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Supplement of

Chemical footprints of anthropogenic nitrogen deposition on recent soil C : N ratios in Europe

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SUPPLEMENT

The N deposition values used in this paper originate from the EMEP atmospheric dispersion model(s) that provide results on a $150 \text{ km} \times 150 \text{ m}$ grid (older model version) and a compatible 3×3 subdivision of $50 \text{ km} \times 50 \text{ km}$, both in a polar stereographic projection (see Fig. S1). For more information on the EMEP grid see www.emep.int/grid and Posch et al. (*Calculation and mapping of critical thresholds in Europe: Status Report 1999*, RIVM, Bilthoven).

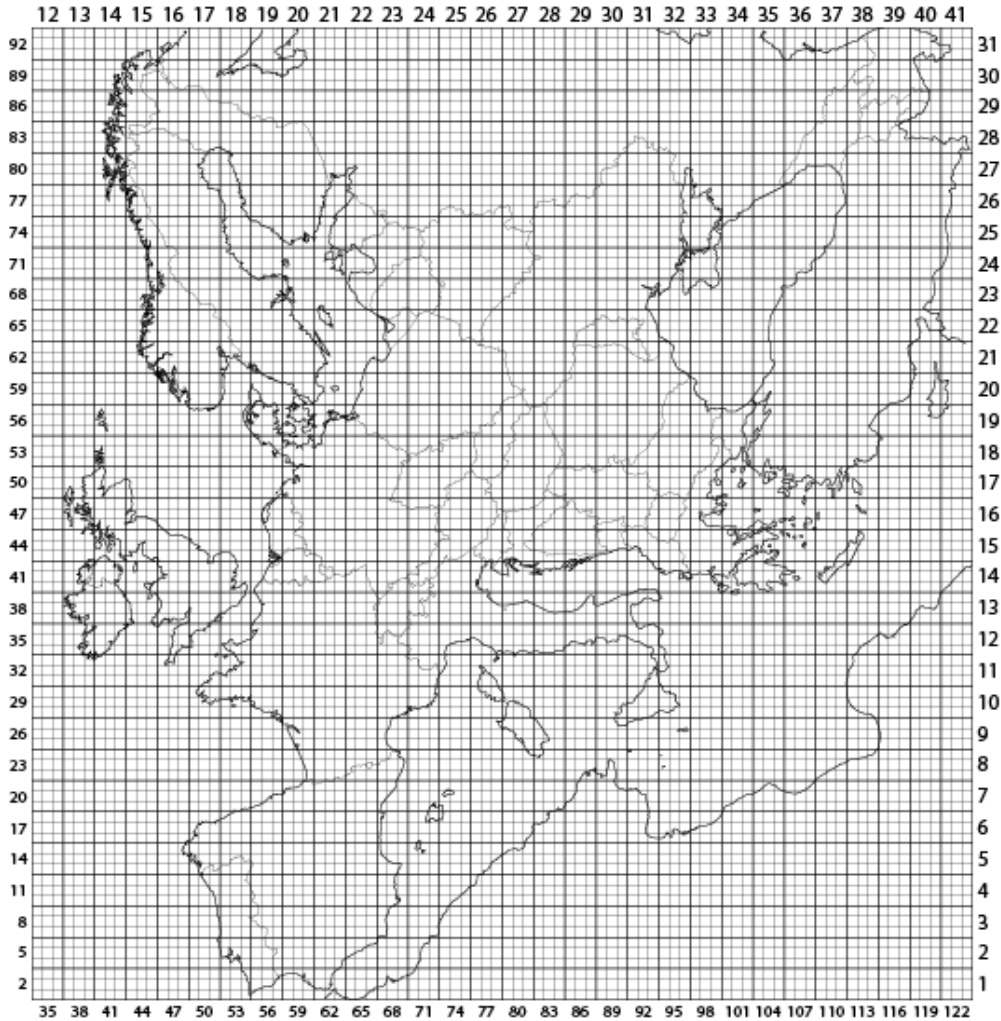


Fig. S1. The EMEP150 grid (thick lines) and the EMEP50 grid (additional thin lines). The labels at the bottom and at the left are the EMEP50 grid indices (every third cell labeled) and the labels at the top and at the right are the EMEP150 grid indices.

A bilinear interpolation is called that way because it is the product of two linear functions. To obtain this kind of interpolation (i.e. the value of the deposition field) at an arbitrary point (x,y) in a given grid cell (irrespective of its size), when it is known at the four corner points (x_1,y_1) , (x_2,y_1) , (x_2,y_2) and (x_1,y_2) (see Fig. S2), we firstly interpolate linearly in the x-direction:

$$(1a) f(x, y_1) \approx (1 - \lambda)f(x_1, y_1) + \lambda f(x_2, y_1) \quad \text{and}$$

$$(1b) f(x, y_2) \approx (1 - \lambda)f(x_1, y_2) + \lambda f(x_2, y_2) \quad \text{with} \quad \lambda = \frac{x - x_1}{x_2 - x_1}$$

Then we interpolate linearly between these two values in the y -direction to obtain the desired estimate:

$$(2) f(x, y) \approx (1 - \mu)f(x, y_1) + \mu f(x, y_2) \quad \text{with} \quad \mu = \frac{y - y_1}{y_2 - y_1}$$

Inserting eqs. 1a,b into eq. 2, this results in the bilinear interpolation formula:

$$(3) f(x, y) \approx (1 - \lambda)(1 - \mu)f(x_1, y_1) + \lambda(1 - \mu)f(x_2, y_1) + (1 - \lambda)\mu f(x_1, y_2) + \lambda\mu f(x_2, y_2)$$

Note that the same result is obtained if the interpolation is firstly done along the y -direction and then along the x -direction.

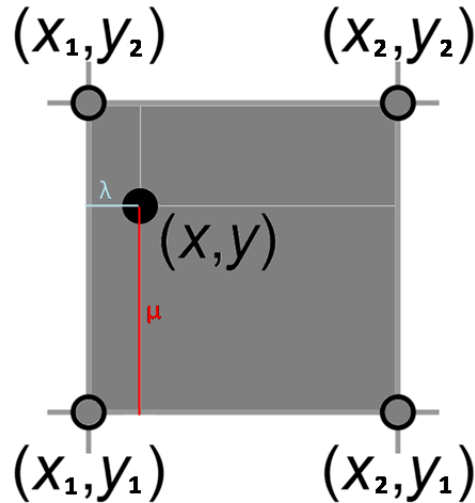


Fig. S2. Graphical representation of the notation used for the bilinear interpolation.

Table S1. Clustering NH₃

Nitrogen Variables

1880NH3 1885NH3 1890NH3 1895NH3 1900NH3 1905NH3 1910NH3 1915NH3 1920NH3 1925NH3 1930NH3 1935NH3 1940NH3
 1945NH3 1950NH3 1955NH3 1960NH3 1965NH3 1970NH3 1975NH3 1980NH3 1985NH3 1990NH3 1991NH3 1992NH3 1993NH3
 1994NH3 1995NH3 1996NH3 1997NH3 1998NH3 1999NH3 2000NH3 2001NH3 2002NH3 2003NH3 2004NH3 2005NH3 2006NH3
 2007NH3 2008NH3 2009NH3 2010NH3

Other Variables/Categories

LAT LONG SOIL2009C2N NATURE CROPLANDS AT BE CZ DE DK EE ES FI FR GR HU IE IT LT LU LV NL PL PT SE SI SK UK

Cluster Distribution

		N	% of Total
Cluster	Dep. Cluster I	1966	10.1%
	Dep. Cluster II	17492	89.9%
Total		19458	100.0%

Auto-Clustering

Number of Clusters	Akaike's Information Criterion (AIC)	AIC Change(a)	Ratio of AIC Changes(b)	Ratio of Distance Measures(c)
1	10338845.606			
2	10159324.403	-179521.202	1.000	2.461
3	10152583.943	-6740.461	.038	1.288
4	10144239.062	-8344.880	.046	1.115
5	9887604.407	-256634.655	1.430	1.299
6	9883536.784	-4067.623	.023	1.275
7	9883671.148	134.364	-.001	1.055
8	9883030.657	-640.491	.004	1.463
9	9882149.697	-880.961	.005	1.061
10	9881637.258	-512.438	.003	1.043
11	9881565.742	-71.517	.000	1.231
12	9881315.047	-250.695	.001	1.032
13	9858636.053	-22678.994	.126	1.072
14	9858732.023	95.971	-.001	1.030
15	9858710.377	-21.647	.000	1.067
16	9855987.262	-2723.114	.015	1.074
17	9851773.248	-4214.014	.023	1.133
18	9851904.244	130.996	-.001	1.014
19	9830028.150	-21876.093	.122	1.015
20	9829147.668	-880.482	.005	1.032
21	9829283.418	135.750	-.001	1.054
22	9829324.232	40.814	.000	1.045
23	9829334.484	10.252	.000	1.004
24	9829247.632	-86.851	.000	1.035
25	9829374.037	126.405	-.001	1.094
26	9829434.604	60.567	.000	1.000
27	9829587.361	152.757	-.001	1.013
28	9829732.352	144.990	-.001	1.019
29	9829869.404	137.052	-.001	1.005
30	9829976.122	106.718	-.001	1.011
31	9829955.815	-20.307	.000	1.042
32	9829705.691	-250.124	.001	1.002
33	9829719.339	13.648	.000	1.009
34	9829858.248	138.909	-.001	1.014
35	9829864.308	6.060	.000	1.008
36	9814184.573	-15679.735	.087	1.010
37	9814273.389	88.816	.000	1.301
38	9814237.520	-35.869	.000	.854
39	9813919.944	-317.577	.002	1.032
40	9814085.488	165.544	-.001	1.041

a The changes are from the previous number of clusters in the table.

b The ratios of changes are relative to the change for the two cluster solution.

c The ratios of distance measures are based on the current number of clusters against the previous number of clusters.

Table S2. Clustering NO_x

Nitrogen Variables

1880NO_x 1885NO_x 1890NO_x 1895NO_x 1900NO_x 1905NO_x 1910NO_x 1915NO_x 1920NO_x 1925NO_x 1930NO_x 1935NO_x 1940NO_x
 1945NO_x 1950NO_x 1955NO_x 1960NO_x 1965NO_x 1970NO_x 1975NO_x 1980NO_x 1985NO_x 1990NO_x 1991NO_x 1992NO_x 1993NO_x
 1994NO_x 1995NO_x 1996NO_x 1997NO_x 1998NO_x 1999NO_x 2000NO_x 2001NO_x 2002NO_x 2003NO_x 2004NO_x 2005NO_x 2006NO_x
 2007NO_x 2008NO_x 2009NO_x 2010NO_x

Other Variables/Categories

LAT LONG SOIL2009C2N NATURE CROPLANDS AT BE CZ DE DK EE ES FI FR GR HU IE IT LT LU LV NL PL PT SE SI SK UK

Cluster Distribution

		N	% of Total
Cluster	Dep. Cluster III	10691	54.9%
	Dep. Cluster IV	8359	43.0%
	Dep. Cluster V	408	2.1%
Total		19458	100.0%

Auto-Clustering

Number of Clusters	Akaike's Information Criterion (AIC)	AIC Change(a)	Ratio of AIC Changes(b)	Ratio of Distance Measures(c)
1	8948406.515			
2	8615207.947	-333198.568	1.000	1.173
3	8594765.051	-20442.897	.061	1.770
4	8509224.562	-85540.489	.257	1.009
5	8508934.323	-290.239	.001	1.153
6	8507044.402	-1889.921	.006	1.203
7	8507117.447	73.045	.000	1.647
8	8501619.088	-5498.359	.017	1.028
9	8501771.293	152.205	.000	1.265
10	8501048.873	-722.420	.002	1.026
11	8464202.886	-36845.987	.111	1.271
12	8447395.238	-16807.647	.050	1.140
13	8447028.722	-366.516	.001	.944
14	8438030.186	-8998.537	.027	1.050
15	8436141.198	-1888.987	.006	1.008
16	8435562.443	-578.755	.002	1.020
17	8435557.625	-4.818	.000	1.026
18	8434018.146	-1539.479	.005	1.176

19	8433909.262	-108.884	.000	1.007
20	8431167.895	-2741.367	.008	1.054
21	8430981.590	-186.305	.001	1.019
22	8428448.332	-2533.258	.008	1.204
23	8425099.865	-3348.467	.010	1.061
24	8425172.499	72.634	.000	1.024
25	8424192.875	-979.624	.003	1.053
26	8424352.498	159.623	.000	1.019
27	8424484.745	132.247	.000	1.000
28	8424487.087	2.342	.000	1.006
29	8424466.224	-20.863	.000	1.036
30	8420783.191	-3683.033	.011	1.027
31	8420915.630	132.439	.000	1.038
32	8421039.020	123.390	.000	1.005
33	8416530.458	-4508.562	.014	1.008
34	8416691.617	161.159	.000	1.012
35	8416525.564	-166.052	.000	1.015
36	8416539.265	13.700	.000	1.019
37	8416492.218	-47.047	.000	1.048
38	8413792.363	-2699.855	.008	1.008
39	8410730.232	-3062.131	.009	1.004
40	8410719.097	-11.135	.000	1.004

- a The changes are from the previous number of clusters in the table.
- b The ratios of changes are relative to the change for the two cluster solution.
- c The ratios of distance measures are based on the current number of clusters against the previous number of clusters.

Table S3. Clustering Nr (NO_x+NH₃)

Nitrogen Variables

1880Nr 1885Nr 1890Nr 1895Nr 1900Nr 1905Nr 1910Nr 1915Nr 1920Nr 1925Nr 1930Nr 1935Nr 1940Nr 1945Nr 1950Nr 1955Nr 1960Nr 1965Nr 1970Nr 1975Nr 1980Nr 1985Nr 1990Nr 1991Nr 1992Nr 1993Nr 1994Nr 1995Nr 1996Nr 1997Nr 1998Nr 1999Nr 2000Nr 2001Nr 2002Nr 2003Nr 2004Nr 2005Nr 2006Nr 2007Nr 2008Nr 2009Nr 2010Nr

Other Variables/Categories

LAT LONG SOIL2009C2N NATURE CROPLANDS AT BE CZ DE DK EE ES FI FR GR HU IE IT LT LU LV NL PL PT SE SI SK UK

Cluster Distribution

		N	% of Total
Cluster	Dep. Cluster VI	16604	85.3%
	Dep. Cluster VII	2854	14.7%
Total		19458	100.0%

Auto-Clustering

Number of Clusters	Akaike's Information Criterion (AIC)	AIC Change(a)	Ratio of AIC Changes(b)	Ratio of Distance Measures(c)
1	10911057.785			
2	10759640.334	-151417.451	1.000	2.127
3	10753459.490	-6180.843	.041	1.257
4	10497516.191	-255943.300	1.690	1.127
5	10488058.314	-9457.877	.062	1.123
6	10484819.361	-3238.953	.021	2.038
7	10484642.271	-177.090	.001	1.002
8	10420376.984	-64265.286	.424	1.121
9	10419275.670	-1101.315	.007	1.230
10	10419365.664	89.994	-.001	1.050
11	10418350.481	-1015.184	.007	1.109
12	10418488.467	137.986	-.001	1.146
13	10418437.396	-51.071	.000	1.062
14	10415380.509	-3056.887	.020	1.064
15	10415374.429	-6.079	.000	1.055
16	10415300.509	-73.920	.000	1.026
17	10415448.698	148.189	-.001	1.085
18	10398599.487	-16849.211	.111	1.043
19	10398727.473	127.986	-.001	1.046
20	10398869.402	141.929	-.001	1.002
21	10397745.656	-1123.746	.007	1.089
22	10397765.086	19.430	.000	1.014
23	10389611.669	-8153.417	.054	1.023
24	10389645.863	34.194	.000	1.027
25	10389779.351	133.488	-.001	1.004
26	10389886.268	106.917	-.001	1.038
27	10389885.982	-.286	.000	1.018
28	10389975.807	89.826	-.001	1.086
29	10390125.519	149.712	-.001	1.014
30	10389634.640	-490.879	.003	1.039
31	10388964.020	-670.620	.004	1.015
32	10388971.217	7.197	.000	1.027
33	10389037.965	66.748	.000	1.035
34	10388950.373	-87.593	.001	1.040
35	10382975.625	-5974.747	.039	1.027
36	10383119.261	143.635	-.001	1.029
37	10383103.250	-16.010	.000	1.236
38	10383228.917	125.667	-.001	.854
39	10383355.777	126.860	-.001	1.081
40	10383524.741	168.964	-.001	1.029

a The changes are from the previous number of clusters in the table.

b The ratios of changes are relative to the change for the two cluster solution.

c The ratios of distance measures are based on the current number of clusters against the previous number of clusters.

Table S4. Soil and atmospheric nitrogen.

Table S4. Correlations between soil C:N ratios as measured in 2009 and the cumulative Nr deposition in 130, 125, 120, ... 10, and 5 years. Pearson Correlation Coefficients, $n = 19,458$
 $\text{Prob} > |r| \text{ under } H_0: \rho = 0, \text{ all variables significantly correlated with each other}$

Nr Deposition	soilC:Nratio	since1880	since1885	since1890	since1895	since1900	since1905	since1910	since1915	since1920	since1925	since1930	since1935	since1940	since1945	since1950	since1955	since1960	since1965	since1970	since1975	since1980	since1985	since1990	since1995	since2000				
since1880	-0.2925	<0.0001																												
since1885	-0.29232	0.99998	<0.0001																											
since1890	-0.29217	0.99993	0.99998	<0.0001																										
since1895	-0.29208	0.99984	0.99993	0.99998	<0.0001																									
since1900	-0.29205	0.99971	0.99984	0.99993	0.99998	<0.0001																								
since1905	-0.29209	0.99955	0.99972	0.99984	0.99993	0.99998	<0.0001																							
since1910	-0.29222	0.99936	0.99956	0.99971	0.99984	0.99993	0.99998	<0.0001																						
since1915	-0.29243	0.99913	0.99936	0.99955	0.99971	0.99983	0.99992	0.99998	<0.0001																					
since1920	-0.29273	0.99885	0.99911	0.99934	0.99953	0.99969	0.99982	0.99992	0.99998	<0.0001																				
since1925	-0.29365	0.99817	0.99849	0.99878	0.99904	0.99927	0.99948	0.99965	0.9998	0.99991	0.99998	<0.0001																		
since1930	-0.29433	0.99774	0.9981	0.99842	0.99872	0.99899	0.99923	0.99944	0.99963	0.99978	0.9999	0.99998	<0.0001																	
since1935	-0.29522	0.99725	0.99764	0.998	0.99833	0.99863	0.99891	0.99916	0.9994	0.9996	0.99976	0.99989	0.99997	<0.0001																
since1940	-0.29748	0.99604	0.99649	0.99691	0.99731	0.99768	0.99804	0.99837	0.99869	0.99898	0.99925	0.99948	0.99968	0.99985	0.99996	<0.0001														
since1945	-0.29875	0.99524	0.99572	0.99618	0.99661	0.99703	0.99742	0.9978	0.99816	0.99851	0.99882	0.99911	0.99938	0.99962	0.99981	0.99995	<0.0001													
since1950	-0.3	0.99412	0.99464	0.99514	0.99562	0.99607	0.99652	0.99695	0.99736	0.99776	0.99814	0.9985	0.99885	0.99918	0.99948	0.99973	0.99991	<0.0001												
since1955	-0.30136	0.99259	0.99316	0.9937	0.99422	0.99473	0.99522	0.99571	0.99618	0.99664	0.99709	0.99753	0.99797	0.99841	0.99884	0.99922	0.99957	0.99986	<0.0001											
since1960	-0.30311	0.99056	0.99117	0.99175	0.99231	0.99286	0.9934	0.99383	0.99446	0.99499	0.9955	0.99602	0.99656	0.99712	0.99769	0.99823	0.99876	0.99931	0.99978	<0.0001										
since1965	-0.30477	0.98728	0.98793	0.98855	0.98915	0.98974	0.99033	0.99092	0.99151	0.99211	0.9927	0.99332	0.99397	0.99468	0.99543	0.99616	0.99693	0.99781	0.99873	0.99966	<0.0001									
since1970	-0.30532	0.98124	0.98194	0.9826	0.98324	0.98388	0.98452	0.98518	0.98585	0.98652	0.98722	0.98795	0.98876	0.98966	0.99064	0.99162	0.9927	0.99404	0.99559	0.99729	0.99902	<0.0001								
since1975	-0.30875	0.96826	0.96897	0.97066	0.97134	0.97203	0.97274	0.97348	0.97426	0.97507	0.97592	0.97684	0.97786	0.97905	0.98035	0.98168	0.98319	0.98515	0.98757	0.99046	0.99399	0.99779	<0.0001							
since1980	-0.31707	0.95608	0.95677	0.95744	0.95813	0.95884	0.9596	0.96043	0.96131	0.96226	0.96328	0.96437	0.96559	0.96699	0.96851	0.97005	0.97181	0.97417	0.97714	0.98078	0.98552	0.99132	0.99742	<0.0001						
since1985	-0.32107	0.94632	0.94685	0.94757	0.94821	0.94889	0.94963	0.95045	0.95136	0.95235	0.95341	0.95455	0.9558	0.95723	0.9588	0.96035	0.96213	0.96458	0.96772	0.97163	0.97691	0.98373	0.99203	0.99829	<0.0001					
since1990	-0.32327	0.91775	0.9184	0.91906	0.91974	0.92048	0.92129	0.9222	0.92322	0.92434	0.92557	0.92692	0.92843	0.93022	0.93221	0.93422	0.93657	0.93983	0.94411	0.94958	0.95713	0.9674	0.98105	0.99154	0.99574	<0.0001				
since2000	-0.33445	0.90695	0.90765	0.90834	0.90907	0.90987	0.91075	0.91174	0.91286	0.91409	0.91545	0.91693	0.91861	0.92059	0.92278	0.92501	0.92761	0.9312	0.93586	0.94172	0.94973	0.96047	0.97498	0.9866	0.99135	0.99804	<0.0001			
since2005	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	<0.0001	