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MODEL EVALUATION WITH RESPECT TO DEPOSITION PROCESSES

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Abstract: The Chemistry Transport Model REM_Calgrid (RCG) has been used to simulate air pollution concentrations and depositions. The fluxes have been evaluated using dry deposition measurements from Augustendorf and wet deposition fluxes from UBA over Germany. Meteorological input fields were provided by COSMO-EU of the German Weather Service (DWD). COSMO-EU friction velocities have been compared to turbulence measurements at the Lindenberg site. Simulated cloud liquid water content has been evaluated using Cloudnet data. Since UBA wet deposition measurement stations also provide precipitation amounts modelled precipitation has been compared to these independent observations. Friction velocities are simulated well with correlation coefficients over 0.8 and reproducing correctly the landscape. Resultant dry deposition fluxes over a forest site were reasonable for most nitrogen species. COSMO-EU precipitation is in sound agreement with most UBA stations. Simulated cloud liquid water content underestimates Cloudnet measurements. Modelled NHx wet depositions are less affected by this underestimation than SOx and NOy wet depositions since NHx is less mixed up. Hence, RCG NHx wet depositions compare very well to UBA observations while SOx and NOy wet depositions are underestimated.

Key words: Atmospheric dispersions, pollutants deposition, model evaluation.

INTRODUCTION

Air Pollution model evaluation needs to consider not only the final air pollution concentration fields comparing the simulated species with airborne measurements, but must validate all evaluable processes within the air pollution model in order to obtain reliable predictions. Deposition processes are of paramount importance in the air pollution budget acting as one of the fundamental parts in the accumulation/removal equilibrium. Furthermore, deposited pollutants damage ecosystems eutrophising and acidifying soils, reduce crop yields and corrode cultural monuments. Considered pathways of pollutants from the air towards the ground are wet deposition processes where pollutants are incorporated in cloud or rain droplets in or below a cloud and scavenged, and dry deposition processes where gaseous and solid pollutants are assimilated by the ground. While wet deposition is easily measured by collecting precipitation and analysing its chemical contents, dry deposition measurements are difficult to realise due to the variety of different surface land types and due to the different mechanisms the pollutant is absorbed by ecosystems. Thus, often chemistry transport models' ability to predict dry deposited fluxes correctly is based on model-to-model inter-comparisons, only. In this paper, independent dry deposition measurements obtained with the covariance eddy technique for nitrogen species in a forest in the North-Western part of Germany have been compared to an ordinary output of the Aerosol-Chemistry-Transport-Model RCG, while wet deposition fluxes simulations were compared to rain water pollutant concentration observations. The aim of this validation is to validate the contributing processes to modelled dry and wet deposition fluxes, i.e. the airborne transport of the pollutant from the atmosphere towards the ground checking turbulence parameters, airborne concentrations and final dry deposition fluxes, separately. For modelled wet deposition flux validation, precipitation and cloud liquid water content have been analysed next to the final scavenged pollutant flux. As measurements have not been available for all processes contemporaneously, single processes have been compared to available data also at different times and different locations.

METHODS AND DATA

The off-line Eulerian grid model RCG simulates air pollution concentrations solving the advection-diffusion equation on a regular lat-lon-grid with a horizontal resolution of approximately 7x7 km² and up to 5000 m height with 20 vertical layers. A comprehensive model description is given in Beekmann *et al.*, 2007. Emissions for Germany were delivered from local and national inventories, while European emissions are based on EMEP data post-processed at TNO (Klotz *et al.*, 2009). Meteorological fields are provided by the COSMO-EU (Doms *et al.* 2007) analyses from German Weather Service (DWD), while mixing heights and friction velocities were post-processed following the approach of Fay *et al.* (1997). RCG was evaluated within many urban and regional applications and within the framework of several European model inter-comparison studies (e.g. Stern *et al.*, 2008 and references therein)

Dry deposition

Dry deposition velocity is parameterised in RCG following the resistance approach proposed by Erisman *et al.* (1994). The atmospheric resistance R_a and the sublayer resistance R_b , explicitly written in equations (1), are driven by the friction velocity u_* and the atmospheric stability Ψ_m , which is parameterised with the Monin-Obukhov-Length (L).

$$R_a = \frac{1}{\kappa u_*} \ln \frac{z-d}{z_0} - \Psi_m \left(\frac{z}{L} \right) \quad R_b = \frac{2}{\kappa u_*} \left(\frac{Sc}{Pr} \right)^{2/3} \quad (1)$$

The viscous sub-layer resistance for gases is dependent also on the pollutant related molecular diffusivities incorporated in the Schmidt number (Sc). Pr is the Prandtl-number and takes the value 0.72, and κ is the von Kármán-constant. The canopy resistance for gases depends largely on the surface humidity and on plant physiological parameters, and is not evaluated in this study, separately. Friction velocity u_* is one of the most prominent parameters in simulating dry deposition processes. It has been calculated from the COSMO-EU-first-level z_1 wind speed $u(z_1)$ and the turbulent momentum transfer coefficient t_{cm} . (see equation (2))

$$u_* = t_{cm} u(z_1) \quad (2)$$

Microphysical meteorological measurements are seldom and difficult to carry out over a long period. Therefore, within the “Global Energy and Water Cycle Experiment“ (GEWEX) a “Coordinated Enhanced Observing Period” (CEOP) has been initiated to obtain long-lasting measurements. At Lindenberg in the South-East of Berlin, DWD has been performing turbulence measurements for more than 10 years at two different sites, one at a meadow and a second 10 km away in a forest. A 99 m tower at the meadow-site and 28.30 m measurement tower at the forest site where equipped with meteorological measurement devices at different levels. At both sites sampling times were for temperature, humidity, wind speed and direction one second. (Beyrich and Adam, 2007). Turbulent momentum fluxes were determined from the high resolution measurements of the three wind components by computing mean eddy covariances and used to compute the friction velocity following the relation in equation (3):

$$u^* = \left(\overline{u'w'}^2 + \overline{v'w'}^2 \right)^{1/4} \quad (3)$$

The nitrogen species deposition measurements at the forest site in Augustendorf in the North-Western plain land of Germany, have been derived using the micrometeorological method described in Dämmgen *et al.* (2005) which uses the eddy covariance assumption coded in the PLATIN-model (Grünhage *et al.*, 2008). The model calculates the exchange of trace gases and fine-particle constituents. The vertical transport between an above-canopy reference height, for which air properties and concentrations of matter must be known, and the sinks and/or sources of the plant/soil-surface system is estimated. The air pollution concentrations were measured using in series denuder tubes (Dämmgen *et al.*, 2005) for gaseous NH₃, HNO₂, HNO₃, SO₂ and HCL and for particles NH₄-N, NO₃-N, SO₄-S, Cl and Na. Wind speed and direction, air temperature and humidity were measured at 25 m as well as at 22 m above ground, with an average sampling period of 15 minutes (Dämmgen *et al.*, 2005).

Wet deposition

The RCG wet deposition scheme has been improved recently following the approach of Seinfeld and Pandis, 1998. The approach describes the physical processes in detail and distinguishes between in-cloud and below-cloud scavenging. Moreover, the in-cloud scavenging coefficient is dependant on the cloud liquid water content. The gas in-cloud scavenging coefficient $\lambda_{icg} = \lambda_{aq} + \lambda_g$ consists of a factor for the aqueous phase scavenging λ_{aq} (equation (4)) and a factor for scavenging of ambient gases λ_g (equations (5) and (6)).

$$\lambda_{aq} = \frac{4.2 \cdot 10^{-7} \cdot E_g \cdot P \cdot H \cdot c_g \cdot cwc}{d_d \cdot c \cdot \rho_w} \quad (4)$$

For reversibly soluble gases equation (5) is taken:

$$\lambda_g = \frac{2.8 \cdot 10^{-7} \cdot P \cdot H \cdot c_g}{c \cdot \Delta z} \cdot (1 - \exp(-\frac{6K_c \Delta z}{d_d v_d H})) \quad (5)$$

and for irreversibly soluble gases equation (6) is used:

$$\lambda_g = 1.67 \cdot 10^{-6} \frac{K_c P}{d_d v_d} \quad (6)$$

λ_g and λ_{aq} are the gas-phase and aqueous-phase scavenging coefficients, respectively, P is the precipitation rate, H the Henry constant, c_g the gas concentration, cwc the cloud water content, d_d the drop diameter, ρ_w the water density, K_c the mass transfer coefficient, Δz the layer depth, and v_d the mean drop fall speed.

Since below the cloud the ambient gas is subject to scavenging the below-cloud scavenging coefficient λ_{bcg} is equal λ_g . As the change in gas concentration Δc is relaxed toward the difference between the maximum possible gas in solution for the given conditions c_{eq} and the amount of pre-existing gas in solution from layers above c_0 (equation (7), see CAMx, 2010):

$$\Delta c = (c_{eq} - c_0)(1 - \exp(-\lambda \Delta c)) \quad (7)$$

where c_{eq} is the maximum possible gas concentration and c_0 the pre-existing gas in solution.

Within cloud layers all aerosols are assumed to exist within the cloud water. Thus the particle in-cloud scavenging coefficient λ_{icp} is (equation (8))

$$\lambda_{icp} = \frac{4.2 \cdot 10^{-7} \cdot E_g \cdot P}{d_d} \quad (8)$$

with E_g the collection efficiency for gases. The particle below-cloud scavenging coefficient λ_{bcp} is expressed in equation (9).

$$\lambda_{bcp} = \frac{4.2 \cdot 10^{-7} \cdot E_p \cdot P}{d_d} \quad (9)$$

with E_p the collection efficiency for particles. E_p is a function of d_p and is described in Seinfeld and Pandis, 1998. COSMO-EU meteorological fields of cloud liquid water content and precipitation have been compared to measurements. For the evaluation of the COSMO-EU model cloud liquid water content Cloudnet liquid water content (LWC) measurements (Illingworth *et al.*, 2007) at the Lindenberg site were applied. LWC is derived by using radar and lidar cloud boundaries followed by using dual-wavelength microwave radiometers to scale the liquid water content values to yield the correct liquid water path. During rainfall no LWC measurements are available because the cloud extent is difficult to ascertain and liquid water path is uncertain. For evaluation of COSMO-EU precipitation and RCG wet deposition fluxes UBA (Umweltbundesamt, German Federal Ministry for Environment) station measurements (UBA, 2004) over Germany were used.

RESULTS AND DISCUSSION

Dry deposition

Figure 1 shows the hourly performance of the friction velocity simulations, derived from the dynamic model COSMO-EU analyses and post-processed following equation 2 (y-axis) compared to the two measurements sites in Lindenberg (x-axis), one over the meadow (left panel) and the other in the forest (right panel). Taking into account that COSMO-EU is a Eulerian-Grid-Model with a resolution of approximately 7 km and thus comprising more than one different land-use-type in one cell, friction velocities are well reproduced temporally with correlation coefficients around 0.8 for both sites and also spatially being slightly overestimated compared to the meadow site (slope 1.3, intercept 0.1 m/s) and slightly underestimated compared to the forest site (slope 0.6 and intercept 0.13 m/s).

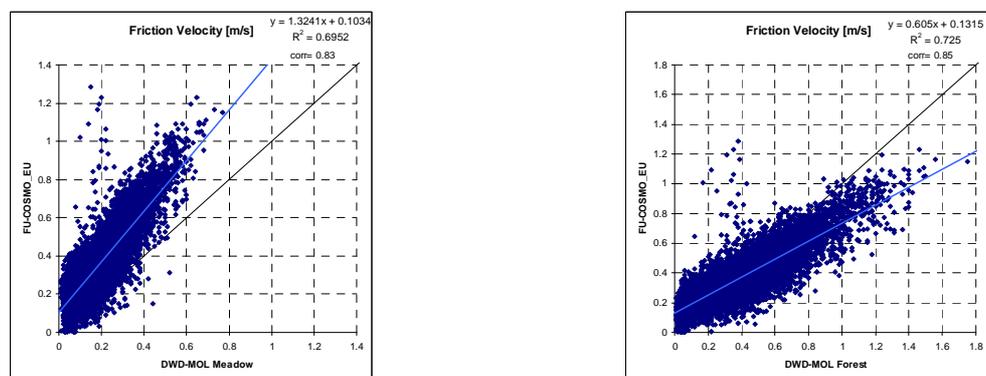


Figure 1. Model (y-axis) friction velocity compared to measurements (x-axis) at meadow (left) and forest (right) site.

Figure 2 shows the comparison between measured and simulated airborne pollutants concentrations for the forest station Augustendorf for HNO_3 (left panel) and NH_3 (right panel). The RCG simulations have been averaged with a moving mean of 30 days, while measurements are monthly means. RCG is able to reproduce the annual slope of all nitrogen components in the air with the right order of magnitude. The seasonal variability of the monthly means is captured well, modelling higher nitric acid concentrations in summer than in winter and being more or less constant over the whole year for ammonia. Nitrogen aerosol components were simulated at the correct order of magnitude compared to the forest data (not shown) and also the seasonality was reproduced acceptably.

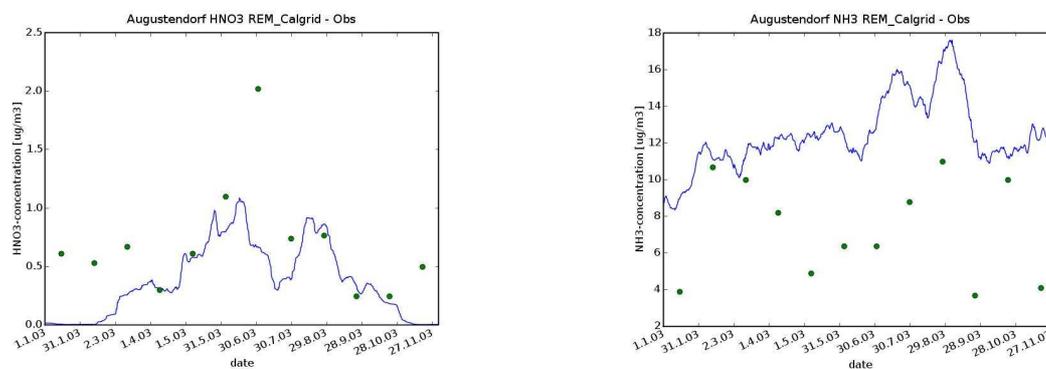


Figure 2. Measured monthly (green dots) and moving averaged (30 days window) RCG simulations (blue line) for HNO_3 (left panel) and NH_3 (right panel) at Augustendorf. Units [$\mu\text{g}/\text{m}^3$].

Table 1 gives accumulated observed and simulated gaseous nitrogen dry deposition fluxes at Augustendorf for the year 2003. Ammonia on the yearly mass basis is modelled very well while HNO_3 is underestimated almost by a factor of 5. Accumulated annual NO_2 dry deposition on the other hand is reproduced very well.

Table 1. Measured and simulated accumulated annual deposition fluxes for gaseous N-species in Augustendorf – Germany.

species	observed [kg-N/ha-a]	simulated [kg-N/ha-a]
$\text{NH}_3\text{-N}$	16.2	16.1
$\text{HNO}_3\text{-N}$	2.9	0.6
$\text{NO}_2\text{-N}$	1.5	1.9
$\text{HNO}_2\text{-N}$	0.8	0.0

Eddy covariance technique measurements have shown that nitrous acid has been contributing to the overall nitrogen dry deposition flux by a small percentage while RCG has attributed to that species a vanishing part. The almost one-to-one correspondence of the $\text{NH}_3\text{-N}$ simulations with the measurements is surprising since the observation should be representative

for the forest site, mainly, while the modelled value should represent a 7x7 km² mesh. Thus, we suppose that NH₃-N deposition fluxes might be overestimated. HNO₃-N at the other hand may be less underestimated as it might seem from table 1 for the same reasons as above. Also, comparing simulated airborne NH₃ concentrations with observations there was a substantially uniform overestimation for most months. This confirms the supposition of a too high simulated ammonia flux.

Wet deposition

For evaluation the RCG modelled wet deposition fluxes are compared to UBA measurements over Germany. The investigation period has been limited to September 2005. Besides an accurate parameterization of the wet deposition process itself reliable meteorological fields -especially those of precipitation and cloud liquid water content- are a key requirement for a successful simulation of the wet deposition flux. Thus, the applied COSMO-EU meteorological fields of precipitation and cloud liquid water content have been compared to independent datasets: non-SYNOP UBA precipitation network and Cloudnet observations at Lindenberg. Figure 3 (left panel) shows the comparison of the considered UBA stations monthly precipitation sums for September 2005 and the corresponding COSMO-EU grid point precipitation sums. COSMO-EU is in good agreement with most of the UBA stations concerning the precipitation sum.

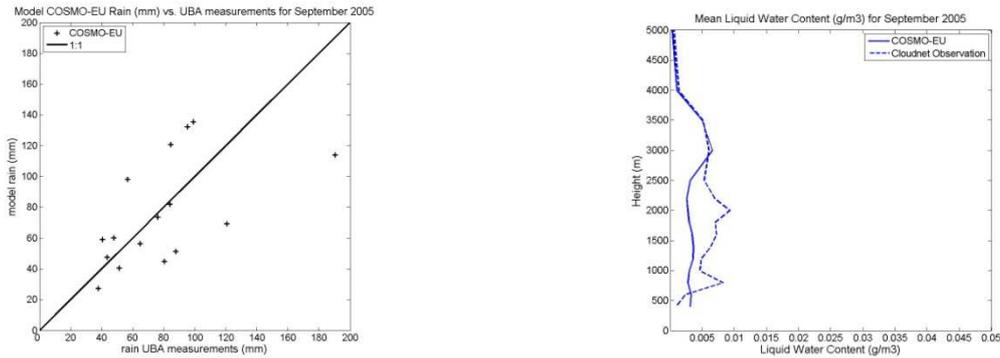


Figure 3. COSMO-EU (y-axis) precipitation compared to UBA measurements (x-axis) (left panel) in mm and COSMO-EU (continuous line) mean liquid water content compared to Cloudnet (dashed line) measurements (right panel) in g/m³.

If the model precipitation sum differs from the measurement the model cannot be able to simulate the wet deposition adequately. As can be seen at Schauinsland station the COSMO-EU precipitation underestimates with 114 mm the measured precipitation of 190 mm by far. Thus, at that site for the comparison of the modelled and measured wet deposition we expect a considerable underestimation of the simulated flux caused by an underestimation of precipitation. The vice versa should be observed when comparing simulated and measured wet deposition at Brotjacklriegel station where COSMO-EU overestimates with 135 mm the measured precipitation of 99 mm. Figure 3 – right panel- shows the results of the comparison between Cloudnet LWC measurements and COSMO-EU model cloud liquid water content. Above about 2500 m the COSMO-EU cloud liquid water content is very close to the Cloudnet measurements. Below 2500 m COSMO-EU underestimates the LWC measurements by up to a factor of 3. This underestimation leads to an underestimation of the in-cloud scavenging (see equation (4)) and thus to an underestimation of the amount of modelled wet deposition.

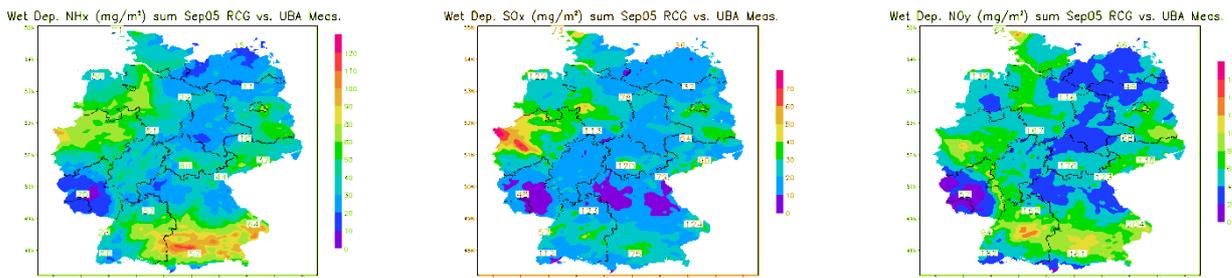


Figure 4. RCG modelled NHx (left panel), SOx (middle panel) and NOy (right panel), expressed in mg/m². UBA measurements are shown as numbers in white boxes inside the plots

Using the Chemistry Transport Model RCG wet depositions of reduced nitrogen (NHx), oxidized sulphur (SOx) and oxidized nitrogen (NOy) have been simulated for September 2005. Figure 4 shows the simulated wet deposition sums (colour plot) compared to measurements (numbers). Concerning NHx wet depositions (Figure 4, left panel) the model results are in good agreement with most station measurements. Especially at stations where COSMO-EU precipitation simulations and observations conform to each other the RCG wet deposition simulation compares well to the measurements. At the site Solling (51.8N; 9.6E) e.g. COSMO-EU precipitation of 82 mm for September 2005 is very close to the UBA precipitation measurement of 84 mm. RCG simulated 48 mg/m² of NHx wet deposition in Solling while UBA measured 51 mg/m². As predicted for Station Schauinsland (47.9N; 7.9E) RCG underestimates with 34 mg/m² the measured wet deposition of 50 mg/m² caused by a too low COSMO-EU precipitation. In contrast, due to an overestimation of COSMO-EU precipitation at the site Brotjacklriegel (48.8N; 13.2E) RCG overestimates with 88 mg/m² the NHx wet deposition measurements of

64 mg/m². For SO_x (Figure 4 middle panel) RCG wet depositions underestimate the measured values by a factor of 2 to 5. One reason for the strong underestimation is the underestimation of the COSMO-EU cloud liquid water content compared to measurements as discussed above. A look at the vertical distribution of SO_x and NH_x (not shown here) shows that SO_x is mixed up much higher into cloud levels than NH_x. Thus, for SO_x the influence of in-cloud scavenging on wet deposition is more significant than for NH_x. Hence, the underestimation of COSMO-EU cloud liquid water content did not lead to a noteworthy underestimation of NH_x wet depositions. However, the cloud liquid water content may not be the only reason for the underestimation of RCG SO_x wet depositions. There may also be deficits within the models' sulphate production or within the emission estimates. Investigations on this issue will be continued. Figure 4 – right panel - shows the modelled NO_y wet depositions compared to the measurements. The model underestimates the NO_y wet deposition by up to a factor of 2. Since NO_y is also mixed up higher than NH_x the main reason for the underestimation is the -compared to LWC measurements- low COSMO-EU cloud liquid water content. A sensitivity study with RCG on the cloud liquid water content has shown that multiplying the cloud liquid water content by two increases the modelled NO_y wet deposition substantially.

CONCLUSION

A thorough model evaluation with main attention on deposition processes has been conducted. Focus has been put on validating the involved parameters like mechanical turbulence factor friction velocity, precipitation and cloud liquid water content. The final deposition fluxes have been interpreted and compared to measurements. COSMO-EU friction velocities are in very good agreement with high resolved observations from Lindenberg. Correlation coefficients of 0.8 are found. Dry deposition measurements are based on eddy covariance techniques; the resultant values are comparable to RCG simulated amounts. COSMO-EU precipitations compared reasonably to most UBA measurement stations, considering that rain has a high spatial variability. The wet deposition evaluation showed that especially, at sites with high disagreements between modelled and simulated rainfalls wet deposition fluxes differ substantially for all investigated species. RCGs wet deposition module cannot be liable for this deviation as input fields must be correct. Following precipitation cloud liquid water content affects the amount of scavenged mass. The influence of cloud liquid water content on the wet deposition amount increases with increasing vertical mixing of the considered component. Since NH_x is less mixed up in the vertical than NO_y and SO_x NH_x is less affected by cloud liquid water content variability. This work has shown that it is not sufficient to compare pollutants concentrations in air or in water with observations but to check meteorology comprehensively.

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