

## Supporting information

### Quantification of the enhanced effectiveness of NO<sub>x</sub> control from simultaneous reductions of VOC and NH<sub>3</sub> for reducing air pollution in Beijing-Tianjin-Hebei region, China

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Table S1. Control matrix of out-of-sample datasets

#	Beijing					Tianjin					HebeiN					HebeiE					HebeiS									
	NO <sub>x</sub>	SO <sub>2</sub>	NH <sub>3</sub>	VOCs	POA	NO <sub>x</sub>	SO <sub>2</sub>	NH <sub>3</sub>	VOCs	POA	NO <sub>x</sub>	SO <sub>2</sub>	NH <sub>3</sub>	VOCs	POA	NO <sub>x</sub>	SO <sub>2</sub>	NH <sub>3</sub>	VOCs	POA	NO <sub>x</sub>	SO <sub>2</sub>	NH <sub>3</sub>	VOCs	POA					
base	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
Out-of-sample 100 (OOS100)																														
1	0.073	0.704	0.122	0.324	0.359	0.073	0.704	0.122	0.324	0.359	0.073	0.704	0.122	0.324	0.359	0.073	0.704	0.122	0.324	0.359	0.073	0.704	0.122	0.324	0.359	0.073	0.704	0.122	0.324	0.359
2	0.182	1.041	0.250	1.015	0.028	0.182	1.041	0.250	1.015	0.028	0.182	1.041	0.250	1.015	0.028	0.182	1.041	0.250	1.015	0.028	0.182	1.041	0.250	1.015	0.028	0.182	1.041	0.250	1.015	0.028
3	0.636	0.197	0.981	0.431	0.721	0.636	0.197	0.981	0.431	0.721	0.636	0.197	0.981	0.431	0.721	0.636	0.197	0.981	0.431	0.721	0.636	0.197	0.981	0.431	0.721	0.636	0.197	0.981	0.431	0.721
4	0.522	0.351	0.178	0.107	0.162	0.522	0.351	0.178	0.107	0.162	0.522	0.351	0.178	0.107	0.162	0.522	0.351	0.178	0.107	0.162	0.522	0.351	0.178	0.107	0.162	0.522	0.351	0.178	0.107	0.162
5	0.583	0.994	0.702	0.259	0.572	0.583	0.994	0.702	0.259	0.572	0.583	0.994	0.702	0.259	0.572	0.583	0.994	0.702	0.259	0.572	0.583	0.994	0.702	0.259	0.572	0.583	0.994	0.702	0.259	0.572
6	0.837	0.431	0.385	0.082	0.644	0.837	0.431	0.385	0.082	0.644	0.837	0.431	0.385	0.082	0.644	0.837	0.431	0.385	0.082	0.644	0.837	0.431	0.385	0.082	0.644	0.837	0.431	0.385	0.082	0.644
7	0.814	0.081	0.420	1.065	1.186	0.814	0.081	0.420	1.065	1.186	0.814	0.081	0.420	1.065	1.186	0.814	0.081	0.420	1.065	1.186	0.814	0.081	0.420	1.065	1.186	0.814	0.081	0.420	1.065	1.186
8	0.114	0.886	0.440	0.935	0.439	0.114	0.886	0.440	0.935	0.439	0.114	0.886	0.440	0.935	0.439	0.114	0.886	0.440	0.935	0.439	0.114	0.886	0.440	0.935	0.439	0.114	0.886	0.440	0.935	0.439
9	0.573	0.825	0.403	0.991	0.714	0.573	0.825	0.403	0.991	0.714	0.573	0.825	0.403	0.991	0.714	0.573	0.825	0.403	0.991	0.714	0.573	0.825	0.403	0.991	0.714	0.573	0.825	0.403	0.991	0.714
10	0.688	1.007	1.104	0.676	0.268	0.688	1.007	1.104	0.676	0.268	0.688	1.007	1.104	0.676	0.268	0.688	1.007	1.104	0.676	0.268	0.688	1.007	1.104	0.676	0.268	0.688	1.007	1.104	0.676	0.268
11	0.223	0.153	0.547	0.186	0.038	0.223	0.153	0.547	0.186	0.038	0.223	0.153	0.547	0.186	0.038	0.223	0.153	0.547	0.186	0.038	0.223	0.153	0.547	0.186	0.038	0.223	0.153	0.547	0.186	0.038
12	0.411	0.086	0.798	1.144	0.181	0.411	0.086	0.798	1.144	0.181	0.411	0.086	0.798	1.144	0.181	0.411	0.086	0.798	1.144	0.181	0.411	0.086	0.798	1.144	0.181	0.411	0.086	0.798	1.144	0.181
13	0.723	0.116	1.140	0.911	1.122	0.723	0.116	1.140	0.911	1.122	0.723	0.116	1.140	0.911	1.122	0.723	0.116	1.140	0.911	1.122	0.723	0.116	1.140	0.911	1.122	0.723	0.116	1.140	0.911	1.122
14	0.231	0.904	0.517	0.494	0.931	0.231	0.904	0.517	0.494	0.931	0.231	0.904	0.517	0.494	0.931	0.231	0.904	0.517	0.494	0.931	0.231	0.904	0.517	0.494	0.931	0.231	0.904	0.517	0.494	0.931
15	0.992	0.810	0.899	0.687	1.029	0.992	0.810	0.899	0.687	1.029	0.992	0.810	0.899	0.687	1.029	0.992	0.810	0.899	0.687	1.029	0.992	0.810	0.899	0.687	1.029	0.992	0.810	0.899	0.687	1.029
16	0.622	0.123	0.083	0.529	0.592	0.622	0.123	0.083	0.529	0.592	0.622	0.123	0.083	0.529	0.592	0.622	0.123	0.083	0.529	0.592	0.622	0.123	0.083	0.529	0.592	0.622	0.123	0.083	0.529	0.592
17	0.287	1.115	0.105	0.773	0.408	0.287	1.115	0.105	0.773	0.408	0.287	1.115	0.105	0.773	0.408	0.287	1.115	0.105	0.773	0.408	0.287	1.115	0.105	0.773	0.408	0.287	1.115	0.105	0.773	0.408
18	0.913	0.626	0.162	1.074	0.954	0.913	0.626	0.162	1.074	0.954	0.913	0.626	0.162	1.074	0.954	0.913	0.626	0.162	1.074	0.954	0.913	0.626	0.162	1.074	0.954	0.913	0.626	0.162	1.074	0.954
19	0.328	1.084	0.820	1.174	0.796	0.328	1.084	0.820	1.174	0.796	0.328	1.084	0.820	1.174	0.796	0.328	1.084	0.820	1.174	0.796	0.328	1.084	0.820	1.174	0.796	0.328	1.084	0.820	1.174	0.796
20	0.589	0.741	1.172	0.961	0.423	0.589	0.741	1.172	0.961	0.423	0.589	0.741	1.172	0.961	0.423	0.589	0.741	1.172	0.961	0.423	0.589	0.741	1.172	0.961	0.423	0.589	0.741	1.172	0.961	0.423
21	0.299	0.310	0.766	0.801	0.872	0.299	0.310	0.766	0.801	0.872	0.299	0.310	0.766	0.801	0.872	0.299	0.310	0.766	0.801	0.872	0.299	0.310	0.766	0.801	0.872	0.299	0.310	0.766	0.801	0.872
22	0.025	0.454	0.868	0.442	0.557	0.025	0.454	0.868	0.442	0.557	0.025	0.454	0.868	0.442	0.557	0.025	0.454	0.868	0.442	0.557	0.025	0.454	0.868	0.442	0.557	0.025	0.454	0.868	0.442	0.557
23	0.144	0.255	0.857	1.188	0.314	0.144	0.255	0.857	1.188	0.314	0.144	0.255	0.857	1.188	0.314	0.144	0.255	0.857	1.188	0.314	0.144	0.255	0.857	1.188	0.314	0.144	0.255	0.857	1.188	0.314
24	0.844	0.347	0.234	0.851	0.138	0.844	0.347	0.234	0.851	0.138	0.844	0.347	0.234	0.851	0.138	0.844	0.347	0.234	0.851	0.138	0.844	0.347	0.234	0.851	0.138	0.844	0.347	0.234	0.851	0.138
25	0.265	0.971	0.642	0.980	0.603	0.265	0.971	0.642	0.980	0.603	0.265	0.971	0.642	0.980	0.603	0.265	0.971	0.642	0.980	0.603	0.265	0.971	0.642	0.980	0.603	0.265	0.971	0.642	0.980	0.603
26	0.424	0.566	0.445	0.203	0.476	0.424	0.566	0.445	0.203	0.476	0.424	0.566	0.445	0.203	0.476	0.424	0.566	0.445	0.203	0.476	0.424	0.566	0.445	0.203	0.476	0.424	0.566	0.445	0.203	0.476
27	0.384	1.171	0.310	0.874	0.208	0.384	1.171	0.310	0.874	0.208	0.384	1.171	0.310	0.874	0.208	0.384	1.171	0.310	0.874	0.208	0.384	1.171	0.310	0.874	0.208	0.384	1.171	0.310	0.874	0.208
28	1.105	0.768	0.924	0.273	1.116	1.105	0.768	0.924	0.273	1.116	1.105	0.768	0.924	0.273	1.116	1.105	0.768	0.924	0.273	1.116	1.105	0.768	0.924	0.273	1.116	1.105	0.768	0.924	0.273	1.116
29	0.962	0.298	0.272	0.818	1.098	0.962	0.298	0.272	0.818	1.098	0.962	0.298	0.272	0.818	1.098	0.962	0.298	0.272	0.818	1.098	0.962	0.298	0.272	0.818	1.098	0.962	0.298	0.272	0.818	1.098
30	0.673	0.383	1.180	0.163	1.056	0.673	0.383	1.180	0.163	1.056	0.673	0.383	1.180	0.163	1.056	0.673	0.383	1.180	0.163	1.056	0.673	0.383	1.180	0.163	1.056	0.673	0.383	1.180	0.163	1.056
31	0.471	0.591	0.669	0.391	0.446	0.471	0.591	0.669	0.391	0.446	0.471	0.591	0.669	0.391	0.446	0.471	0.591	0.669	0.391	0.446	0.471	0.591	0.669	0.391	0.446	0.471	0.591	0.669	0.391	0.446
32	0.553	1.059	1.119	0.402	0.076	0.553	1.059	1.119	0.402	0.076	0.553	1.059	1.119	0.402	0.076	0.553	1.059	1.119	0.402	0.076	0.553	1.059	1.119	0.402	0.076	0.553	1.059	1.119	0.402	0.076
33	0.863	0.729	0.149	0.650	0.688	0.863	0.729	0.149	0.650	0.688	0.863	0.729	0.149	0.650	0.688	0.863	0.729	0.149	0.650	0.688	0.863	0.729	0.149	0.650	0.688	0.863	0.729	0.149	0.650	0.688
34	0.449	0.509	0.355	0.704	0.650	0.449	0.509	0.355	0.704	0.650	0.449	0.509	0.355	0.704	0.650	0.449	0.509	0.355	0.704	0.650	0.449	0.509	0.355	0.704	0.650	0.449	0.509	0.355	0.704	0.650
35	0.172	0.875	0.753	0.467	0.099	0.172	0.875	0.753	0.467	0.099	0.172	0.875	0.753	0.467	0.099	0.172	0.875	0.753	0.467	0.099	0.172	0.875	0.753	0.467	0.099	0.172	0.875	0.753	0.467	0.099
36	1.191	0.891	0.317	0.134	0.398	1.191	0.891	0.317	0.134	0.398	1.191	0.891	0.317	0.134	0.398	1.191	0.891	0.317	0.134	0.398	1.191	0.891	0.317	0.134	0.398	1.191	0.891	0.317	0.134	0.398
37	0.089	0.142	0.186	0.299	0.977	0.089	0.142	0.186	0.299	0.977	0.089	0.142	0.186	0.299	0.977	0.089	0.142	0.186	0.											

40	0.257	0.065	0.198	1.185	0.876	0.257	0.065	0.198	1.185	0.876	0.257	0.065	0.198	1.185	0.876	0.257	0.065	0.198	1.185	0.876	0.257	0.065	0.198	1.185	0.876
41	0.165	0.051	0.572	0.354	0.992	0.165	0.051	0.572	0.354	0.992	0.165	0.051	0.572	0.354	0.992	0.165	0.051	0.572	0.354	0.992	0.165	0.051	0.572	0.354	0.992
42	1.132	1.050	0.970	0.280	0.467	1.132	1.050	0.970	0.280	0.467	1.132	1.050	0.970	0.280	0.467	1.132	1.050	0.970	0.280	0.467	1.132	1.050	0.970	0.280	0.467
43	1.042	1.018	0.829	0.448	0.071	1.042	1.018	0.829	0.448	0.071	1.042	1.018	0.829	0.448	0.071	1.042	1.018	0.829	0.448	0.071	1.042	1.018	0.829	0.448	0.071
44	0.301	0.243	0.930	0.237	1.075	0.301	0.243	0.930	0.237	1.075	0.301	0.243	0.930	0.237	1.075	0.301	0.243	0.930	0.237	1.075	0.301	0.243	0.930	0.237	1.075
45	1.016	0.746	1.152	0.004	0.760	1.016	0.746	1.152	0.004	0.760	1.016	0.746	1.152	0.004	0.760	1.016	0.746	1.152	0.004	0.760	1.016	0.746	1.152	0.004	0.760
46	0.714	0.316	0.064	0.958	0.058	0.714	0.316	0.064	0.958	0.058	0.714	0.316	0.064	0.958	0.058	0.714	0.316	0.064	0.958	0.058	0.714	0.316	0.064	0.958	0.058
47	0.106	0.788	1.076	0.218	0.509	0.106	0.788	1.076	0.218	0.509	0.106	0.788	1.076	0.218	0.509	0.106	0.788	1.076	0.218	0.509	0.106	0.788	1.076	0.218	0.509
48	0.758	0.326	0.612	0.147	0.745	0.758	0.326	0.612	0.147	0.745	0.758	0.326	0.612	0.147	0.745	0.758	0.326	0.612	0.147	0.745	0.758	0.326	0.612	0.147	0.745
49	0.040	0.803	0.731	0.477	1.002	0.040	0.803	0.731	0.477	1.002	0.040	0.803	0.731	0.477	1.002	0.040	0.803	0.731	0.477	1.002	0.040	0.803	0.731	0.477	1.002
50	0.899	0.693	0.495	0.643	0.624	0.899	0.693	0.495	0.643	0.624	0.899	0.693	0.495	0.643	0.624	0.899	0.693	0.495	0.643	0.624	0.899	0.693	0.495	0.643	0.624
51	0.530	0.983	0.537	0.579	0.364	0.530	0.983	0.537	0.579	0.364	0.530	0.983	0.537	0.579	0.364	0.530	0.983	0.537	0.579	0.364	0.530	0.983	0.537	0.579	0.364
52	0.511	0.520	0.772	0.242	0.805	0.511	0.520	0.772	0.242	0.805	0.511	0.520	0.772	0.242	0.805	0.511	0.520	0.772	0.242	0.805	0.511	0.520	0.772	0.242	0.805
53	0.023	0.758	0.780	0.366	0.768	0.023	0.758	0.780	0.366	0.768	0.023	0.758	0.780	0.366	0.768	0.023	0.758	0.780	0.366	0.768	0.023	0.758	0.780	0.366	0.768
54	0.950	0.651	0.911	1.030	1.088	0.950	0.651	0.911	1.030	1.088	0.950	0.651	0.911	1.030	1.088	0.950	0.651	0.911	1.030	1.088	0.950	0.651	0.911	1.030	1.088
55	0.791	0.927	0.879	0.116	0.855	0.791	0.927	0.879	0.116	0.855	0.791	0.927	0.879	0.116	0.855	0.791	0.927	0.879	0.116	0.855	0.791	0.927	0.879	0.116	0.855
56	0.933	0.231	0.951	1.127	0.221	0.933	0.231	0.951	1.127	0.221	0.933	0.231	0.951	1.127	0.221	0.933	0.231	0.951	1.127	0.221	0.933	0.231	0.951	1.127	0.221
57	0.550	0.362	0.045	0.490	0.524	0.550	0.362	0.045	0.490	0.524	0.550	0.362	0.045	0.490	0.524	0.550	0.362	0.045	0.490	0.524	0.550	0.362	0.045	0.490	0.524
58	0.780	0.035	1.193	0.069	0.682	0.780	0.035	1.193	0.069	0.682	0.780	0.035	1.193	0.069	0.682	0.780	0.035	1.193	0.069	0.682	0.780	0.035	1.193	0.069	0.682
59	0.011	0.394	0.514	0.590	0.542	0.011	0.394	0.514	0.590	0.542	0.011	0.394	0.514	0.590	0.542	0.011	0.394	0.514	0.590	0.542	0.011	0.394	0.514	0.590	0.542
60	0.482	0.407	0.029	0.213	0.232	0.482	0.407	0.029	0.213	0.232	0.482	0.407	0.029	0.213	0.232	0.482	0.407	0.029	0.213	0.232	0.482	0.407	0.029	0.213	0.232
61	1.100	0.945	0.712	0.310	0.841	1.100	0.945	0.712	0.310	0.841	1.100	0.945	0.712	0.310	0.841	1.100	0.945	0.712	0.310	0.841	1.100	0.945	0.712	0.310	0.841
62	1.031	0.620	0.673	0.612	0.740	1.031	0.620	0.673	0.612	0.740	1.031	0.620	0.673	0.612	0.740	1.031	0.620	0.673	0.612	0.740	1.031	0.620	0.673	0.612	0.740
63	1.060	1.093	0.485	0.129	0.244	1.060	1.093	0.485	0.129	0.244	1.060	1.093	0.485	0.129	0.244	1.060	1.093	0.485	0.129	0.244	1.060	1.093	0.485	0.129	0.244
64	0.945	0.713	0.218	0.035	0.829	0.945	0.713	0.218	0.035	0.829	0.945	0.713	0.218	0.035	0.829	0.945	0.713	0.218	0.035	0.829	0.945	0.713	0.218	0.035	0.829
65	0.663	0.046	0.998	1.080	1.066	0.663	0.046	0.998	1.080	1.066	0.663	0.046	0.998	1.080	1.066	0.663	0.046	0.998	1.080	1.066	0.663	0.046	0.998	1.080	1.066
66	0.816	0.546	0.985	0.834	0.480	0.816	0.546	0.985	0.834	0.480	0.816	0.546	0.985	0.834	0.480	0.816	0.546	0.985	0.834	0.480	0.816	0.546	0.985	0.834	0.480
67	1.149	0.670	0.654	0.523	0.092	1.149	0.670	0.654	0.523	0.092	1.149	0.670	0.654	0.523	0.092	1.149	0.670	0.654	0.523	0.092	1.149	0.670	0.654	0.523	0.092
68	0.877	1.030	0.287	1.005	0.177	0.877	1.030	0.287	1.005	0.177	0.877	1.030	0.287	1.005	0.177	0.877	1.030	0.287	1.005	0.177	0.877	1.030	0.287	1.005	0.177
69	0.748	0.538	0.340	0.505	1.014	0.748	0.538	0.340	0.505	1.014	0.748	0.538	0.340	0.505	1.014	0.748	0.538	0.340	0.505	1.014	0.748	0.538	0.340	0.505	1.014
70	0.982	0.101	1.092	0.946	0.148	0.982	0.101	1.092	0.946	0.148	0.982	0.101	1.092	0.946	0.148	0.982	0.101	1.092	0.946	0.148	0.982	0.101	1.092	0.946	0.148
71	0.141	0.213	1.086	1.040	0.613	0.141	0.213	1.086	1.040	0.613	0.141	0.213	1.086	1.040	0.613	0.141	0.213	1.086	1.040	0.613	0.141	0.213	1.086	1.040	0.613
72	0.502	0.415	0.844	1.128	0.373	0.502	0.415	0.844	1.128	0.373	0.502	0.415	0.844	1.128	0.373	0.502	0.415	0.844	1.128	0.373	0.502	0.415	0.844	1.128	0.373
73	1.049	1.123	0.331	0.732	0.946	1.049	1.123	0.331	0.732	0.946	1.049	1.123	0.331	0.732	0.946	1.049	1.123	0.331	0.732	0.946	1.049	1.123	0.331	0.732	0.946
74	0.438	1.147	0.382	0.889	1.155	0.438	1.147	0.382	0.889	1.155	0.438	1.147	0.382	0.889	1.155	0.438	1.147	0.382	0.889	1.155	0.438	1.147	0.382	0.889	1.155
75	0.216	0.846	0.210	0.542	1.173	0.216	0.846	0.210	0.542	1.173	0.216	0.846	0.210	0.542	1.173	0.216	0.846	0.210	0.542	1.173	0.216	0.846	0.210	0.542	1.173
76	1.181	1.074	0.613	0.750	0.888	1.181	1.074	0.613	0.750	0.888	1.181	1.074	0.613	0.750	0.888	1.181	1.074	0.613	0.750	0.888	1.181	1.074	0.613	0.750	0.888
77	0.631	0.857	1.035	0.326	1.191	0.631	0.857	1.035	0.326	1.191	0.631	0.857	1.035	0.326	1.191	0.631	0.857	1.035	0.326	1.191	0.631	0.857	1.035	0.326	1.191
78	1.081	0.917	0.010	0.665	1.042	1.081	0.917	0.010	0.665	1.042	1.081	0.917	0.010	0.665	1.042	1.081	0.917	0.010	0.665	1.042	1.081	0.917	0.010	0.665	1.042
79	0.462	1.194	0.022	0.919	0.699	0.462	1.194	0.022	0.919	0.699	0.462	1.194	0.022	0.919	0.699	0.462	1.194	0.022	0.919	0.699	0.462	1.194	0.022	0.919	0.699
80	0.909	0.267	0.473	0.711	0.120	0.909	0.267	0.473	0.711	0.120	0.909	0.267	0.473	0.711	0.120	0.909	0.267	0.473	0.711	0.120	0.909	0.267	0.473	0.711	0.120
81	0.866	0.679	1.031	0.343	0.533	0.866	0.679	1.031	0.343	0.533	0.866	0.679	1.031	0.343	0.533	0.866	0.679	1.031	0.343	0.533	0.866	0.679	1.031	0.343	0.533
82	0.699	0.471	0.052	0.863	0.021	0.699	0.471	0.052	0.863	0.021	0.699	0.471	0.052	0.863	0.021	0.699	0.471	0.052	0.863	0.021	0.699	0.471	0.052	0.863	0.021
83	0.124	0.642	0.261	0.756	0.498	0.124	0.642	0.261	0.756	0.498	0.124	0.642	0.261	0.756	0.498	0.124	0.642	0.261	0.756	0.498	0.124	0.642	0.261	0.756	0.498
84	1.116	0.004	0.629	0.045	0.310	1.116	0.004	0.629	0.045	0.310	1.116	0.004	0.629	0.045	0.310	1.116	0.004	0.629	0.045	0.310	1.116	0.004	0.629	0.045	0.310

85	0.061	0.838	0.113	0.603	0.825	0.061	0.838	0.113	0.603	0.825	0.061	0.838	0.113	0.603	0.825	0.061	0.838	0.113	0.603	0.825	0.061	0.838	0.113	0.603	0.825		
86	0.606	0.176	0.694	0.058	0.300	0.606	0.176	0.694	0.058	0.300	0.606	0.176	0.694	0.058	0.300	0.606	0.176	0.694	0.058	0.300	0.606	0.176	0.694	0.058	0.300		
87	0.242	0.157	0.143	0.018	0.262	0.242	0.157	0.143	0.018	0.262	0.242	0.157	0.143	0.018	0.262	0.242	0.157	0.143	0.018	0.262	0.242	0.157	0.143	0.018	0.262		
88	0.996	0.558	0.092	0.885	0.960	0.996	0.558	0.092	0.885	0.960	0.996	0.558	0.092	0.885	0.960	0.996	0.558	0.092	0.885	0.960	0.996	0.558	0.092	0.885	0.960		
89	1.162	0.181	0.946	1.053	0.906	1.162	0.181	0.946	1.053	0.906	1.162	0.181	0.946	1.053	0.906	1.162	0.181	0.946	1.053	0.906	1.162	0.181	0.946	1.053	0.906		
90	0.360	0.957	0.742	0.373	1.143	0.360	0.957	0.742	0.373	1.143	0.360	0.957	0.742	0.373	1.143	0.360	0.957	0.742	0.373	1.143	0.360	0.957	0.742	0.373	1.143		
91	1.073	0.609	1.160	0.629	0.119	1.073	0.609	1.160	0.629	0.119	1.073	0.609	1.160	0.629	0.119	1.073	0.609	1.160	0.629	0.119	1.073	0.609	1.160	0.629	0.119		
92	0.401	1.154	0.560	0.094	0.791	0.401	1.154	0.560	0.094	0.791	0.401	1.154	0.560	0.094	0.791	0.401	1.154	0.560	0.094	0.791	0.401	1.154	0.560	0.094	0.791		
93	0.344	0.485	0.414	0.575	0.667	0.344	0.485	0.414	0.575	0.667	0.344	0.485	0.414	0.575	0.667	0.344	0.485	0.414	0.575	0.667	0.344	0.485	0.414	0.575	0.667		
94	0.795	0.499	0.579	0.807	0.342	0.795	0.499	0.579	0.807	0.342	0.795	0.499	0.579	0.807	0.342	0.795	0.499	0.579	0.807	0.342	0.795	0.499	0.579	0.807	0.342		
95	0.054	0.584	1.016	1.161	0.324	0.054	0.584	1.016	1.161	0.324	0.054	0.584	1.016	1.161	0.324	0.054	0.584	1.016	1.161	0.324	0.054	0.584	1.016	1.161	0.324		
96	0.648	1.139	0.369	0.417	0.009	0.648	1.139	0.369	0.417	0.009	0.648	1.139	0.369	0.417	0.009	0.648	1.139	0.369	0.417	0.009	0.648	1.139	0.369	0.417	0.009		
97	0.381	0.225	1.053	1.111	0.919	0.381	0.225	1.053	1.111	0.919	0.381	0.225	1.053	1.111	0.919	0.381	0.225	1.053	1.111	0.919	0.381	0.225	1.053	1.111	0.919		
98	0.197	0.437	1.062	0.787	0.581	0.197	0.437	1.062	0.787	0.581	0.197	0.437	1.062	0.787	0.581	0.197	0.437	1.062	0.787	0.581	0.197	0.437	1.062	0.787	0.581		
99	1.174	0.286	0.465	0.562	0.388	1.174	0.286	0.465	0.562	0.388	1.174	0.286	0.465	0.562	0.388	1.174	0.286	0.465	0.562	0.388	1.174	0.286	0.465	0.562	0.388		
100	0.737	1.183	0.815	0.170	1.131	0.737	1.183	0.815	0.170	1.131	0.737	1.183	0.815	0.170	1.131	0.737	1.183	0.815	0.170	1.131	0.737	1.183	0.815	0.170	1.131		
Out-of-sample 15 (OOS15)																											
1	0.1	0.1	0.1	0.1	0.1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	
2	0.5	0.5	0.5	0.5	0.5	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	
3	1.15	1.15	1.15	1.15	1.15	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	
4	1	1	1	1	1	0.1	0.1	0.1	0.1	0.1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	
5	1	1	1	1	1	0.5	0.5	0.5	0.5	0.5	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	
6	1	1	1	1	1	1.15	1.15	1.15	1.15	1.15	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	
7	1	1	1	1	1	1	1	1	1	1	0.1	0.1	0.1	0.1	0.1	1	1	1	1	1	1	1	1	1	1	1	
8	1	1	1	1	1	1	1	1	1	1	0.5	0.5	0.5	0.5	0.5	1	1	1	1	1	1	1	1	1	1	1	
9	1	1	1	1	1	1	1	1	1	1	1.15	1.15	1.15	1.15	1.15	1	1	1	1	1	1	1	1	1	1	1	
10	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	0.1	0.1	0.1	0.1	0.1	0.1	1	1	1	1	1	
11	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	0.5	0.5	0.5	0.5	0.5	0.5	1	1	1	1	1	
12	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1.15	1.15	1.15	1.15	1.15	1.15	1	1	1	1	1	
13	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	0.1	0.1	0.1	0.1	0.1	
14	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	0.5	0.5	0.5	0.5	0.5	
15	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1.15	1.15	1.15	1.15	1.15	

Table S2. Performance of PM<sub>2.5</sub> and O<sub>3</sub> prediction using pf-RSM across grid cells

PM <sub>2.5</sub>		MaxNE		MeanNE					All
		Pollution levels							
		<25	25-50	50-75	75-100	100-125	>125		
Case1	0.88%	0.16%	0.16%	0.08%	0.07%	0.06%	0.03%	0.12%	
Case2	5.64%	0.76%	0.71%	0.79%	1.12%	2.22%	1.73%	0.90%	

O <sub>3</sub>		MaxNE		MeanNE					All
		Pollution levels							
		<60	60-70	70-80	80-90	90-100	>100		
Case1	0.79%	0.02%	0.05%	0.08%	0.09%	0.08%	0.16%	0.07%	
Case2	1.95%	0.08%	0.26%	0.30%	0.29%	0.22%	0.29%	0.26%	

\*PM<sub>2.5</sub> and O<sub>3</sub> concentrations are calculated based on monthly averaged concentrations at each grid cell

Table S3. Performance of PM<sub>2.5</sub> and O<sub>3</sub> prediction using pf-RSM across days

PM <sub>2.5</sub>		MaxNE	MeanNE						All
			Pollution levels						
			<25	25-50	50-75	75-100	100-125	>125	
Case1	Beijing	0.69%	0.23%	0.32%	0.21%	0.23%	0.36%	0.44%	0.28%
	Tianjin	1.23%	-	0.37%	0.13%	0.37%	0.37%	0.31%	0.30%
	Hebei N	0.65%	0.19%	0.24%	0.34%	-	-	-	0.23%
	Hebei E	0.51%	-	0.28%	0.13%	0.15%	0.20%	0.29%	0.21%
	Hebei S	0.40%	-	0.40%	0.17%	0.10%	0.10%	0.14%	0.14%
Case2	Beijing	12.69%	0.52%	0.36%	1.66%	3.37%	4.86%	6.74%	3.00%
	Tianjin	10.12%	-	0.95%	3.25%	4.46%	3.51%	3.63%	3.64%
	Hebei N	3.91%	1.61%	1.80%	1.00%	-	-	-	1.63%
	Hebei E	7.47%	-	0.58%	2.81%	2.65%	2.89%	3.62%	3.00%
	Hebei S	7.40%	-	0.48%	2.11%	3.42%	3.48%	3.29%	3.19%

O <sub>3</sub>		MaxNE	MeanNE						All
			Pollution levels						
			<60	60-70	70-80	80-90	90-100	>100	
Case1	Beijing	2.40%	0.29%	0.10%	1.39%	0.04%	0.85%	0.32%	0.46%
	Tianjin	5.32%	1.26%	0.12%	0.52%	0.18%	0.48%	1.55%	0.59%
	Hebei N	0.31%	0.03%	0.07%	0.07%	0.09%	0.17%	-	0.07%
	Hebei E	0.87%	0.52%	0.34%	0.15%	0.16%	0.18%	0.50%	0.26%
	Hebei S	0.54%	0.29%	0.19%	0.12%	0.23%	0.15%	0.38%	0.20%
Case2	Beijing	6.42%	0.68%	1.06%	4.00%	0.42%	1.25%	1.06%	1.24%
	Tianjin	4.72%	1.74%	0.45%	1.12%	0.22%	1.01%	1.00%	0.93%
	Hebei N	1.03%	0.21%	0.35%	0.05%	0.33%	0.54%	-	0.26%
	Hebei E	2.86%	1.83%	0.84%	0.93%	0.55%	0.52%	1.40%	0.87%
	Hebei S	2.61%	0.70%	0.27%	0.50%	0.46%	0.61%	1.17%	0.60%

\*PM<sub>2.5</sub> and O<sub>3</sub> concentrations are calculated based on daily averaged concentrations at averages of urban sites in each region

Table S4. Fitting coefficients of terms for PM<sub>2.5</sub> and O<sub>3</sub> in BTH

Pollutant	Month	Region	1*	2	3	4	5	6	7	8	9	10	11	12	13	14	15
PM <sub>2.5</sub>	January	Beijing	-70.66	-75.01	-5.53	-1.34	2.48	-24.33	22.29	-1.16	-2.14	5.44	20.42	13.94	3.96	6.06	13.40
		Tianjin	-60.70	-58.47	-8.04	-10.03	3.21	-10.58	-8.51	-15.22	0.02	10.22	36.62	29.14	9.56	10.77	22.70
		HebeiN	-12.13	-9.79	-0.37	-0.58	1.14	-6.68	3.76	-1.06	-0.37	2.00	8.93	6.52	2.97	2.09	5.26
		HebeiE	-95.99	-95.56	-3.52	-1.65	2.08	-31.97	29.28	-1.03	-2.94	4.74	28.76	19.70	5.57	7.46	24.03
		HebeiS	-82.06	-84.44	-3.53	-3.27	1.95	-28.66	28.79	-0.94	-4.58	4.72	27.26	17.01	4.41	8.58	26.72
	July	Beijing	-27.64	-29.09	-10.51	6.73	8.17	-6.41	5.11	4.55	-0.86	4.57	10.98	2.85	6.60	7.10	3.75
		Tianjin	-31.62	-30.77	-6.49	5.69	6.96	-8.64	6.99	2.69	-1.07	6.13	10.83	2.52	7.58	5.00	4.32
		HebeiN	-10.24	-14.19	-5.66	5.58	4.20	-1.70	2.09	2.30	-0.68	2.49	1.12	-2.45	3.18	4.40	1.91
		HebeiE	-27.35	-27.96	-10.04	6.70	5.78	-2.86	-1.68	1.59	-0.09	5.93	30.63	20.87	8.03	6.76	5.25
		HebeiS	-33.85	-37.48	-12.03	4.76	6.85	-8.76	6.88	2.02	-1.46	5.15	10.19	1.93	5.75	5.90	6.71
O <sub>3</sub>	January	Beijing	164.46	213.89	53.32	12.99	-15.10	28.82	36.10	-1.19	-1.87	0.29	-0.57	4.00	0.50	-2.67	0.16
		Tianjin	114.98	149.97	41.72	12.94	-17.77	22.77	21.12	-4.47	-0.58	0.17	-0.12	5.03	0.50	-2.19	-0.04
		HebeiN	21.80	11.73	-4.78	3.92	-6.01	12.43	8.14	-1.29	-1.11	-0.04	0.00	2.39	0.23	-1.01	-0.05
		HebeiE	130.94	167.06	48.38	12.69	-15.04	25.45	24.37	-1.12	-2.85	0.13	-0.32	5.36	-0.32	-3.34	0.02
		HebeiS	109.76	134.7	34.02	12.16	-17.35	24.55	21.26	-4.56	-2.21	0.18	-0.18	5.81	-0.08	-3.09	-0.08
	July	Beijing	102.76	138.300	62.14	-12.18	-10.61	10.05	-9.88	2.29	-1.41	0.17	-0.31	18.37	-3.78	-1.93	-0.05
		Tianjin	-2.30	-38.89	-13.37	-3.60	-16.67	9.17	-9.03	0.06	-3.53	0.27	-0.31	14.05	-1.70	-1.84	-0.05
		HebeiN	28.07	44.95	27.48	-2.94	2.87	1.87	-4.84	1.39	-2.21	-0.05	-0.29	6.19	-1.03	-1.29	0.01
		HebeiE	24.69	23.24	14.04	-6.69	-5.32	7.83	-3.72	3.74	-0.76	0.14	-0.26	11.77	-3.05	-2.32	-0.07
		HebeiS	59.88	74.92	35.79	-8.14	-12.52	4.75	-7.62	1.55	-0.84	0.14	-0.28	11.80	-1.72	-0.91	-0.04

\*The orders of terms are followed as Figure 4.

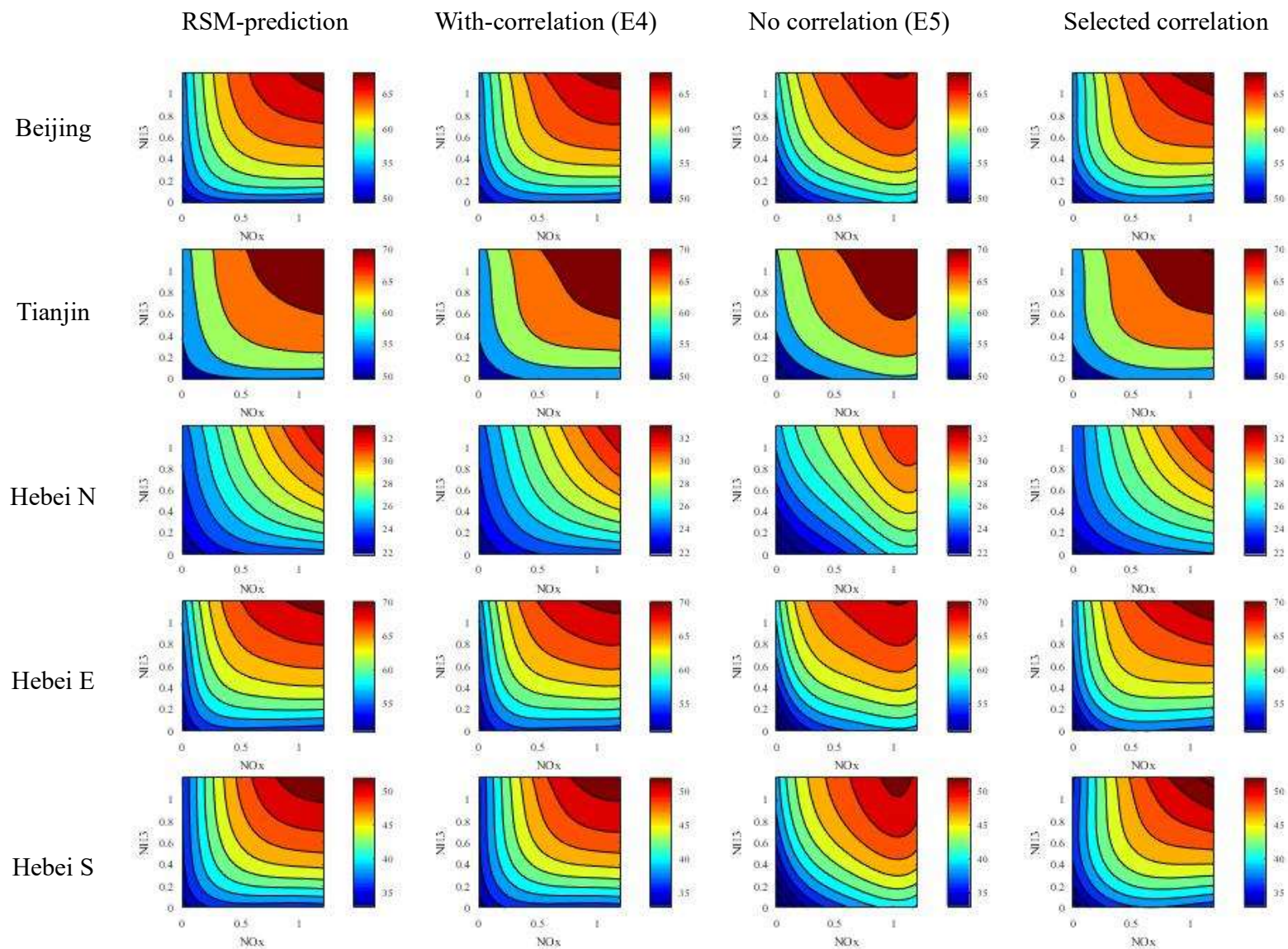


Figure S1. Interaction examination between  $NO_x$  and  $NH_3$  for  $PM_{2.5}$  (monthly averages in July 2014, The x and y axes shows  $1+E_{NOx}$  and  $1+E_{NH3}$ , The different colors represent  $PM_{2.5}$  concentrations, unit:  $\mu g m^{-3}$ )



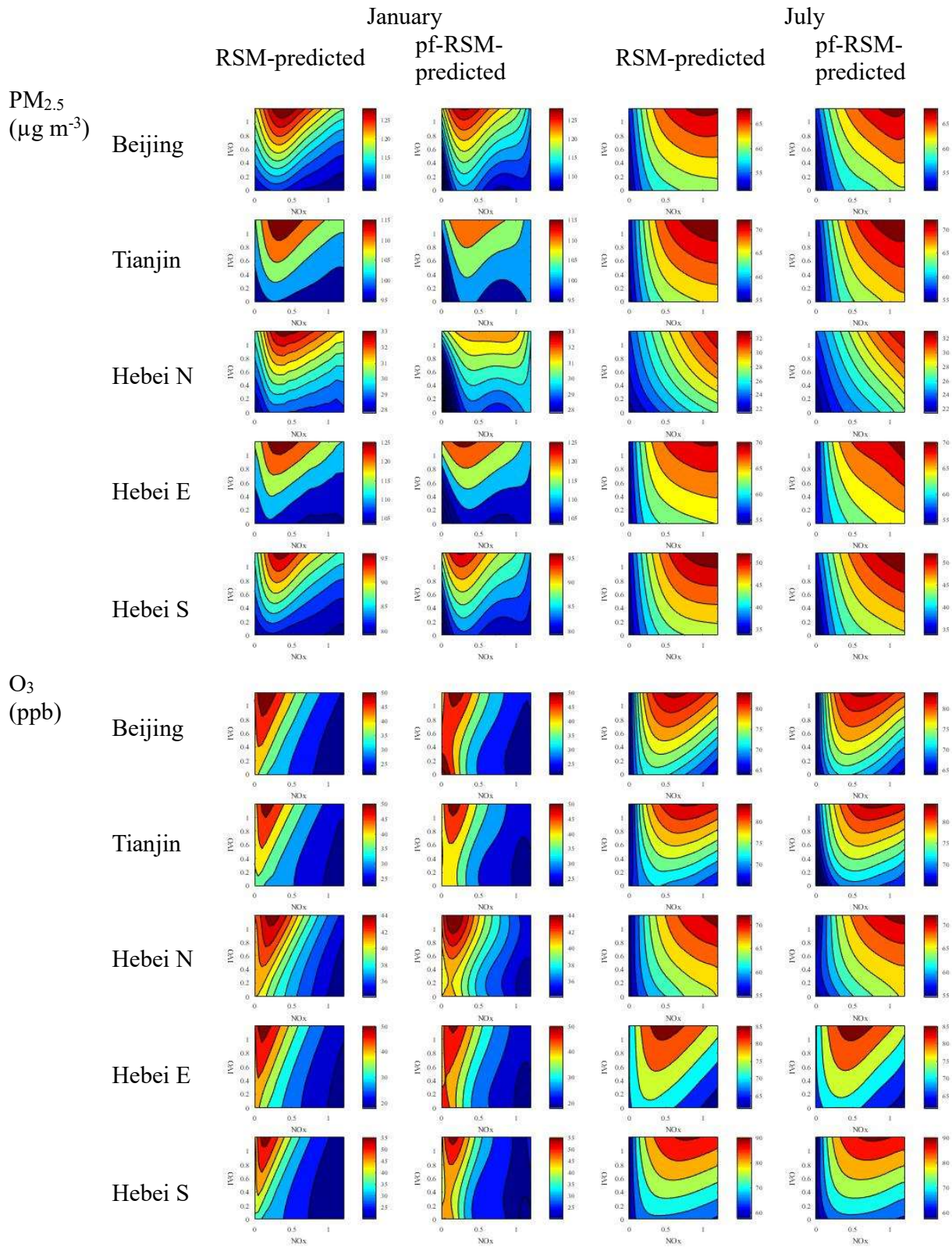


Figure S2. Isopleth validation of polynomial function with 20 training samples and even distributions (The x and y axes shows  $1+E_{\text{NO}_x}$  and  $1+E_{\text{VO}_x}$ , The different colors represent PM<sub>2.5</sub> or O<sub>3</sub> concentrations)



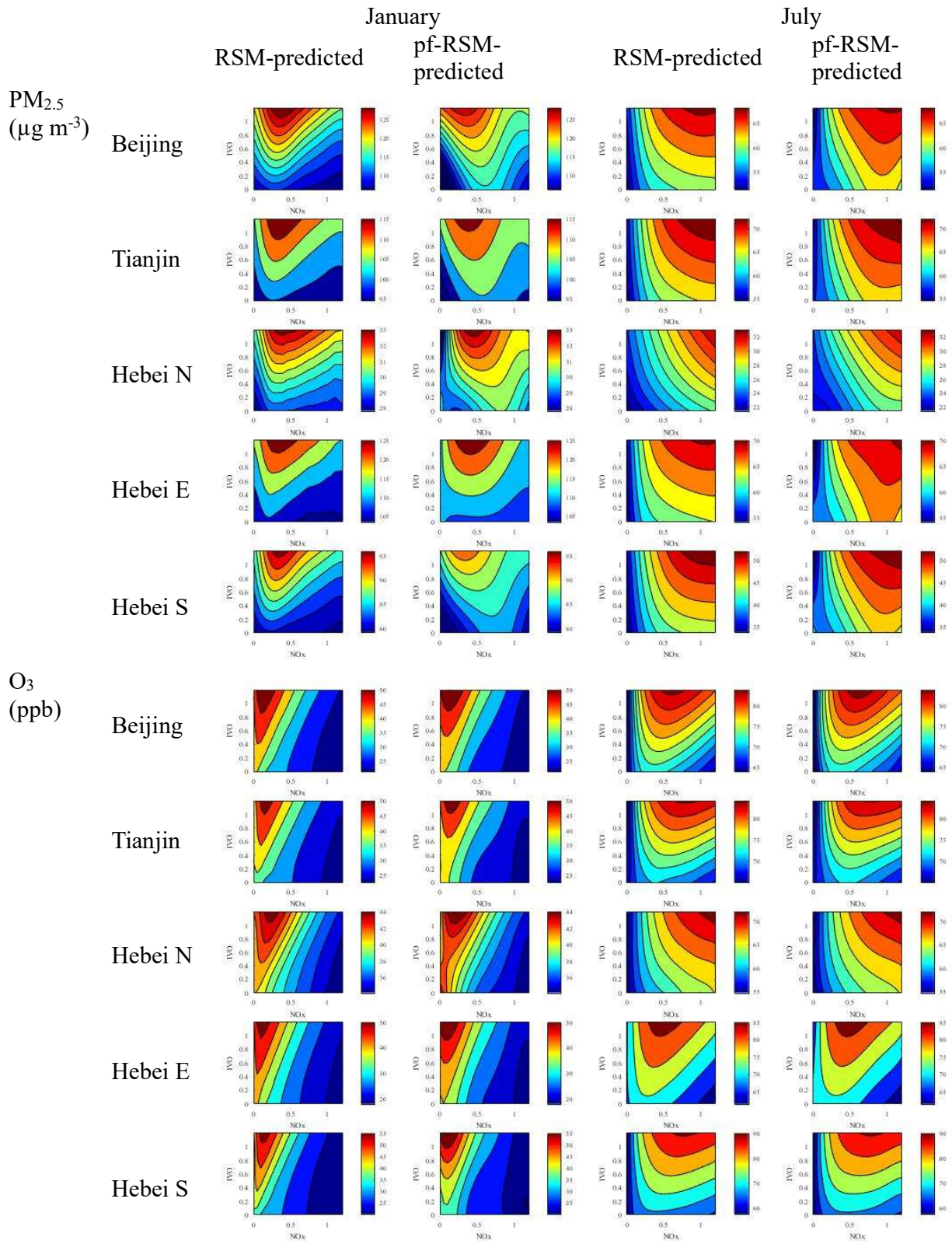


Figure S3. Same as Figure S1, for 20 training samples and marginal distributions

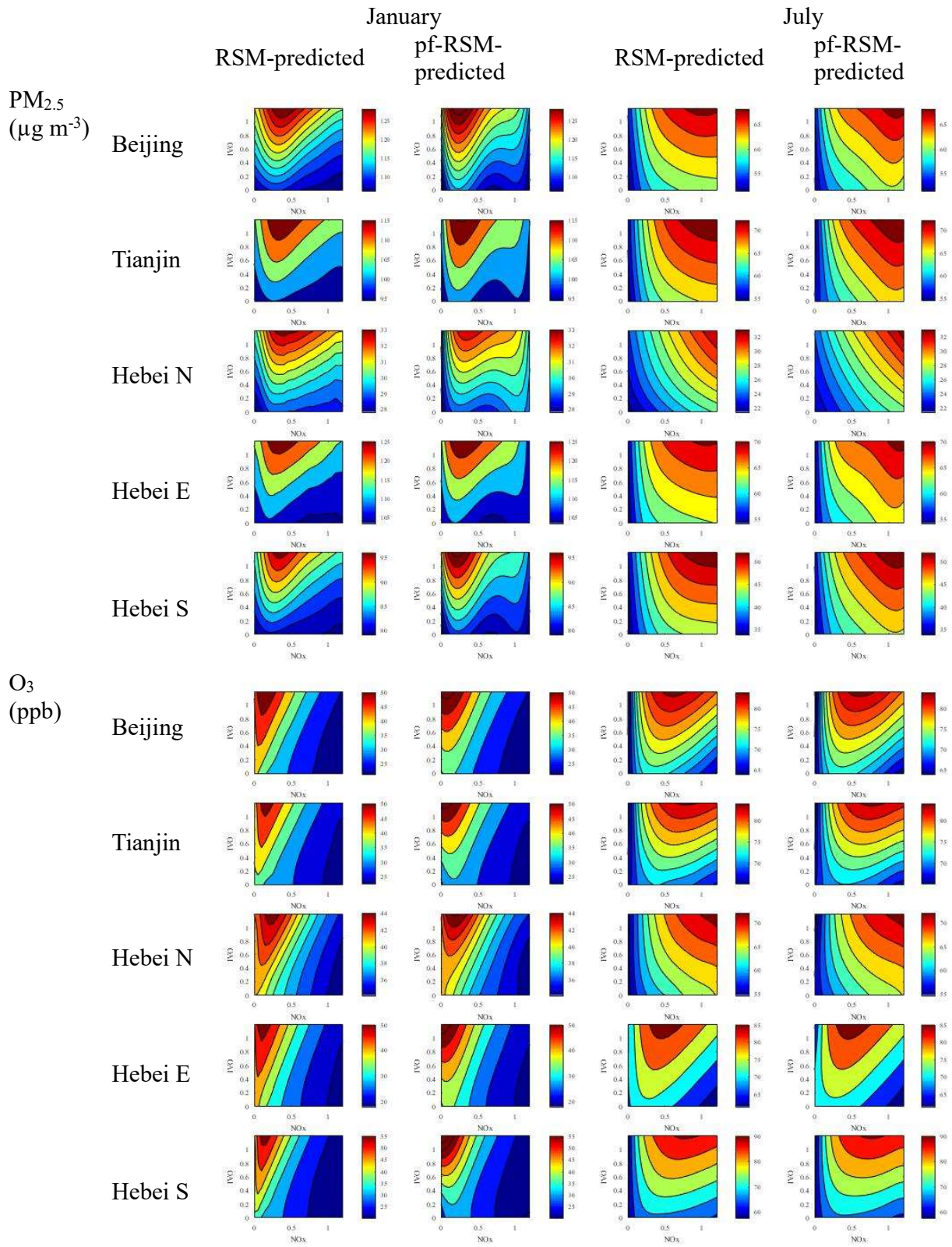


Figure S4. Same as Figure S1, for 30 training samples and even distributions



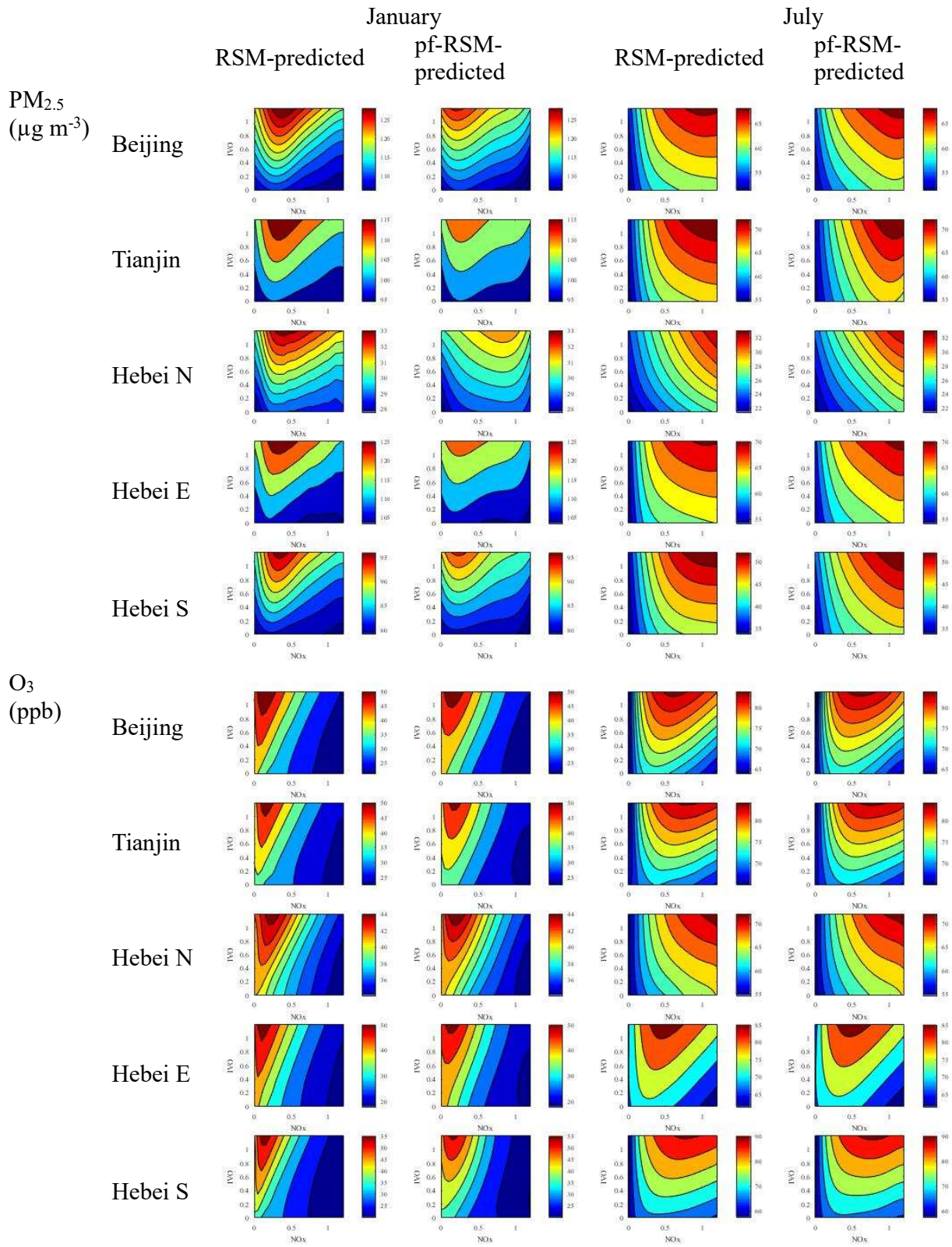


Figure S5. Same as Figure S1, for 30 training samples and marginal distributions

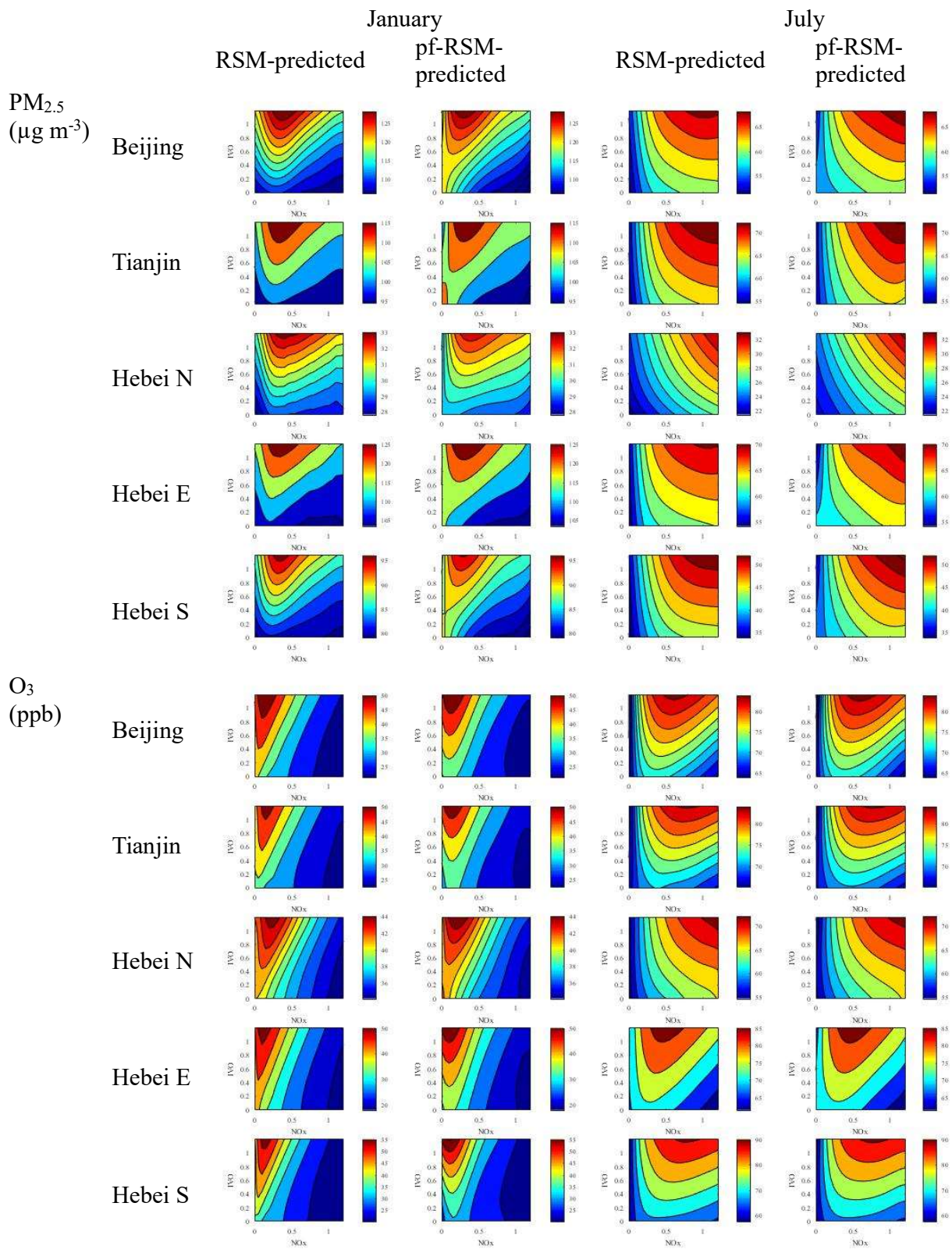


Figure S6. Same as Figure S1, for 40 training samples and even distributions



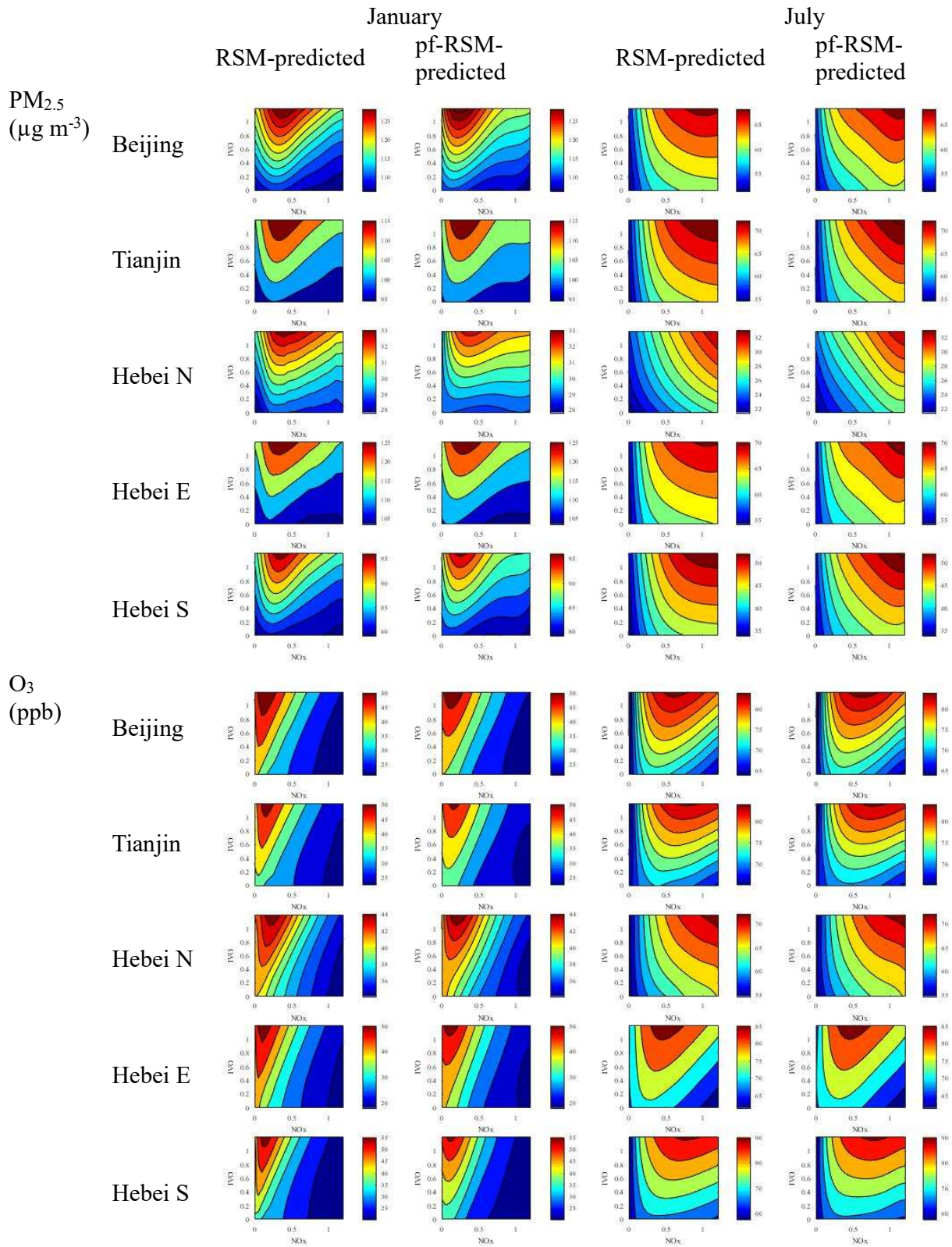


Figure S7. Same as Figure S1, for 40 training samples and marginal distributions

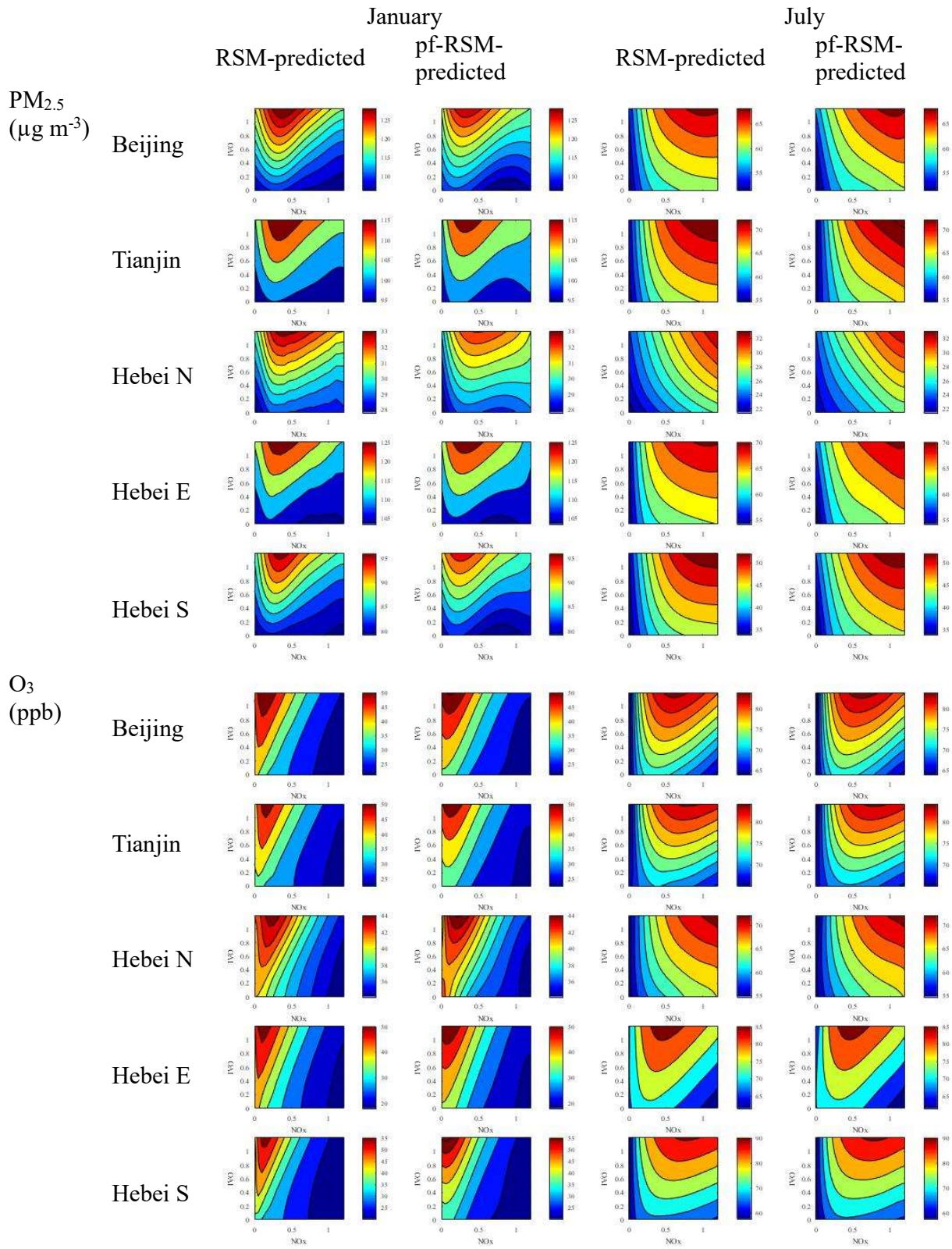


Figure S8. Same as Figure S1, for 50 training samples and even distributions



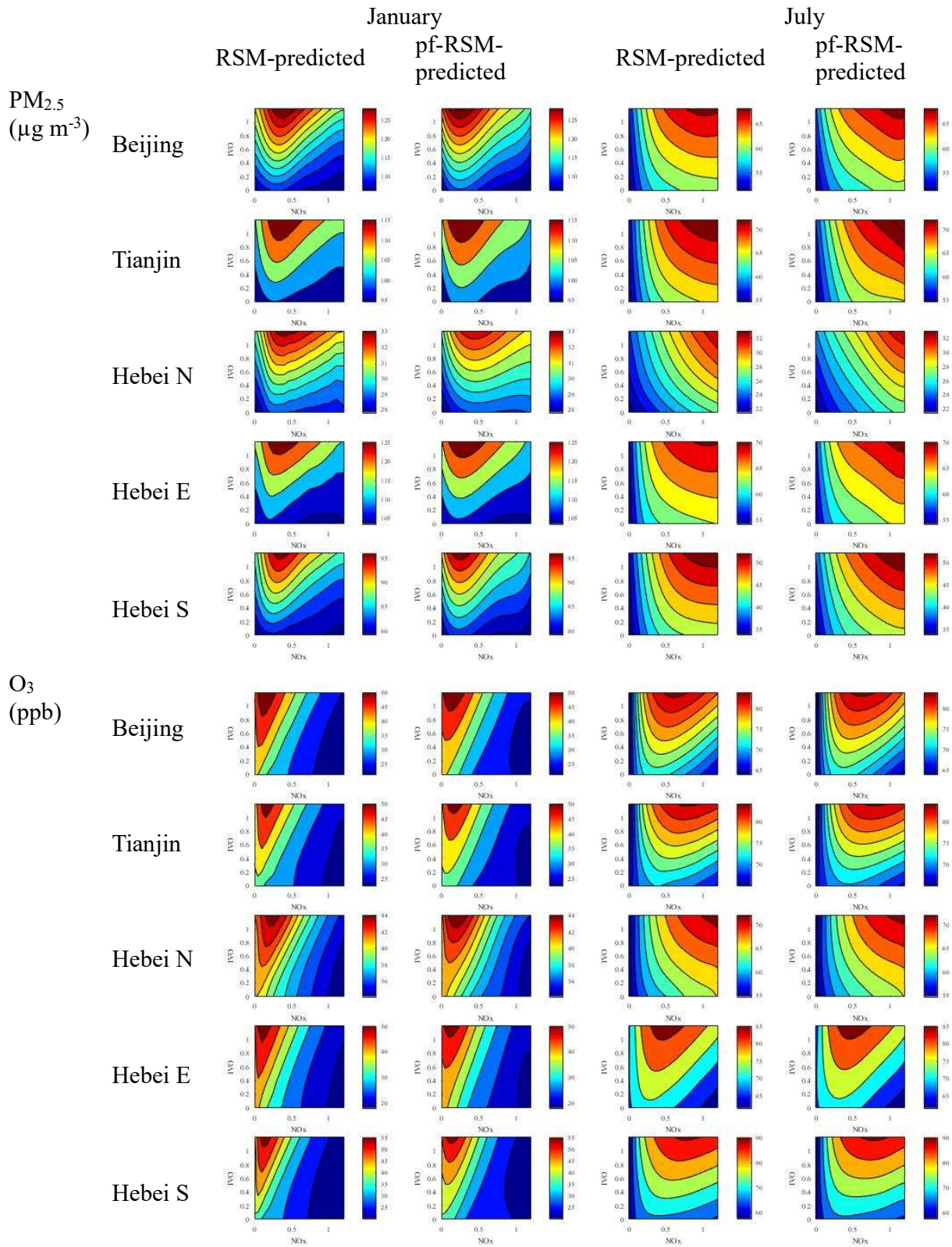


Figure S9. Same as Figure S1, for 50 training samples and marginal distributions