1. The calculation for total N_2O emissions in 1860 and 2006.

Total N_2O emission = Terrestrial direct emission from global soils + Other sources + Marine N_2O emissions + Atmospheric chemistry source

Other sources = Anthropogenic biological N_2O source + Terrestrial indirect N_2O emission from riverine

Taking "baseline" as an example: in 1860, total emission = 6.20 + 3.5 + 0.77 + 0.6 = 11.07 Tg N yr^{-1};

in 2006, total emission = 6.2 + 4.5 + 0.77 + 0.6 + 7.2 = 19.27 Tg N yr^{-1}.

Table 1S. The description of field measurements from natural vegetation in different sites.

PFT	location		Year	References
	Longitude	Latitude		
Forest: Spruce	11°25'E	48°46'N	1993-1995	Butterbach-Bahl et al., 1998
Forest: Spruce	09°34'E	51°46'N	2007-2008	Eickenscheidt and Brumme, 2012
Forest: Liana canopy	55°31'W	3°59'S	1998-2000	Davidson et al., 2004
Forest: Douglas-fir	124°30'W	44°00'N	2007-2008	Erickson and Perakis, 2014
Grassland	09°42'E	51°46'N	2008-2009	Hoeft et al., 2012
Forest	156°14'W	20°48'N	2000-2001	Holtgrieve et al., 2006
Forest: Spruce &Oak	19°57'–58'E	47°53'N	2002-2003	Horváth et al., 2006
Forest: Beech	16°15'E	48°14'N	2002-2004	Kitzler et al., 2006
Grassland	104°42'W	40°50'N	1997-2000	Mosier et al., 2002
Tropical rain forest	145°30'E	17°30'S	1997-1999	Breuer et al., 2000
Tropical rain forest	63°00'W	10°00'S	–	Stehfest and Bouwman, 2006
Savanna	28°30'E	24°30'S	1994	Scholes et al., 1997
Tropical forest	47°30'W	3°00'S	1987	Luizão et al., 1989
Tropical forest	115°30'E	2°00'S	1998-1999	Hadi et al., 2000
Tropical forest	84°00'W	10°26'N	1990-1991	Keller and Reiners, 1994
Subtropical forest	66°00'W	18°00'N	1995-1996	Erickson et al. 2001
Temperate forest	116°30'E	39°30'N	1997-1998	Sun and Xu, 2001
Temperate forest	89°00'W	43°00'N	1979-1981	Goodroad and Keeney, 1983
Grassland	116°04'E	43°26'N	1995	Chen et al., 2000
Temperate forest	126°55'E	41°23'N	1994-1995	

Table 2S. All results of the global-, continental-, and biome-level N_2O emission from 100 sets of DLEM simulations in 1860. The unit is Tg N yr^{-1}.

No.	Total	Europe	North America	South America	Southern Asia	Northern Asia	Oceania	Africa	Boreal Forest	Tropical Forest	Shrubland	Grassland	Cropland	Tundra
1	6.986	0.339	0.774	2.323	1.315	0.175	0.348	1.645	0.695	4.467	0.925	0.225	0.479	0.010
2	6.546	0.292	0.684	2.244	1.225	0.166	0.325	1.545	0.594	4.281	0.845	0.199	0.428	0.013
3	5.121	0.243	0.554	1.724	0.959	0.124	0.254	1.213	0.504	3.309	0.672	0.159	0.341	0.005
4	6.699	0.350	0.772	2.167	1.248	0.179	0.339	1.578	0.734	4.180	0.926	0.219	0.443	0.012
5	5.386	0.257	0.585	1.807	1.004	0.137	0.268	1.275	0.536	3.466	0.715	0.165	0.347	0.009
6	6.178	0.317	0.701	2.016	1.152	0.163	0.312	1.458	0.661	3.882	0.846	0.199	0.408	0.011
7	6.029	0.268	0.627	2.073	1.129	0.150	0.298	1.424	0.545	3.953	0.775	0.182	0.396	0.010
8	6.966	0.303	0.720	2.405	1.306	0.176	0.346	1.643	0.612	4.583	0.892	0.211	0.455	0.014
9	6.033	0.289	0.659	2.019	1.132	0.150	0.300	1.426	0.595	3.878	0.796	0.190	0.406	0.008
10	5.666	0.259	0.602	1.932	1.067	0.134	0.278	1.338	0.530	3.703	0.728	0.175	0.381	0.005
11	5.796	0.267	0.616	1.970	1.088	0.142	0.287	1.369	0.545	3.770	0.751	0.179	0.389	0.008
12	4.945	0.218	0.508	1.713	0.929	0.117	0.244	1.167	0.442	3.258	0.628	0.149	0.333	0.006
13	7.677	0.376	0.855	2.536	1.437	0.205	0.386	1.806	0.773	4.873	1.033	0.246	0.511	0.017
14	5.347	0.236	0.551	1.851	1.003	0.129	0.264	1.263	0.478	3.523	0.681	0.161	0.355	0.007
15	6.257	0.321	0.709	2.041	1.167	0.168	0.317	1.474	0.667	3.924	0.859	0.201	0.416	0.013
16	7.443	0.335	0.787	2.540	1.414	0.181	0.371	1.742	0.666	4.847	0.955	0.237	0.528	0.011
17	6.384	0.287	0.670	2.184	1.188	0.167	0.318	1.509	0.590	4.164	0.833	0.192	0.404	0.014
18	6.019	0.303	0.675	1.979	1.123	0.158	0.303	1.421	0.629	3.803	0.818	0.192	0.400	0.011
19	5.520	0.251	0.587	1.886	1.042	0.127	0.269	1.304	0.514	3.615	0.706	0.171	0.376	0.003
20	6.455	0.291	0.673	2.205	1.206	0.170	0.324	1.523	0.589	4.197	0.842	0.196	0.422	0.016
21	6.145	0.253	0.580	2.090	1.170	0.128	0.278	1.583	0.480	4.105	0.805	0.185	0.443	0.015
22	6.886	0.343	0.770	2.266	1.287	0.183	0.347	1.623	0.708	4.354	0.933	0.221	0.459	0.014
23	6.927	0.300	0.714	2.395	1.313	0.166	0.344	1.627	0.596	4.563	0.876	0.214	0.480	0.010
24	7.927	0.381	0.866	2.635	1.482	0.221	0.403	1.862	0.774	5.038	1.067	0.251	0.524	0.024
25	8.045	0.388	0.885	2.669	1.501	0.224	0.407	1.892	0.795	5.117	1.085	0.253	0.521	0.023
26	4.766	0.219	0.507	1.627	0.898	0.108	0.233	1.129	0.451	3.120	0.610	0.147	0.323	0.002
27	5.459	0.253	0.578	1.856	1.025	0.134	0.271	1.288	0.514	3.547	0.708	0.168	0.369	0.007
28	5.181	0.252	0.567	1.730	0.964	0.134	0.260	1.226	0.526	3.314	0.696	0.160	0.334	0.008

No.	Total	Europe	North America	South America	Southern Asia	Northern Asia	Oceania	Africa	Boreal Forest	Tropical Forest	Shrubland	Grassland	Cropland	Tundra
29	6.774	0.314	0.724	2.290	1.267	0.176	0.339	1.597	0.642	4.373	0.891	0.210	0.447	0.015
30	6.805	0.325	0.741	2.275	1.268	0.184	0.341	1.605	0.671	4.357	0.911	0.211	0.436	0.016
31	6.258	0.260	0.603	2.121	1.204	0.125	0.282	1.598	0.494	4.168	0.822	0.191	0.465	0.012
32	7.660	0.370	0.839	2.543	1.438	0.205	0.388	1.802	0.749	4.877	1.024	0.245	0.517	0.019
33	6.276	0.319	0.711	2.054	1.167	0.167	0.316	1.481	0.670	3.955	0.860	0.199	0.404	0.012
34	6.591	0.294	0.691	2.261	1.242	0.160	0.326	1.551	0.595	4.315	0.845	0.204	0.447	0.010
35	5.087	0.224	0.525	1.763	0.963	0.115	0.249	1.198	0.451	3.360	0.639	0.156	0.354	0.003
36	8.133	0.381	0.878	2.728	1.521	0.222	0.411	1.911	0.772	5.213	1.081	0.255	0.536	0.024
37	6.066	0.293	0.665	2.026	1.144	0.147	0.301	1.431	0.597	3.895	0.796	0.194	0.421	0.007
38	4.979	0.243	0.545	1.660	0.930	0.125	0.249	1.178	0.505	3.185	0.664	0.155	0.331	0.007
39	5.230	0.249	0.569	1.760	0.980	0.124	0.257	1.240	0.519	3.386	0.685	0.163	0.345	0.004
40	6.075	0.275	0.641	2.076	1.135	0.153	0.301	1.435	0.566	3.964	0.788	0.185	0.394	0.010
41	6.822	0.266	0.681	2.228	1.223	0.264	0.519	1.577	0.550	4.225	1.074	0.233	0.440	0.050
42	5.872	0.242	0.582	2.075	1.113	0.135	0.288	1.380	0.477	3.931	0.724	0.176	0.407	0.006
43	6.743	0.298	0.702	2.317	1.261	0.172	0.335	1.592	0.607	4.416	0.870	0.204	0.438	0.014
44	7.161	0.380	0.832	2.300	1.338	0.195	0.365	1.682	0.788	4.435	0.996	0.237	0.483	0.016
45	5.090	0.224	0.525	1.764	0.954	0.121	0.250	1.203	0.457	3.358	0.647	0.153	0.337	0.006
46	5.912	0.253	0.596	2.061	1.108	0.147	0.294	1.395	0.506	3.908	0.749	0.176	0.393	0.011
47	6.370	0.332	0.731	2.066	1.191	0.165	0.321	1.502	0.692	3.987	0.873	0.209	0.428	0.010
48	5.153	0.214	0.513	1.821	0.967	0.120	0.252	1.216	0.432	3.452	0.640	0.151	0.339	0.006
49	6.057	0.290	0.665	2.030	1.138	0.146	0.298	1.431	0.600	3.906	0.795	0.192	0.407	0.006
50	4.905	0.217	0.507	1.699	0.922	0.114	0.241	1.158	0.441	3.236	0.621	0.148	0.331	0.004
51	4.862	0.227	0.521	1.650	0.912	0.115	0.239	1.153	0.470	3.165	0.631	0.149	0.322	0.004
52	7.706	0.355	0.825	2.603	1.441	0.206	0.387	1.814	0.724	4.972	1.016	0.239	0.504	0.020
53	6.758	0.306	0.708	2.304	1.266	0.176	0.339	1.593	0.617	4.390	0.881	0.207	0.448	0.016
54	5.321	0.248	0.565	1.806	0.993	0.136	0.266	1.255	0.508	3.443	0.699	0.162	0.349	0.009
55	5.758	0.281	0.635	1.914	1.076	0.146	0.288	1.362	0.584	3.678	0.769	0.181	0.382	0.008
56	5.814	0.247	0.586	2.033	1.078	0.150	0.288	1.375	0.503	3.850	0.742	0.169	0.362	0.014

No.	Total	Europe	North America	South America	Southern Asia	Northern Asia	Oceania	Africa	Boreal Forest	Tropical Forest	Shrubland	Grassland	Cropland	Tundra
57	7.764	0.400	0.887	2.516	1.452	0.215	0.396	1.822	0.823	4.840	1.070	0.254	0.523	0.020
58	5.386	0.230	0.545	1.884	1.019	0.124	0.265	1.268	0.459	3.581	0.672	0.162	0.370	0.005
59	5.222	0.232	0.543	1.801	0.970	0.131	0.259	1.236	0.482	3.427	0.676	0.154	0.328	0.009
60	6.341	0.290	0.667	2.159	1.182	0.168	0.318	1.495	0.589	4.110	0.832	0.192	0.411	0.016
61	7.150	0.326	0.761	2.430	1.354	0.175	0.356	1.678	0.653	4.645	0.922	0.227	0.500	0.011
62	5.007	0.229	0.531	1.709	0.947	0.113	0.245	1.183	0.469	3.275	0.639	0.156	0.348	0.002
63	5.877	0.259	0.610	2.030	1.112	0.138	0.289	1.383	0.521	3.869	0.744	0.181	0.406	0.006
64	5.175	0.231	0.539	1.786	0.976	0.119	0.254	1.220	0.468	3.409	0.655	0.158	0.353	0.004
65	6.739	0.324	0.740	2.244	1.255	0.179	0.338	1.591	0.674	4.307	0.905	0.210	0.433	0.014
66	5.383	0.253	0.577	1.819	1.006	0.134	0.268	1.273	0.521	3.483	0.706	0.165	0.353	0.008
67	6.911	0.344	0.772	2.276	1.296	0.181	0.347	1.628	0.705	4.376	0.930	0.223	0.469	0.013
68	5.503	0.219	0.536	1.967	1.031	0.128	0.268	1.299	0.440	3.719	0.675	0.158	0.357	0.007
69	6.429	0.297	0.684	2.181	1.201	0.166	0.321	1.516	0.606	4.164	0.843	0.197	0.420	0.013
70	7.045	0.333	0.766	2.361	1.326	0.179	0.351	1.660	0.678	4.531	0.927	0.223	0.479	0.012
71	6.688	0.309	0.720	2.263	1.268	0.159	0.330	1.573	0.626	4.338	0.864	0.214	0.470	0.007
72	5.739	0.280	0.633	1.911	1.077	0.141	0.285	1.357	0.579	3.675	0.760	0.183	0.390	0.006
73	4.956	0.232	0.529	1.679	0.925	0.123	0.247	1.172	0.480	3.207	0.651	0.152	0.326	0.007
74	5.216	0.252	0.566	1.746	0.974	0.134	0.261	1.233	0.519	3.342	0.695	0.161	0.345	0.009
75	5.450	0.258	0.592	1.838	1.031	0.125	0.267	1.287	0.529	3.534	0.704	0.173	0.379	0.002
76	5.734	0.238	0.570	2.021	1.069	0.141	0.282	1.357	0.481	3.828	0.720	0.167	0.367	0.010
77	5.783	0.273	0.621	1.948	1.086	0.144	0.289	1.366	0.557	3.731	0.759	0.180	0.391	0.009
78	5.469	0.223	0.541	1.938	1.027	0.127	0.267	1.291	0.450	3.673	0.676	0.160	0.360	0.007
79	7.412	0.352	0.802	2.480	1.387	0.200	0.374	1.744	0.714	4.743	0.987	0.233	0.491	0.020
80	5.884	0.281	0.641	1.972	1.107	0.144	0.292	1.390	0.576	3.789	0.772	0.186	0.402	0.007
81	5.102	0.218	0.519	1.786	0.961	0.116	0.249	1.203	0.442	3.398	0.637	0.152	0.342	0.003
82	6.319	0.291	0.673	2.144	1.189	0.156	0.314	1.491	0.592	4.101	0.821	0.197	0.429	0.009
83	5.903	0.276	0.637	1.995	1.120	0.136	0.290	1.391	0.560	3.829	0.760	0.189	0.418	0.004
84	5.845	0.250	0.599	2.037	1.102	0.136	0.285	1.378	0.508	3.880	0.734	0.176	0.391	0.006

No.	Total	Europe	North America	South America	Southern Asia	Northern Asia	Oceania	Africa	Boreal Forest	Tropical Forest	Shrubland	Grassland	Cropland	Tundra
85	6.217	0.300	0.683	2.075	1.162	0.158	0.309	1.469	0.622	3.986	0.827	0.195	0.408	0.010
86	6.938	0.312	0.729	2.371	1.307	0.173	0.346	1.633	0.627	4.525	0.894	0.215	0.471	0.013
87	5.874	0.276	0.631	1.981	1.093	0.154	0.294	1.387	0.572	3.785	0.779	0.180	0.377	0.012
88	5.571	0.276	0.619	1.847	1.043	0.138	0.277	1.318	0.573	3.556	0.743	0.177	0.372	0.006
89	5.908	0.260	0.608	2.040	1.099	0.153	0.294	1.396	0.531	3.875	0.764	0.175	0.375	0.013
90	6.759	0.304	0.708	2.307	1.259	0.181	0.339	1.595	0.619	4.394	0.885	0.204	0.433	0.018
91	6.084	0.306	0.685	2.002	1.141	0.151	0.303	1.438	0.637	3.862	0.816	0.197	0.411	0.007
92	5.669	0.279	0.626	1.880	1.058	0.148	0.285	1.339	0.579	3.608	0.763	0.178	0.372	0.011
93	6.102	0.278	0.644	2.080	1.139	0.157	0.304	1.440	0.570	3.967	0.797	0.185	0.396	0.012
94	6.085	0.281	0.646	2.065	1.136	0.157	0.304	1.436	0.576	3.939	0.800	0.186	0.397	0.012
95	8.086	0.378	0.872	2.716	1.519	0.214	0.408	1.899	0.760	5.194	1.067	0.256	0.547	0.021
96	7.247	0.342	0.784	2.429	1.354	0.195	0.365	1.707	0.700	4.648	0.964	0.226	0.474	0.018
97	7.495	0.344	0.797	2.537	1.406	0.197	0.377	1.764	0.695	4.845	0.982	0.234	0.500	0.018
98	7.284	0.365	0.822	2.389	1.358	0.196	0.367	1.716	0.760	4.597	0.992	0.233	0.476	0.016
99	6.607	0.341	0.756	2.148	1.232	0.175	0.333	1.557	0.714	4.141	0.908	0.214	0.436	0.012
100	7.233	0.334	0.775	2.446	1.351	0.190	0.361	1.705	0.683	4.678	0.951	0.223	0.469	0.016

2. The increase amount of the human-induced N_2O emission during 1860–2006

According to Syakila and Kroeze (2011), the anthropogenic emission is the sum from energy, biomass burning, agriculture, and ocean. As their estimates were not continuous during 1860–2006, we used the linear interpolation to get the annual increase, which was also the approach they used to get the continuous N_2O concentration trends.

Table 3S. The net increase of N_2O emission from Syakila and Kroeze (2011).

Year	Net increase between two-time period (Tg N yr^{-1})	Total (Tg N yr^{-1})
1850	0	7.2
1900	0.2	
1930	0.6	
1950	0.6	
1960	0.8	
1970	1.2	
1975	0.7	
1980	0.7	
1985	0.7	
1990	0.6	
1994	0.3	
2000	0.3	
2002	0.1	
2004	0.3	
2006	0.1	

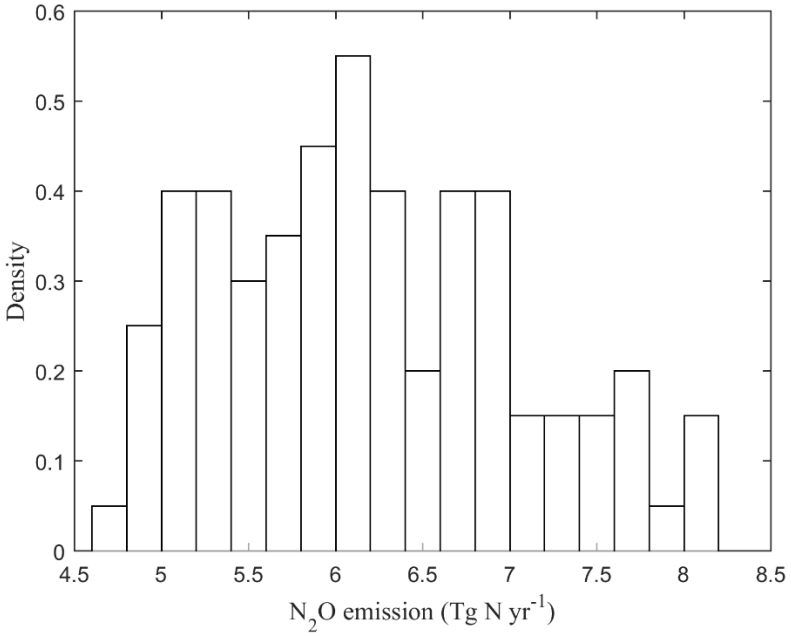

Figure 1S. The distribution of 100 sets of results from DLEM simulations.

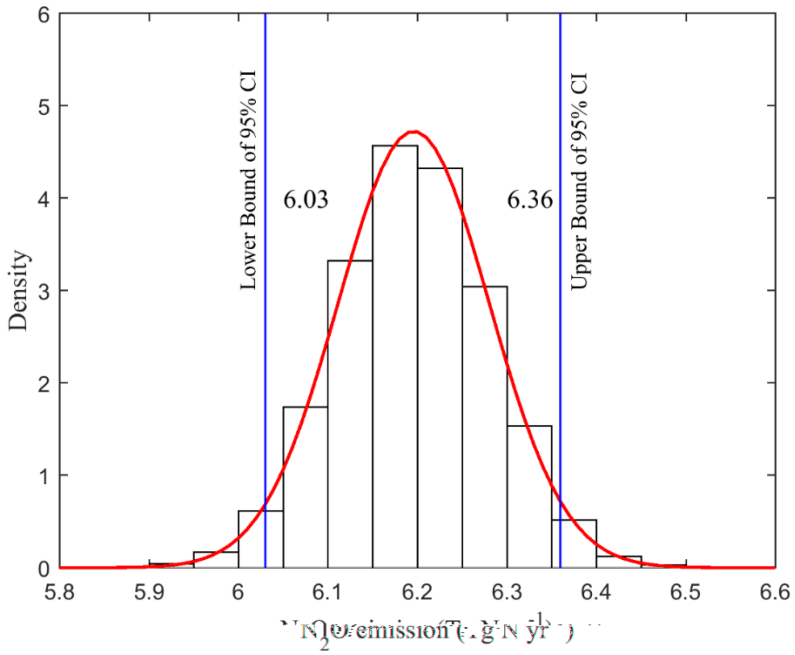

Figure 2S. The 95% confidence intervals of the mean pre-industrial N_2O emission using the Bootstrap resampling method with 10,000 replicates.