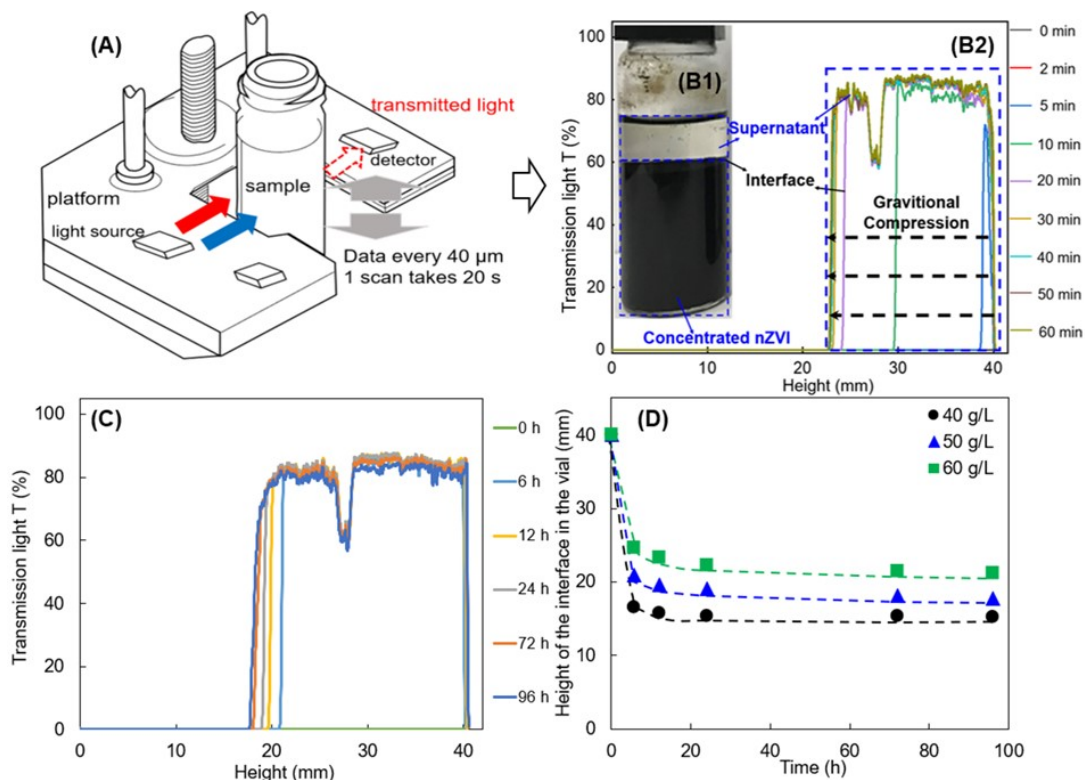


Figure S2



Study of the gravitational compression. nZVI aggregates during gravitational compression were studied using Turbiscan stability analyzer, of which principle is shown in Figure S2A. In brief, the stability analyzer measures the transmitted light (T, %) passing through the sample slurry in a 20-mL cylindrical glass sample vial (Figure S2A and S2B1) that has an effective height of 40 mm. In the analyzer, the vial is scanned using a moving light source ($\lambda = 880 \text{ nm}$); the light passing through the slurry is recorded by a detector that moves synchronously with the light source. The light source and the detector move along the vial and one T value is acquired at every $40 \mu\text{m}$ during the moving. One entire scan from the vial bottom to the top takes 20 s and is repeated at the preset intervals. DI water in the vial was used as

a blank, which had an average T of ~90%. Scanning temperature was set at 20 °C.

To study the gravitational compression of nZVI, a 20-mL nZVI slurry, at the nZVI concentrations of 40 g/L, 50 g/L and 60 g/L respectively, was first injected into the vial; the vial was then sonicated for 10 s and inserted into the machine. A steep T line was observed immediately after the scan started, e.g., at 10 min at the height of ~30 mm (Figure S2B2): right to the steep line was the part of high T value (~80%) that corresponded to the supernatant in the vial top; left to the steep line was the part of the zero T that corresponded to the formed concentrated nZVI at the vial bottom; the steep line corresponded to the interface between the supernatant and the concentrated nZVI. The steep line kept moving to the left (the vial bottom) during the settling: for example, the steep line of the 50 g/L slurry reached at 23 mm at 40 min (Figure S2B2) and 18 mm at 72 h (Figure S2C). This suggested that the concentrated nZVI at the bottom was shrinking in volume and the compression occurred. From the height of the steep line, volume of the concentrated nZVI was deduced and then used for the calculation of the volume per unit mass, the bulk density, and the porosity.

Figure S3

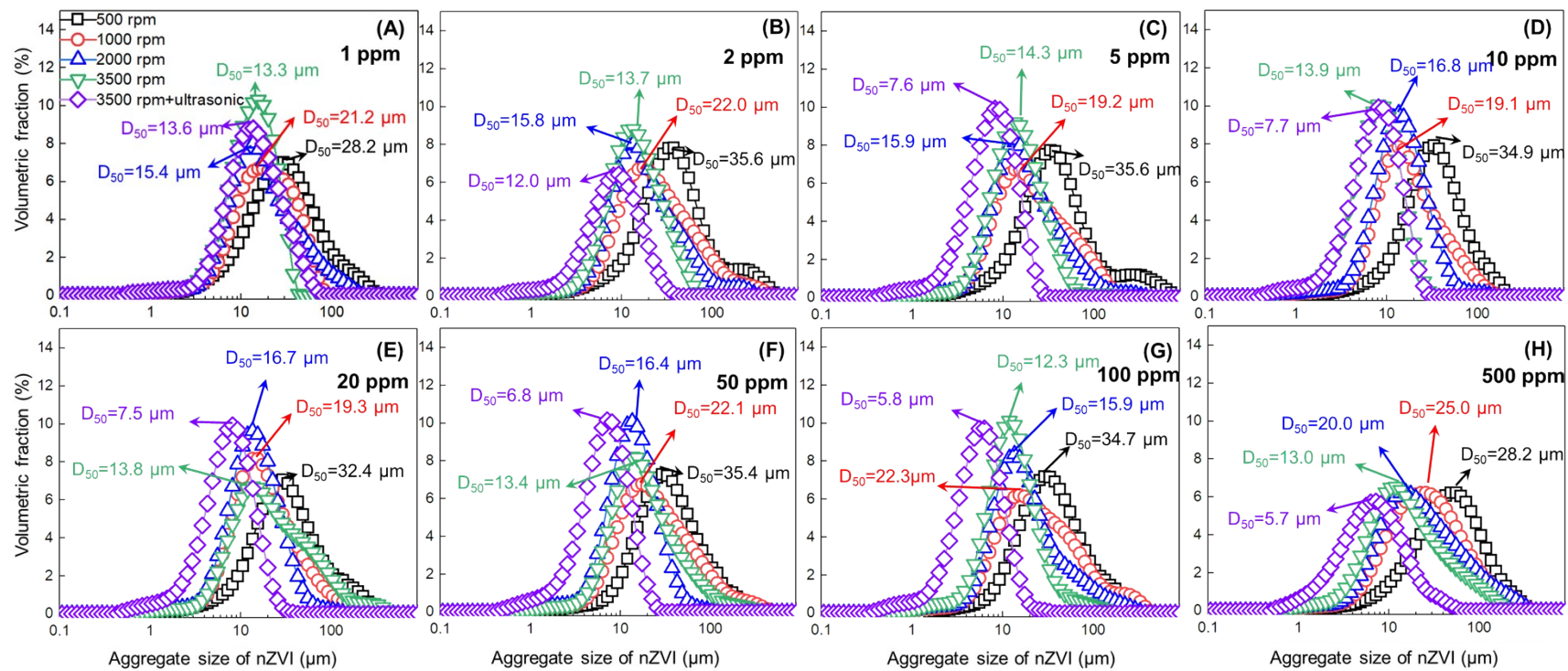


Figure S4

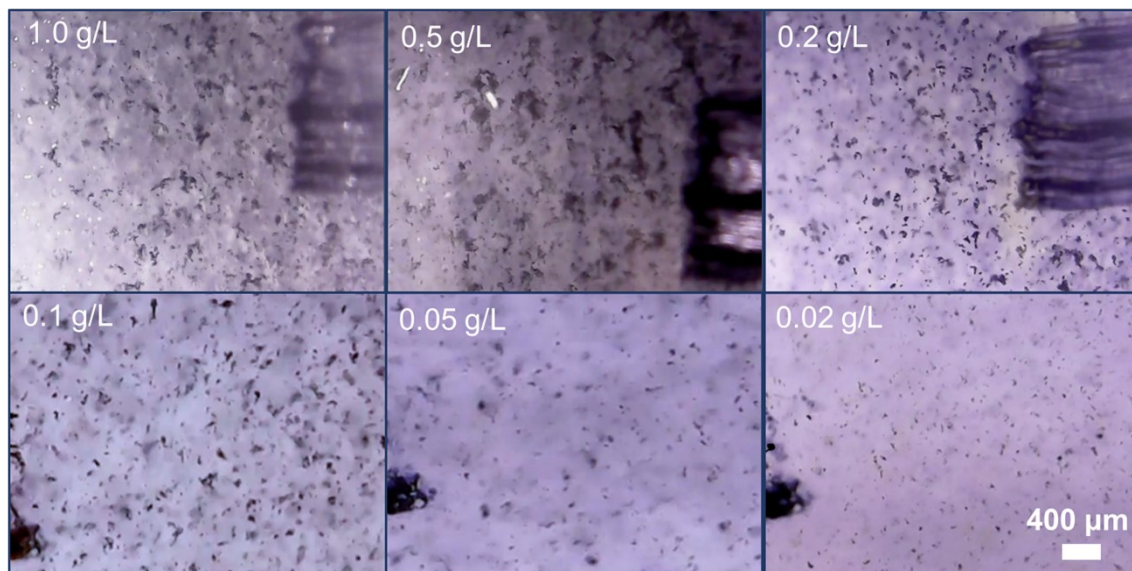


Figure S5

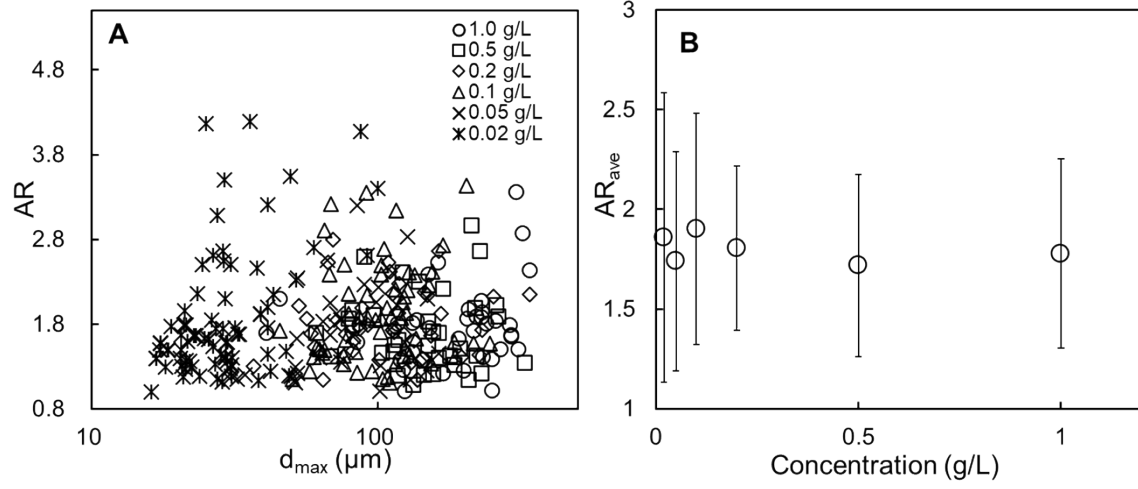


Figure S6

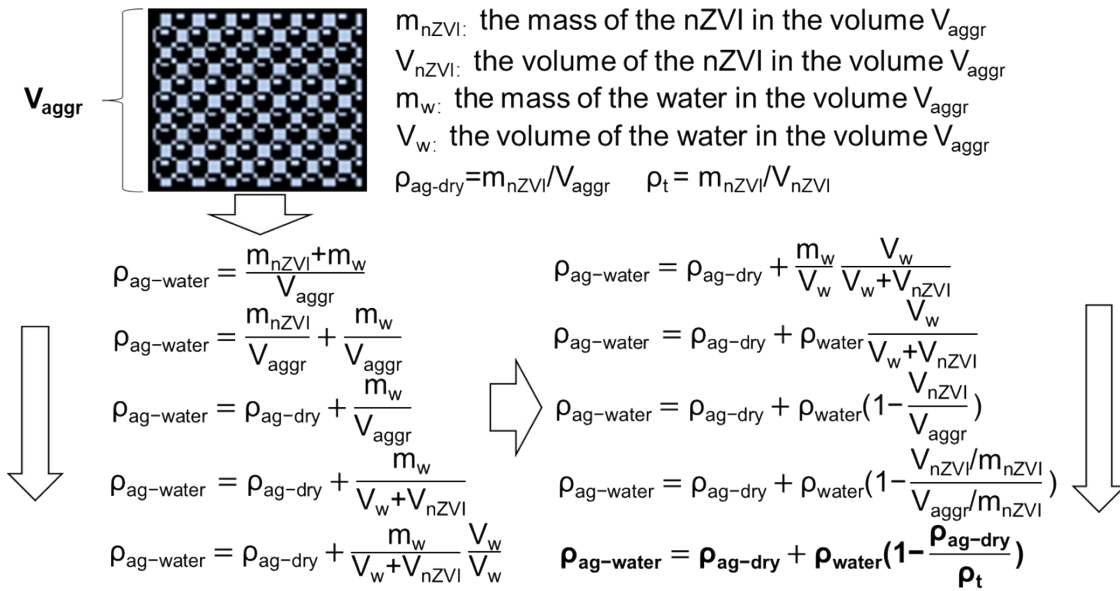


Table S1 Solution pH at different nZVI concentrations (20 °C)

concentration (g/L)	pH	concentration (g/L)	pH
0 (DI water)	6.9	1	9.5
0.001	7.0	2	9.9
0.002	7.1	5	10.3
0.005	7.2	10	10.6
0.01	7.4	20	10.9
0.02	7.4	30	11.0
0.05	7.5	40	11.0
0.1	7.6	50	11.1
0.2	8.0	60	11.2
0.5	9.1		

Table S2 Raw data about the aggregate morphology and structure
presented in Figure 5

	PA (μm^2)	d_{max}	d_{min}	P	L	W	AR	SI	v_s (mm/s)
	(μm)								
1	27311	339	93	973	349	143	2.4	0.4	
2	20100	321	80	803	322	112	2.9	0.4	0.32
3	29367	310	101	1082	305	203	1.5	0.3	
4	14100	304	79	736	310	92	3.4	0.3	0.15
5	27578	293	79	812	293	177	1.7	0.5	
6	23989	293	79	963	293	175	1.7	0.3	
7	25222	289	69	883	291	163	1.8	0.4	
8	34297	269	152	828	272	180	1.5	0.6	
9	23318	257	107	840	267	145	1.8	0.4	
10	27744	251	121	995	277	200	1.4	0.4	0.34
11	27643	251	98	1086	268	263	1.0	0.3	
12	19437	239	97	643	247	136	1.8	0.6	0.28
13	19722	233	88	678	218	154	1.4	0.5	0.25
14	16892	231	75	644	227	117	1.9	0.5	0.17
15	17611	230	79	661	236	125	1.9	0.5	0.25
16	16321	230	70	606	243	117	2.1	0.6	0.26
17	13956	221	24	705	240	127	1.9	0.4	0.24
18	14389	216	54	687	237	138	1.7	0.4	0.18
19	9944	208	61	690	212	107	2.0	0.3	0.10
20	9467	206	18	612	212	113	1.9	0.3	0.16
21	18090	199	107	607	195	156	1.3	0.6	0.26
22	13514	192	84	538	197	121	1.6	0.6	0.14
23	13137	169	85	541	166	136	1.2	0.6	0.23
24	6757	162	49	394	162	64	2.5	0.5	0.16
25	7670	152	62	429	151	86	1.8	0.5	0.12
26	6044	150	52	364	150	63	2.4	0.6	0.18
27	10411	144	94	379	145	105	1.4	0.9	0.21
28	5467	137	53	354	134	72	1.8	0.5	0.17
29	6403	132	31	392	132	73	1.8	0.5	0.11
30	10044	132	89	461	171	116	1.5	0.6	0.11
31	7476	124	65	418	116	115	1.0	0.5	0.10
32	9022	123	78	381	125	99	1.3	0.8	0.15
33	6756	120	70	313	121	88	1.4	0.9	0.18
34	5578	117	60	282	118	65	1.8	0.9	0.13
35	2922	84	34	254	91	57	1.6	0.6	0.15
36	1056	45	24	118	49	24	2.1	1.0	0.12
37	878	41	24	102	41	24	1.7	1.0	0.07
38	29176	327	58	1182	329	245	1.3	0.3	0.06
39	15704	265	46	780	265	141	1.9	0.3	0.24
40	14230	259	51	789	273	135	2.0	0.3	0.12

41	21212	230	43	919	231	189	1.2	0.3	0.27
42	12879	227	76	538	234	88	2.7	0.6	0.15
43	24590	221	124	652	234	163	1.4	0.7	0.19
44	18284	218	109	546	227	114	2.0	0.8	0.19
45	8190	213	55	544	220	74	3.0	0.3	0.19
46	14394	194	80	564	198	131	1.5	0.6	0.16
47	12223	185	57	575	183	139	1.3	0.5	0.12
48	16154	179	96	508	175	122	1.4	0.8	0.26
49	9234	169	65	384	169	76	2.2	0.8	0.05
50	8640	163	45	493	164	97	1.7	0.4	
51	13042	163	94	455	166	118	1.4	0.8	
52	9132	156	67	418	167	97	1.7	0.7	0.19
53	11957	156	63	487	151	125	1.2	0.6	0.12
54	14940	208	62	696	156	136	1.1	0.4	
55	11056	150	73	465	153	83	1.8	0.6	0.19
56	6040	150	29	425	145	94	1.5	0.4	0.12
57	7371	145	63	367	141	118	1.2	0.7	0.07
58	8436	139	49	480	133	58	2.3	0.5	0.11
59	5487	133	51	304	122	112	1.1	0.7	0.11
60	9029	122	83	361	117	49	2.4	0.9	0.09
61	4279	118	29	278	115	71	1.6	0.7	0.10
62	6429	117	59	297	115	96	1.2	0.9	0.11
63	8108	114	79	335	119	77	1.6	0.9	0.15
64	4832	113	35	330	114	77	1.5	0.6	0.04
65	5364	103	59	323	99	67	1.5	0.6	0.13
66	5262	97	63	258	90	47	1.9	1.0	0.05
67	3808	90	42	225	82	32	2.6	0.9	0.12
68	1925	82	23	187	89	45	2.0	0.7	0.06
69	3010	82	35	217	81	45	1.8	0.8	0.12
70	2764	79	41	202	68	36	1.9	0.9	0.11
71	2211	68	42	167	61	43	1.4	1.0	
72	2334	61	42	166	42	25	1.7	1.0	
73	22327	340	68	1182	345	160	2.2	0.2	0.22
74	14019	253	65	714	251	118	2.1	0.3	0.08
75	17076	232	76	632	230	133	1.7	0.5	0.10
76	19293	226	60	929	239	166	1.4	0.3	0.18
77	14743	196	73	651	205	145	1.4	0.4	0.12
78	9675	166	44	549	170	88	1.9	0.4	0.08
79	5700	164	32	420	165	62	2.7	0.4	0.13
80	7871	153	53	427	156	94	1.7	0.5	0.11
81	5022	149	27	429	146	68	2.1	0.3	0.11
82	7285	146	55	376	145	67	2.2	0.6	0.10
83	8928	143	82	407	143	103	1.4	0.7	0.18
84	7952	140	61	385	141	103	1.4	0.7	0.06
85	9388	138	76	412	129	113	1.1	0.7	0.09
86	3355	134	49	363	139	86	1.6	0.3	

87	9147	131	55	401	133	99	1.3	0.7	0.12
88	6872	123	62	358	141	84	1.7	0.7	0.15
89	4539	118	41	303	125	55	2.3	0.6	0.05
90	6377	118	44	356	140	72	1.9	0.6	0.06
91	5860	115	56	308	119	68	1.8	0.8	0.06
92	5010	114	44	298	113	66	1.7	0.7	0.09
93	4987	113	37	322	118	68	1.7	0.6	0.06
94	4746	113	47	267	113	52	2.2	0.8	0.07
95	4137	110	41	258	112	46	2.4	0.8	0.07
96	3194	109	27	256	108	43	2.5	0.6	
97	5619	101	43	369	123	89	1.4	0.5	0.09
98	3206	92	38	238	95	53	1.8	0.7	
99	3321	88	42	210	88	44	2.0	0.9	
100	3355	86	41	226	90	51	1.8	0.8	0.09
101	3286	86	37	227	84	47	1.8	0.8	
102	3424	82	48	214	83	52	1.6	0.9	0.09
103	3194	82	43	252	91	62	1.5	0.6	
104	2689	77	32	198	79	47	1.7	0.9	
105	1298	70	29	191	79	28	2.8	0.4	
106	1517	67	24	152	67	26	2.5	0.8	
107	3172	64	54	195	65	57	1.1	1.0	0.05
108	2022	60	39	160	62	41	1.5	1.0	
109	1816	59	31	153	58	34	1.7	1.0	0.06
110	1505	56	31	140	57	31	1.9	1.0	
111	1264	53	24	128	53	26	2.0	1.0	
112	931	37	27	105	38	29	1.3	1.0	0.03
113	770	31	26	89	32	26	1.2	1.0	
114	28368	246	129	646	250	159	1.6	0.9	
115	16701	216	99	777	258	164	1.6	0.3	
116	7006	204	41	455	204	59	3.4	0.4	
117	14928	187	79	539	188	134	1.4	0.6	0.15
118	17244	185	98	545	198	125	1.6	0.7	
119	7464	169	55	390	171	62	2.7	0.6	
120	8264	155	56	369	158	65	2.4	0.8	0.13
121	10924	153	70	510	158	118	1.3	0.5	0.09
122	6949	151	48	370	154	68	2.3	0.6	0.09
123	8179	136	79	335	137	84	1.6	0.9	0.12
124	6034	135	54	316	135	57	2.4	0.8	0.08
125	7864	129	61	392	139	96	1.4	0.6	0.10
126	4804	129	33	307	131	54	2.4	0.6	0.09
127	5433	127	49	309	130	59	2.2	0.7	
128	6835	125	65	319	131	68	1.9	0.8	
129	8007	124	74	335	131	87	1.5	0.9	0.08
130	8036	123	70	341	122	86	1.4	0.9	0.09
131	4232	123	34	370	125	59	2.1	0.4	0.08
132	6348	122	61	306	131	63	2.1	0.9	

133	5920	121	49	316	121	65	1.9	0.7	0.09
134	3946	116	34	265	120	38	3.1	0.7	
135	6920	115	67	320	120	90	1.3	0.8	
136	5062	115	43	288	115	52	2.2	0.8	0.08
137	6091	112	57	294	117	62	1.9	0.9	0.06
138	7836	110	65	376	119	107	1.1	0.7	0.06
139	7778	108	70	331	108	92	1.2	0.9	
140	4947	107	48	282	117	59	2.0	0.8	
141	3317	105	34	231	105	39	2.7	0.8	0.06
142	7550	104	80	305	103	89	1.2	1.0	
143	4232	103	42	253	105	44	2.4	0.8	0.07
144	3460	103	48	252	108	43	2.5	0.7	
145	5176	103	55	270	106	57	1.9	0.9	
146	5062	102	50	285	109	64	1.7	0.8	
147	4461	98	48	238	100	54	1.8	1.0	0.13
148	6205	95	61	329	114	91	1.2	0.7	
149	3718	92	43	244	91	49	1.9	0.8	
150	3088	92	37	210	91	43	2.1	0.9	
151	2088	91	17	203	91	27	3.4	0.6	
152	3088	86	36	232	92	49	1.9	0.7	
153	4432	85	57	237	84	68	1.2	1.0	0.09
154	3632	84	48	213	82	55	1.5	1.0	
155	2888	79	45	192	85	44	1.9	1.0	0.09
156	2745	79	34	193	81	38	2.2	0.9	0.06
157	3203	77	53	205	79	49	1.6	1.0	0.09
158	2145	77	27	185	77	31	2.5	0.8	
159	3603	76	56	207	76	57	1.3	1.0	
160	3546	76	53	203	79	55	1.4	1.0	
161	1344	68	21	150	68	21	3.2	0.7	0.07
162	1945	68	34	175	71	30	2.4	0.8	0.05
163	2517	65	42	169	65	44	1.5	1.0	
164	1716	65	21	155	64	22	2.9	0.9	0.05
165	2574	64	43	169	65	44	1.5	1.0	0.08
166	2717	63	43	175	69	46	1.5	1.0	0.11
167	2374	61	38	167	64	38	1.7	1.0	0.08
168	2231	61	55	155	60	39	1.5	1.0	
169	2459	60	45	163	62	44	1.4	1.0	
170	2459	58	46	162	59	47	1.2	1.0	
171	2088	50	41	147	53	46	1.2	1.0	0.08
172	1458	45	29	122	53	31	1.7	1.0	0.04
173	7305	163	77	425	161	111	1.5	0.5	0.13
174	7709	149	60	350	148	71	2.1	0.8	0.15
175	13910	149	115	418	151	121	1.2	1.0	
176	8410	130	65	378	122	106	1.1	0.7	
177	4821	127	31	298	127	45	2.8	0.7	0.09
178	5160	117	51	286	122	60	2.0	0.8	0.06

179	4821	113	42	298	122	51	2.4	0.7	0.13
180	5564	102	58	322	109	83	1.3	0.7	0.10
181	6031	102	46	332	96	95	1.0	0.7	0.08
182	3398	101	37	249	100	45	2.2	0.7	0.09
183	3334	99	23	257	101	49	2.1	0.6	0.09
184	2846	89	31	213	89	39	2.3	0.8	0.06
185	1975	85	23	193	85	27	3.2	0.7	0.06
186	3462	82	50	207	82	49	1.7	1.0	0.05
187	2739	74	41	186	76	41	1.9	1.0	0.09
188	2803	74	34	201	75	55	1.4	0.9	0.09
189	2145	72	32	171	71	37	1.9	0.9	0.10
190	2251	70	37	172	71	40	1.8	1.0	0.06
191	2017	68	33	165	73	35	2.0	0.9	0.07
192	2336	68	34	172	68	41	1.7	1.0	0.06
193	1656	66	23	158	69	27	2.6	0.8	0.04
194	1975	65	33	165	66	36	1.8	0.9	0.11
195	2591	63	42	173	66	47	1.4	1.0	0.06
196	2400	53	46	159	58	47	1.2	1.0	0.07
197	1189	53	28	127	55	24	2.3	0.9	0.05
198	1699	52	37	143	56	34	1.6	1.0	0.05
199	1784	52	36	142	52	40	1.3	1.0	0.06
200	2039	52	41	152	51	46	1.1	1.0	0.07
201	1975	51	46	145	51	45	1.1	1.0	0.03
202	1911	50	42	146	51	43	1.2	1.0	0.05
203	765	34	28	84	28	23	1.2	1.0	0.06
204	1041	32	28	106	34	30	1.2	1.0	0.05
205	1967	100	31	238	107	32	3.4	0.4	
206	2418	92	27	230	92	35	2.6	0.6	0.13
207	1530	87	23	187	87	21	4.1	0.6	0.08
208	2943	72	48	189	73	51	1.4	1.0	0.09
209	874	60	8	143	61	23	2.7	0.5	0.06
210	1107	52	41	127	57	24	2.3	0.9	0.08
211	539	50	24	104	52	15	3.5	0.6	0.09
212	1530	49	34	134	48	41	1.2	1.0	0.08
213	1369	48	30	128	50	33	1.5	1.0	0.09
214	830	43	23	107	43	20	2.2	0.9	0.04
215	1326	42	35	126	43	35	1.2	1.0	0.09
216	874	41	19	102	44	22	2.0	1.0	0.03
217	860	41	36	104	43	25	1.8	1.0	0.05
218	554	41	14	96	48	15	3.2	0.8	0.04
219	1151	41	28	117	44	31	1.4	1.0	0.04
220	787	39	21	99	38	20	1.9	1.0	0.05
221	685	39	21	90	39	21	1.9	1.0	0.05
222	1209	38	34	121	43	38	1.1	1.0	0.08
223	626	38	12	92	41	16	2.5	0.9	0.09
224	248	36	9	66	31	7	4.2	0.7	0.08

225	510	33	20	73	31	18	1.7	1.0	0.05
226	495	32	19	74	27	16	1.7	1.0	0.06
227	466	32	19	71	30	17	1.8	1.0	0.07
228	539	31	22	76	29	21	1.4	1.0	0.08
229	918	31	27	96	35	30	1.2	1.0	0.03
230	830	31	26	92	33	25	1.3	1.0	0.07
231	350	31	14	63	27	11	2.5	1.0	0.06
232	918	31	28	103	36	28	1.3	1.0	0.02
233	568	30	30	77	32	20	1.6	1.0	
234	524	30	23	79	28	20	1.4	1.0	
235	422	29	18	66	27	17	1.6	1.0	
236	466	29	20	72	26	18	1.4	1.0	
237	495	29	22	74	29	21	1.4	1.0	
238	335	29	15	54	26	12	2.1	1.0	
239	262	29	11	54	27	8	3.5	1.0	
240	277	29	12	49	24	9	2.5	1.0	
241	262	29	11	50	24	9	2.7	1.0	
242	335	29	15	60	27	15	1.7	1.0	
243	554	29	25	76	27	24	1.1	1.0	0.02
244	437	28	20	58	26	18	1.4	1.0	0.04
245	160	27	8	47	22	7	3.1	0.9	
246	510	27	24	72	26	22	1.1	1.0	0.06
247	350	27	17	60	25	14	1.7	1.0	
248	670	27	27	80	32	24	1.3	1.0	
249	350	27	17	59	24	15	1.5	1.0	
250	160	26	9	48	22	8	2.6	0.9	
251	291	26	14	52	22	12	1.9	1.0	
252	320	26	16	52	22	14	1.6	1.0	
253	320	26	16	54	22	14	1.6	1.0	
254	160	25	8	38	19	5	4.2	1.0	
255	219	25	13	48	20	12	1.6	1.0	
256	233	24	12	43	19	8	2.5	1.0	
257	364	24	20	62	22	19	1.2	1.0	
258	204	23	11	44	20	9	2.2	1.0	
259	291	23	16	50	19	11	1.7	1.0	
260	262	23	14	45	19	11	1.7	1.0	
261	262	23	15	47	19	11	1.7	1.0	
262	306	22	17	53	19	15	1.3	1.0	
263	262	22	15	52	19	14	1.4	1.0	
264	248	22	15	47	19	12	1.6	1.0	
265	262	21	15	52	19	14	1.4	1.0	
266	189	21	12	41	17	9	1.8	1.0	
267	277	21	16	50	17	14	1.3	1.0	
268	160	21	10	33	16	8	2.0	1.0	
269	204	21	12	39	16	9	1.8	1.0	
270	291	21	18	52	18	16	1.2	1.0	

271	248	21	15	45	16	12	1.3	1.0
272	219	21	14	39	16	11	1.5	1.0
273	189	21	12	40	17	10	1.7	1.0
274	233	20	14	42	17	12	1.4	1.0
275	160	19	11	34	15	9	1.8	1.0
276	204	18	14	44	16	11	1.5	1.0
277	189	18	13	41	16	11	1.5	1.0
278	204	18	14	37	16	12	1.3	1.0
279	160	17	12	33	14	9	1.6	1.0
280	146	17	11	31	13	8	1.6	1.0
281	175	17	13	38	11	8	1.5	1.0
282	160	17	12	33	13	9	1.4	1.0
283	160	17	12	37	13	9	1.4	1.0
284	204	16	16	39	11	11	1.0	1.0
