

Interactive comment on “Chemical footprints of anthropogenic nitrogen deposition on recent soil C : N ratios in Europe” by C. Mulder et al.

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Dear Board, In addition to the previous reply (where only short answers were provided as we focused more on the statistical part), given your request for more details on all the used methodologies, we add a new M&M part and two additional figures illustrating the grids and explaining the used interpolation. Here the enhanced complete M&M:

2.1 Nitrogen deposition Annual-average deposition time series of oxidized and reduced nitrogen were obtained from simulations with the Eulerian atmospheric dispersion model (Simpson et al., 2012 for a detailed description), 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). This model output is provided on a grid covering Europe with

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a resolution of 50 km × 50 km 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 on a 150 km × 150 km grid (Supplement). 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 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 points were chosen for this field sampling with a single standardized procedure, resulting in geo-referenced points classified according to land-cover types (Tóth et al., 2013). 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 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 locations were assigned in situ 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 (incomplete documentation), and all the remaining locations (including two organic soil outliers with C:N > 200) were assigned in situ to either unmanaged woodlands, shrublands, or grasslands (lumped together as ‘nature’).

2.3 Cluster analysis To explore the similarities of the time series from 1880 up to 2010,

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we used the TwoStep Clustering method 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 with the distance ratio measuring the most reliable current number of clusters against alternative solutions. The TwoStep Clustering method became rapidly accepted when Chiu et al. (2001) demonstrated that such 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, 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 (refer to the Tables S1–S3 in the Supplement).

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

Please also note the supplement to this comment:

<http://www.biogeosciences-discuss.net/12/C2059/2015/bgd-12-C2059-2015-supplement.pdf>

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