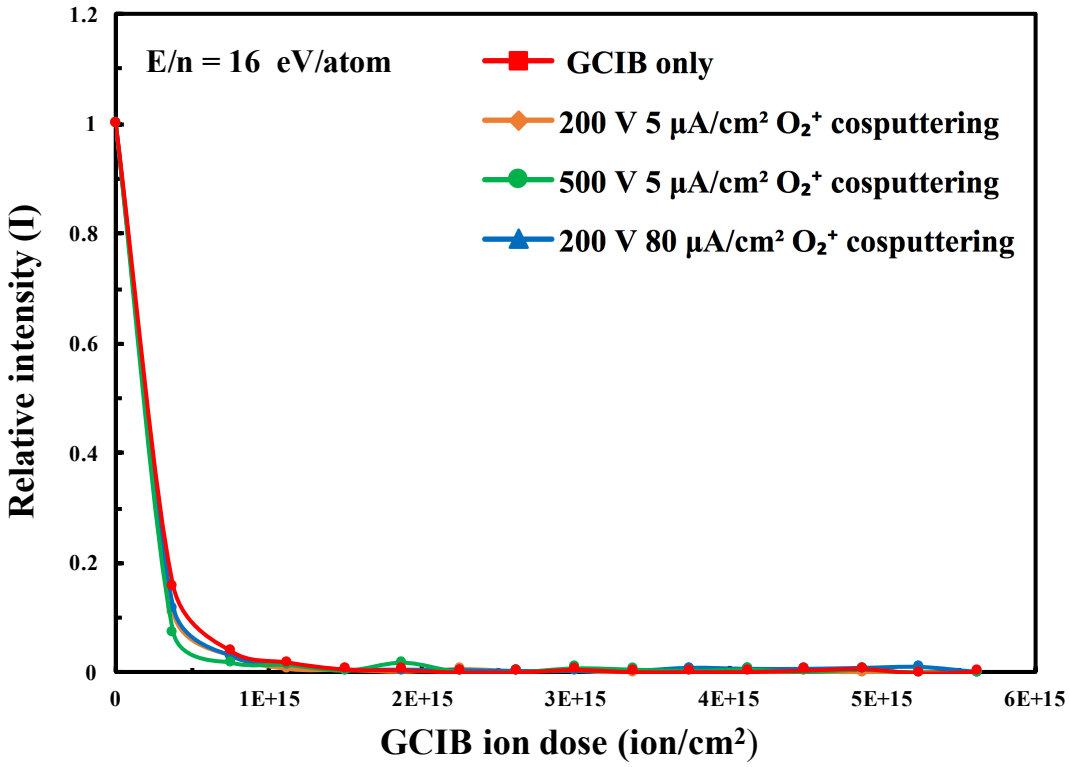

Figure S1. The SIMS depth profile of parent $C_{10}H_9O_4^+$ ($[M + H]^+$) at m/z 193 generated with 16 kV Ar_{1000}^+ sputtering and various O_2^+ cosputtering.

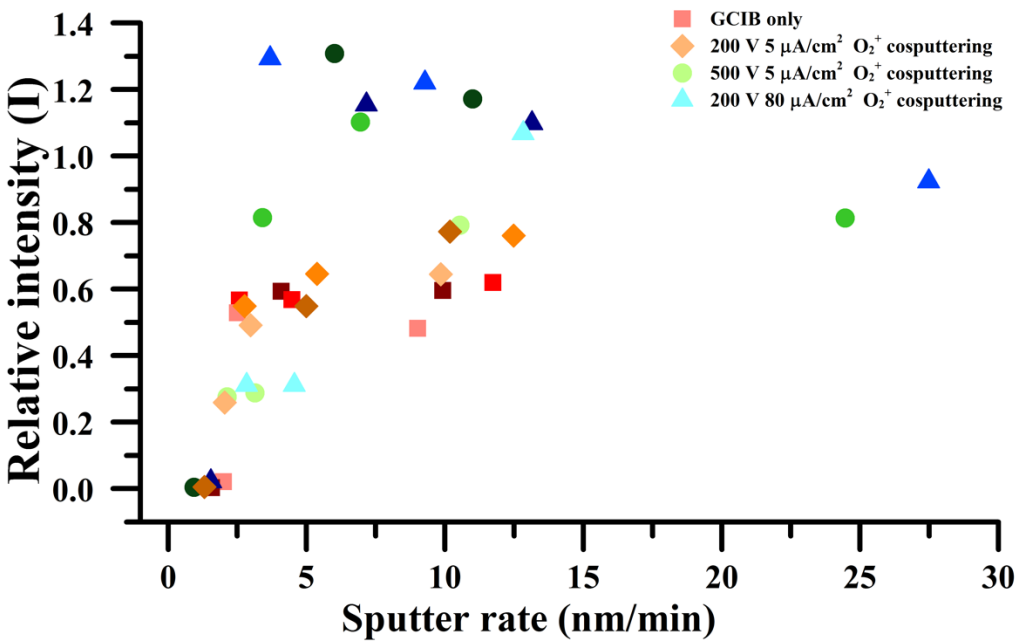

Figure S2. The steady-state ion intensity (normalized with respect to the intensity on pristine surface) of $[M+H]^+$ after GCIB-O_2^+ cosputtering with various parameters versus the sputter rate. The shade of each color (light/normal/dark) represents the usage of 10/15/20 keV GCIB, respectively.

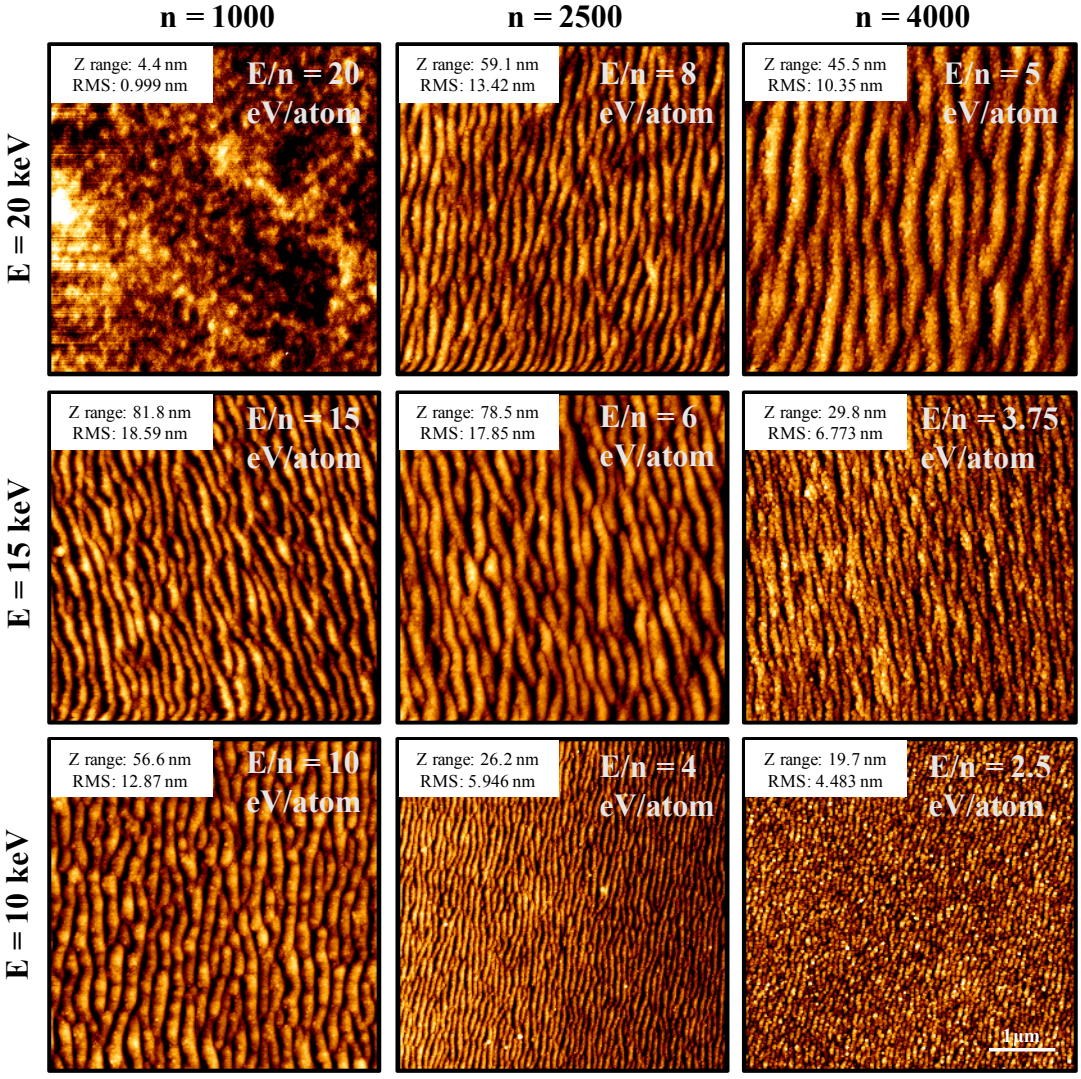

Figure S3. Zero-force AFM images of surfaces sputtered by single GCIB sputtering at fluence of 1.09×10^{16} ion/cm^2. Field of view: 5 µm × 5 µm.

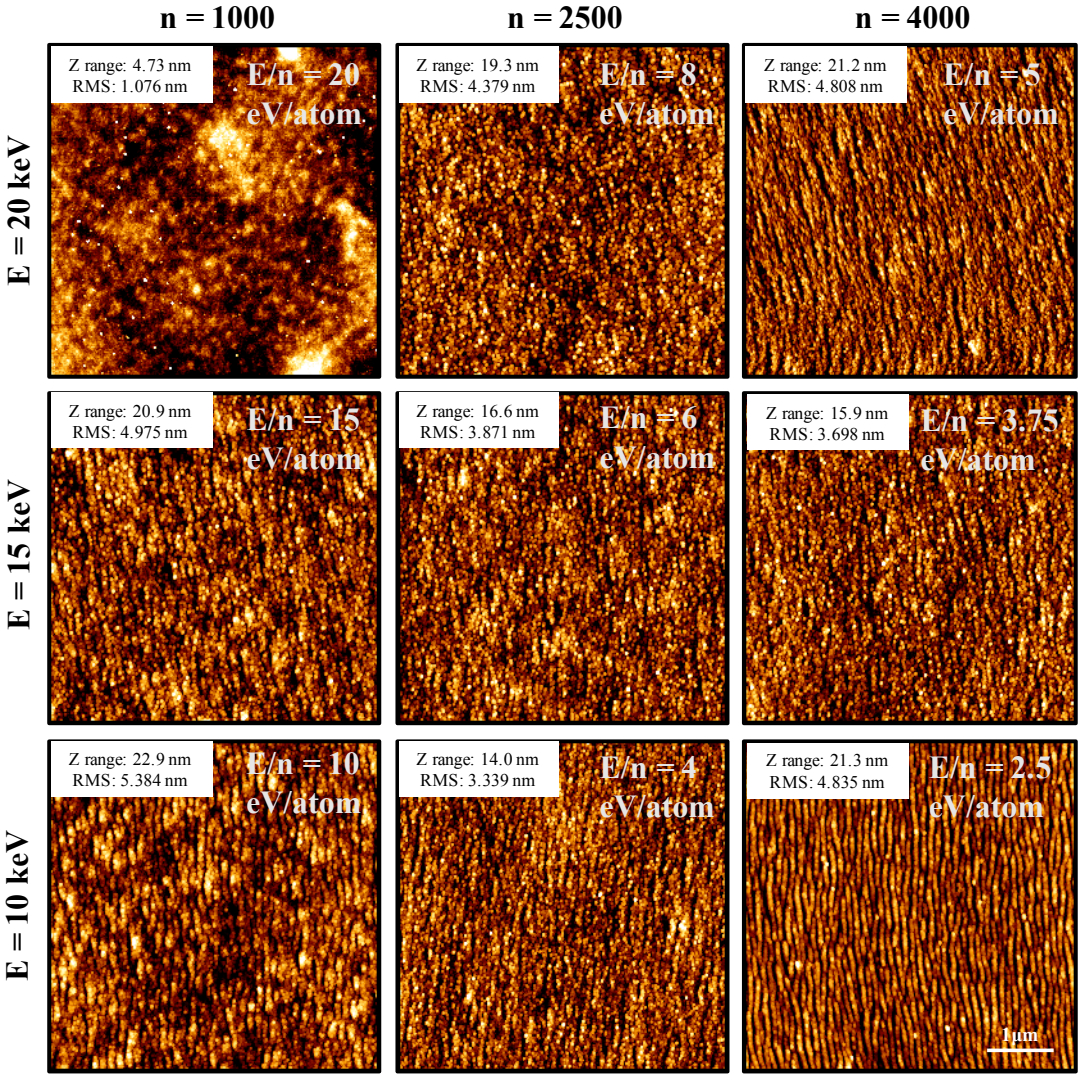

Figure S4. Zero-force AFM of surfaces cosputtered by 200 V, 5 µA/cm^2 O_2^+ with GCIB fluence of 1.09 × 10^{16} ion/cm^2. Field of view: 5 µm × 5 µm.

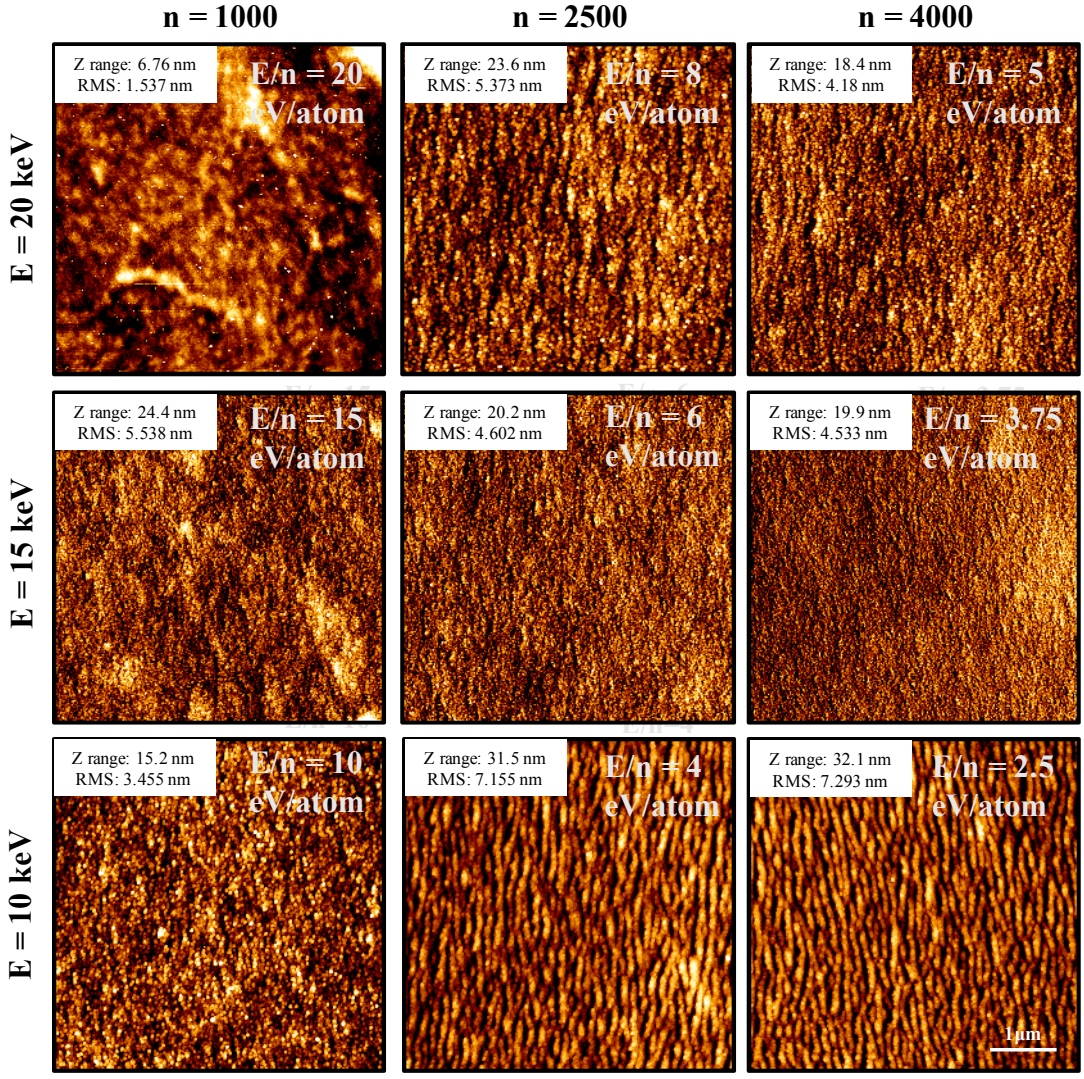

Figure S5. Zero-force AFM images of surfaces cosputtered by 500 V, 5 µA/cm^2 O_2^+ with GCIB fluence of 1.09 × 10^{16} ion/cm^2. Field of view: 5 µm × 5 µm.

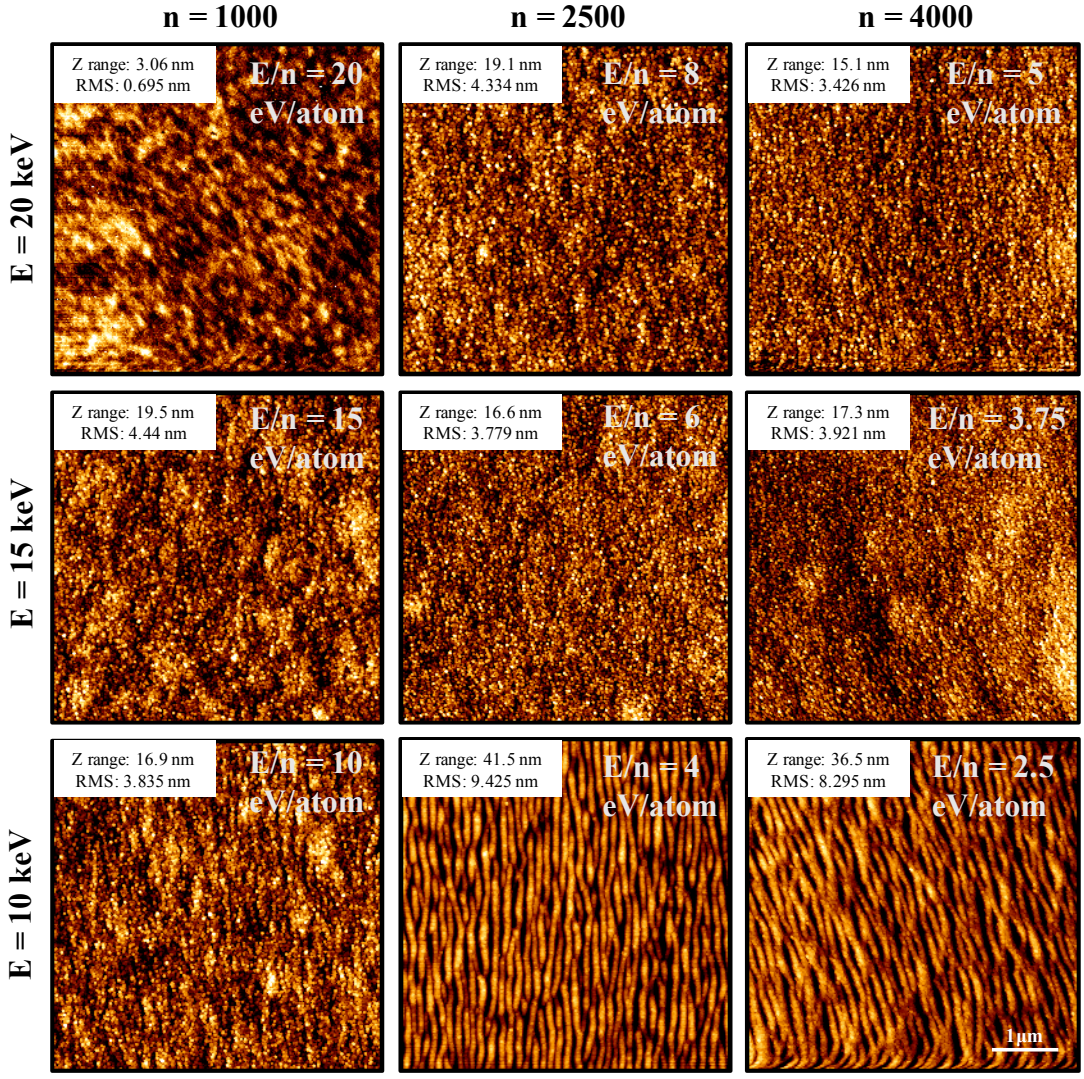

Figure S6. Zero-force AFM images of surfaces cosputtered by 200 V, 80 µA/cm^2 O_2^+ with GCIB fluence of 1.09 × 10^{16} ion/cm^2. Field of view: 5 µm × 5 µm.

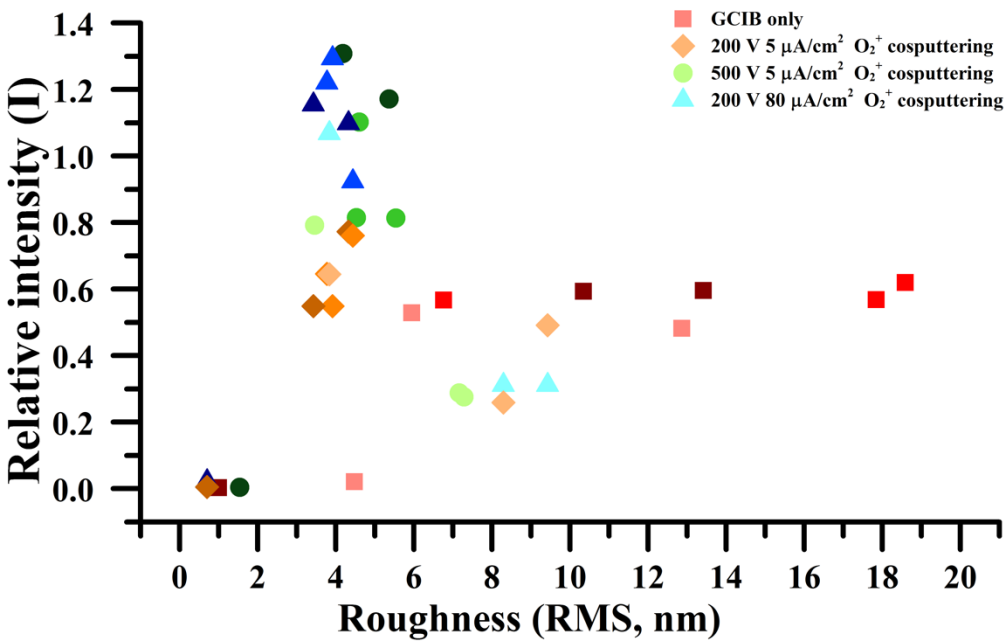

Figure S7. The steady-state ion intensity (normalized with respect to the intensity on pristine surface) of $[M+H]^+$ versus the surface roughness after GCIB-O_2^+ cosputtering with various parameters. The shade of each color (light/normal/dark) represents the usage of 10/15/20 keV GCIB, respectively.

Table S1. Sputter rate, relative intensity, Young's modulus and RMS roughness measured with various beam parameters.

E/n	GCIB (kV, n)	O_2 beam (V, μA/cm^2)	Sputter rate (nm/min)	Relative intensity (a.u.)	Young's modulus (GPa)	RMS roughness (nm)
2.5	10, 4000	0, 0	2.00	0.02±0.03	2.50±0.26	4.48
		200, 5	2.05	0.26±0.03	2.42±0.13	4.84
		500, 5	2.13	0.28±0.08	2.23±0.21	7.29
		200, 80	2.84	0.31±0.06	2.74±0.36	8.30
3.75	15, 4000	0, 0	2.58	0.57±0.08	1.96±0.18	6.77
		200, 5	2.76	0.55±0.07	2.03±0.27	3.70
		500, 5	3.42	0.82±0.17	1.64±0.26	4.53
		200, 80	3.68	1.29±0.16	2.22±0.34	3.92
4	10, 2500	0, 0	2.51	0.53±0.07	2.29±0.22	5.95
		200, 5	2.98	0.49±0.11	2.27±0.17	3.34
		500, 5	3.15	0.29±0.09	2.18±0.14	7.16
		200, 80	4.56	0.31±0.08	2.66±0.36	9.43
5	20, 4000	0, 0	4.08	0.59±0.08	1.95±0.18	10.4
		200, 5	4.99	0.55±0.16	1.89±0.28	4.81
		500, 5	6.02	1.31±0.33	1.84±0.13	4.18
		200, 80	7.16	1.15±0.18	2.11±0.12	3.43
6	15, 2500	0, 0	4.48	0.57±0.11	1.84±0.23	17.9
		200, 5	5.38	0.65±0.18	1.90±0.26	3.87
		500, 5	6.95	1.10±0.15	1.59±0.27	4.60
		200, 80	9.28	1.22±0.09	2.02±0.16	3.78
8	20, 2500	0, 0	9.92	0.60±0.03	1.79±0.13	13.4
		200, 5	10.2	0.77±0.05	1.69±0.25	4.38
		500, 5	11.0	1.17±0.28	1.67±0.10	5.37
		200, 80	13.2	1.10±0.01	1.92±0.18	4.33
10	10, 1000	0, 0	9.03	0.48±0.13	1.93±0.22	12.9
		200, 5	9.85	0.65±0.07	1.89±0.23	5.38
		500, 5	10.5	0.79±0.09	1.73±0.13	3.46
		200, 80	12.8	1.07±0.01	1.89±0.27	3.84
15	15, 1000	0, 0	11.7	0.62±0.07	1.52±0.10	18.6
		200, 5	12.5	0.76±0.17	1.67±0.11	4.98
		500, 5	24.5	0.81±0.11	1.26±0.24	5.54
		200, 80	27.5	0.92±0.15	1.73±0.12	4.44
20	20, 1000	0, 0	1.58	0.003±0.00	14.2±0.72	1.00
		200, 5	1.32	0.005±0.00	19.1±1.26	1.08
		500, 5	0.94	0.004±0.03	22.5±1.96	1.54
		200, 80	1.54	0.022±0.01	20.5±2.30	0.70