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

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Abstract. Long-term human interactions with the natural landscape have produced a plethora of trends and patterns of environmental disturbances across time and space. Nitrogen deposition, closely tracking energy and land use, is known to be among the main drivers of pollution, affecting both freshwater and terrestrial ecosystems. We present a statistical approach for investigating the historical and geographical distribution of nitrogen deposition and the impacts of accumulation on recent soil carbon-to-nitrogen ratios in Europe. After the second Industrial Revolution, large swaths of land emerged characterized by different atmospheric deposition patterns caused by industrial activities or intensive agriculture. Nitrogen deposition affects soil C : N ratios in a still recognizable way despite the abatement of oxidized and reduced nitrogen emissions during the last 2 decades. Given a 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 through atmospheric deposition.

1 Introduction

The global cycle of nitrogen is 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 the 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, atmospheric nitrogen deposition is also a major source of concern because it induces soil acidification by decreasing the exchangeable cation pools (Bowman et al., 2008). Moreover, 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 focused on the loss of biodiversity and the impacts on ecosystem services related to nitrogen deposition (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 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.

To investigate such correlations, we used 19 458 sites in 23 European countries to quantify the effect of atmospheric deposition of nitrogen compounds on soil C : N measurements. We 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, with agriculture 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

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sites are located in France (2950 sites), Spain (2693 sites), Sweden (2254 sites) and Germany (1888 sites).

Given the rapid expansion in Europe of industrial technology and intensive agriculture during the second Industrial Revolution (Mokyr, 1990), we chose 1880 as the starting point of our time series under the hypothesis that accumulated nitrogen deposition since 1880 may contribute the most to the spatial variability of recent soil C : N ratios. The statistical relationship 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.

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 models. Annual-average deposition time series of total (wet and dry) oxidized and reduced nitrogen were obtained from simulations with a Eulerian atmospheric dispersion model (Simpson et al., 2012; for a comparison with measurements see Simpson et al., 2006), 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 oxidized 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 is comprised of NH_3 and NH_4 aerosols. The model output is provided on a grid covering Europe with a resolution of 50 km × 50 km in a polar stereographic projection (see Fig. S1 in the Supplement). Deposition fields are provided for 1990 and after. 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 on a 150 km × 150 km grid (see thick lines in Fig. S1). Results from the overlapping years (1990–1996) were used to adjust the older (Lagrangian) simulations to ensure a smooth transition in the deposition time series (see Schöpp et al., 2003 for details). Depositions at the C : N measurement sites were bilinearly interpolated from the four nearest grid values (Fig. S2).

2.2 Soil data

We collected data from a recent European Soil Survey study known as LUCAS (Land Use/Cover Area frame Survey): ∼ 20 000 geo-referenced points were chosen for this field sampling with the same standardized procedure, 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 Joint Research Centre (Ispra, Italy) and stored in the European Soil Archive Facility in order to obtain a coherent pan-European data set 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 a complete categorical site description: 8010 (intensively) fertilized locations were assigned in situ to fodder crops, annual crops and permanent crops (here as croplands), 14 locations could not be assigned to any specific land use (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 unfertilized locations were assigned in situ to woodlands, shrublands or grasslands (here as nature).

2.3 Cluster analysis

To explore the similarities of the time series from 1880 to 2010, we used the TwoStep Clustering method implemented in SPSS, which is suitable for very large data sets. 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 with the distance ratio measuring the most reliable current number of clusters against alternative solutions.

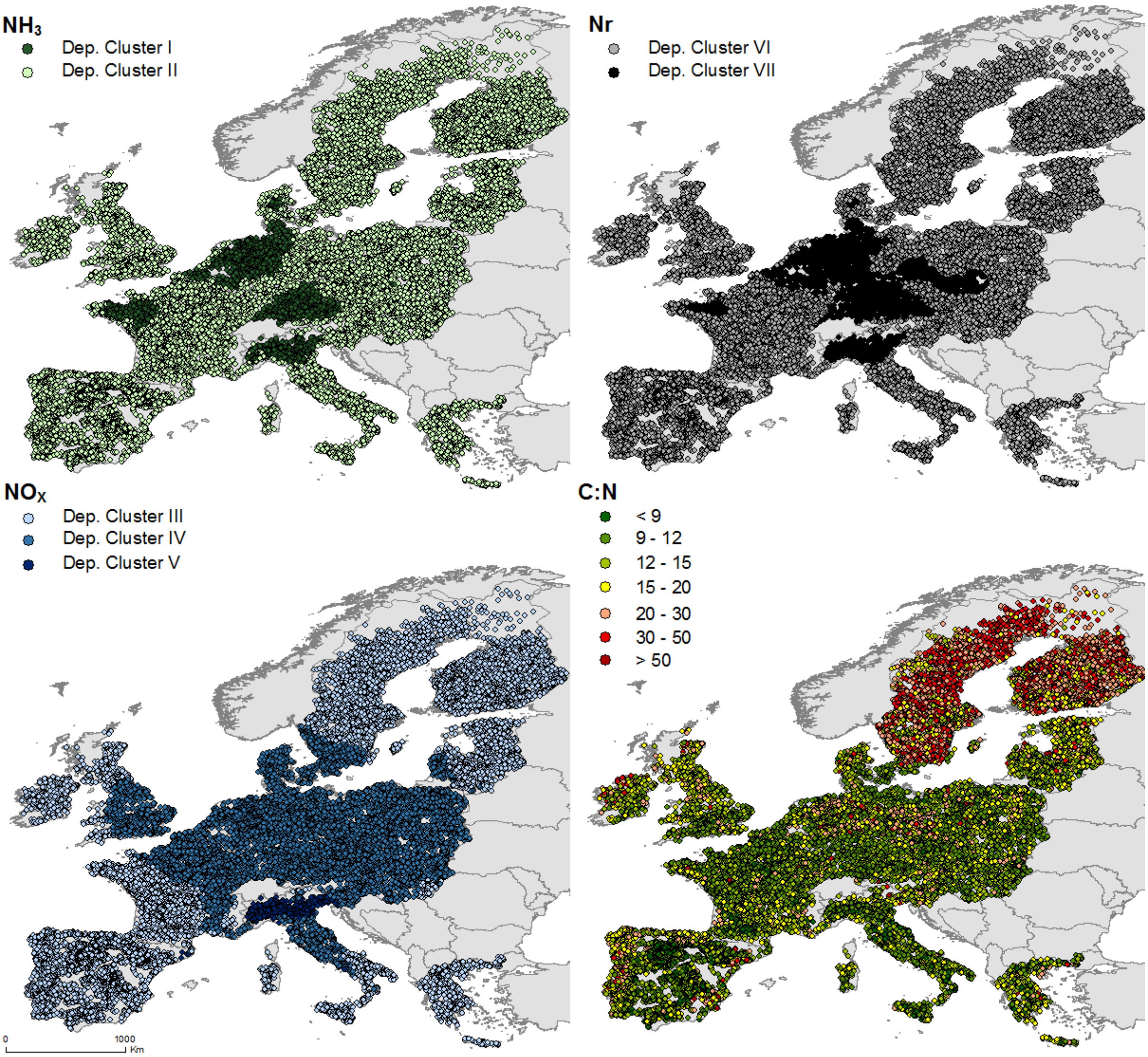

Figure 1. Nitrogen deposition and the recent soil C : N ratios (mass units). Spatial clusters (shown clockwise) of NO_x, NH_3 and Nr (NO_x and NH_3) 1880–2010 depositions at the 19 458 sites of the soil C : N in 2009. The darker the colour of a cluster, the higher the nitrogen load for NH_3, NO_x and Nr. Deposition Cluster IV reveals a high degree of homogeneity in 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).

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

3 Results and discussion

Our statistical clustering enables the objective detection 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 of hotspots and spatial aggregations in all forms of nitrogen deposition across Europe. 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

Table 1. Soil C : N values clearly differ by nitrogen deposition cluster. The soil C : N ratios are given as cluster-specific averages (± standard error); Roman numerals (I–VII) as in Figs. 1 and 2. Both the three-factor ANOVA with NH_3, NO_x and Nr (NO_x and 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) V: 12.67 (±0.23)	III: 17.89 (±0.09)
Nr Deposition Clusters	VII: 14.26 (±0.14)	VI: 16.51 (±0.07)

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, Germany, Brittany and the Po Valley (Fig. 1, upper right).

There are multiple fates for atmospheric N, and its sources have changed substantially (Holtgrieve et al., 2011; Steffen et al., 2015). Within a century, the average of Nr increased more than two-fold everywhere between 1880 and 1980. In 2010 the Nr deposition was still much higher than in 1880, and only 16 sites (0.082 %) exhibited a lower Nr deposition in 2010 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 was a tipping point for nitrogen deposition, and beginning with the 1999 Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone, the deposition of oxidized and reduced nitrogen have simultaneously decreased (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 significant correlations between the nitrogen of mineral soils and the anthropogenic nitrogen deposition are either weak or far from causal (Nadelhoffer et al., 1999; Aber et al., 2003). Our soil C : N ratio 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 a 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 recent 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 nega-

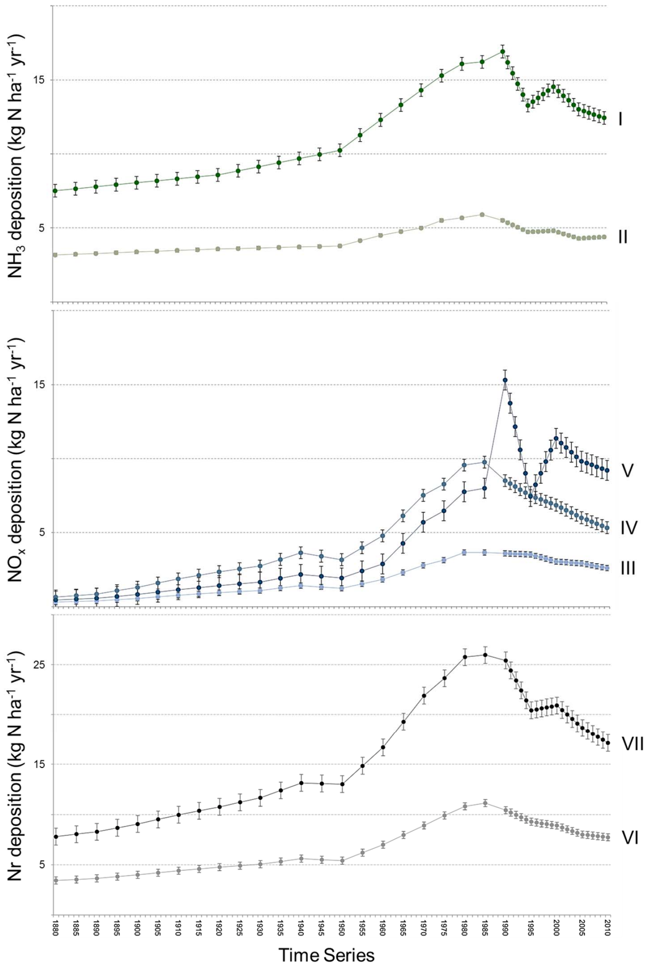

Figure 2. Temporal cluster vector means (averages and standard errors of the series) of the depositions of NH_3 (upper panel), NO_x (middle panel) and Nr (lower panel) across Europe. The colours and Roman numerals correspond to those used for the clusters in Fig. 1. Nitrogen deposition did not increase 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 data set increases from 5-year calculations (1880–1990) to yearly observations (1990–2009).

tive associations between the soil C : N ratio and the nitrogen deposition clusters (the Mantel asymptotic method exhibits $t = -12.23$ for the 11 434 (semi-)natural non-agricultural ecosystems but only a slight $t = -0.59$ for the 8010 agroecosystems). Given the computational independence of our matrices, this Mantel analysis determined that the associations between the nitrogen deposition during 130 years and the recent soil C : N ratios were much stronger in 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 definitely 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 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 these areas are associated with intense human activity, high emissions and soil saturation due to elevated nitrogen loads. Soil C : N of (semi-)natural sites seem to be the most sensitive to 5-year pulses of atmospheric nitrogen supply; short-term deposition is clearly the best predictor for recent soil C : N ratios under chronic nitrogen deposition ($R^2 = 89.2$ %, $F = 66.09$).

In summary, 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 (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 the 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 nitrogen supply through atmospheric Nr deposition affects (semi-)natural ecosystems. As examining the soil black box is now at the "front line" of research (Schmidt et al., 2011; Amundson et al., 2015), mapping the soil and the air compartments together can con-

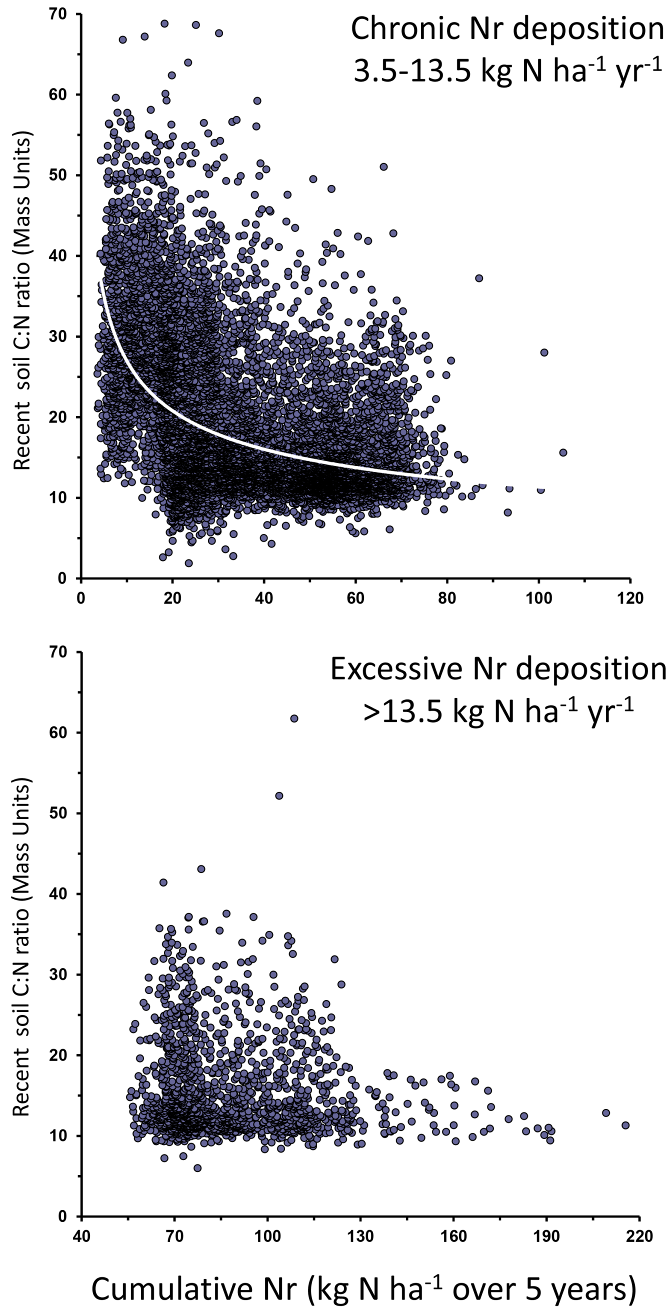

Figure 3. Cumulative Nr deposition of the last 5 years prior to sampling and soil C : N ratios: negative power functions of soil C : N ratios in nature (measured in 2009) as predicted by cumulative Nr deposition. Upper panel: 9888 unmanaged sites belonging to the cluster with low Nr load but chronic 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 (see Table S4).

tribute to a better conservation of our unmanaged environment.

It is challenging to find a mechanistical explanation of why the atmospheric nitrogen supply does not also seem to affect managed ecosystems: 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 has until now hampered the detection of robust effects directly attributable to nitrogen deposition.

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Author contributions. C. Mulder and J.-P. Hettelingh developed 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 had input on the composition of the manuscript.

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