Biogeosciences Discuss., 12, 4315–4330, 2015
www.biogeosciences-discuss.net/12/4315/2015/
doi:10.5194/bgd-12-4315-2015

Biogeosciences
Discussions
Open Access

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

C. Mulder[1], J.-P. Hettelingh[1], L. Montanarella[2], M. R. Pasimeni[3], M. Posch[1], W. Voigt[4], and G. Zurlini[3]

[1]National Institute for Public Health and the Environment, Bilthoven, the Netherlands
[2]European Commission, DG JRC, Ispra, Italy
[3]Biotechnology and Environmental Science, University of Salento, Lecce, Italy
[4]Institute of Ecology, Friedrich Schiller University, Jena, Germany

Received: 16 December 2014 – Accepted: 23 February 2015 – Published: 11 March 2015

Correspondence to: C. Mulder (christian.mulder@rivm.nl)

Published by Copernicus Publications on behalf of the European Geosciences Union.

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 as terrestrial ecosystems. We investigated the 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 evidence for a rapid response of nature to chronic nitrogen supply by atmospheric deposition.

1 Introduction

The global cycle of nitrogen (N) is unique and highly sensitive to human activities (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; Reich, 2009), affecting the heterotrophic component of ecosystem respiration. 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 related to nitrogen

BGD
12, 4315–4330, 2015

deposition, and the impacts on biodiversity and ecosystem services have caught the attention of many ecologists (Reis et al., 2012; Sutton et al., 2014).

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. We used 19 458 sites in 23 countries from a European Soil Survey (Tóth et al., 2013) to quantify the effect of atmospheric deposition on soil C : N measurements in 2009 and separately investigated 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). More than half of the investigated sites are located in either France (2950 sites), Spain (2693 sites), Sweden (2254 sites) or Germany (1888 sites).

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 (Sect. 2.1). The relation between long-term nitrogen deposition and recent soil C : N ratio was tested by exploring whether clusters of accumulated nitrogen deposition exist and if chemical footprints on soil C : N occur. This large-scale 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 (Sect. 2.2).

BGD
12, 4315–4330, 2015

2 Methods

2.1 Nitrogen deposition

Annual-average deposition time series of oxidized and reduced nitrogen were computed with the former EMEP Lagrangian 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), which computes outputs on a compatible 50 km × 50 km grid (http://emep.int/mscw). These newer results were also used to adjust the older simulations to ensure a smooth transition in our deposition time series. Depositions at the C : N measurement sites were obtained by bi-linear interpolation from the four nearest grid values.

2.2 Soil data

We collected data from a recent European Soil Survey known as LUCAS (Land Use/Cover Area frame Survey): ∼ 20 000 points were chosen for this harmonized sampling with one standardized procedure, resulting in geo-referenced points classified according to land-cover types. Soil samples were collected in 2009 from 23 European countries and all samples, weighing ∼ 11 tons, were sent to one central laboratory at the JRC (Ispra, Italy) and stored in the European Soil Archive Facility where the soil C : N was measured (Tóth et al., 2013). We have selected 19 458 locations with complete categorical site description: 8010 locations were assigned to "croplands" (cereal fields, winter farms with annual or permanent crops, orchards, vineyards, etc.), twelve locations could not be assigned to one specific land use/cover, and the remaining unmanaged locations (including two organic soil outliers with C : N $>$ 200) were woodlands, shrublands or grasslands (lumped together as "nature").

BGD
12, 4315–4330, 2015

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 pre-clusters are merged stepwise until all locations hierarchically close to each other fall within the same cluster. 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 good hierarchical solution will have a reasonably large ratio of AIC changes and the distance ratio measures the current number of clusters against the previous number of clusters. The TwoStep Clustering method became rapidly accepted when Chiu et al. (2001) noted that this approach was able to find the correct number of clusters for ~ 98 % of the generated data sets. Running TwoStep Clustering separately for nitrogen oxides, atmospheric ammonia and reactive nitrogen, the correct numbers of deposition clusters were determined (please refer to the Tables S1–S3 in the Supplement).

3 Results and discussion

Clustering enables detecting 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

BGD
12, 4315–4330, 2015

left). Also Nr shows a clear distinction between its two clusters, where the high annual load (averaging $\sim 15.2\,\mathrm{kg\,N\,ha^{-1}}$, Deposition Cluster VII), covers the former Austro–Hungarian Empire, Western Germany, Brittany and the Po Valley (Fig. 1, upper right).

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 – effects due to nitrogen deposition are far from causal (Nadelhoffer et al., 1999; 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

BGD
12, 4315–4330, 2015

Soil C:N response to recent nitrogen deposition in Europe

C. Mulder et al.

($p < 0.0001$). We analysed the clusters separately with high vs. 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 11 434 natural ecosystems but only a slight $t = -0.59$ for 8010 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 ecosystems than could result from chance.

Focusing on natural 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 natural 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 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 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,

BGD
12, 4315–4330, 2015

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

Spatial 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 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.

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 natural ecosystems is highly-responsive to Nr. We detected where and why nitrogen supply through atmospheric Nr deposition affects natural ecosystems. 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. 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. As the

BGD
12, 4315–4330, 2015

soil black box is now in the “front line” (Schmidt et al., 2011), 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/bgd-12-4315-2015-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.

Acknowledgements. This research was performed for the Dutch Ministry of Infrastructure and the Environment, the Working Group on Effects within the trust fund for the effect-oriented activities under the UNECE Convention on Long-range Transboundary Air Pollution and the 7th EU Framework Programme, Theme ENV.2011.1.1.2-1, Grant Agreement No. 282910 “Effects of Climate Change on Air Pollution Impacts and Response Strategies for European Ecosystems”. The EMEP MSC-W at the Norwegian Meteorological Institute is acknowledged for providing deposition calculations over the last three decades.

References

Bowman, W. D., Cleveland, C. C., Halada, Ĺ., Hresko, J., and Baron, J. S: Negative impact of nitrogen deposition on soil buffering capacity, Nat. 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, in: Proc. 7th ACM SIGKDD International Conference on Knowledge Discovery and Data Mining, San Francisco, CA, 26–29 August, 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, JAPCA J. Air Waste Ma., 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, P. Natl. Acad. Sci. USA, 104, 14580–14585, 2007.

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. Cy., 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, Philos. T. Roy. Soc. B, 368, 20130164, doi:10.1098/rstb.2013.0164, 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. Cy., 24, GB1001, doi:10.1029/2008GB003336, 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.

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, Glob. Change 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, Climate Change, 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.
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, doi:10.5194/hess-7-436-2003, 2003.
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, doi:10.5194/acp-12-7825-2012, 2012.
SPSS: The SPSS TwoStep Cluster Component. A Scalable Component Enabling More Efficient Customer Segmentation, White paper – Technical report, Chicago, IL, 2001.
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.

BGD
12, 4315–4330, 2015

Title Page
Abstract | Introduction
Conclusions | References
Tables | Figures
◀| |▶
◀ ▶
Back | Close
Full Screen / Esc
Printer-friendly Version
Interactive Discussion

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 $1^{\circ} \times 1^{\circ}$ resolution data set of historical anthropogenic trace gas emissions for the period 1890–1990, Global Biogeochem. Cy., 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. Cy., 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.

BGD
12, 4315–4330, 2015

Soil C:N response to recent nitrogen deposition in Europe

C. Mulder et al.

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)

BGD
12, 4315–4330, 2015

Soil C:N response to recent nitrogen deposition in Europe

C. Mulder et al.

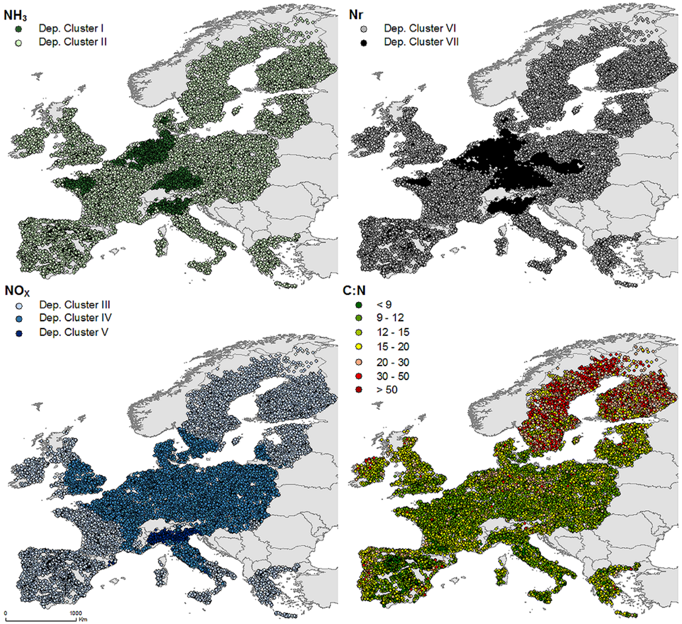

Figure 1. Nitrogen deposition and the recent soil C : N ratios (mass units). Spatial clusters (clockwards) of NO_x, NH_3, and Nr (= NO_x + NH_3) 1880–2010 depositions at the 19 458 sites of the soil C : N in 2009. The darker the colour of a cluster, the higher the nitrogen load for NH_3, NO_x, and Nr. 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 Fig. 3.

BGD
12, 4315–4330, 2015

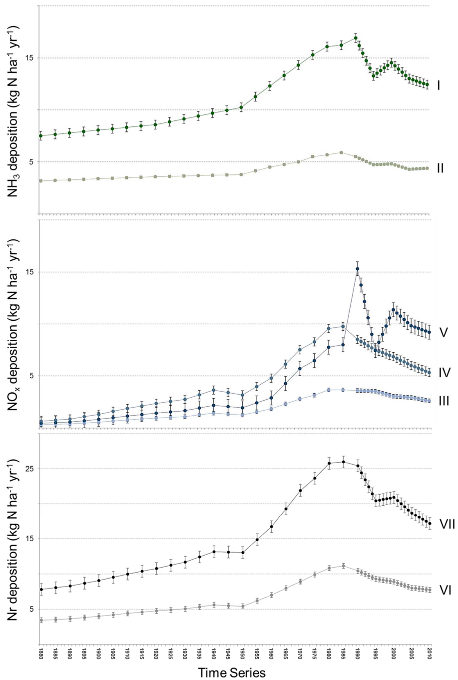

Figure 2. Temporal cluster vector means (averages and standard errors of the series) of the depositions of NH_3 (upper panel), NO_x (middle panel), and Nr (lower panel) across Europe. The colours and Roman numbers correspond to those used for the clusters in Fig. 1. The Nr deposition did not increase 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 year calculations (1880–1990) to yearly observations (1990–2009).

BGD
12, 4315–4330, 2015

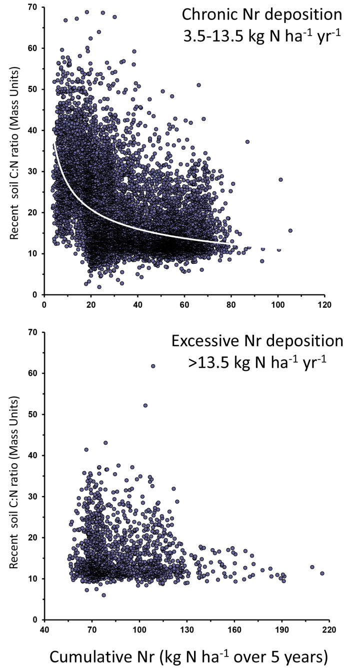

Figure 3. Cumulative Nr deposition of the last five years prior to sampling and soil C : N ratios: negative power functions of soil C : N ratios in nature (measured in 2009) as predicted by cumulative Nr deposition. Upper panel: 9888 unmanaged sites belonging to the cluster with low Nr load but chronic exposure to nitrogen (Deposition Cluster VI); lower panel: 1546 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).

BGD
12, 4315–4330, 2015