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Can further mitigation of ammonia emissions reduce exceedances of particulate matter air quality standards?

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ABSTRACT

Several studies point out the importance of agricultural emissions to particulate matter (PM) concentrations, and particularly of NH_3 emissions to $PM_{2.5}$. Our study used three different chemical transport models (CHIMERE, EMEP and LOTOS-EUROS) to quantify the reductions of $PM_{2.5}$ and PM_{10} concentrations due to reductions of NH_3 emissions beyond the Gothenburg Protocol (GP), as well as due to the GP alone compared to 2009. Simulations of $PM_{2.5}$ and PM_{10} concentrations using 2009 meteorology were undertaken for five emission scenarios: 2009 emissions (as the reference simulation), GP emissions in 2020, and further 10%, 20% and 30% NH_3 emission reductions in EU27 beyond the GP. The modelling results for the scenarios with further 10%, 20% and 30% NH_3 agriculture emission reductions in EU27 beyond the GP show that the reduction achieved in PM concentrations is not linear with the emission reductions. In fact, the results from the study show that the impact of ammonia emissions reduction is significantly more efficient when the emission reduction rises. Moreover, based on the evaluation on 2009, the modelling study shows that the expected impact of ammonia emissions on the formation of particulate ammonium was underestimated by all models. This would imply that the role of ammonia on PM concentration and exceedances of $PM_{2.5}$ and PM_{10} limit values is likely to be even larger than quantified in this study. This study shows that the implementation of the emission reductions imposed by the revised GP for 2020 will not suffice to achieve compliance with PM limit values everywhere in Europe; hence further European and local measures may be considered. NH_3 emissions from agriculture can be further reduced with the implementation of proven and feasible measures (substitution of fertilizers, improved storage of manure, way fertilizer injections, *etc.*, . . .), in order to reduce PM concentrations and their impacts on human health across Europe.

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1. Introduction

One of the most noxious problems in air quality is the persistence of high concentrations of particulate matter (PM) particularly in urban areas (EEA, 2012). In Europe, about one third of PM_{10} and half of $PM_{2.5}$ have an inorganic chemical speciation consisting of ammonium (NH_4^+), nitrate (NO_3^-) or sulphate (SO_4^{2-}). These species are the product of oxidation of PM precursor gases: ammonia (NH_3), nitrogen oxides (NO_x) and sulphur oxides (SO_x). Although NH_3 by itself makes a small fraction of the PM mass it plays a decisive role in PM formation chemistry by determining the amounts of ammonium sulphate ($(NH_4)_2SO_4$)) and ammonium nitrate (NH_4NO_3) as PM constituents. Historically, the sources of SO_x and NO_x were not too difficult to control, targeting in particular the industrial sector. Emissions of the precursor gases such as SO_x and NO_x declined by 54% and 26% in the period 2001–2010 while ammonia emissions have fallen less by 10% during this period. The agricultural sector was responsible for 94% of the total NH_3 emissions in the EU in 2010. Ammonia emissions are largely from animal excreta and fertilizers. Continued reductions in SO_x and NO_x emissions are likely to decrease in efficiency (and increase in costs) in terms of PM abatements, if they are not accompanied by relevant strategies on NH_3 emission treatments.

According to Erisman and Schaap (2004) inorganic PM concentrations can only be effectively reduced if all three precursor gases NO_x, SO_x and NH_x are reduced to the same extent. Since that study, the ''chemical landscape'' of Europe has changed; it is time to re-evaluate the potential for NH_3 emission reductions in the control strategy for PM concentrations. So far, the Gothenburg Protocol (Protocol to the 1979 Convention on Long Range Transboundary Air Pollution – LRTAP – to abate acidification, eutrophication and ground level ozone) and the European National Emission Ceilings Directive (NEC; EC, 2001) set emission reduction targets for NH_3 primarily with the aim of reducing acidification and eutrophication. Abatement of NH_3 emissions is also required by the Directive 2010/75/EU (EU, 2010) on industrial emissions, the Nitrates and Water Framework Directives respectively 91/676/EEC (EEC, 1991) and 2000/60/EC (EC, 2000), as well as EURO VI emission limits for heavy duty vehicles, becoming mandatory for all new registrations from 2014 (Regulation 595/2009/EC; EC, 2009).

The NH_3 emission reduction expected by the Gothenburg Protocol by 2020, compared to 2005 is lower than 10% in most of EU countries. The average reduction of ammonia emissions for the EU27 is 6%. Although there is less ambition in reducing NH_3 emissions compared to other PM precursors, there are proven and feasible methods to control and mitigate ammonia emissions from agriculture, including for the major sources of agricultural ammonia emissions (*e.g.* animal manure and urea fertilizer application). Furthermore and because of learning effects, the practical functioning of these techniques has been improved and their costs have declined. The available measures could reduce ammonia emissions in the EU27 by about 30% on top of current legislation in 2020 (Amman, 2012).

Several studies point out the importance of agricultural emissions to PM concentrations, and particularly of NH_3 emissions to $PM_{2.5}$ (Thunis et al., 2008; Erisman et al., 2008; Erisman and Schaap, 2004; Derwent et al., 2009; Harrison et al., 2013; Megariti et al., 2012). Erisman et al. (2008) estimate that NH_3 emissions from agriculture in EU15 give a substantial contribution to PM formation in Europe (13%). Harrison et al. (2013) and Derwent et al. (2009) give very similar results, 30% emission reduction of NH_3 over the whole Europe leads to the highest reduction in secondary inorganic aerosol (SIA) concentrations (9%) at a modelled site in England. Comparatively, a 30% reduction in NO_x and SO_2 emissions leads to a 5% and 6% reduction in SIA concentrations, respectively. In the EURO-DELTA II study, Thunis et al. (2008) showed however that the relative effectiveness of various emission reductions (NO_x, SO_2, NMVOC as Non Methanic Volatil Organic Compounds and NH_3) for $PM_{2.5}$ concentration reduction may vary considerably from model to model, justifying multi-model assessment of mitigation effectiveness.

In view of future negotiations of the revision of the NEC directive and to design strategies to deal with the recurrent PM exceedances in Europe, policy makers need to be informed about the options for reducing emissions of NH_3 and the other inorganic precursors which can lead to the desired reductions of inorganic PM concentrations. The objective of this study is to analyse the impact of the Gothenburg Protocol emission reductions and additional reductions in ammonia emissions, on European PM concentrations and on the exceedances of the air quality limit values (LV) for PM. To tackle this issue, an ensemble set of chemistry transport models (CTM) is performed to investigate several emission reduction scenarios using as reference the year 2009. The ensemble is constituted of: (i) the CHIMERE model (Menut et al., 2013) which is being used in France (Bessagnet et al., 2005) and Europe (EEA, 2013) for policy relevant scenario analysis and sensitivity studies, (ii) LOTOS-EUROS (Schaap et al., 2008; Sauter et al., 2012) used in support to Dutch authorities and (iii) the EMEP model (Simpson et al., 2012) which is widely used for policy support in Europe, especially in the framework of the CLRTAP. The use of several models results can provide valuable information on the uncertainty of the analysis. The calculations of PM concentrations including NH_4^+, NO_3^- and SO_4^{2-} are performed using the 2009 meteorology for the whole year. Emission inputs to chemical transport models consist of a European emission inventory with a spatial resolution of $0.125^\circ \times 0.0625^\circ$. The model predictions of speciated inorganic PM concentrations calculated in the baseline run (using current emissions) is compared with observations in order to assess their skills. Acronyms CHIM, EMEP and LOTO respectively refer to CHIMERE, EMEP, and LOTOS-EUROS in this study.

2. Emission scenarios

The reference scenario was based on official EMEP emissions for 2009 (update in 2012). Four alternative scenarios were built, one representing the Gothenburg 2020 protocol emissions and three scenarios which are expected to show the effect of an additional effort on ammonia emission reductions within the EU27. The Gothenburg protocol emission scenarios were built with the national emission ceiling figures reported in Annex II issued from UNECE (2012). Coarse primary PM emissions have