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Modelling agricultural ammonia emissions: impact on particulate matter formation

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Overview

In March and April 2007, the forecasts delivered in France by the chemical transport model (CTM) CHIMERE, within the operational platform for air quality monitoring and forecasting PREV'AIR, were significantly underestimating PM10 levels in the boundary layer during intense episodes. The origin of such event was first investigated analyzing observations of the PM10 speciation. It was found a lack of ammonium nitrate in the modeled aerosol composition. This was interpreted as a wrong estimate of ammonia emissions.

Agriculture is the main source of anthropogenic ammonia emissions. Ammonia reacts with sulfuric and nitric acids to form ammonium sulfate and nitrate aerosols. Under favourable meteorological conditions, ammonium nitrate can contribute to high PM2.5 thus to PM10 concentrations (Walker et al., 2004, Deutsch et al., 2008), which have been linked to a range of adverse health effects (Pope et al., 2009). The estimation of ammonia emissions is associated with large uncertainties and its availability is considered to be the most uncertain factor controlling the model performance in the calculation of secondary nitrate aerosol (Hass et al., 2003, Schaap et al., 2003, Schaap et al., 2004).

To improve the existent approach which is not fully satisfying in terms of model performances regarding PM10 in spring, a coupling process has been built between CHIMERE and a mechanistic VOLT'AIR model computing the NH₃ volatilization.

Methods

To calculate the ammonia emissions after N application on agricultural lands in France, a mechanistic model "VOLT'AIR" (Générumont and Cellier, 1997) is used offline with CHIMERE. It describes the action of environmental conditions and soil properties on ammonia volatilization. The calculation of ammonia fluxes is made over agricultural plot of land on an hourly basis depending on collocated input data. The NH₃ volatilization process is highly dependent on the meteorological conditions: air/soil temperature, air/soil humidity, wind speed and rainfall are provided to Volt'Air by the outputs of the meteorological mesoscale model WRF (Weather Research and Forecasting; <http://www.wrf-model.org>). The physicochemical properties of the soil (texture and pH) (INRA infosol; http://www.orleans.inra.fr/les_unites/us_infosol) are available at local scale and have been interpolated on the chosen grid scale (0.15° X 0.10°). Data about crop and grassland areas are available at cantonal level from the national agricultural census. Using a Geographic Information System (GIS), data are intersected with the corresponding land cover types of the Corine Land Cover database (*UE- SOeS, CLC 2006*) and spatialized on a regular grid. Fertilizer types considered are ammonium nitrates, nitrogen solutions and urea. They constitute the bulk of the synthetic fertilizers used in France (UNIFA, 2007). Fertilization rates and dates of spreading are available at regional scales.

The chemistry-transport model CHIMERE is used to simulate the observed PM concentrations for France. The meteorological input fields are computed using the WRF mesoscale model. Boundary conditions are provided by a large-scale simulation covering Western Europe with a 0.5° horizontal resolution. We carried out two simulations. The first is the reference simulation using emissions provided by EMEP inventory (<http://www.emep.int/>). The second simulation is the same as the first one but ammonia emissions are calculated as the sum of the NH₃ emissions due to fertilization (VOLT'AIR), animal husbandry and manure spreading. These two last NH₃ emissions are provided by

the National Spatialized Inventory (NSI) for France. For the rest of the domain, EMEP NH₃ emissions are used. The CHIMERE-VOLT'AIR runs were carried out on the period February-April 2007 over France with a resolution of 0.15° X 0.10° (~ 10 km).

Results

Ammonia emissions: The emissions from fertilizers calculated by Volt'air model present a significant spatiotemporal variation. The spatial variation of the emissions depends especially on soil pH and the agricultural practices (type and amount of fertilizers). The temporal variation depends on the soil temperature (diurnal variations of the emissions) and on the dates of the fertilization varying with different regions for different crops.

Ammonia concentrations: The ammonia concentrations are calculated with CHIMERE using the corresponding emissions. They vary between 0 and ~ 80 ppb when the Volt'air plus NSI emissions are used and between 0 and ~ 50 ppb when EMEP emissions are used. The results show significant differences in the spatial variations using the two methods (*fig. 1.a*).

PM10 concentrations: The PM10 concentrations are calculated with CHIMERE CTM using ammonia emissions provided by Volt'air + NSI and using those provided by EMEP. The differences are significant in some regions. In the north, north-east, centre and south-west of France, the PM10 mean levels calculated using the new method are the highest. The maximum of this difference is about 4 μg.m⁻³ (*fig. 1.b*) on the total spring period and the increase of the PM10 mean levels varying from 0 to 16 %. For the episode of PM10 pollution of April 17, the daily mean difference can reach locally 15 μg.m⁻³ (*fig. 1.c*) using the new emission method and the difference can range from 0 to 30 % of PM10 mean concentrations. These results confirm the impact of high NH₃ emissions on PM formation at favourable meteorological conditions and thus the importance of ammonia modelling in CTMs. The comparison of PM concentrations with observations will be shown.

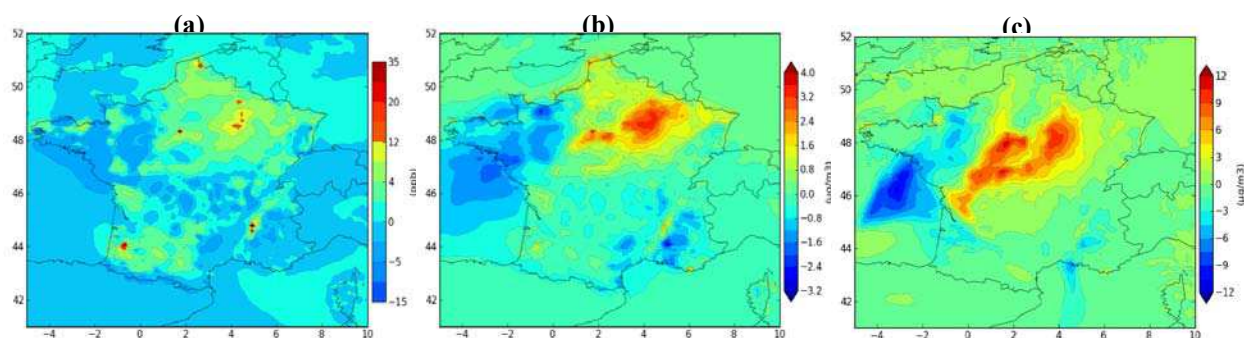


Fig.1. The difference of NH₃ [ppbv] (a) and PM10 [μg.m⁻³] mean concentrations on the period February-April (b), and on April 17 2007 (c) between Voltair_NSI method and EMEP method of NH₃ emissions implemented in CHIMERE CTM.

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