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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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EC: The paper tries to bring light into an interesting topic that seems trivial at a first glance. Does the C:N ratio in the top soil reflect the systematic increase of atmospheric N deposition? The authors present a cluster analysis and try to combine information on the spatial trend of N-deposition based on the EMEP model with about 20,000 soil profiles across Europe.

REPLY: Indeed, we investigated the geographical distribution of nitrogen deposition in Europe after the Second Industrial Revolution (1880–2010) and the impacts of accumulation on the C:N ratio in the top soil. Obviously, the nitrogen deposition affected the soil C:N ratio but (most important), focusing on unmanaged ecosystems, we provided unexpected evidence for a rapid response of nature to the chronic nitrogen supply by

atmospheric deposition.

EC: The statistical analysis needs to bring together two different things, a spatial distribution of soil C:N ratio at a given time with a N-deposition trend (divided into three categories) on a rather coarse grid over Europe. C:N ratio in a specific soil core reflect an integrated time history on a very local scale. N deposition processes are necessarily over smoothed by the used model approach. The idea of the statistical analysis needs to be presented in a way that it is transparent also for interested reader without a strong statistical background.

REPLY: We thank the Editor-in-Chief for these comments. Due to the very long period on N-deposition taken into account (1880-2010), we were forced to use two EMEP grids, the one (the rather coarse grid of 150*150 km you mentioned, shown in Fig. S1) was used only until 1990, and the other, much finer grid described by Simpson et al. (Atmos. Chem. Phys., 12, 7825–7865, 2012) was used afterwards. In both cases, regardless of the grid, the depositions at the C:N measurement sites were obtained by bi-linear interpolation from the four nearest grid values. Hence, the spatial resolution is high even with a coarse grid, seen that for each of the 19,458 sites a bi-linear interpolation from the four nearest grid values was computed for each of the three N categories (here, NOx, NH3 and Nr). What changes is not the resolution of the grid alone, but the temporal resolution of the atmospheric deposition for each of the three N categories, as we switched from 5-year calculations (1880–1990) to yearly observations (1990–2009). For 5-year calculations, the resolution of the first EMEP grid seems to be quite high, but for yearly observations we really need the much finer 50*50 km grid (Fig. S2).

It is generally found that such models capture broad features over large areas, although it should be noted that comparisons of observed and modelled trends in the last decades are extremely consistent. We deliberately kept such methodological background short since it was not the focus of our paper, but we will be pleased to add more information in response to this comment. In particular the TwoStep Clustering deserves more details and references, and therefore would like to rewrite the section

2.3 as follows:

"To explore the similarities of the time series from 1880 up to 2010, 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 (Lazary et al., 2014) to medicine (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 (please refer to the Tables S1–S3 in the Supplement)."

EC: How you can convince the readers that the observed spatial pattern of C:N ratios in the soil is causally linked with the N-deposition? Especially the figures need to be better explained, how are the clusters defined, what are the mass units?

REPLY: While an inverse correlation between anthropogenic nitrogen input and soil C:N seems to be intuitive, the extent to which this relationship holds has never been investigated before. 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 the

short-term supply of atmospheric nitrogen in their recent past. This is the novelty of our communication. The clustering allowed to identify and to separate the soils under low pressure from the soils under high pressure. Being a ratio, the soil C:N is adimensional and we added the adjective "mass" to units because with molar units the ratio would have been different. We computed the soil C:N ratio as gram C / kg soil divided by grams N / kg soil, resulting in grams C / grams N, thus C/N (no units).

EC: Half of the 20,000 soil sites are coming from soils that are classified as unmanaged, thus not from intensively managed agricultural land. I guess that many of the so called unmanaged sites have been affected by anthropogenic activities, e.g. alp meadows etc.

REPLY: The Editor-in-Chief correctly pointed to a remaining challenge in vegetation science. For instance, Gundersen (1991) claims independence of the C:N ratio from N deposition suggesting that soil C:N of forests is driven by factors such as climate and history. This holds for sure for plant species, hence directly for litter quality, and thus indirectly also for soil C:N. However, anthropic disturbance during the late Holocene (many even use the term Anthropocene) affected the entire European continent, even the Alpine meadows. For a geographical comparison with a comparable area, Liu et al. (2013) stated that "recent studies indicate that Nr deposited by China may be moving to surrounding marine ecosystems and perhaps to tropical and subtropical forests. . .. Clearly, N deposition has increased significantly in China and has affected both non-agricultural and agricultural ecosystems." Nitrogen exhibits a strong global pattern, affecting both human-managed (fertilized) as unmanaged ecosystems worldwide, as can be seen by the current status of the biogeochemical flows of N very recently described by Steffen et al. (2015).

In our paper, we analyzed the managed and unmanaged ecosystems separately and identified with the the Mantel's asymptotic method different negative associations between the soil C:N and the nitrogen deposition clusters in the 11,434 natural ecosystems and the 8,010 agroecosystems, demonstrating for the first time that the association between nitrogen deposition and recent soil C:N ratios is much stronger in natural ecosystems (and this is the first novelty) and that the chemical footprint of the accumulated nitrogen in all forms during the last century is much less than expected (and this is the second novelty).

EC: It is important to give more details on the soil sampling procedure, what was sampled, the top soil, profiles etc.

REPLY: We thought that a reference to the ISO methods applied to LUCAS by Tóth et al. (Environ. Monit. Assess., 185, 7409–7425, 2013) would have been enough, but we will be pleased to add details and enlarge that part as well.

EC: The paper addresses an important topic well in scope of BG. The presented analyses are showing promising results, but the paper needs to be substantially improved in order to be published in BG.

REPLY: We are happy that the Editon-in-Chief agrees with our choice for BG. We believe that many readers will be interested in the aforementioned novel results over the rapid response of "nature" to chronic nitrogen deposition.

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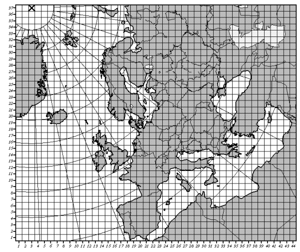

Fig. S1. The first grid used. http://emep.int/mscw/index_mscw.html

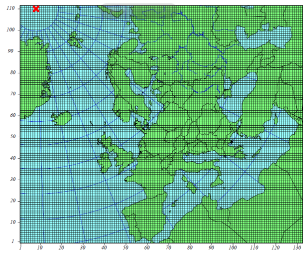

Fig. S2. The second grid used. http://emep.int/mscw/index_mscw.html

Fig. 1. Composite plate (Fig. S1 and S2) showing the used grids