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SENSITIVITY OF AMMONIA AND PARTICULATE NITRATE IN EUROPE TO DRY DEPOSITION PARAMETERIZATION

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Abstract: Chemical transport models use different parameterizations for deposition processes. In the previous versions of the air quality model CAMx, the scaling parameter for surface resistance in dry deposition velocity calculations was set to zero for ammonia, leading to a fast deposition. In order to estimate ammonia removal rates more appropriately, in the new CAMx version 6.50, the scaling parameter was activated to include surface resistance in the deposition velocity calculations. The tests with the new parameterization in the United States showed that ammonia concentrations increased significantly leading to a better model performance. In this study, we simulated the European air quality in 2010 to investigate the effect of changing surface resistance parameter on the concentrations of ammonia, particulate nitrate, ammonium, sulfate and nitric acid. The results showed a significant increase in ammonia (30-60%) in central Europe, leading to a better model performance at most of the measurement sites. As a result of increased ammonia levels, nitric acid concentrations decreased by about 20-50% while nitrate and ammonium increased by 20 and 10%, respectively. The increase in sulphate was less than 10%.

***Keywords**: dry deposition, ammonia, particulate nitrate, CAMx, Europe*

INTRODUCTION

Ammonia is an important precursor of particulate nitrate and sulphate and it is a threat to health and environment (Erisman et al., 2008; Behera et al., 2013). In Europe, NO_x, SO_2 and VOC emissions have decreased significantly over the last few decades while ammonia emissions remained constant or even increased (EEA, 2017). It is therefore very important to simulate the fate of ammonia in the atmosphere correctly. Modelling ammonia however, is one of the challenges in air quality modelling studies because of high uncertainties in its emissions as well as its rapid deposition (Simpson et al., 2011). Ammonia is efficiently dry deposited close to the source areas. Dry deposition is an important loss process and can be a significant part of total deposition estimates calculated for critical loads.

Air quality models use different parameterizations for dry deposition processes which have high uncertainties (Saylor et al., 2019). In an earlier study in the United States, the model performance of CAMx for ammonia was found to be very poor with large negative biases and the dry deposition was identified as the largest ammonia sink due to too high deposition velocities (Rodriguez et al., 2011). Several sensitivity simulations with modified dry deposition velocities improved the model performance for ammonia slightly. Nopmongcol et al. (2018) tested the bidirectional NH_3 dry deposition scheme in CAMx over a domain covering the United States and found increases in NH_3, NO_3^- and NH_4^+, as expected.

CAMx currently employs unidirectional dry deposition schemes. In order to estimate ammonia removal rates more appropriately, the scaling factor for ammonia was modified in the latest CAMx version 6.50, enabling the surface resistance for dry deposition velocity calculations. In this study, we investigated how the recent change in dry deposition parameterization affects the model results for ammonia and relevant aerosol species in Europe.

METHOD

In this study we used the air quality model CAMx v6.50 (http://www.camx.com) on a domain covering Europe with a horizontal resolution of 0.250° x 0.125°. The meteorological parameters were generated using the meteorological model WRF (v3.7.1) (Skamarock et al., 2008). There were 14 layers and the first layer was 20 m thick. The gas-phase mechanism used in this study was CB6r2 (Hildebrandt Ruiz and Yarwood, 2013). We simulated the particle concentrations using CAMx's fine/coarse options and calculated the organic aerosol concentrations using the SOAP model (Strader et al., 1999). CAMx uses the ISORROPIA (Nenes et al., 1998, 1999) model for inorganic thermodynamics and gas–aerosol partitioning. Anthropogenic emissions were based on TNO-MACC-III European emission inventory for 2010 (Kuenen et al., 2014) and biogenic emissions were calculated by the PSI-Model (Jiang et al., 2019). The initial and boundary conditions for the chemical species were obtained from the Model of Ozone and Related Chemical Tracers (MOZART) global model data for 2010 with a time resolution of 6 h (Horowitz et al., 2003).

We selected the dry deposition scheme based on the algorithms of Zhang et al. (2003). The two simulations for 2010 were performed with CAMx version 6.50: i) using the new scaling factor for NH_3 dry deposition (Rscale=1), ii) using the scaling factor as in the previous CAMx versions (Rscale=0). All the other parameterizations were kept the same for both simulations.

RESULTS AND DISCUSSION

The modelled annual concentrations of ammonia and particulate nitrate using the new dry deposition parameters are shown in Figure 1. The highest ammonia concentrations were predicted in regions close to the emission sources such as Benelux area and northern Italy. Concentrations of particulate nitrate which is formed from gaseous ammonia and nitric acid were higher over a larger area in central Europe and along the coastal areas where there are high NO_x emissions from ships and ammonia emissions from land.

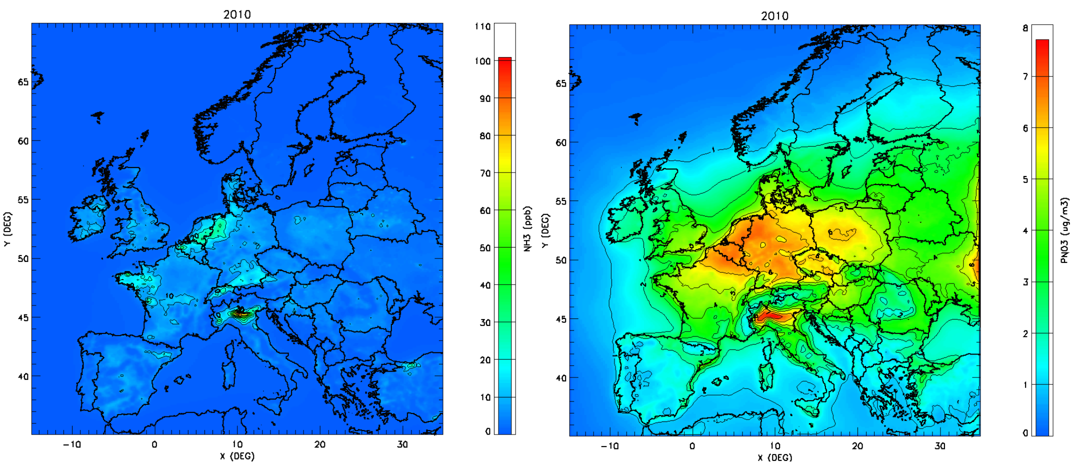

Figure 1. Annual NH_3 (left, in ppb) and particulate NO_3^- concentrations (right, in µg m^{-3}) in 2010 modelled by CAMx 6.50 using the new dry deposition parameterization.

The results of the tests were analyzed as annual differences between the two simulations. The new scaling parameter which enabled the surface resistance for deposition velocity calculations of ammonia, led to a decrease in deposition and an increase in concentration, as expected (Figure 2, left panel). The increase in ammonia concentrations was between 30 and 50% over central Europe. On the other hand, particulate nitrate and ammonium concentrations increased over the land as well as along the coastal regions (Figure 3) by about 20 and 10%, respectively while HNO_3 concentrations decreased by about 20-30% (Figure 3, right panel) indicating that with more ammonia available, more HNO_3 was consumed to form ammonium nitrate. The increase in sulphate particles was less than 10% (not shown).

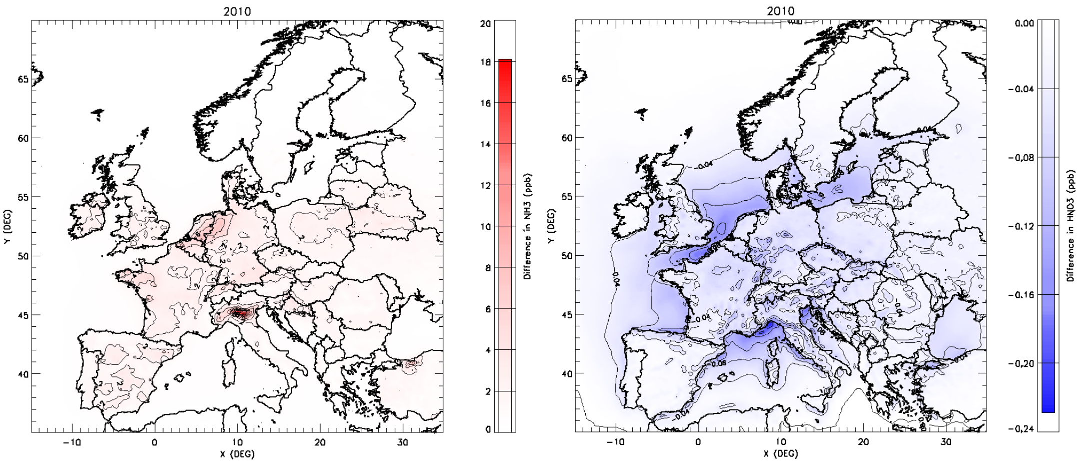

Figure 2. Change in annual concentrations (ppb) of NH_3 (left) and HNO_3 (right) due to the new dry deposition parameterization.

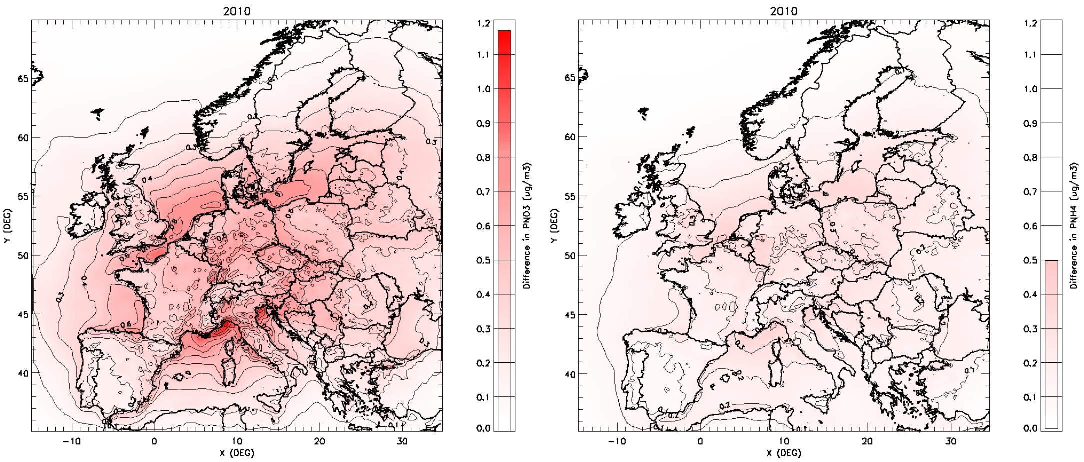

Figure 3. Change in annual concentrations (μg m^{-3}) of NO_3^- (left) and NH_4^+ (right) due to the new dry deposition parameterization.

The modelled ammonia concentrations from both simulations using old and new parameterization were compared with measurements at 13 stations extracted from the European air quality database (AirBase v7) (Mol and Leeuw, 2005). These comparisons suggested that the model performance for ammonia concentrations became better at most of the measurement sites (4 in the Netherlands, 2 in Italy, 2 in Bulgaria and 1 in Croatia) where ammonia was underestimated previously (Figure 4). At the remaining 4 sites in the Netherlands on the other hand, overestimation of ammonia with the old parameterization was enhanced with the new parameterization. The fact that the measurement sites in the Netherlands are very close to each other might be a problem for the model domain resolution.

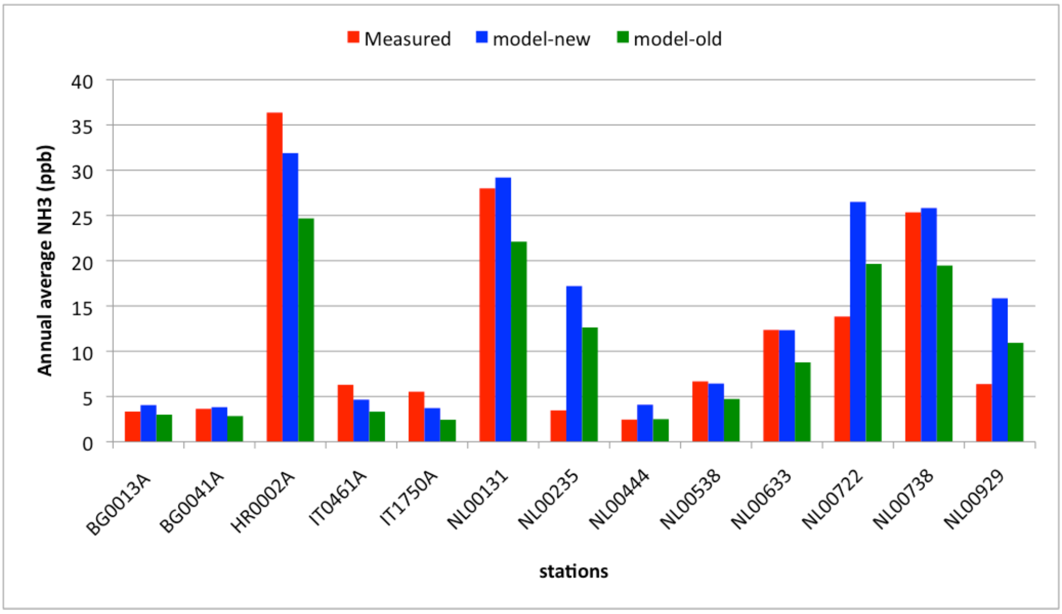

Figure 4. Comparison of modelled annual NH_3 concentrations using the old (green) and new (blue) dry deposition parameterization with measurements (red) at 13 stations in Europe (BG: Bulgaria, HR: Croatia, IT: Italy, NL: The Netherlands).

CONCLUSIONS

In this study, the effects of a change in the dry deposition parameter for ammonia in the latest CAMx model version (6.50) were investigated over the European domain in 2010. The modification of the scaling parameter for surface resistance in dry deposition velocity calculations led to an increase in gaseous ammonia concentrations by about 30-60%, leading to a better model performance at most of the measurement sites where ammonia was previously underestimated. At a few locations overestimation of ammonia was enhanced. As a result of the increased ammonia, particulate NO_3^- and NH_4^+ concentrations increased as well by 20 and 10%, respectively while gaseous HNO_3 concentrations decreased by 20-50% over Europe. On the other hand, the increase in SO_4^{2-} particles was less than 10%. These tests suggested that the new parameterization of ammonia dry deposition in CAMx v6.50 improved the model performance at most of the European sites. The implementation of bidirectional ammonia dry deposition scheme in the next CAMx version is expected to lead to a further improvement in ammonia modelling.

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