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Biogeosciences Discussions

Interactive comment on "Governing processes for reactive nitrogen compounds in the atmosphere in relation to ecosystem, climatic and human health impacts" *by* O. Hertel et al.

Anonymous Referee #1

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General Comments The manuscript by Hertel et al. provides a useful review of the reactive nitrogen (Nr) emissions, transformations, and deposition with an emphasis on observation and modeling research in the EU. Nr is controlled by a number of complex processes, which determine transport pathways in the environment. The authors address chemical, physical, and biological interactions of the oxidized and reduced species (e.g., NOx, NOy, and NHx), including organics. They also consider issues related to spatial and temporal distribution of Nr compounds and how these affect ecosystem and human health. Lastly, the authors identify specific areas of need that could improve the understanding of Nr governing processes. Overall, the authors offer a constructive view of Nr that places the issue in a broader context of global climate

and ecosystems. This reviewer recommends publication of the manuscript after careful consideration of the specific comments addressed below, including notes regarding additional relevant references.

Specific Comments Page 9352 Ln 8: Quantify the 'fast' dry deposition rate or give a range of values. Is this higher than 2 cm/sec, for example? A reference from Phillips et al. (Atm Env, 2004) might be useful. Ln 13: Quantify the 'slow' deposition velocity. Perhaps a value on the order of 0.10 – 1 cm/sec? References by Duyzer et al (JGR, 1994) and Horvath (Atm Env, 2003) might be useful. Ln 17: There is some uncertainty regarding atmospheric residence time of NH3 in the atmosphere (1-2 days). The authors should cite their source for the listed 24 h. Page 9353 Ln 7: There is another US study of oxidized nitrogen in urban areas (Luke et al, Atm Env. 2010) that might provide additional context given that the broader applicability of the mountain study may be limited. Ln 19: Instead of disregarding certain compounds in Fig. 1, it would be more useful to include them in a sidebar or caption. Page 9355 Ln 20: Provide appropriate references. Ln 23: It is interesting that landfills are listed with non-anthropogenic sources such as natural fires. It would seem that landfills, though a smaller sources, might be more appropriately listed with sewage systems or perhaps generalized under 'other anthropogenic waste sources' or a similar phrase. Page 9357 Ln 24: For readers unfamiliar with regional differences of European emissions, it would be helpful to provide context for these differences between Northern Europe and Southern Europe, as this seems to be central to the authors' suppositions about manure/fertilizer emissions. Are the types of crops, growing seasons, etc. vastly different? Page 9359 Insertion of a research program table would be useful for the reader. The table should list program/database name; resolution; web link; years of data collection; brief description, etc. Page 9360 Ln 4: Identify the 'national experts.' Are these academic, government or industry groups? Page 9361 Ln 22: Quantify the height imposed by the regulations (in meters) and provide references that substantiate the statement that local urban areas are 'unaffected.' Page 9362 Sec 2.2.2: There is an unacceptable paucity of references in this paragraph that should be corrected, in particular for statements regarding

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transport and seasonal variations observed. Ln 24: Have other countries or regions experienced increases in NOx as a result of fewer nuclear energy sources? It would improve the argument if such data and references were included. Page 9364 Ln 11: Quantify the magnitude of the decrease in H2SO4. Ln 13: What time period does 'in those days' refer to? Page 9365 Ln 4: The reversible nature of the reaction should be explored in more detail. What are the atmospheric conditions that promote the return of NH3? How common is this process in air masses over Europe? Ln 15: It seems that authors are attempting to indicate that HNO3 is therefore more abundant and available for chemical reactions than HCl, but this is not explicitly stated. Ln 19: Referring to Pg. 9360, Ln 26, it seems as if the SO2 emission growth is contradictory to earlier statements. Please clarify. Page 9366 Ln 19: Given that O3 is a major reacting species, what are the O3 trends in European countries? Page 9367 There is an unacceptable paucity of references in this paragraph that should be corrected, in particular for statements regarding conversion of NO to NO2 and ubiquitous nature of OH in the polluted atmosphere. Page 9369 Ln 3: What is the height (in meters) of the tower? Ln 27: Provide references for the nighttime/daytime HONO chemistry. Page 9370 Ln 3: Have any of the 'few studies' been published in the literature? Page 9371 Ln 13: Additional reference to DON measurements by Walker et al. (Atm Env, 2012) should be considered. Ln 17: Additional reference to organic nitrogen work by Lin et al. (ACP, 2010) should be considered. Page 9372 There is an unacceptable paucity of references that should be corrected as there are several papers that could inform statements in this section. Some useful references include Gaffney et al (EST, 1993) on PAN and NO2 measurements; Tanner et al. (EST, 1988) on aldehydes; Schrimpf et al. (GRL, 1996) on PAN dry deposition; and Doskey et al (JGR, 2004) on PAN deposition. Ln 26 and 27: Add references for NADP (Lamb et al., Atm Env, 2000) and EANET (Totsuka et al, 2005, book chapter). Page 9373 Ln 1: For inferential models (e.g. used by NADP), a reference to Meyers et al. (JGR, 1998) should be included. Ln 4: These spare datasets are based on short term campaigns or long-term monitoring studies. Ln 11: There is an excellent comparison study by Milford et al. (BG, 2009) that describes a number

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of techniques and their performance for NH3 flux measurements. Ln 22: The comprehensiveness can only 'presently' be achieved with modeling would be a more accurate statement. Ln 25: These contributions also depend on local and regional meteorology and emissions in addition to climate. Page 9374 Ln 3: Additional references to a paper by Walker et al (BGD, 2010) would be useful as it describes some source/sink characterization and details the affect of vegetative state (open/closed stomata, etc.) on NH3 flux. Ln 9: NH3 modeling work by Yu et al (AgFMet, 2009) and Pleim et al. (report published by Springer, 2012) should be referenced here. Both these papers note some challenges that might support the information presented here by the authors. Page 9376 The description of resistances, in particular the behavior of HNO3 and NH3 deposition across the layers, could be supplemented by references to Cellier and Brunet (AFMet, 1992); Spokes and Jickells (CSR, 2005), and Sorensen et al (AE, 2003), which provide useful information about the roughness sublayer over various terrain. A schematic of the resistances (in parallel and series) would be beneficial to the reader. Many such schematics have been published in the literature. Page 9379 Ln 7: It would be useful to include the Henry's Law coefficient for NH3 here. Page 9385 Ln 25: A list of the EU precipitation networks or at least the total number should be included to provide the reader with some context.

Figure 1: The sketch does not include sufficient information to truly illustrate the complexity of the system. Percentages of compounds that continues through each pathway should be included. As transport and dispersion exist throughout the systems, move this notation to a sidebar and eliminate the multiple blue boxes. Add symbols to indicate the phases (e.g. g for gaseous, p for particulate, etc.). Insert an external sidebar that lists other species, such as PAN, N2O, etc.

Fig. 2: The graph does not add value to the material presented as is adequately described in the text. It is not needed.

Technical Corrections General Ensure that all acronyms (including model names) are fully identified at first use. At times, geography of observations and models mentioned

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in the text was clearly evident. However, there are other instances where it was difficult to discern whether statements referred to work in specific European countries or to international efforts. Identified In the abstract, the first sentence should be reworded to avoid double use of different/differences. Another double use occurs on Page 9357, Ln 21 (affect).

Interactive comment on Biogeosciences Discuss., 9, 9349, 2012.