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Ammonia emission time profiles based on manure transport data improve ammonia modelling across north western Europe

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HIGHLIGHTS

- Time profiles of ammonia emissions from manure application were modelled using manure transport data for north western Europe.
- Meteorological dependent emission characteristics for ammonia increased model performance for ammonia.
- Source attribution modelling shows a 1–2 μg/m^3 contribution of Flemish agriculture to domestic PM concentrations.

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ABSTRACT

Accurate modelling of mitigation measures for nitrogen deposition and secondary inorganic aerosol (SIA) episodes requires a detailed representation of emission patterns from agriculture. In this study the meteorological influence on the temporal variability of ammonia emissions from livestock housing and application of manure and fertilizer are included in the chemistry transport model LOTOS-EUROS. For manure application, manure transport data from Flanders (Belgium) were used as a proxy to derive the emission variability. Using improved ammonia emission variability strongly improves model performance for ammonia, mainly by a better representation of the spring maximum. The impact on model performance for SIA was negligible as explained by the limited, ammonia rich region in which the emission variability was updated. The contribution of Flemish agriculture to modelled annual mean ammonia and SIA concentrations in Flanders were quantified at respectively 7–8 and 1–2 μg/m^3. A scenario study was performed to investigate the effects of reducing ammonia emissions from manure application during PM episodes by 75%, yielding a maximum reduction in modelled SIA levels of 1–3 μg/m^3 during episodes. Year-to-year emission variability and a soil module to explicitly model the emission process from manure and fertilizer application are needed to further improve the modelling of the ammonia budget.

1. Introduction

Ammonia (NH_3) is the primary form of reactive nitrogen in the environment (Sutton et al., 2013). NH3 is lost to the environment at different stages of the nitrogen cascade: during and after application of fertilizer to land, from senesces of plants, animal excretion in housing systems, during grazing and after application of manure, in food processing, at industries using NH_3 and as a byproduct from car exhausts equipped with a three-way catalyst (Erisman et al., 2007; Galloway et al., 2003). The atmospheric lifetime of ammonia is limited to several hours as it is effectively removed by dry and wet deposition. Once deposited, the reduced nitrogen components contribute to acidification and eutrophication of vulnerable terrestrial and aquatic ecosystems that can lead to reduced biodiversity (Bobbink et al., 1998; Krupa, 2003). Recently, also the role of reduced nitrogen in the fixation of carbon dioxide has emerged as a new research topic (Reay et al., 2008).

Ammonia readily reacts with sulfuric and nitric acid to form its particulate ammonium salts (Fowler et al., 2009). These particles play an important role in the radiation balance of the earth as they contribute effectively to light scattering and they alter the number, size and hygroscopic properties of cloud condensation nuclei (Xu

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and Penner, 2012). Moreover, particulate ammonium salts provide a means of long range transport of reduced nitrogen due to their longer atmospheric lifetime than ammonia. In terms of mass, ammonium salts contribute largely (40–50%) to the levels of fine particulate matter (Putaud et al., 2004), which is thought to cause adverse health effects (Brunekreef and Holgate, 2002). Especially ammonium nitrate concentrations are increased during particulate matter episodes in Europe (Weijers et al., 2011; Vercauteren et al., 2011). The potential to mitigate particulate matter concentrations through ammonia emission reductions has been highlighted by several modelling studies (e.g. Banzhaf et al., 2013; Bessagnet et al., 2014; Erisman and Schaap, 2004). In comparison to mitigating emissions of sulphur dioxide and nitrogen oxides, taking measures aimed at reducing ammonia emissions is considered to be cost-effective (Pinder et al., 2007).

Despite its central role in many environmental threats, our knowledge about the ammonia budget is rather poor. Agriculture largely dominates the ammonia emissions to air in Europe, of which livestock excretion is the most important source (European Union, 2010). However, there are large uncertainties in the emission estimates for ammonia with errors over 50% on the global emission budget and even higher uncertainties on regional/local scales (e.g. Galloway et al., 2008; Sutton et al., 2013). In north western Europe, livestock housing and manure application are the agricultural activities leading to the largest ammonia emissions (Velthof et al., 2015). Emissions from livestock housing in principle depend on the total inorganic nitrogen content of manure, the temperature and ventilation conditions as well as housing type (Groot Koerkamp et al., 1998). Manure and fertilizer application occurs mainly at the start of the growing season for summer and winter crops, which occurs at different times in different climate zones and may differ between years (Geels et al., 2012). Besides, the timing of manure application depends on soil conditions such as soil water content and non-frozen conditions, as well as agricultural practice, crop type and legislative limitations (Sommer and Hutchings, 2001; Webb et al., 2010). Although the annual emissions of manure application are lower than those from livestock housing, the limited time period of application causes these emissions to dominate during springtime in north western Europe.

Designing mitigation strategies for particulate matter relies on the use of chemistry transport models (CTMs) (Kiesewetter et al., 2015; Simpson et al., 2012). The emission information used in CTMs consists of inventories obtained by combining activity data and activity specific emission factors. In most CTMs, the annual total emission of ammonia is translated into hourly fluxes using relatively simple approaches (Hutchings et al., 2001; Pouliot et al., 2012). Hence, the intrinsic dependence of ammonia emissions on meteorological conditions is neglected, which may be a severe limitation (Sutton et al., 2013). Several validation studies have highlighted the underestimation of the temporal variability in ammonia concentrations in comparison to observations (Menut and Bessagnet, 2010; Pouliot et al., 2012; Skjøth and Geels, 2013). Moreover, evaluation of seasonal cycles have highlighted that the month with maximum ammonia levels in spring is often not predicted correctly (e.g. Banzhaf et al., 2013). A first attempt to model NH_3 emissions dynamically has been presented for Denmark (Skjøth et al., 2011). Application of the emission model including temperature effects but neglecting impacts of soil conditions improved the CTM skill. A second study related agricultural cropping information and a process based description of the ammonia evaporation from soils after fertilizer application (Hamaoui-Laguel et al., 2014). This study showed that the more explicit approach affected the spatial distribution of the ammonia emissions in France. Moreover, they reported a systematic effect on modelled SIA concentrations for a 3-month period in 2007. The temporal variability in manure application is not yet included in this study. Paulot et al. (2014) use an inverse modelling approach to establish ammonia emission timing characteristics from wet deposition flux measurements while Bash et al. (2013) use a process-based approach by coupling a CTM to an agroecosystem model. These recent efforts show that the need to improve the temporal variation of ammonia emissions is widely recognized.

For some countries with a large livestock farming, detailed data on manure production, transport and/or application are available in order to monitor and regulate the amount of manure applied to the land. This is for example the case for Flanders, where in 2013 82% of ammonia emissions came from animal manure (VMM, 2014). In this study we examine the use of Flemish manure transportation data to model the temporal variability in ammonia emissions from manure application. Moreover, we assess if the modified temporal variability affects modelled ammonia and SIA concentrations using the CTM LOTOS-EUROS. Using the updated variability, we investigate the impact of reducing emissions from manure application on PM levels during episodes.

2. Ammonia emission variability

The CTM LOTOS-EUROS uses sector-specific time profiles to model the temporal variation of anthropogenic emissions over the year. For ammonia from agriculture, the seasonal and hourly variation that is used is shown in Fig. 1. Standard practice in CTMs is to use these fixed time profiles for each year, independent of climatological variables (Flechard et al., 2013). In reality, however, meteorological conditions have a large impact on emissions from agriculture. The start of the growing season and soil conditions determine when farmers work and fertilize their land. Also, emissions of volatile ammonia from manure and livestock housing increase with temperature. As the fate of reactive nitrogen after emission is also highly dependent on concurring meteorological conditions, it is important to use meteorological dependent time profiles for ammonia emissions in LOTOS-EUROS.

To investigate the importance of a correct representation of temporal variability in ammonia emissions, updated emission profiles for manure application, fertilizer application and emissions from livestock housing are used in this study. These time profiles are described in the following sections.

2.1. Manure application

Manure transportation data were used as a proxy to estimate the variability in ammonia emissions from manure application in Flanders during year. The Flemish Landmaatschappij (VLM) provided manure transportation data for 2007–2011 comprising manure transports between farmers on a province basis. For these transports, the provinces in which sending and receiving farmer reside and the amount of manure, including its nitrogen content, are reported. Two types of transport data were used. Daily data are available for manure transports across multiple municipality boundaries. For manure that is transported across less than two municipality boundaries, longer running contracts can be reported (these were not available for 2007). The VLM divides these evenly over the weeks in the contract period. For the years 2007–2011, daily reports of manure transport accounted for 70% of the total nitrogen content of the manure, while the longer running contracts contributed only 30%.

Fig. 2 shows the amount of manure transport (in kg nitrogen) reported on a daily basis for each receiving province for 2008. In this year, manure transport peaked in early spring (February-March), late spring (April-May) and late summer (August-September). In April-May, the amount of transported manure is