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Joint analysis of deposition fluxes and atmospheric concentrations of inorganic nitrogen and sulphur compounds predicted by six chemistry transport models in the frame of the EURODELTAIII project

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HIGHLIGHTS

- The estimates of N and S deposition by six regional models are evaluated.
- The inclusion of sea salt sulfate emissions was found to be important.
- Formation of $NH3+NH4^+$ is generally underestimated in summer.
- There is a general underestimation of wet deposition of reduced N by most models.
- Different performance was found for the different models and pollutants.

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ABSTRACT

In the framework of the UNECE Task Force on Measurement and Modelling (TFMM) under the Convention on Long-range Transboundary Air Pollution (LRTAP), the EURODELTAIII project is evaluating how well air quality models are able to reproduce observed pollutant air concentrations and deposition fluxes in Europe. In this paper the sulphur and nitrogen deposition estimates of six state-of-the-art regional models (CAMx, CHIMERE, EMEP MSC-W, LOTOS-EUROS, MINNI and CMAQ) are evaluated and compared for four intensive EMEP measurement periods (25 Feb–26 Mar 2009; 17 Sep–15 Oct 2008; 8 Jan–4 Feb 2007 and 1–30 Jun 2006).

For sulphur, this study shows the importance of including sea salt sulphate emissions for obtaining better model results; CMAQ, the only model considering these emissions in its formulation, was the only model able to reproduce the high measured values of wet deposition of sulphur at coastal sites. MINNI and LOTOS-EUROS underestimate sulphate wet deposition for all periods and have low wet deposition efficiency for sulphur.

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For reduced nitrogen, all the models underestimate both wet deposition and total air concentrations (ammonia plus ammonium) in the summer campaign, highlighting a potential lack of emissions (or incoming fluxes) in this period. In the rest of campaigns there is a general underestimation of wet deposition by all models (MINNI and CMAQ with the highest negative bias), with the exception of EMEP, which underestimates the least and even overestimates deposition in two campaigns. This model has higher scavenging deposition efficiency for the aerosol component, which seems to partly explain the different behaviour of the models.

For oxidized nitrogen, CMAQ, CAMx and MINNI predict the lowest wet deposition and the highest total air concentrations (nitric acid plus nitrates). Comparison with observations indicates a general underestimation of wet oxidized nitrogen deposition by these models, as well as an overestimation of total air concentration for all the campaigns, except for the 2006 campaign. This points to a low efficiency in the wet deposition of oxidized nitrogen for these models, especially with regards to the scavenging of nitric acid, which is the main driver of oxidized N deposition for all the models. CHIMERE, LOTOS-EUROS and EMEP agree better with the observations for both wet deposition and air concentration of oxidized nitrogen, although CHIMERE seems to overestimate wet deposition in the summer period. This requires further investigation, as the gas-particle equilibrium seems to be biased towards the gas phase (nitric acid) for this model.

In the case of MINNI, the frequent underestimation of wet deposition combined with an overestimation of atmospheric concentrations for the three pollutants indicates a low efficiency of the wet deposition processes. This can be due to several reasons, such as an underestimation of scavenging ratios, large vertical concentration gradients (resulting in small concentrations at cloud height) or a poor parameterization of clouds.

Large differences between models were also found for the estimates of dry deposition. However, the lack of suitable measurements makes it impossible to assess model performance for this process. These uncertainties should be addressed in future research, since dry deposition contributes significantly to the total deposition for the three deposited species, with values in the same range as wet deposition for most of the models, and with even higher values for some of them, especially for reduced nitrogen.

1. Introduction

Atmospheric deposition of air pollutants can lead to a range of detrimental impacts to terrestrial and aquatic ecosystems. Nitrogen (N) deposition is currently considered a major threat to European biodiversity, including sensitive habitats and species listed under the European Commission Habitats Directive (92/43/EEC) (Sutton et al., 2011; Ochoa et al., 2014). N deposition can lead to the replacement of local plant communities of species adapted to low-nutrient environments by nitrophilous species able to thrive under high-N conditions (Stevens et al., 2004). On the other hand, an alteration of soil N and carbon storage could contribute to either mitigate or reinforce the effects of climate change (Reich et al., 2006). The deposition of both sulphur (S) and nitrogen (N) can lead to the acidification of soils as well as freshwater and marine ecosystems (Longhurst, 1991). Acidification makes forests and other ecosystems more vulnerable to stress factors such as frost, drought and pests (Bouwman et al., 2002; Heij and Schneider, 1991).

It is generally difficult and expensive to measure the components of atmospheric deposition, especially dry deposition fluxes, and thus the use of deposition estimates simulated by chemical transport models (CTMs) has become a common practice. Nowadays modelled deposition is commonly used to evaluate a range of environmental impacts. For example, modelled deposition fluxes of nitrogen and sulphur can be used to evaluate potential ecosystem damage by comparing annual deposition rates with habitat-specific thresholds, such as critical loads for acidification and nutrient nitrogen (Nilsson et al., 1988). Maps of the exceedances of critical loads in Europe for last decades can be found in annual EMEP Status Reports (http://www.emep.int). A robust evaluation of model capabilities to correctly predict atmospheric deposition rates is, therefore, necessary, beyond the evident importance of correctly calculating air pollutant concentrations.

Atmospheric deposition can occur through dry or wet mechanisms. Wet deposition refers to the processes of scavenging of air pollutant by hydrometeors, *i.e.* cloud and fog droplets, rain or solid precipitation. One of these processes is the dissolution into cloud-drops of soluble gases such as NH_3, HNO_3 and SO_2, present in the interstitial cloud air. A proportion of aerosol particles (nitrates, sulphates) can also be removed within clouds by incorporation into the liquid phase. Below clouds, pollutants can be scavenged by precipitation elements between the cloud base and the surface. Soluble gas species can dissolve into falling raindrops during rain, while airborne particles can be collected by raindrops through collisions. Dry deposition includes a downward transport and the subsequent uptake of the atmospheric pollutant species by surfaces, in the absence of precipitation. Models generally use an approach based on an electrical resistance analogy, defining a “resistance” to deposition, for the turbulent transport, molecular diffusion and surface processes, adding them in the same way as electrical resistances. Downward fluxes for particles can also be increased by sedimentation.

The presence of NH_3, HNO_3 and H_2SO_4 in the atmosphere is the result of a combination of processes. Whereas ammonia is directly emitted, nitric acid (HNO_3) and sulphuric acid (H_2SO_4) can be formed through the oxidation of nitrogen dioxide (NO_2) and sulphur dioxide (SO_2). Anthropogenic SO_2 emissions mainly come from the combustion of fossil fuels (primarily coal and oil), whereas natural sources of atmospheric S include volcanoes and marine algae, mainly in the form of dimethyl sulphide (DMS). Nitric oxide (NO) and NO_2 emissions are mainly from fossil fuel combustion, biomass burning, and microbiological emissions from soils (Lee et al., 1997). In Europe NH_3 mostly comes from agricultural practices such as the volatilization from animal waste and synthetic fertilizers with contributions from other sources such as biomass burning, emissions from oceans and soils under natural vegetation, emissions from waste industrial processes and transport