Bilthoven, June 19, 2015

Thank you, Dr. Neftel, for offering to reconsider it after major revision. We have addressed all referees’ comments, including the comments of the fifth peer, in a new version, and their comments helped to improve the paper. We have endeavored to address all the suggestions of the referees as well as your suggestions, as already anticipated in BGD.

New material in our revision includes:

1) a clear statistical focus as the EC recommended – we defined better our main focus (assessment of the actual relationship between N deposition and soil C:N), as the topic is complex and several implications are beyond the scope of the current manuscript;
2) explanation of NO_y deposition and NH_x deposition in Material&Methods, with definition of NO_y deposition as the sum of dry and wet deposition of NO_x ($NO + NO_2$) and reaction products (HNO_3 and NO_3 aerosol), as recommended by the fifth referee;
3) as this manuscript has never aimed to be a paper on N deposition, we also prefer to refer for more details to Simpson et al. (2006 –as new ref.–, 2012) and Schöpp et al. (2003);
4) in this new version, we attempt to reinforce the statistical evidence explaining shortly the kind of interpolations used in space in our improved supplementary material, linking the modeling background more clearly with the other main points of the paper;
5) a transparent discussion of Material&Methods 2.1 and 2.3, further justifying the choices of mathematical models made in our study;
6) an explanation of the historical and agicultural backgrounds beyond the terms managed (i.e., fertilized) croplands and unmanaged (i.e., unfertilized) semi-natural sites in Material&Methods 2.2, without entering in details on the plethora of vegetation units in Europe;
7) much improved organization of the manuscript, as recommended by referee #4;
8) additional references, as suggested by referees #3 and #4 (Thank you for making us aware of these studies, we have cited most of them), and removal of possibly confusing references.

We have also reorganized the manuscript for greater clarity. We would be glad for the paper to be considered for publication in *BIOGEOSCIENCES*. Thanks again for offering us the opportunity to improve our work.

Yours sincerely,

Christian Mulder

THIS REPLY INCLUDES:

THE COMPLETE MANUSCRIPT REVISION IN TRACK CHANGES AND A NEW SUPPLEMENT

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

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Abstract

Long-term human interactions with landscape and nature produced a plethora of trends and patterns of environmental disturbances in time and space. Nitrogen deposition, closely tracking energy and land use, is known to be among the main pollution drivers, affecting both freshwater and~~s~~ terrestrial ecosystems. We present a statistical approach to~~We~~ investigate~~d~~ the historical and geographical distribution of nitrogen deposition and the impacts of accumulation on recent soil carbon to nitrogen ratios over Europe. After the Second Industrial Revolution (1880–2010), large landscape stretches characterized by different atmospheric deposition caused either by industrialized areas or by intensive agriculture emerged. Nitrogen deposition affects in a still recognizable way recent soil C:N ratios despite the emission abatement of oxidized and reduced nitrogen during the last two decades. Given the seemingly disparate land-use history, we focused on ~10,000 unmanaged ecosystems, providing statistical evidence for a rapid response of nature to the chronic nitrogen supply by atmospheric deposition.

1 Introduction

The global cycle of nitrogen ~~(N)~~ is ~~unique and~~ highly sensitive to human activities (Galloway et al., 2004; Costanza et al., 2007; Doney et al., 2007; Fowler et al., 2013). Shifts in nitrogen availability alter carbon cycle and litter decomposition (Vitousek et al., 1997; Stevens et al., 2004; Reich, 2009), affecting the heterotrophic component of ecosystem respiration (Janssens et al., 2010). In terrestrial ecosystems, the atmospheric nitrogen deposition is also a major source of concern because it induces soil acidification by decreasing the exchangeable cations pools (Bowman et al., 2008). Moreover, the nutrient enrichment directly influences the biodiversity and ecological stoichiometry of vascular plants through the soil (Stevens et al., 2004; Mulder et al., 2013). Public and political concerns for current agricultural and environmental policies have emphasized ~~increased species~~ loss of biodiversity ~~related to nitrogen deposition,~~ and ~~the~~ impacts on ~~biodiversity and~~ ecosystem services related to nitrogen deposition~~have caught the attention of many ecologists~~ (Reis et al., 2012~~,~~; Sutton et al., 2014). It is widely accepted that correct relative proportions of physiologically-required nutrients will promote the growth of plant species, influence their diversity, and finally drive the vegetation succession (Sterner and Elser, 2002; Hillebrand et al., 2014). Among such chemical elements, carbon (C) and nitrogen (N) are the most important, which makes the determination of relationships between soil C:N and nitrogen deposition interesting.

~~Given the rapid expansion in Europe of industrial technology and intensive agriculture during the late XIX Century (Mokyr, 1990), we chose 1880 as the starting point under the hypothesis that accumulated nitrogen deposition since 1880 might have contributed most to the spatial variability of recent soil C:N ratios.~~ To investigate such correlations, w~~W~~e used 19,458 sites in 23 European countries~~ from a European Soil Survey (Tóth et al., 2013)~~ to quantify the effect of atmospheric deposition of nitrogen compounds on soil C:N measurements~~ in 2009~~. We~~ and~~ separately investigated the effects of nitrogen oxides (NO_x, sum of NO and NO_2), atmospheric ammonia (NH_3), and reactive nitrogen (Nr, defined as the sum of NO_x and NH_3). NO_x is mostly emitted from fossil fuel combustion in industry and transport, whereas NH_3 reflects the use of fertilizers, agriculture being the causal agent of such emissions (Dignon and Hameed, 1989; Williams et al., 1992; Vitousek et al., 1997; Doney et al., 2007; Woodward et al., 2012; Liu et al., 2013). More than half of the investigated sites are located in either France (2950 sites), Spain (2693 sites), Sweden (2254 sites) or Germany (1888 sites).

Given the rapid expansion in Europe of industrial technology and intensive agriculture during the late XIX Century (Mokyr, 1990), we chose 1880 as the starting point of our time series under the hypothesis that accumulated nitrogen deposition since 1880 might have contributed most to the spatial variability of recent soil C:N ratios. ~~Between 1880 and 2010, estimated nitrogen emissions in each country for every 5 years until 1990 and each year afterwards were translated to depositions with the aid of an atmospheric dispersion model (Section 2.1).~~ The statistical relation between long-term nitrogen deposition and recent soil C:N ratio was tested by exploring whether spatial clusters of accumulated nitrogen deposition exist and if chemical footprints on soil C:N occur. This large-scale statistical comparison was made possible by using consistent data from one single survey in which all soils were sampled according to the same protocol and analysed in the same laboratory~~ (Section 2.2)~~.

2 Methods

2.1 Nitrogen deposition

Between 1880 and 2010, estimated nitrogen emissions in each country for every 5 years until 1990 and each year afterwards were used to compute depositions with the aid of atmospheric dispersion model(s). Annual-average deposition time series of total (= wet + dry) oxidized and reduced nitrogen were ~~computed~~ obtained from simulations with the ~~former EMEP Lagrangian~~ Eulerian atmospheric dispersion model ~~on a 150 km × 150 km grid covering Europe (Schöpp et al., 2003) and using the 12-years average climatology of 1985 to 1996. For the years after 1990, the depositions were updated with results from the current EMEP Eulerian dispersion model~~ (Simpson et al., 2012; for a comparison with measurements see Simpson et al., 2006)~~, which computes outputs~~, operated and maintained by the European Monitoring and Evaluation Programme (EMEP) at the Norwegian Meteorological Institute and routinely used in European air pollution assessments (www.emep.int/mscw).

Total oxidised N deposition is the sum of NO_2, HNO_3, nitrous acid (HONO), particulate NO_3, peroxyacetyl nitrate (PAN) and peroxymethacryloyl nitrate (MPAN), whereas total reduced N deposition comprises NH_3 and NH_4 aerosols. The model output is provided on a ~~compatible~~

grid covering Europe with a resolution of 50 km × 50 km ~~grid (http://emep.int/mscw). These newer~~in a polar stereographic projection (see Fig. S1 in the Supplement). Deposition fields are provided for the years 1990 and later. For the years up to 1996, the results from the former (Lagrangian) version of the EMEP model were used (Eliassen and Saltbones, 1983). This former model version produced results ~~were also~~on a 150 km × 150 km grid (see thick lines in Fig. S1 in the Supplement). Results from the overlapping years (1990–1996) were used to adjust the older (Lagrangian) simulations to ensure a smooth transition in ~~our~~the deposition time series (see Schöpp et al., 2003 for details). Depositions at the C:N measurement sites were ~~obtained by bi-linear interpolation~~bilinearly interpolated from the four nearest grid values (Fig. S2 in the Supplement).

2.2 Soil data

We collected data from a recent European Soil Survey known as LUCAS (Land Use/Cover Area frame Survey): ~20,000 geo-referenced points were chosen for this ~~harmonized~~ field sampling with ~~one~~ the same standardized procedure~~, resulting in geo-referenced points classified according to land-cover types.~~, covering several ecosystem types, from unfertilized ‘grasslands’ (steppes, wet or saline grasslands, (sub)alpine forb grasslands, arctic meadows and abandoned pastures), ‘shrublands’ (tundra and heathlands) and ‘woodlands’ (broadleaved, evergreen, coniferous and mixed forests) up to fertilized ‘croplands’ (cereal fields, winter farms, orchards, vineyards, etc.), Soil samples were collected in 2009 from 23 European countries and all samples, weighing ~11 tons, were sent to one central ISO-certified laboratory at the JRC (Ispra, Italy) and stored in the European Soil Archive Facility ~~where the soil C:N was measured~~in order to obtain a coherent pan-European dataset with harmonized analytical methods (Tóth et al., 2013).

Total soil carbon (g C kg^{-1}) and total soil nitrogen (g N kg^{-1}) were determined simultaneously by dry combustion with a quantification limit of 50 mg kg^{-1} (Richard and Proix, 2009). Then, every soil C:N ratio was computed in mass units (g C / g N) for the upper part of each of these soil profiles (i.e., 0–30 cm). We have selected 19,458 locations with complete categorical site description: 8,010 (intensively) fertilized locations were assigned *in situ* to fodder crops, annual crops and permanent crops (here as ‘croplands’~~ (cereal fields, winter farms with annual or permanent crops, orchards, vineyards, etc.~~), ~~twelve~~ 14 locations could not be assigned to ~~o~~an~~e~~y specific land use~~/cover~~ (incomplete documentation for 12 sites) or were outliers (soil C:N > 200 for two sites) and were excluded from further analysis, and the remaining 11,434 unfertiliz~~manag~~ed locations ~~(including two organic soil outliers with C:N > 200)~~ were assigned *in situ* to woodlands, shrublands or grasslands (~~lumped toget~~here as 'nature').

2.3 Cluster analysis

To explore the similarities of the time series from 1880 up to 2010, we used the TwoStep Clustering method ~~(SPSS, 2001). This method implies a pre-clustering of cases with a sequential approach and then a model-based hierarchical technique similar to agglomerative techniques, where the~~implemented in SPSS that is suitable for very large datasets. The first step of the two-step algorithm is a BIRCH algorithm to define pre-clusters (Zhang et al., 1996, 1997); in the second step, using an agglomerative hierarchical algorithm, these pre-clusters are merged stepwise until all locations hierarchically close to each other fall within the same cluster (SPSS, 2001). The numbers of clusters are determined with a two phase estimator like the Akaike's Information Criterion (AIC) and a (ratio of) distance measure in both pre-cluster and cluster steps. AIC is a relative measure of goodness of fit and is used to compare different hierarchical solutions with different numbers of clusters: any "correct" good hierarchical solution will have a reasonably large ratio of AIC changes ~~and~~with the distance ratio measur~~es~~ing the most reliable current number of clusters against ~~the previous number of clusters.~~ alternative solutions.

The TwoStep Clustering method became rapidly accepted when Chiu et al. (2001) ~~noted~~demonstrated that ~~this approach~~such technique was able to ~~find~~identify objectively the correct number of clusters for ~~∼~~more than 98 % of ~~the generated~~a large number of simulated data sets. This clustering method for very large databases has been used in many different fields, from biochemistry, genetics and molecular biology (e.g., Lazary et al., 2014) to medicine (e.g., Kretzschmar and Mikolajczyk, 2009). Here we identified seven clusters running TwoStep Clustering separately for the three N categories: nitrogen oxides, atmospheric ammonia and reactive nitrogen~~, the correct numbers of deposition clusters were determined~~ (~~please~~ refer to the Tables S1~~ to ~~–S3 in the Supplement).

3 Results and ~~D~~discussion

Our statistical c~~C~~lustering enables an objective detection~~g~~ of sites with similar historical paths of nitrogen deposition, showing how much sites respond to nitrogen supply through atmospheric deposition over time. Figure 1 shows the distribution across Europe of hotspots and spatial aggregations in all forms of nitrogen deposition. The ammonia clusters are distinct (the high load is more than two-fold the low load) and Deposition Cluster I visualizes an emerging cocktail of manure and synthetic fertilizers due to intensive agriculture (Fig. 1, upper left). In contrast, long-term deposition of NO_x reflects demographic pressure and industrial boundaries and needs three clusters to be fully characterized (Fig. 1, bottom left). Also Nr shows a clear distinction between its two clusters, where the high annual load (averaging ~15.2 kg N ha^{-1}, Deposition Cluster VII), covers the former Austro-Hungarian Empire, Western Germany, Brittany and the Po Valley (Fig. 1, upper right).

Atmospheric N has multiple fates and sources have changed substantially (Holtgrieve et al., 2011; Steffen et al., 2015). Within one century, the average of Nr increased everywhere more than two-fold between 1880 and 1980. In 2010 the Nr deposition was still much higher than in 1880, and only 16 sites (0.082 %) exhibited in 2010 a lower Nr deposition than in 1880, with the highest increase in southern Europe (up to 8 times the Nr deposition of 1880). Shortly after World War II, NH_3 and NO_x started to rise rapidly in Europe (Fig. –2), as agricultural production surpassed pre-war levels and industrial production recovered (van Aardenne et al., 2001). The 1980s were tipping points for nitrogen deposition and since the 1999 Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone, the deposition of oxidized and reduced nitrogen started to decrease simultaneously (Fig. -2), with the most pronounced reductions in Eastern Europe (Rafaj et al., 2014).

Clustering highly increased the discrimination power to establish historical shifts in recent soil C:N ratios (Table 1). We used these nitrogen deposition clusters to assess the spatial distribution of recent soil C:N assuming the existence a long-term footprint in soil C:N ratios due to atmospheric deposition, although some authors claim that ~~– at least in forests –~~ significant correlations between the nitrogen of mineral soils and the anthropogenic~~effects due to~~ nitrogen deposition are either weak or far from causal (Nadelhoffer et al., 1999; Aber et al., 2003~~Cools et al., 2014~~). Our soil C:N ratio ~~in mass units~~ averages 16.18 (± 8.38 SD) and the coefficient of variation is 51.8 % (Fig. 1, bottom right).

To investigate the extent to which atmospheric nitrogen deposition affects terrestrial ecosystems, we compared geospatial patterns of recent soil C:N ratios with temporal trends in nitrogen deposition, keeping in mind that time is one-dimensional and directional, whereas space is two-dimensional and non-directional (White, 2007). Overall, a generalized linear model (here as GLM with normal distribution, identity link) for soil C:N as function of historical depositions showed a temporal increase in Wald's χ^2 from 1814.9 (in 1905) to 2450.7 (in 2005), suggesting the short-term supply of nitrogen through atmospheric deposition as primarily responsible for soil C:N ($p < 0.0001$).

We analysed the clusters separately with high versus low nitrogen loads as classification variables, and detected a comparable χ^2 increase in time. We also analysed the unmanaged and managed ecosystems separately and detected negative associations between the soil C:N ratio and the nitrogen deposition clusters (the Mantel's asymptotic method exhibits $t = -12.23$ for the 11,434 (semi-)natural non-agricultural ecosystems but only a slight $t = -0.59$ for the 8,010 agroecosystems). Given the computational independence of our matrices, this Mantel analysis assessed that the associations between the nitrogen deposition during 130 years and the recent soil C:N ratios were much stronger in ~~natural~~ less-disturbed ecosystems than could result from chance.

Focusing on unmanaged ecosystems, the same type of GLM was performed for the recent soil C:N as function of accumulated Nr, assuming that all locations sampled in 2009 and classified as 'nature' were surely unmanaged 5 years before sampling and most probably even unmanaged 50 years before sampling. For the soil C:N ratios of the unmanaged ecosystems under chronic pollution there was a significant increase of explanatory power by reduced time-spans of accumulated Nr deposition ($p = 0.00004$). In these ~~natural~~ ecosystems – all located within Deposition Cluster VI (Fig. 3, upper panel) – almost half of the variation of the soil C:N ratio is likely to be explained by chronic nitrogen pollution at the site ($R^2 = 46.3$ %).

Such a conclusion is indirectly supported by the lack of any significant trend in the other (semi-)natural ecosystems, all located within Deposition Cluster VII (Fig. 3, lower panel), given that their area is associated with intense human activity, high emissions and soil saturation due to elevated nitrogen loads. Soils C:N of (semi-)natural sites seem to be the most sensitive to five-year pulses of atmospheric nitrogen supply, short-term deposition clearly being the best predictor for recent soil C:N ratios under chronic nitrogen deposition (R^2 = 89.2 %, F = 66.09). ~~Although sudden increases in nitrogen availability enhance carbon cycling rates and carbon nitrogen feedbacks are mostly related to harvest (Gerber et al., 2010), forests play a major role in the uptake and storage of carbon (Gerber et al., 2010; Fleischer et al., 2013) and act, like grasslands, as a sink for anthropogenic CO_2 (Pregitzer et al., 2008; Johnson et al., 2013). Hence, atmospheric nitrogen deposition affected nature in Europe both directly and through secondary effects, contributing via N-saturated soils to enhanced leaching of nitrogen to rivers and finally to the sea (Galloway et al., 2004; Doney et al., 2007; Woodward et al., 2012).~~

~~4 Conclusions~~

Summarizing, s~~S~~patial clustering reveals long-term effects of atmospheric nitrogen deposition on the recent soil C:N ratios in Europe. While an inverse correlation between this anthropogenic input and soil C:N seems to be intuitive, the extent to which this relationship holds has never been investigated before. Our results show that the C:N ratio varies more across the soils of (semi-)natural ecosystems with a history of low (chronic) nitrogen pollution and that it remains surprisingly constant elsewhere. Moreover, despite the investigated deposition of nitrogen since the 1880s, it turns out that soils supposed to be under low pressure are not only the most affected by nitrogen accumulation, but also the most responsive to a short-term supply of atmospheric nitrogen in their recent past.

Statistical signals from responsive chronic nitrogen pollution became detectable only after clustering the nitrogen deposition, and we were able to provide novel evidence that the soil C:N of (semi-)natural ecosystems is highly-responsive to Nr. We detected where ~~and why~~ nitrogen supply through atmospheric Nr deposition affects (semi-)natural ecosystems. It will be challenging to determine a mechanistical explanation of why atmospheric nitrogen supply does not seem to affect managed ecosystems as well: for instance, are many exploited soils N-saturated? How much anthropogenic nitrogen becomes mediated through soil processes has to be addressed in the future, given the long history of land (ab)use in Europe that hampered until now the detection of robust effects directly attributable to the nitrogen deposition.

We are better equipped than ever before and big data can visualize such global changes, making forecasting of large-scale data-driven evidence for chemical footprints possible (e.g., Liu et al., 2013; Steffen et al., 2015). Among others, this paper demonstrates that clustering big data on broad spatial and temporal scales allows successful exploration of the long-term relevance of atmospheric nitrogen deposition on measured soil C:N ratios at continental level. As the soil black box is now in the "front line" (Schmidt et al., 2011; Amundson et al., 2015), mapping soil and air compartments together can provide more valuable inputs and contribute to a much better management and conservation of our environment.

The Supplement related to this article is available online at doi:10.5194/bg-12~~1~~-**-2015-supplement.**

Author contributions

C. Mulder and J.-P. Hettelingh conceived the study. L. Montanarella and M. Posch collected soil C:N coverage and atmospheric deposition data. M. R. Pasimeni and G. Zurlini contributed nitrogen deposition clusters. C. Mulder, W. Voigt and G. Zurlini analysed the data. All authors commented on the composition of the manuscript.

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References

Aber, J. D., Goodale, C. L., Ollinger, S. V., Smith, M.-L., Magill, A. H., Martin, M. E., Hallett, R. A., and Stoddard, J. L.: Is nitrogen deposition altering the nitrogen status of northeastern forests?, BioScience 53, 375–389, 2003.

Amundson, R., Berhe, A. A., Hopmans, J. W., Olson, C., Sztein, A. E., and Sparks, D. L.: Soil and human security in the 21st century, Science 348, 1261071, 2015.

Bowman, W. D., Cleveland, C. C., Halada, Ĺ., Hresko, J., and Baron, J. S: Negative impact of nitrogen deposition on soil buffering capacity, Nature Geosci. 1, 767–770, 2008.

Chiu, T., Fang, D., Chen, J., Wang, Y., and Jeris, C.: A robust and scalable clustering algorithm for Mixed Type Attributes in large database environment, Proc. 7th ACM SIGKDD International Conference on Knowledge Discovery and Data Mining, pp. 263–268, 2001.

~~Cools, N., Vesterdal, L., De Vos, B., Vanguelova, E., and Hansen, K.: Tree species is the major factor explaining C:N ratios in European forest soils, Forest Ecol. Manage. 311, 3–16, 2014.~~

Costanza, R., Graumlich, L., Steffen, W., Crumley, C., Dearing, J., Hibbard, K., Leemans, R., Redman, C., and Schimel, D.: Sustainability or collapse: What can we learn from integrating the history of humans and the rest of nature?, Ambio 36, 522–527, 2007.

Dignon, J. and Hameed, S.: Global emissions of nitrogen and sulfur oxides from 1860 to 1980, J. Air Pollut. Control Assoc. 39, 180–186, 1989.

Doney, S. C., Mahowald, N., Lima, I., Feely, R. A., Mackenzie, F. T., Lamarque, J.-F., and Rasch, P. J.: Impact of anthropogenic atmospheric nitrogen and sulfur deposition on ocean acidification and the inorganic carbon system, Proc. Natl. Acad. Sci. USA 104, 14580–14585, 2007.

Eliassen, A., and Saltbones, J.: Modelling of long-range transport of sulphur over Europe: A two-year model run and some model experiments, Atmos. Environ. 17, 1457–1473, 1983.

~~Fleischer, K., Rebel, K. T., van der Molen, M. K., Erisman, J. W., Wassen, M. J., van Loon, E. E., Montagnani, L., Gough, C. M., Herbst, M., Janssens, I. A., Gianelle, D., and Dolman, A. J.: The contribution of nitrogen deposition to the photosynthetic capacity of forests, Global Biogeochem. Cycles 27, 187–199, 2013.~~

Fowler, D., Coyle, M., Skiba, U., Sutton, M. A., Cape, J. N., Reis, S., Sheppard, L. J., Jenkins, A., Grizzetti, B., Galloway, J. N., Vitousek, P., Leach, A., Bouwman, A. F., Butterbach-Bahl, K., Dentener, F., Stevenson, D., Amann, M., and Voss, M.: The global nitrogen cycle in the twenty-first century, Phil. Trans. R. Soc. London 368B, 20130164, 2013.

Galloway, J. N., Dentener, F. J., Capone, D. G., Boyer, E. W., Howarth, R. W., Seitzinger, S. P., Asner, G. P., Cleveland, C. C., Green, P. A., Holland E. A., Karl, D. M., Michaels, A. F., Porter, J. H., Townsend, A. R., and Vöosmarty, C. J.: Nitrogen cycles: Past, present, and future, Biogeochemistry 70, 153–226, 2004.

~~Gerber, S., Hedin, L. O., Oppenheimer, M., Pacala, S. W., and Shevliakova, E.: Nitrogen cycling and feedbacks in a global dynamic land model, Global Biogeochem. Cycles 24, GB1001, 2010.~~

Hillebrand, H., Cowles, J. M., Lewandowska, A., Van de Waal, D. B., and Plum, C.: Think ratio! A stoichiometric view on biodiversity–ecosystem functioning research, Basic Appl. Ecol. 15, 465-474, 2014.

Holtgrieve, G. W., Schindler, D. E., Hobbs, W. O., Leavitt, P. R., Ward, E. J., Bunting, L., Chen, G., Finney, B. P., Gregory-Eaves, I., Holmgren, S., Lisac, M. J., Lisi, P. J., Nydick, K., Rogers, L. A., Saros, J. E., Selbie, D. T., Shapley, M. D., Walsh, P. B., and Wolfe, A. P.: A coherent signature of anthropogenic nitrogen deposition to remote watersheds of the northern hemisphere, Science 334, 1545–1548, 2011.

Janssens, I. A., Dieleman, W., Luyssaert, S., Subke, J.-A., Reichstein, M., Ceulemans, R., Ciais, P., Dolman, A. J., Grace, J., Matteucci, G., Papale, D., Piao, S. L., Schulze, E.-D., Tang, J., and Law, B. E.: Reduction of forest soil respiration in response to nitrogen deposition, Nature Geosci. 3, 315–322, 2010.

~~Johnson, N. C., Angelard, C., Sanders, I. R., and Kiers, E. T.: Predicting community and ecosystem outcomes of mycorrhizal responses to global change, Ecol. Lett. 16, 140-153, 2013.~~

Lazary, J., Dome, P., Csala, I., Kovacs, G., Faludi, G., Kaunisto, M., and Dome, B.: Massive withdrawal symptoms and affective vulnerability are associated with variants of the *CHRNA4* gene in a subgroup of smokers, PLoS ONE 9, e87141, 2014.

Liu, X., Zhang, Y., Han, W., Tang, A., Shen, J., Cui, Z., Vitousek, P., Erisman, J. W., Goulding, K., Christie, P., Fangmeier, A., and Zhang, F.: Enhanced nitrogen deposition over China, Nature 494, 459–462, 2013.

Kretzschmar, M., and Mikolajczyk, R. T.: Contact profiles in eight European countries and implications for modelling the spread of airborne infectious diseases, PLoS ONE 4, e5931, 2009.

Mokyr, J.: The Lever of Riches: Technological creativity and economic progress, Oxford University Press, New York, 1990.

Mulder, C., Ahrestani, F. S., Bahn, M., Bohan, D. A., Bonkowski, M., Griffiths, B. S., Guicharnaud, R. A., Kattge, J., Krogh, P. H., Lavorel, S., Lewis, O. T., Mancinelli, G., Naeem, S., Peñuelas, J., Poorter, H., Reich, P. B., Rossi, L., Rusch, G. M., Sardans, J., and Wright, I. J.: Connecting the green and brown worlds: Allometric and stoichiometric predictability of above- and belowground networks, Adv. Ecol. Res. 49, 69–175, 2013.

Nadelhoffer, K. J., Emmett, B. A., Gundersen, P., Kjønaas, O. J., Koopmans, C. J., Schleppi, P., Tietema, A., and Wright, R. F.: Nitrogen deposition makes a minor contribution to carbon sequestration in temperate forests, Nature 398, 145–148, 1999.

~~Pregitzer, K. S., Burton, A. J., Zak, D. R., and Talhelm, A. F.: Simulated chronic nitrogen deposition increases carbon storage in northern temperate forests, Global Cha. Biol. 14, 142–153, 2008.~~

Rafaj, P., Amann, M., Siri, J., and Wuester, H.: Changes in European greenhouse gas and air pollutant emissions 1960-2010: Decomposition of determining factors, Clim. Cha. 124, 477–504, 2014.

Reich, P. B.: Elevated CO_2 reduces losses of plant diversity caused by nitrogen deposition, Science 326, 1399–1402, 2009.

Reis, S., Grennfelt, P., Klimont, Z., Amann, M., ApSimon, H., Hettelingh, J.-P., Holland, M., LeGall, A.-C., Maas, R., Posch, M., Spranger, T., Sutton, M. A., and Williams, M.: From acid rain to climate change, Science 338, 1153–1154, 2012.

Richard, A., and Proix, N.: Biosoil Project – Soil samples analysis technical report, INRA, Laboratoire d'analyses des sols, Arras, France, 1–132, 2009.

Schmidt, M. W. I., Torn, M. S., Abiven, S., Dittmar, T., Guggenberger, G., Janssens, I. A., Kleber, M., Kögel-Knabner, I., Lehmann, J., Manning, D. A. C., Nannipieri, P., Rasse, D. P., Weiner, S., and Trumbore, S. E.: Persistence of soil organic matter as an ecosystem property, Nature 478, 49–56, 2011.

Schöpp, W., Posch, M., Mylona, S., and Johansson, M.: Long-term development of acid deposition (1880–2030) in sensitive freshwater regions in Europe, Hydrol. Earth Syst. Sci. 7, 436–446, 2003.

Simpson, D., Fagerli, H., Hellsten, S., Knulst, J. C., and Westling, O.: Comparison of modelled and monitored deposition fluxes of sulphur and nitrogen to ICP-forest sites in Europe, Biogeosciences 3, 337–355, 2006.

Simpson, D., Benedictow, A., Berge, H., Bergström, R., Emberson, L. D., Fagerli, H., Flechard, C. R., Hayman, G. D., Gauss, M., Jonson, J. E., Jenkin, M. E., Nyíri, A., Richter, C., Semeena, V. S., Tsyro, S., Tuovinen, J.-P., Valdebenito, Á., and Wind, P.: The EMEP MSC-W chemical transport model – technical description, Atmos. Chem. Phys. 12, 7825–7865, 2012.

SPSS: The SPSS TwoStep Cluster Component. A scalable component enabling more efficient customer segmentation, White paper – Technical report, Chicago, IL, 2001.

Steffen, W., Richardson, K., Rockström, J., Cornell, S. E., Fetzer, I., Bennett, E. M., Biggs, R., Carpenter, S. R., de Vries, W., de Wit, C. A., Folke, C., Gerten, D., Heinke, J., Mace, G. M., Persson, L., M., Ramanathan, V., Belinda Reyers, B., and Sörlin, S.: Planetary boundaries: Guiding human development on a changing planet, Science 347, 1259855, 2015.

Sterner, R. W., and Elser, J. J.: Ecological Stoichiometry: The biology of elements from molecules to the biosphere, Princeton University Press, Princeton, 2002.

Stevens, C. J., Dise, N. B., Mountford, J. O., and Gowing, D. J.: Impacts of nitrogen deposition on the species richness of grasslands, Science 303, 1876–1879, 2004.

Sutton, M. A., Mason, K. E., Sheppard, L. J., Sverdrup, H., Haeuber, R., and Hicks, W. K. (eds.): Nitrogen Deposition, Critical Loads and Biodiversity, Springer, Dordrecht, 2014.

Tóth, G., Jones, A., and Montanarella, L.: The LUCAS topsoil database and derived information on the regional variability of cropland topsoil properties in the European Union, Environ. Monit. Assess. 185, 7409–7425, 2013.

van Aardenne, J. A., Dentener, F. J., Olivier, J. G. J., Klein Goldewijk, C. G. M., and Lelieveld, J.: A 1° × 1° resolution data set of historical anthropogenic trace gas emissions for the period 1890–1990, Global Biogeochem. Cycles 15, 909–928, 2001.

Vitousek, P. M., Aber, J. D., Howarth, R. W., Likens, G. E., Matson, P. A., Schindler, D. W., Schlesinger, W. H., and Tilman, D. G.: Human alterations of the global nitrogen cycle: Sources and consequences, Ecol. Appl. 7, 737–750, 1997.

White, E. P.: Spatiotemporal scaling of species richness: Patterns, processes and implications, Ecol. Rev. 4, 325–346, 2007.

Williams, E. J., Hutchinson, G. L., and Fehsenfeld F. C.: NO_x and NO_2 emissions from soil, Global Biogeochem. Cycles 6, 351–388, 1992.

Woodward, G., Gessner, M. O., Giller, P. S., Gulis, V., Hladyz, S., Lecerf, A., Malmqvist, B., McKie, B. G., Tiegs, S. D., Cariss, H., Dobson, M., Elosegi, A., Ferreira, V., Graça, M. A. S., Fleituch, T., Lacoursière, J. O., Nistorescu, M., Pozo, J., Risnoveanu, G., Schindler, M., Vadineanu, A., Vought, L. B.-M., and Chauvet, E.: Continental-scale effects of nutrient pollution on stream ecosystem functioning, Science 336, 1438–1440, 2012.

Zhang, T., Ramakrishnon, R., and Livny, M.: BIRCH: An efficient data clustering method for very large data bases, in: Proc. SIGMOD/PODS '96 Joint ACM SIGMOD International Conference on Management of Data and ACM Symposium on Principles of Database Systems, Montreal, QC, Canada, 103–114, 1996.

Zhang, T., Ramakrishnon, R., and Livny, M.: BIRCH: A new data clustering algorithm and its applications, Data Mining and Knowledge Discovery 1(2), 141–182, 1997.

TABLE

Table 1. Soil C:N values clearly differ per nitrogen deposition cluster. The soil C:N ratios are given as cluster-specific averages (± standard error); Roman numbers (I–VII) as in Fig. 1 and 2. Both the three-factor ANOVA with NH_3, NO_x, and Nr (=NO_x+NH_3) and the nested ANOVA with NH_3(Nr) and NO_x(Nr) are significant for their long-term effects on the soil C:N ratios (all share $p < 0.0001$).

	High Nitrogen Loads	**Low Nitrogen Loads**
NH_3 Deposition Clusters	**I**: 13.97 (± 0.15)	**II**: 16.43 (± 0.06)
NO_x Deposition Clusters	**IV**: 14.16 (± 0.07)	**III**: 17.89 (± 0.09)
	V: 12.67 (± 0.23)	
Nr Deposition Clusters	**VII**: 14.26 (± 0.14)	**VI**: 16.51 (± 0.07)

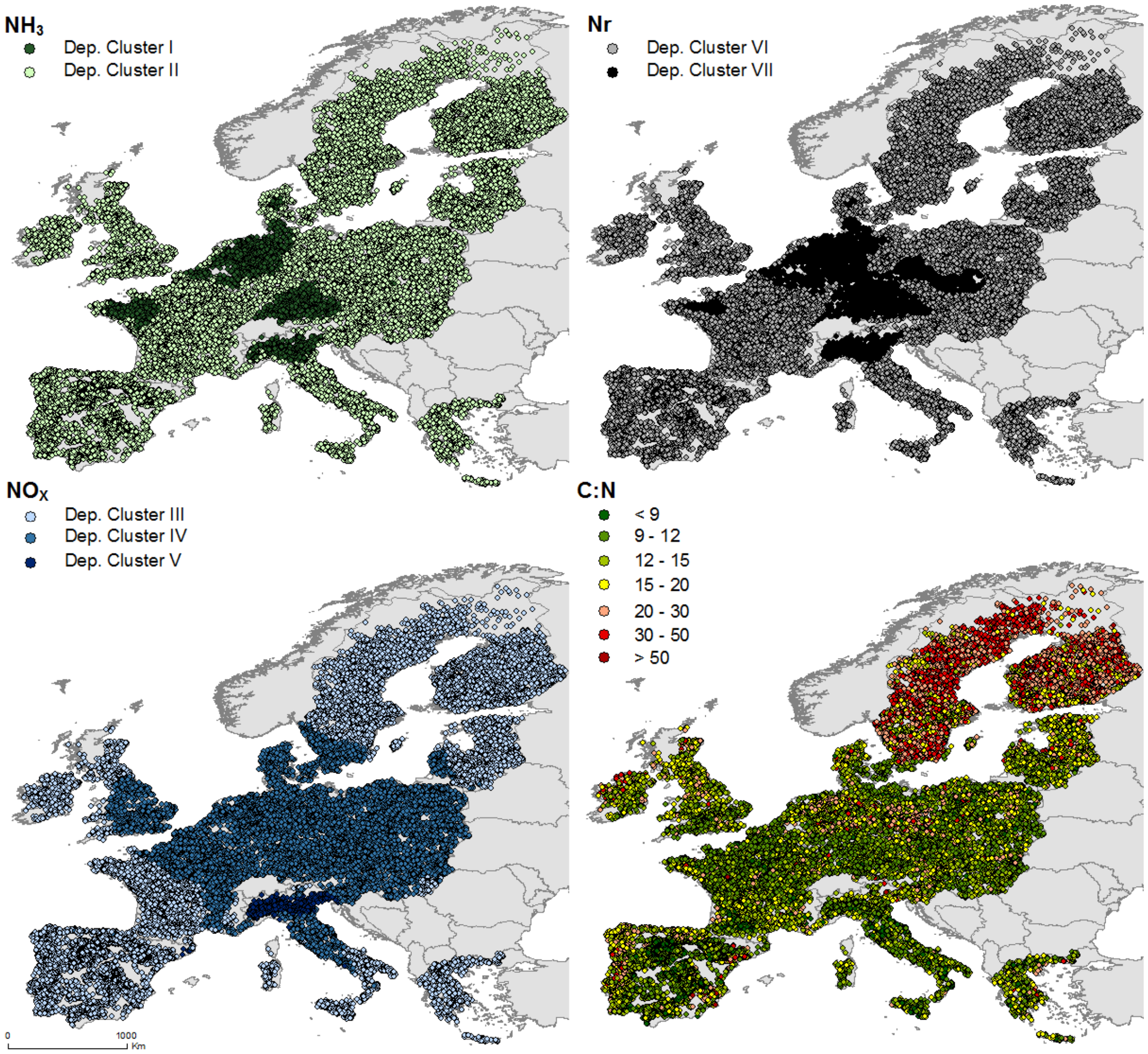

Figure 1. Nitrogen deposition and the recent soil C:N ratios (mass units). Spatial clusters (clockwards) of NO_x, NH_3, and Nr (= $NO_x + NH_3$) 1880–2010 depositions at the 19,458 sites of the soil C:N in 2009. The darker the colour of a cluster, the higher the nitrogen load for NH_3, NO_x, and Nr. Deposition Cluster IV reveals a high degree of homogeneity in the NO_x deposition, in contrast to the patchiness of Deposition Cluster I (NH_3). However, NH_3 deposition accounts the most for the aggregation of Deposition Cluster VII (Nr). ~~Correlations in natural ecosystems between recent soil C:N ratios and short-term Nr deposition in Figure 3.~~

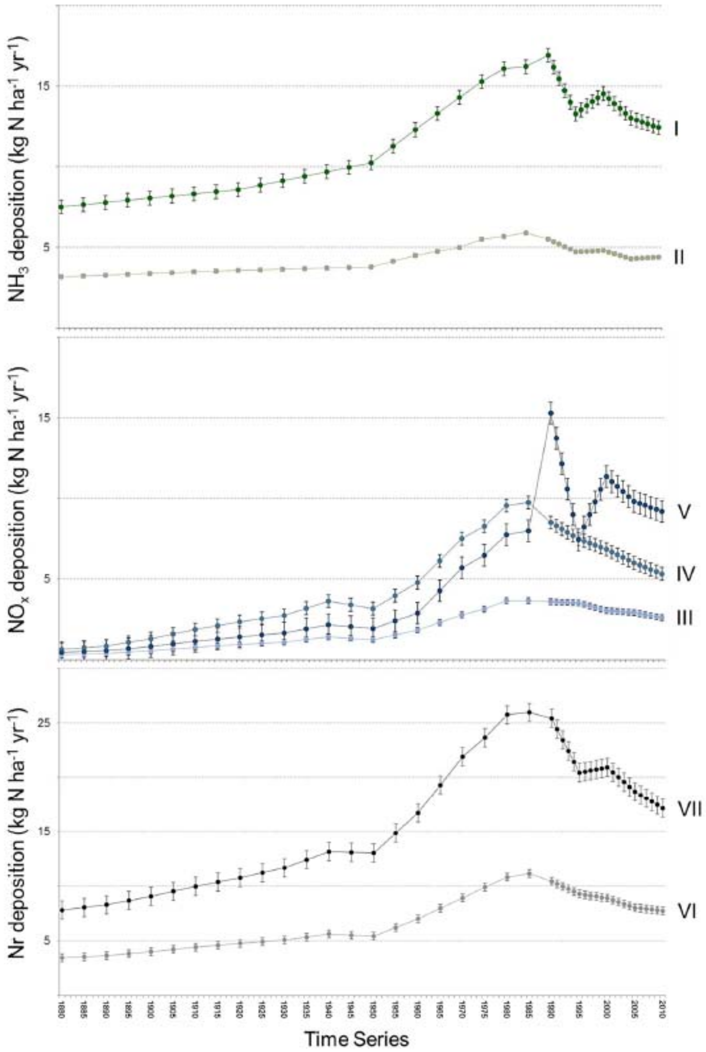

Figure 2. Temporal cluster vector means (averages and standard errors of the series) of the depositions of NH_3 (upper panel), NO_x (middle panel), and Nr (lower panel) across Europe. The colours and Roman numbers correspond to those used for the clusters in Fig. 1. The Nr deposition did not increase during the 1940s and started to rise again shortly after the introduction of the Marshall Plan in Europe. The time series for Deposition Cluster V (NO_x), encompassing 408 sites located in the Po Valley (Italy) subject to local thermic inversion, is the only trend that suddenly intercepts other trends when the resolution of the dataset increases from 5-yr calculations (1880–1990) to yearly observations (1990–2009).

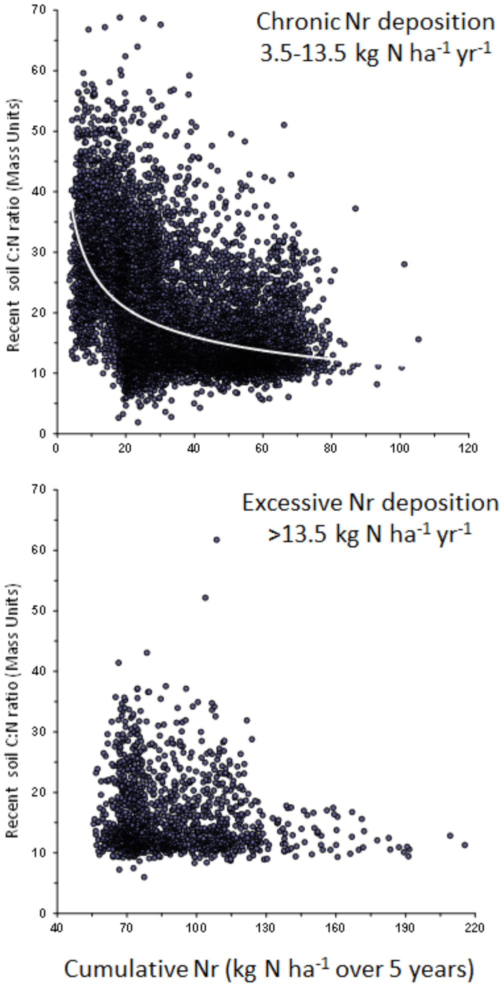

Figure 3. Cumulative Nr deposition of the last five years prior to sampling and soil C:N ratios: negative power functions of soil C:N ratios in nature (measured in 2009) as predicted by cumulative Nr deposition. Upper panel: 9,888 unmanaged sites belonging to the cluster with low Nr load but chronic exposure to nitrogen (Deposition Cluster VI); Lower panel: 1,546 unmanaged sites under excessive Nr load (Deposition Cluster VII). This last cluster acts as a kind of envelope which incorporates sites with low soil C:N ratios. We were not able to extract a significant deposition effect for managed ecosystems, although long-term inverse relationships between Nr and soil C:N hold (~~please~~ refer to the Table S4 in the Supplement).

SUPPLEMENT

The N deposition values used in this paper originate from the EMEP atmospheric dispersion model(s) that provide results on a 150 km × 150 m grid (older model version) and a compatible3 × 3 subdivision of 50 km × 50 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).

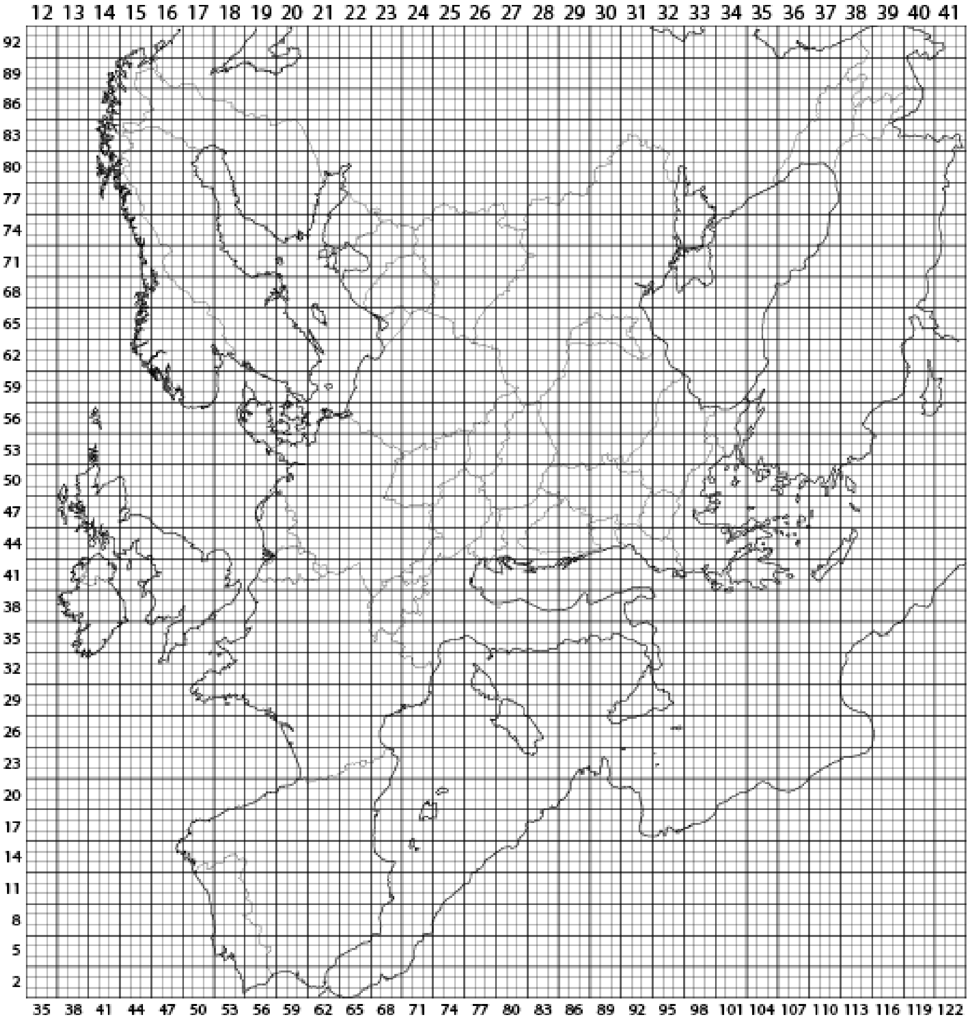

Fig. S1. The EMEP150 grid (thick lines) and the EMEP50 grid (additional thin lines). The labels at the bottom and at the left are the EMEP50 grid indices (every third cell labeled) and the labels at the top and at the right are the EMEP150 grid indices.

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_2,y_1) (see Fig. S2), we firstly interpolate linearly in the x-direction:

(1a) $f(x, y_1) \approx (1-\lambda) f(x_1, y_1) + \lambda f(x_2, y_1)$ and
(1b) $f(x, y_2) \approx (1-\lambda) f(x_1, y_2) + \lambda f(x_2, y_2)$ with $\lambda = \frac{x-x_1}{x_2-x_1}$

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

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

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.

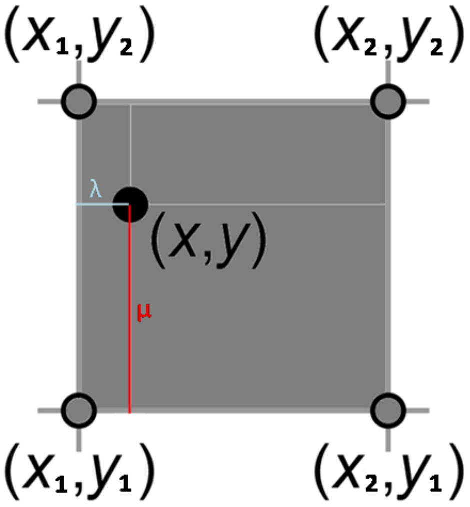

Fig. S2. Graphical representation of the notation used for the bilinear interpolation.

Table S1. Clustering NH_3

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

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

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+NH_3)

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
Prob > |r| under H_0: ρ = 0, all variables significantly correlated with each other

Nr Deposition	soilCNratio	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 <0.0001	0.99998 <0.0001																								
since1890	-0.29217 <0.0001	0.99993 <0.0001	0.99998 <0.0001																							
since1895	-0.29208 <0.0001	0.99984 <0.0001	0.99993 <0.0001	0.99998 <0.0001																						
since1900	-0.29205 <0.0001	0.99971 <0.0001	0.99984 <0.0001	0.99993 <0.0001	0.99998 <0.0001																					
since1905	-0.29209 <0.0001	0.99955 <0.0001	0.99972 <0.0001	0.99984 <0.0001	0.99993 <0.0001	0.99998 <0.0001																				
since1910	-0.29222 <0.0001	0.99936 <0.0001	0.99956 <0.0001	0.99971 <0.0001	0.99984 <0.0001	0.99993 <0.0001	0.99998 <0.0001																			
since1915	-0.29243 <0.0001	0.99913 <0.0001	0.99936 <0.0001	0.99955 <0.0001	0.99971 <0.0001	0.99983 <0.0001	0.99992 <0.0001	0.99998 <0.0001																		
since1920	-0.29273 <0.0001	0.99885 <0.0001	0.99911 <0.0001	0.99934 <0.0001	0.99953 <0.0001	0.99969 <0.0001	0.99982 <0.0001	0.99992 <0.0001	0.99998 <0.0001																	
since1925	-0.29313 <0.0001	0.99853 <0.0001	0.99883 <0.0001	0.99908 <0.0001	0.99931 <0.0001	0.99951 <0.0001	0.99967 <0.0001	0.99981 <0.0001	0.99991 <0.0001	0.99998 <0.0001																
since1930	-0.29365 <0.0001	0.99817 <0.0001	0.99849 <0.0001	0.99878 <0.0001	0.99904 <0.0001	0.99927 <0.0001	0.99948 <0.0001	0.99965 <0.0001	0.9998 <0.0001	0.99991 <0.0001	0.99998 <0.0001															
since1935	-0.29433 <0.0001	0.99774 <0.0001	0.9981 <0.0001	0.99842 <0.0001	0.99872 <0.0001	0.99899 <0.0001	0.99923 <0.0001	0.99944 <0.0001	0.99963 <0.0001	0.99978 <0.0001	0.9999 <0.0001	0.99997 <0.0001														
since1940	-0.29522 <0.0001	0.99725 <0.0001	0.99764 <0.0001	0.998 <0.0001	0.99833 <0.0001	0.99863 <0.0001	0.99891 <0.0001	0.99916 <0.0001	0.9994 <0.0001	0.9996 <0.0001	0.99976 <0.0001	0.99989 <0.0001	0.99997 <0.0001													
since1945	-0.29629 <0.0001	0.99668 <0.0001	0.9971 <0.0001	0.99749 <0.0001	0.99785 <0.0001	0.99819 <0.0001	0.99851 <0.0001	0.99881 <0.0001	0.99908 <0.0001	0.99933 <0.0001	0.99955 <0.0001	0.99973 <0.0001	0.99987 <0.0001	0.99996 <0.0001												
since1950	-0.29748 <0.0001	0.99604 <0.0001	0.99649 <0.0001	0.99691 <0.0001	0.99731 <0.0001	0.99768 <0.0001	0.99804 <0.0001	0.99837 <0.0001	0.99869 <0.0001	0.99898 <0.0001	0.99925 <0.0001	0.99948 <0.0001	0.99968 <0.0001	0.99985 <0.0001	0.99996 <0.0001											
since1955	-0.29875 <0.0001	0.99524 <0.0001	0.99572 <0.0001	0.99618 <0.0001	0.99661 <0.0001	0.99703 <0.0001	0.99742 <0.0001	0.9978 <0.0001	0.99816 <0.0001	0.99851 <0.0001	0.99882 <0.0001	0.99911 <0.0001	0.99938 <0.0001	0.99962 <0.0001	0.99981 <0.0001	0.99995 <0.0001										
since1960	-0.3 <0.0001	0.99412 <0.0001	0.99464 <0.0001	0.99514 <0.0001	0.99562 <0.0001	0.99607 <0.0001	0.99652 <0.0001	0.99695 <0.0001	0.99736 <0.0001	0.99776 <0.0001	0.99814 <0.0001	0.9985 <0.0001	0.99885 <0.0001	0.99918 <0.0001	0.99948 <0.0001	0.99973 <0.0001	0.99991 <0.0001									
since1965	-0.30136 <0.0001	0.99259 <0.0001	0.99316 <0.0001	0.9937 <0.0001	0.99422 <0.0001	0.99473 <0.0001	0.99522 <0.0001	0.99571 <0.0001	0.99618 <0.0001	0.99664 <0.0001	0.99709 <0.0001	0.99753 <0.0001	0.99797 <0.0001	0.99841 <0.0001	0.99884 <0.0001	0.99922 <0.0001	0.99957 <0.0001	0.99986 <0.0001								
since1970	-0.30311 <0.0001	0.99056 <0.0001	0.99117 <0.0001	0.99175 <0.0001	0.99231 <0.0001	0.99286 <0.0001	0.9934 <0.0001	0.99393 <0.0001	0.99446 <0.0001	0.99499 <0.0001	0.9955 <0.0001	0.99602 <0.0001	0.99656 <0.0001	0.99712 <0.0001	0.99769 <0.0001	0.99823 <0.0001	0.99876 <0.0001	0.99931 <0.0001	0.99978 <0.0001							
since1975	-0.30477 <0.0001	0.98728 <0.0001	0.98793 <0.0001	0.98855 <0.0001	0.98915 <0.0001	0.98974 <0.0001	0.99033 <0.0001	0.99092 <0.0001	0.99151 <0.0001	0.99211 <0.0001	0.9927 <0.0001	0.99332 <0.0001	0.99397 <0.0001	0.99468 <0.0001	0.99543 <0.0001	0.99616 <0.0001	0.99693 <0.0001	0.99781 <0.0001	0.99873 <0.0001	0.99956 <0.0001						
since1980	-0.30532 <0.0001	0.98124 <0.0001	0.98194 <0.0001	0.9826 <0.0001	0.98324 <0.0001	0.98388 <0.0001	0.98452 <0.0001	0.98518 <0.0001	0.98585 <0.0001	0.98652 <0.0001	0.98722 <0.0001	0.98795 <0.0001	0.98876 <0.0001	0.98966 <0.0001	0.99064 <0.0001	0.99162 <0.0001	0.9927 <0.0001	0.99404 <0.0001	0.99559 <0.0001	0.99729 <0.0001	0.99902 <0.0001					
since1985	-0.30875 <0.0001	0.96926 <0.0001	0.96997 <0.0001	0.97066 <0.0001	0.97134 <0.0001	0.97203 <0.0001	0.97274 <0.0001	0.97348 <0.0001	0.97426 <0.0001	0.97507 <0.0001	0.97592 <0.0001	0.97684 <0.0001	0.97786 <0.0001	0.97905 <0.0001	0.98035 <0.0001	0.98168 <0.0001	0.98319 <0.0001	0.98515 <0.0001	0.98757 <0.0001	0.99046 <0.0001	0.99399 <0.0001	0.99779 <0.0001				
since1990	-0.31707 <0.0001	0.95608 <0.0001	0.95677 <0.0001	0.95744 <0.0001	0.95813 <0.0001	0.95884 <0.0001	0.9596 <0.0001	0.96043 <0.0001	0.96131 <0.0001	0.96226 <0.0001	0.96328 <0.0001	0.96437 <0.0001	0.96559 <0.0001	0.96699 <0.0001	0.96851 <0.0001	0.97005 <0.0001	0.97181 <0.0001	0.97417 <0.0001	0.97714 <0.0001	0.98078 <0.0001	0.98552 <0.0001	0.99132 <0.0001	0.99742 <0.0001			
since1995	-0.32107 <0.0001	0.94632 <0.0001	0.94695 <0.0001	0.94757 <0.0001	0.94821 <0.0001	0.94889 <0.0001	0.94963 <0.0001	0.95045 <0.0001	0.95136 <0.0001	0.95235 <0.0001	0.95341 <0.0001	0.95455 <0.0001	0.9558 <0.0001	0.95723 <0.0001	0.9588 <0.0001	0.96035 <0.0001	0.96213 <0.0001	0.96458 <0.0001	0.96772 <0.0001	0.97163 <0.0001	0.97691 <0.0001	0.98373 <0.0001	0.99203 <0.0001	0.99829 <0.0001		
since2000	-0.32327 <0.0001	0.91775 <0.0001	0.9184 <0.0001	0.91906 <0.0001	0.91974 <0.0001	0.92048 <0.0001	0.92129 <0.0001	0.9222 <0.0001	0.92322 <0.0001	0.92434 <0.0001	0.92557 <0.0001	0.92692 <0.0001	0.92843 <0.0001	0.93022 <0.0001	0.93221 <0.0001	0.93422 <0.0001	0.93657 <0.0001	0.93983 <0.0001	0.94411 <0.0001	0.94958 <0.0001	0.95713 <0.0001	0.9674 <0.0001	0.98105 <0.0001	0.99154 <0.0001	0.99574 <0.0001	
since2005	-0.33445 <0.0001	0.90695 <0.0001	0.90765 <0.0001	0.90834 <0.0001	0.90907 <0.0001	0.90987 <0.0001	0.91075 <0.0001	0.91174 <0.0001	0.91286 <0.0001	0.91409 <0.0001	0.91545 <0.0001	0.91693 <0.0001	0.91861 <0.0001	0.92059 <0.0001	0.92278 <0.0001	0.92501 <0.0001	0.92761 <0.0001	0.9312 <0.0001	0.93586 <0.0001	0.94172 <0.0001	0.94973 <0.0001	0.96047 <0.0001	0.97498 <0.0001	0.9866 <0.0001	0.99135 <0.0001	0.99804 <0.0001