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MODELING NITROGEN DEPOSITION: SEASONAL VARIATION OF DRY DEPOSITION VELOCITIES ON VARIOUS LAND-USE TYPES IN SWITZERLAND

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Abstract: Nitrogen deposition in Switzerland was analyzed in an air quality modeling study for 2006. The modeled annual nitrogen deposition was 12.2 kg N ha^{-1} a^{-1}, dominated by the deposition of reduced nitrogen compounds (74%). The model results suggest that the largest contribution to nitrogen deposition in Switzerland is from dry deposition of ammonia. Dry deposition velocities of oxidized and reduced nitrogen compounds were calculated for specific land-use types found in Switzerland. The highest deposition velocity for ammonia (4.6 cm s^{-1}) and nitric acid (3.4 cm s^{-1}) was estimated over evergreen shrubs followed by evergreen needleleaf and deciduous broadleaf forests whereas the deposition was slowest over water surfaces (0.8-0.9 cm s^{-1}). Deposition velocities over various land-use types were shown to vary seasonally and the highest values were calculated for spring and summer except for water.

Key words: *nitrogen, deposition velocity, land-use, Switzerland.*

INTRODUCTION

Nitrogen (N) is an essential nutrient for plant growth but excess N deposition has adverse effects such as acidification, eutrophication, and toxicity to plants (Jones et al., 2014). Excess N deposition might also lead to the loss of plant diversity (Roth et al., 2015). Nitrogen oxide (NO) is emitted into the atmosphere by both natural sources like lightning and soil and by human activities related to combustion processes and then it is converted to other oxides of nitrogen. The reduced N compounds, primarily ammonia (NH_3), derive mainly from agricultural activities and after reacting with acids in the atmosphere, it forms particulate ammonium (NH_4^+). Modeling studies indicate that the dry deposition of reduced N (NH_3, NH_4^+) is the most important contributor to N deposition in central Europe (Dentener et al., 2006). The decline in N deposition in the past in Europe was shown to be mainly related to the oxidized fraction due to large reductions in NO_x ($NO+NO_2$) emissions in the past; the deposition of reduced N, however, was predicted to increase further until 2020 (Aksoyoglu et al., 2014). Dry deposition is estimated in models through deposition velocity which is a function of pollutant species, nature of the surface and meteorological conditions. In this study, we report the land-use specific deposition velocity of oxidized and reduced nitrogen compounds and their seasonal variability in Switzerland using the CAMx model.

METHOD

In this study, we used the output of an earlier model simulation for 2006 using the regional air quality model CAMx (comprehensive air quality model with extensions), version 5.40 (http://www.camx.com) with CB05 chemical mechanism (Aksoyoglu et al., 2014). The coarse model domain covered Europe with a horizontal resolution of 0.250° x 0.125° whereas the nested domain covered Switzerland with a higher resolution (0.083° x 0.042°). The meteorological parameters were calculated by the Weather Research and Forecasting Model (WRF-ARW), version 3.2.1 (http://wrf-model.org/index.php) initialized by the ECMWF data (http://www.ecmwf.int/). There were 14 layers in CAMx and the thickness of the first layer was about 20 m. We used the TNO-MACC emission inventory for the anthropogenic emissions in Europe (Denier van der Gon et al., 2010) while biogenic emissions were calculated with our own model (Andreani-Aksoyoglu and Keller, 1995). More details about the model parameterization can be found in Aksoyoglu et al. (2014). We selected the resistance model of Zhang et al. (2003) to calculate the dry deposition. For a given species, particle size and grid cell, CAMx determines a deposition velocity for each land-use type in that grid cell and then linearly combines them according to the fractional

distribution of land-use classes. In this study, using the hourly deposition output of nitrogen species per each grid cell, we extracted the dry deposition velocities over the land-use types found in Switzerland.

RESULTS AND DISCUSSION

Agriculture contributes about 93-95% to the total ammonia emissions in Switzerland (Kupper et al., 2015). The modeled annual ammonia concentrations are shown in Fig. 1 (left panel). Comparison of modeled annual NH_3 concentrations with measurements at 27 sites in Switzerland suggests that the model could reproduce annual ammonia concentrations reasonably well in the Swiss Plateau (Fig.1, right panel). Especially the highest levels at sites with intensive cattle farming were captured very well.

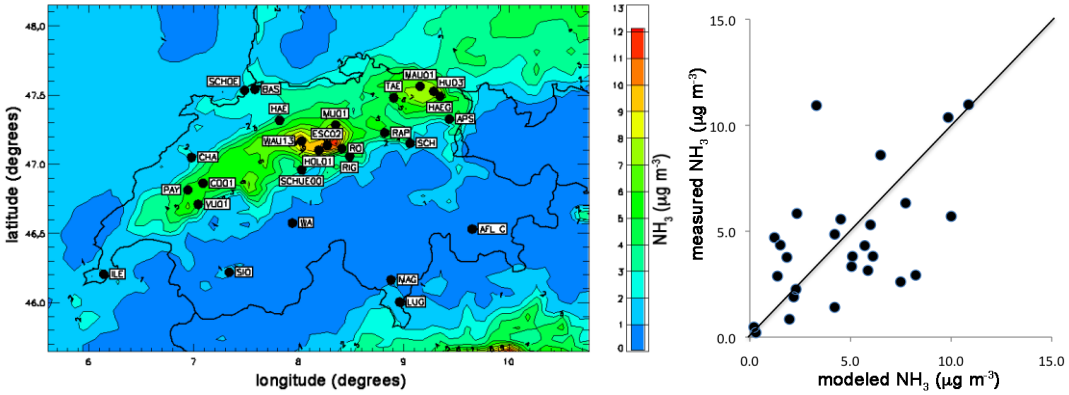


Figure 1. The modeled annual NH_3 concentrations in 2006 (left), measured versus modeled annual NH_3 concentrations (right). Measurements at 27 sites (shown in the left panel) are from FUB.

Analysis of long-term (10 years) measurement data indicated that the seasonal variation of ammonia emissions might vary depending on the meteorological conditions prevailing each year (Thoni and Seitler, 2013). Data showed also that the lowest ammonia emissions in Switzerland occur in winter, highest in spring, and there are often some peaks in summer and fall. In 2006, however, the highest emissions were in summer and emissions in spring were unusually low. This leads to an overestimation of total ammonia (NH_3 + NH_4^+) in spring (Fig. 2) since temporal profiles used in the emission inventory for agricultural emissions are based on the common seasonal variations observed in Europe. Measurements and model results agree quite well with each other in other seasons.

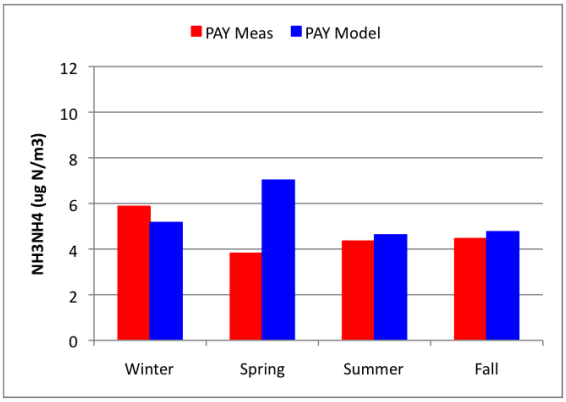


Figure 2. Seasonal variation of measured (red) and modeled (blue) total ammonia at Payerne in 2006.

The average total nitrogen deposition was predicted to be 12.2 kg N ha^{-1} a^{-1} with highest values found in central Switzerland where NH_3 emissions are the highest (Fig. 3, left panel). Deposition of reduced N compounds (blue colors in Fig. 3, right panel) was about 74% of the total nitrogen deposition with dry NH_3 deposition being the dominant fraction. The wet N deposition was underestimated by -30% and -55%, for reduced and oxidized N compounds, respectively, especially in the south of the Alps.

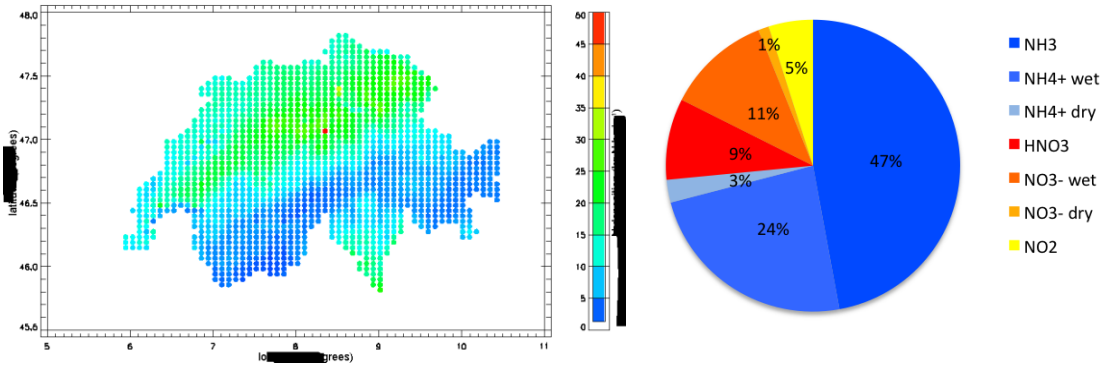


Figure 3. Modeled N deposition in 2006 (kg N ha^{-1} a^{-1}) in Switzerland (left) and relative contribution of oxidized (red, orange, yellow) and reduced (blue tones) components to dry (65%) and wet (35%) deposition (right). The average N deposition is 12.2 kg N ha^{-1} a^{-1} of which 74% reduced components and 26% are oxidized components.

The annual dry deposition velocity of gaseous ammonia (NH_3) was predicted to be the highest in Swiss domain, followed by HNO_3 (Fig. 4). On the other hand, deposition velocities of NO_2 and particulate species (NH_4^+ and NO_3^-) were much lower (not shown). Among the 26 land use categories used in model simulations, deposition velocities of species were determined for 9 land-use types relevant for Switzerland. The fractional distributions of these land-use types in the grid cells as well as the annual average deposition velocities of HNO_3 and NH_3 over these land-use types are shown in Fig. 5. Mixed and evergreen needleleaf forests are the most abundant land-use types while other land-use types are more local, i.e. tundra in the Alpine regions, crops mainly in the Swiss Plateau. The highest annual deposition velocities were predicted for evergreen shrubs (4.6 and 3.4 cm s^{-1}, for NH_3 and HNO_3, respectively) as well as deciduous broadleaf and evergreen needleleaf forests whereas the lowest deposition velocities were predicted over water surfaces (0.9 and 0.8 cm s^{-1} for NH_3 and HNO_3, respectively). In a review paper by Schrader and Brümmer (2014), the following ranges were reported for NH_3 deposition velocities in cm s^{-1}: for water 0.5-0.9, coniferous forests 0.5-3.3, deciduous forests 0.3-1.8, mixed forests 0.4-3.0, urban 0.1-1.1, agricultural land 0.2-7.1. These numbers however, are based on various measurements and models carried out at different times of the day, seasons and regions. It is therefore difficult to make a direct comparison.

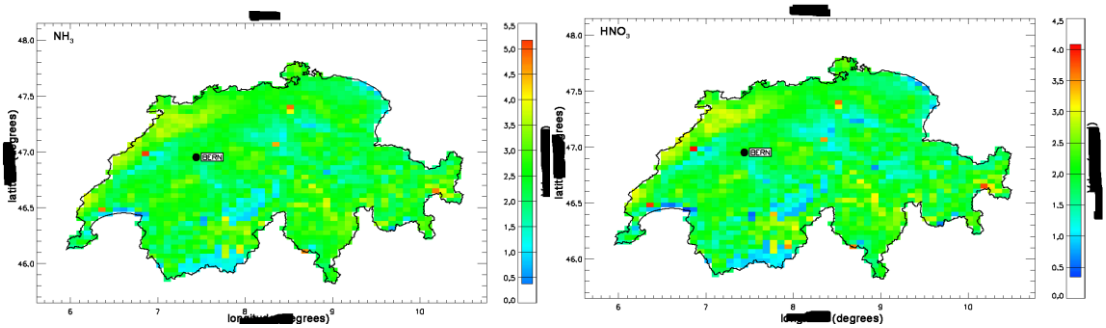
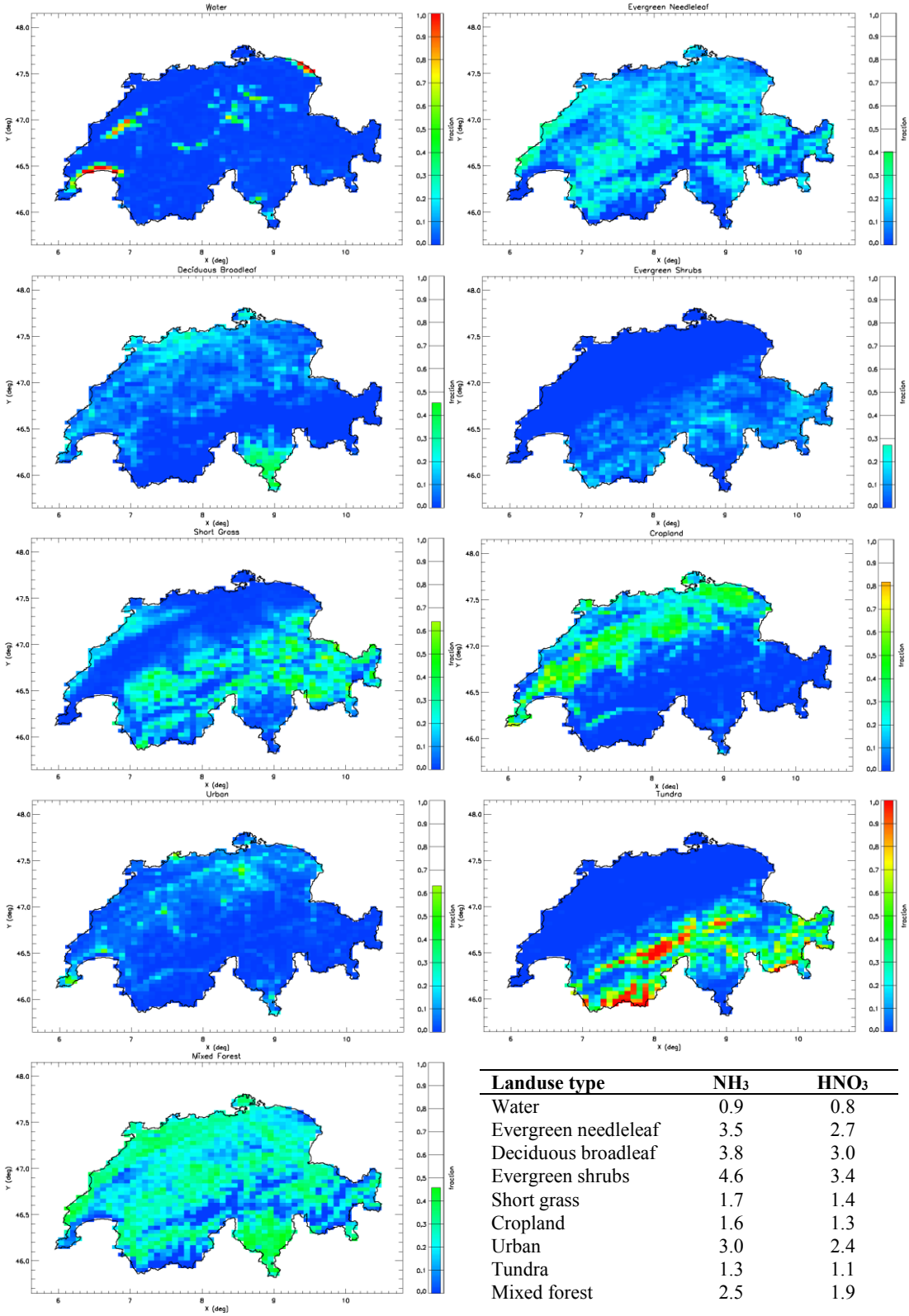


Figure 4. Spatial distribution of annual average dry deposition velocity (cm s^{-1}) of NH_3 (left) and HNO_3 (left).



Landuse type	NH_3	HNO_3
Water	0.9	0.8
Evergreen needleleaf	3.5	2.7
Deciduous broadleaf	3.8	3.0
Evergreen shrubs	4.6	3.4
Short grass	1.7	1.4
Cropland	1.6	1.3
Urban	3.0	2.4
Tundra	1.3	1.1
Mixed forest	2.5	1.9

Figure 5: Fractional distribution of land-use types in grid cells of the Swiss domain and annual dry deposition velocities (cm s^{-1}) of NH_3 and HNO_3 on these land-use types (Table).

As shown in Fig. 6, deposition velocities vary seasonally and the highest values were calculated for spring and summer except over water surfaces. Over evergreen shrubs, the highest deposition velocities of ammonia and nitric acid were in summer (7.1 and 5.4 cm s^{-1}, respectively) while over the forests (coniferous, deciduous, mixed) there was no significant difference between spring and summer.

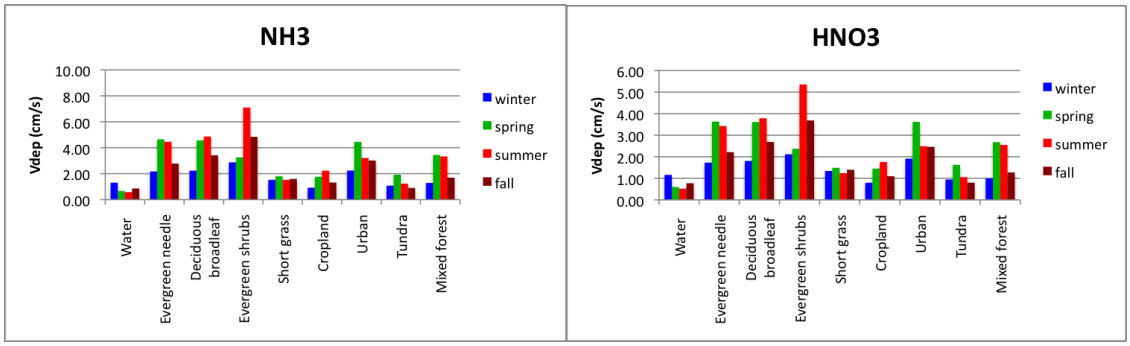


Figure 6. Seasonal variation of dry deposition velocity (cm s^{-1}) for NH_3 (left) and HNO_3 (right) in 2006.

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REFERENCES

Andreani-Aksoyoglu, S., and Keller, J., 1995: Estimates of monoterpene and isoprene emissions from the forests in Switzerland, Journal of Atmospheric Chemistry, 20, 71-87.

Aksoyoglu, S., Keller, J., Barmpadimos, I., Oderbolz, D., Lanz, V. A., Prévôt, A. S. H., and Baltensperger, U., 2011: Aerosol modelling in Europe with a focus on Switzerland during summer and winter episodes, Atmos. Chem. Phys., 11 doi:10.5194/acp-5111-7355-2011, 7355-7373.

Aksoyoglu, S., Keller, J., Ciarelli, G., Prévôt, A. S. H., and Baltensperger, U., 2014: A model study on changes of European and Swiss particulate matter, ozone and nitrogen deposition between 1990 and 2020 due to the revised Gothenburg protocol, Atmos. Chem. Phys., 14, 13081-13095, 10.5194/acp-14-13081-2014.

Denier van der Gon, H., Visschedijk, A., van de Brugh, H., and Droege, R., 2010: A high resolution European emission data base for the year 2005. A contribution to UBA-Projekt: "Strategien zur Verminderung der Feinstaubbelastung" – PAREST: Partikelreduktionsstrategien –Particle Reduction Strategies TNO, Utrecht (NL)TNO-034-UT-2010-01895_RPT-ML.

Dentener, F. et al., 2006: Nitrogen and sulfur deposition on regional and global scales: A multimodel evaluation, Global Biogeochemical Cycles, 20, n/a-n/a, 10.1029/2005GB002672.

Jones, L., Provins, A., Holland, M., Mills, G., Hayes, F., Emmett, B., Hall, J., Sheppard, L., Smith, R., Sutton, M., Hicks, K., Ashmore, M., Haines-Young, R., and Harper-Simmonds, L., 2014: A review and application of the evidence for nitrogen impacts on ecosystem services, Ecosystem Services, 7, 76-88, http://dx.doi.org/10.1016/j.ecoser.2013.09.001.

Kupper, T., Bonjour, C., and Menzi, H., 2015: Evolution of farm and manure management and their influence on ammonia emissions from agriculture in Switzerland between 1990 and 2010, Atmos. Environ., 103, 215-221, http://dx.doi.org/10.1016/j.atmosenv.2014.12.024.

Roth, T., Kohli, L., Rihm, B., Amrhein, V., and Achermann, B., 2015: Nitrogen deposition and multi-dimensional plant diversity at the landscape scale, Royal Society Open Science, 2, 10.1098/rsos.150017.

Schrader, F. and Brümmer, C., 2014: Land use specific ammonia deposition velocities: a review of recent studies (2004-2013), Water Air Soil Pollut., 225:2114

Thoeni, L., and Seitler, E., 2013: Ammoniak-Immissionsmessungen in der Schweiz 2000 bis 2012, Forschungsstelle fuer Umweltbeobachtung, FUB Report, Rapperswil.

Zhang, L., Brook, J. R., and Vet, R., 2003: A revised parameterization for gaseous dry deposition in air-quality models, Atmos. Chem. Phys., 3 2067-2082, 10.5194/acp-3-2067-2003.