INTERACTIONS OF AEROSOLS AND GASES WITH CLOUDS AND PRECIPITATION IN THE ONLINE-COUPLED REGIONAL CHEMISTRY-TRANSPORT MODEL COSMO-ART

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1. INTRODUCTION

Wet scavenging represents a major sink for gaseous and particulate constituents of the atmosphere. This complex of processes is described in current chemistry transport models with varying degree of complexity and accuracy. COSMO-ART, a recently developed online-coupled regional chemistry transport model, has been lacking a comprehensive scheme to describe these processes so far.

In this work we present the coupling of COSMO-ART to the SCAV submodel of the MESSy interface, a module to describe wet scavenging of aerosols and trace gases, aqueous-phase chemistry and cloud processing of aerosols. We describe the coupling method and adaptations that were made to use SCAV with COSMO-ART. We stress the importance of an explicit description of the gas to droplet transfer processes instead of assuming equilibrium.

2D flow-over-hill simulations were made to better understand the effects of the new scavenging scheme, and to assess the accuracy of the implementation. These simulations provide valuable insights into the workings of the module which could not have been revealed in complex but realistic 3D simulations.

2. MOTIVATION

COSMO-ART, as described in Vogel et al. (2009), lacked a wet scavenging scheme for gases and the description of aqueous-phase chemistry. Extensive evaluation of the modeling system in Knote et al. (2011) revealed a consistent tendency of the model to overestimate nitrate aerosols and underestimate sulfate contributions (see Fig. 1). Combined with an overestimation of SO_2 gas-phase concentrations (not shown) this strongly underlined the importance of including a description of wet scavenging of gases and aqueous-phase chemistry.

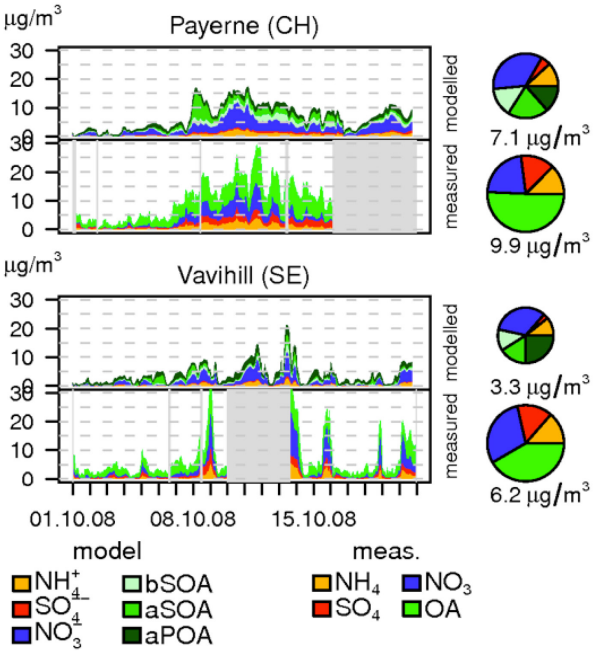

Fig 1: Comparison of Aerosol Mass Spectrometer measurements of aerosol chemical composition in Switzerland (top) and Sweden (bottom) with COSMO-ART simulation results. Pie charts depict mean compositions. Figure adapted from Knote et al. (2011).

3. PROBLEM STATEMENT

"Wet scavenging" is the combined effect of a number of processes:

- transfer of gaseous compounds from the gas-phase into cloud droplets and rain drops
- nucleation of new cloud droplets through the activation of aerosols
- scavenging of aerosols by cloud droplets and precipitation
- aqueous-phase chemistry within droplets
- solution and recombination of aerosols within a droplet
- re-release of matter into the gas- or aerosol-phase on evaporation of a droplet
- deposition of mass onto the ground by precipitation.

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The net outcome is therefore not necessarily a removal of mass from the atmosphere, it can also result in a transformation (e.g. of aerosol characteristics through cloud processing) or even be a source for substances (e.g. oxidation of sulfur to form sulfate aerosols).

Current chemistry transport models (CTM) treat this complex of processes with various degrees of complexity. Very simple approaches only account for the removal of mass from the atmosphere through precipitation, e.g. with size-independent scavenging ratios for aerosols and gas-to-particle transfer by fixed Henrys law coefficients, and they might parameterize aq.-phase sulfate production independently. More detailed schemes include aqueous-phase chemistry, but still assume Henrys law equilibrium – therefore avoiding the need to transport chemical components in the liquid phase. Advanced schemes feature an explicit description of the transport from gas / aerosols to droplets and thereby avoid the assumption of concentration equilibrium.

How CTMs treat those processes depends also on their method in general, where “offline-coupled” models (using pre-calculated meteorology) encounter bigger difficulties in representing such processes (due to the lack of information about cloud microphysics from the driving meteorological core), compared to “online-coupled” models for which this information is available.

Changing point of view and looking at the representation of clouds and precipitation in current regional numerical weather prediction (NWP) models, one can derive the degree of detail and information that could be available for use in a modern wet scavenging scheme. The Consortium of small scale modeling model COSMO (Baldauf et al. 2011) can serve as example: this non-hydrostatic NWP model is employed operationally by several European weather services on grid scales of up to 2 km. Its (bulk) cloud microphysics scheme includes up to 5 prognostic water classes (cloud water, cloud ice, rain, snow, graupel) constantly interacting with each other. Precipitation is therefore a prognostic quantity as well, allowing for horizontal transport and possibly evaporation – this additional effort has been shown to be necessary to improve forecast quality especially for simulations in complex terrain (Baldauf; Schulz 2004). COSMO simulations typically employ timesteps of *O*(<1min).

This leads to several conclusions regarding the design of a wet scavenging scheme:

- the assumption of Henrys law equilibrium for gas-to-particle transfer is no longer valid
- in-cloud transport of chemical components is necessary
- in-precipitation transport is also necessary to be consistent with microphysics

3. MODELING SYSTEM

3.1 COSMO-ART

COSMO-ART (*A*erosols and *R*eactive *T*race gases) (Vogel et al. 2009) is an extension to the COSMO model, recently developed at KIT Karlsruhe, Germany. It includes gas-phase chemistry based on an extended RADM2 (Stockwell et al. 1990) mechanism, a description of aerosols using the MADEsoot extended module (Riemer et al. 2003; Vogel et al. 2006), interactions between gas and aerosol phase for inorganic species using ISORROPIA (Nenes; Fountoukis 2007) and SORGAM (Schell et al. 2001) or (in development) a volatility basis set approach (Donahue et al. 2006) for secondary organic aerosols. Parameterizations are available for emissions of dust, biogenic VOCs, sea salt, pollen and DMS. The whole system is online-coupled to COSMO, allowing for direct (Vogel et al. 2009) and indirect (Bangert et al. 2011) feedbacks of aerosols on meteorology.

3.2 SCAV

SCAV, presented in (Tost et al. 2006), is a submodel for the MESSy interface (Jockel et al. 2005) and has been developed to simulate wet scavenging and aqueous-phase chemistry for global scale simulations. It includes:

* The description of the *transfer process of gaseous compounds* into cloud droplets and precipitation either via Henrys law equilibrium or an explicit description of the transfer kinetics through a system of coupled ordinal differential equations.

* The description of *scavenging of aerosols* by cloud droplets and precipitation. Processes included are Brownian motion, impaction, and interception. A simple activation parameterization is included as well. All processes are formulated in a size-dependent manner usable with modal aerosol schemes.

* *Aqueous-phase chemistry* based on the MECCA mechanism (Sander et al. 2005) with some additions from CAPRAM (Herrmann et al. 2003). Different subsets of this mechanism can be employed to reduce computational costs, with a minimum mechanism recommended by the authors of 35 species in the aqueous-phase and a set of 45 reactions.

* *Cloud processing of aerosols*. In case the driving model has modes / bins in different size ranges available, aerosols scavenged from all modes can be, once a cloud droplet evaporates, released into the largest available soluble mode. This represents a net mass transfer from smaller to larger aerosol sizes (see Fig. 2 for a graphical representation).

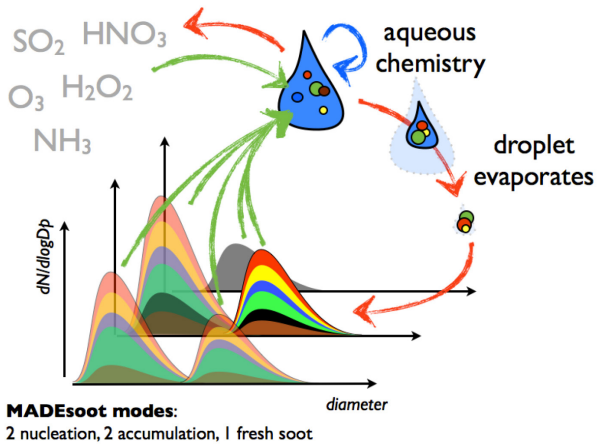

Fig. 2: Schematic description of cloud processing of aerosols in the coupled COSMO-ART-SCAV system.

4. COUPLING STRATEGY

The SCAV scheme needed to be adapted for coupling with COSMO-ART as it had been originally developed for global modeling systems. Two major adaptations will be briefly described below.

4.1 Partial evaporation and condensation

In its original formulation, the whole lifecycle of a cloud (see Fig. 3) took place each time step. This meant the cloud would condensate and evaporate completely. While this was acceptable for time steps in the order of hours, where one could assume the lifetime of a cloud will be shorter, this is not reasonable for COSMO-ART with time steps below 1 minute. One major reason it had been implemented that way originally was that in doing so one could avoid the transport of species in the liquid (cloud) phase. This strongly reduced the computational demand of the scheme.

We have included partial condensation and evaporation in accordance with COSMO microphysics. There, a bulk scheme with 5 different water classes is implemented. The total change in cloud liquid water content (cLWC) can be split into tendencies due to advection and condensation / evaporation. We could therefore estimate the relative contribution of condensation / evaporation to total cloud water. As we assume each grid box to be well mixed we can estimate the fraction of aerosols / gases that are subject to condensation / evaporation. For condensation this results in only a fraction of the aerosol to be subject to the nucleation parameterization (see also next section). For evaporation it has the consequence that some part of the chemical constituents might remain in the cloud-phase at the end of a time step. This part is now transported, i.e. an additional prognostic equation is solved for those components. As COSMO-ART is online-coupled this transport is in its methods consistent with the chosen transport method for the cloud water itself. In the following time step, existing cloud water concentrations are fed into SCAV as initial values for the transfer calculations.

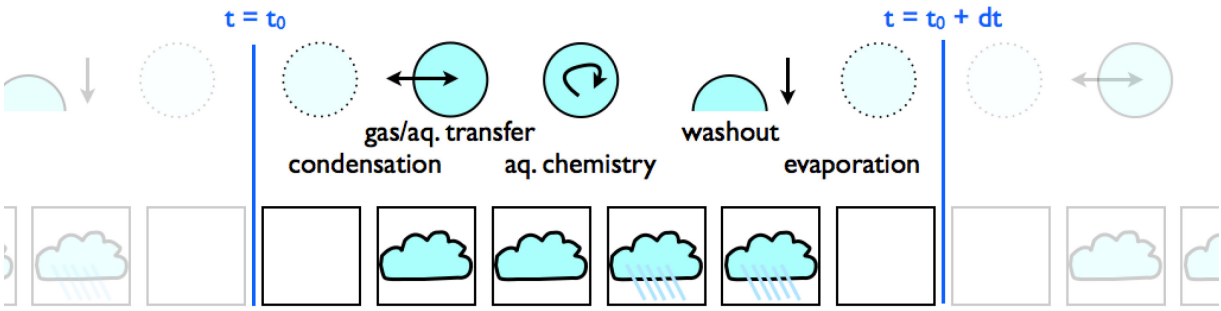

Fig. 3: Schematic depiction of the SCAV module processes. Note that a complete cloud lifecycle is simulated each timestep.

4.2 Time-step independent nucleation parameterization

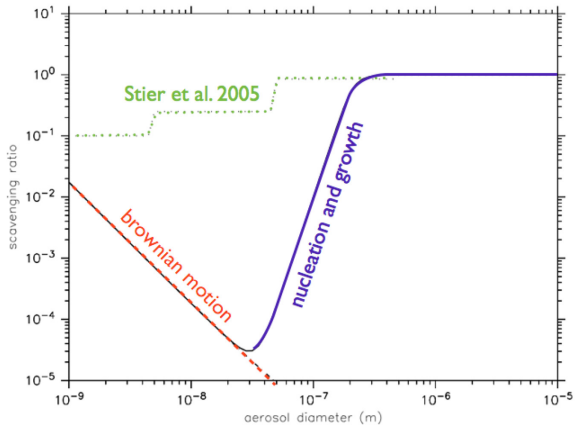

Fig. 4: Aerosol nucleation scavenging as implemented in SCAV. Figure adapted from Tost et al. (2006). The green dotted line shows a comparison with a parameterization from Stier et al. (2005) as it is used in the ECHAM-HAM global model.

In Figure 4 one can the parameterization for nucleation scavenging as done in SCAV. Note that the y-axis represents a scavenging ratio, not a coefficient. This is due to the fact that the nucleation and growth ("activation") parameterization is not formulated in a time-step independent manner – on each call a certain fraction of aerosols would be scavenged. This had to be changed in our adaptation, as it is only valid under the assumption that the complete cloud lifecycle will take place each time step.

As stated before, the different tendencies in cLWC are available from COSMO microphysics. Therefore, the change in cLWC due to condensation could be used as fraction of aerosols that will be subject to this nucleation parameterization, thereby removing the time step dependence.

The more realistic alternative would have been to include a complete, active coupling with the COSMO microphysics and include an activation scheme. While such a coupling is available for COSMO-ART (Bangert et al. 2011) it is not used in the operational version, and additionally very time consuming, so no coupling has been attempted with those modules so far.

5. 2D SIMULATIONS

5.1 Setup

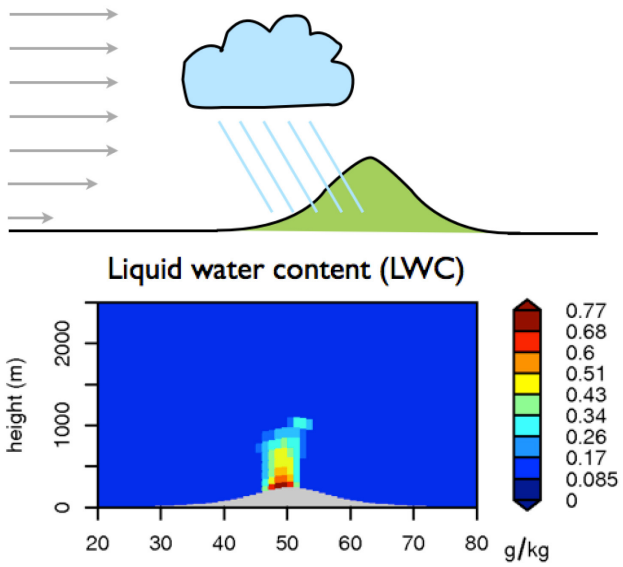

Fig. 5: Schematic of the 2D simulation setup (top) and the resulting mean cloud liquid water content (LWC).

COSMO allows for idealized simulations of a two dimensional flow-over-hill setup resulting in orographically induced cloud and precipitation development. Our setup (see Fig. 5) consists of a grid of 100 grid points in the horizontal (0.0045°, about 500m) and 40 hybrid sigma terrain following layers extending up to 20 hPa. The model is initialized and forced at the boundaries with 20 m/s wind speed from the left hand side, and a temperature / moisture profile following (Weisman; Klemp 1982). The default Runge-Kutta time integration scheme is employed with a timestep of 15 s. Aerosols and trace gases are initialized and forced at the boundaries with a column profile taken from a realistic 3D simulation of COSMO-ART, representing the environmental state of the Swiss midlands (e.g. at Payerne) in summer at 12 UTC. The model is integrated for 6h allowing it to reach steady-state and the results will be plotted as time average over that period. We performed two different simulations, one with SCAV included and one without.

5.2 Results

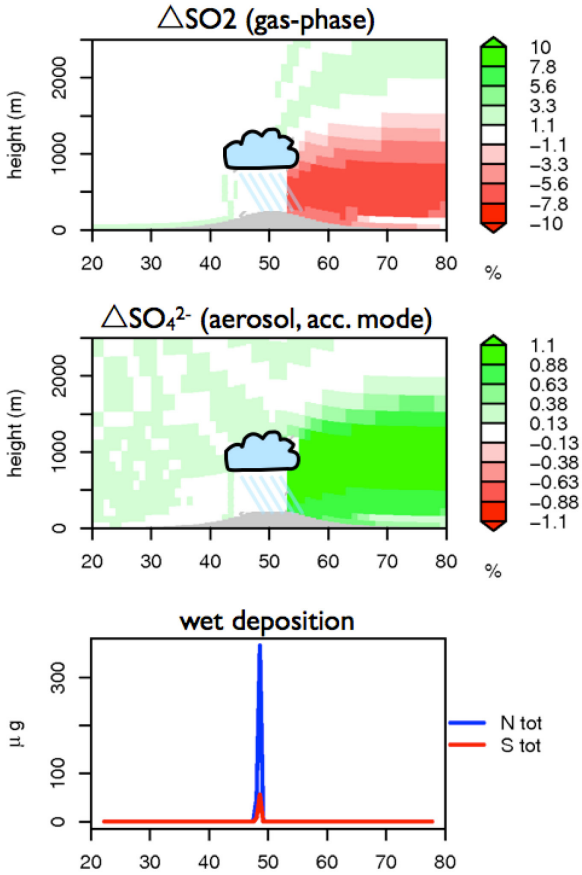

Fig. 6: Changes in gas-phase SO2 concentrations (top), accumulation-mode sulfate aerosol mass (middle) and cumulated deposition of nitrogen (N) and sulfur (S) (bottom plot). Change values are in % mass concentration of a reference simulation without SCAV.

The simulations show (Fig 6) that the inclusion of SCAV efficiently decreases SO_2 gas-phase concentrations leeward of the mountain and increases the SO_4^{2-} aerosol concentration in the accumulation mode. Additionally one can see the deposition of sulfur (and nitrogen) compounds due to washout by precipitation over the mountain.

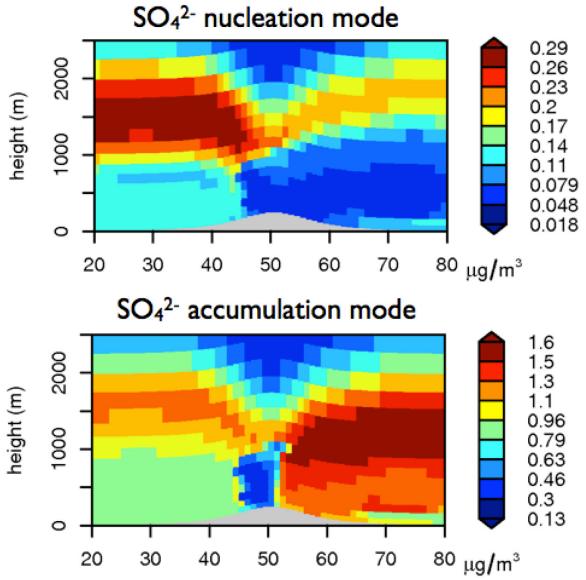

Fig. 7: Sulfate mass concentrations in the nucleation / Aitken and the accumulation mode, showing the consequence of cloud processing.

Figure 7 shows the effects of cloud processing of aerosols. Smaller sized aerosols (in the nucleation / Aitken mode) are scavenged within the cloud but not released into this size range on evaporation of the cloud droplets. Instead, the accumulation mode concentration gains mass from the combined input of scavenged sulfate aerosols from all available modes plus the additional sulfate create through aqueous-phase oxidation of sulfur.

To assess the accuracy of the aqueous-phase chemistry we compared the simulated acidity of the cloud water (negative decadic logarithm of H^+ ion concentration, pH) with measured values. As we have forced our simulations at the boundary with the chemical state of a Swiss midlands station at 12 UTC it should be roughly comparable with the measured values of precipitation acidity from another Swiss midlands station. Mean simulated pH was 5.6, while a mean over summer 2008 at Duebendorf (CH) gave a precipitation pH of 5.9.

6. CONCLUSIONS AND OUTLOOK

We are coupling the comprehensive wet scavenging and aqueous-phase chemistry scheme SCAV with the online-coupled regional chemistry-transport model COSMO-ART. We have already adapted the scheme to be used on the regional scale, including prognostic transport of the chemical composition of cloud water and precipitation, and a time step independent nucleation parameterization. Results from 2D simulations show that the scheme is working as expected; comparisons with station data indicate that the conditions for aqueous-phase chemistry (acidity) are realistic.

Next steps are the assessment of the different coupling methods (no transport, fixed Henrys law coefficients; only transport of cloud water) and evaluation of the impact of the scheme on realistic 3D simulations.

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