

Reviewer 1:

General comments

This manuscript describes the incorporation of a wet scavenging and aqueous chemistry scheme into the COSMO-ART regional online-coupled meteorology and chemical transport model. What is unique about this scheme is that the grid-scale clouds are simulated in a consistent manner between the meteorological and chemical transport components. The model explicitly tracks the concentrations of species in the gas phase, cloud water, and rain water, to simulate the release of aerosols to the gas phase upon cloud/rain droplet evaporation.

This is a well-written paper, and I appreciate the effort the authors have put into actually explaining the algorithms used. I recommend the paper be accepted for publication after the points below are addressed.

We are thankful for the reviewer's efforts and comments on our manuscript.

Specific comments

A1) I could not understand what was meant by the last sentence of the abstract (dominates over what?) until reading Section 4.6. Please expand and clarify so that the statement in the abstract can be understood by itself.

We concluded that the estimation of rain contribution to evaporation is an interesting question, but needs more work which we could not deliver within the review period. Therefore, section 4.6 and references to it in abstract and conclusions have been removed.

A2) I had a hard time following the discussion on pages 26107-26109: 26107 line 24 This sentence does not make sense. I think the first word should be some quantity other than f_wght . If f_wght is a fraction, it can't equal a dimensional quantity like qc_max . Also, the name of the variable f_wght for the fraction of the total flux that is considered to be nucleation is not intuitive.

We have renamed the flux fraction variable to f_flx . The fraction does not equate the upper limit of the cloud liquid water content qc_max . Rather, it scales between 0.0 and 1.0, depending on qc , with f_flx being 1.0 for $qc = 0$ and $f_flx = 0.0$ for $qc = qc_max$ and above. The paragraph has been rephrased and now reads

“Furthermore, saturation adjustment does not distinguish fresh nucleation from condensation on existing droplets. Hence only a fraction (f_{flx}) of the total flux is to be considered nucleation: if no cLWC exists, all of the flux must be nucleation, as no droplets exist to provide a condensation sink. In contrast, for fully developed clouds and high cLWC, all of the flux is assumed to be condensation on existing particles. In our implementation f_{flx} therefore scales linearly between no cloud ($qc = 0.0$, $f_{flx} = 1.0$) and a fully developed cloud represented by an upper limit of cLWC ($qc = qc_{max}$, $f_{flx} = 0.0$), remaining 0.0 for cloud liquid water contents above qc_{max} . The upper limit qc_{max} is determined...”

A3) 26109 lines 7-9: It would help to give equations describing the transfer of 10% and 90% of the number and mass to the the Aitken and accumulation modes. Presumably the surface area is adjusted accordingly?

The aerosol module “MADEsoot extended”, which is used in COSMO-ART, has fixed sigmas (mode widths) for each lognormal mode, while number and mass densities are prognostic quantities. The volume, or more precisely, the 3rd moment, is diagnosed at each timestep from prognosed mass and number densities and known (fixed) densities for each aerosol mass component for further calculations using this quantity. In our implementation of SCAV, the released masses and numbers are both distributed by 10% onto the mixed Aitken mode and by 90% onto the mixed accumulation mode. Finally, the volume (and if required the surface area) is diagnosed again to ensure that subsequent routines which might use aerosol parameters use a consistent set of values.

We have reformulated paragraph 2.3.2 and included a description of the evaporation procedure:

“This treatment of evaporation, i.e. using the mass as scavenged, but assigning a number based on the number of cloud droplets, represents “cloud processing of aerosols” in our model. Under the assumption that each cloud or rain droplet will form a single aerosol particle upon evaporation, the aerosol size distribution may be substantially altered as compared to the size distribution upstream of the cloud (Feingold and Kreidenweis, 2000). The question that arises is how to represent this process in a modal scheme like MADE. There, a number of log-normal modes exist which represent Aitken, accumulation and coarse mode particles. If now cloud processing creates an additional mode, a “from-cloud” mode, and one does not intend to represent this new mode in the aerosol-phase additionally to the existing ones, mass and number released from cloud droplets need to be redistributed onto the existing modes. SCAV in its original formulation put mass back into the largest soluble mode (in our case the “mixed” accumulation mode) upon complete evaporation. We slightly refined this by distributing mass and number by equal fractions onto the mixed Aitken (10 %) and accumulation mode (90 %), assuming that a small fraction of the cloud processed aerosol is still smaller than the accumulation mode size range. The median diameter of a given aerosol mode is proportional to $(M/N)^{1/3}$, where M is the total mass and N the number of particles in the mode. The new aerosol produced by cloud evaporation will tend to have a larger ratio M/N and hence a larger median diameter than the preexisting aerosol since the mass M of previously scavenged material (and additionally converted trace gas precursors) is distributed over a comparatively small number N of evaporated cloud droplets. Due to this treatment both modes are expected to shift to larger diameters. For the anthropogenic coarse mode and all sea salt and dust modes, evaporating aerosol particles are returned to their mode of origin. Table 1 lists the evaporation targets for all modes.

Note that scavenged species that would be released as gases, not aerosols, are in connection with the gas-phase via the kinetic transfer description of Tost et al. (2006) as long as the droplet exists, but are released in their entirety upon droplet evaporation.”

A4) 26112 lines 25-28: If I am interpreting these lines correctly, reduced nitrogen (NR = NH₃ + NH₄⁺) is initially predominantly NH₄⁺. As the air encounters a cloud, some of the NH₃ is processed by the cloud and converted to NH₄⁺, with the result that aerosol phase NR is further increased. But is the model conserving N (and S) mass? Similarly, if SO₂ decreases by 40% from 5 ug/m³, it seems that SO₄ should increase by more than 0.55 ug/m³. The idealized framework is well suited to addressing this question of mass conservation.

We ensured that the scavenging scheme itself is mass conservative down to machine precision. In the idealized setup, the accumulated errors in mass at the right end of the model domain (outflow region) due to the transport formulation (advection, diffusion, deposition) are below 0.01 % (column total).

Note that we have revised the idealized simulations and updated the accompanying text (see our replies B12, B13 to reviewer 2). The reviewer is right, reduced nitrogen is initially mostly NH₄⁺. This is what the model suggests for the chosen point (Swiss plateau, Payerne, 12 UTC 20 day average). We have now modified the advected profile by increasing the contributions of gas-phase ammonia (*2.5), and further (see response B13 to reviewer 2) increasing the concentration of H₂O₂ by a factor of 5 to avoid (at least initially) counter-ion or oxidant limitations.

This had considerable effects on the formation of sulfate in the cloud, as the previous setup was indeed oxidant-limited, and we refer to our replies to reviewer 2 for a list of changes.

A5) Section 4.6: I am not sure what is meant by "diagnostic precipitation." The statement that the meteorological community has already found such a treatment to be inadequate should have a reference. If what is unique in the modeling system here is that chemical composition of rainwater is tracked separately from cloudwater, then it would be nice to show that this makes an important difference in the modeling results. Advecting additional tracers for rainwater constituents is computationally expensive. The argument presented in this section and Figure 10 is not very conclusive.

"Diagnostic" is meant here as the opposite of "prognostic". In numerics, a "diagnostic" quantity is derived only using values **at the same time level**, while "prognostic" quantities are derived employing **also values from previous time levels**. In simple terms, in a diagnostic formulation, all precipitation either has to evaporate or reach the ground at the end of a timestep, but no horizontal transport, diffusion, etc. can happen.

A diagnostic formulation which is computationally less expensive is in preparation, but requires additional work, as the assumptions in the original formulation of SCAV cannot be used 1:1 in a regional model. This additional work was out of scope for this manuscript.

Paragraph 4.6 had been removed (see reply A1). We have however already stated this fact in section 2.3.3, so we have added 3 references for different commonly used modeling system communities (COSMO, WRF, and ARPEGE) there:

“While in many schemes scavenged mass is immediately lost and wet deposited, the treatment of precipitation in today’s mesoscale modeling systems would allow for a more comprehensive treatment including evaporation and 3-dimensional transport of rain droplets out of a vertical column, as presented e.g. for COSMO in Gassmann (2003); Baldauf and Schulz (2004), for WRF in Hong et al. (2004), or in Lopez (2002) for Arpege. This is realized in our approach...”

COSMO: Baldauf and Schulz, 2004 COSMO newsletter 4, www.cosmo-model.org.

WRF: Hong, Song-You, Jimmy Dudhia, Shu-Hua Chen, 2004: A Revised Approach to Ice Microphysical Processes for the Bulk Parameterization of Clouds and Precipitation. *Mon. Wea. Rev.*, 132, 103–120.

ARPEGE: Lopez, P., 2002: Implementation and validation of a new prognostic large-scale cloud and precipitation scheme for climate and data assimilation purposes. *Quart. J. Roy. Meteor. Soc.*, 128, 229–258.

A6) The limitation that only grid-scale clouds are treated, and not subgrid clouds, should be mentioned in Section 2 as well as Section 5.1.

Section 2, Coupling and extension, first paragraph has been extended to include this and now reads:

“As COSMO-ART is an online-coupled modeling system all transport processes applied to rain and precipitation are now likewise applied to its chemical composition, employing exactly the same operators and methods. In this work, the coupling is done for grid-scale clouds and precipitation.”

Technical corrections

A7) In the last paragraph of the introduction, the authors use “the first section” to refer to Section 2, “Section 2” to refer to Section 3, and so on.

We now refer to section 2, 3, and 4.

A8) Athanasopoulou et al. 2012 is not in the list of references. It sounds like an advertisement of a work in progress that does not need to be cited, especially if it has not been submitted.

This work has since been submitted to ACP and finished peer review, the citation has been updated. Furthermore, all simulations have been repeated to include this new development, so the appropriate method section was updated accordingly. We did this, as the new module will be the default for COSMO-ART simulations in the foreseeable future and it was very valuable to show the impact of our coupling to this new version. The new sentence reads:

“Formation of secondary organic aerosol components is realized with a volatility basis set (Donahue et al., 2006) approach described in Athanasopoulou et al. (2013).”

The inclusion of this new method of SOA formation shows a positive (i.e. more comparable with measurements) impact on aerosol chemical composition, with increased SOA contributions and reduced nitrate. See response B15) for an update to section 4.3 to reflect these changes.

A9) In Tables 2 and 3, what is k_0 , and what are the units of k_0 and dH/R ? The value given for water of $1.0E-16$ is not what I expected to see. I suppose the difference is due to rewriting an equilibrium as a kinetic reaction, but please clarify. Should all the quantities on the left side of Table 2 be denoted (aq)?

Firstly, we changed “ k_0 ” to “ K_0 ” in the Table 2 header to state clearly that this is not a rate, but an equilibrium constant. We think the confusion arises from the difference between

$K_w = [H^+][OH^-]$ (ionic product of water, values of about $1e-14$)

and

$K_0 = [OH][H]/[H_2O]$ (equilibrium constant, values $\sim 1e-16$, given in Table 2).

Units have been added to Tables 2 and 3. K_0 is in M, dH/R in Kelvin. All values are based on SCAV (Tost et al., ACP, 2009), which in turn uses the MECCA mechanism. The full MECCA reaction set can be found in the supplement to

Kerkweg, A., Sander, R., Tost, H., Jöckel, P., and Lelieveld, J.: Technical Note: Simulation of detailed aerosol chemistry on the global scale using MECCA-AERO, Atmos. Chem. Phys., 7, 2973-2985, doi:10.5194/acp-7-2973-2007, 2007.

<http://www.atmos-chem-phys.net/7/2973/2007/acp-7-2973-2007-supplement.zip>

Therein it is stated that the value for water is based on

Chameides, W. L.: The photochemistry of a remote marine stratiform cloud, J. Geophys. Res., 89D, 4739-4755, 1984.

We have added the (aq) tag to the components on the left side of Table 2.

A10) 26104 line 1 Please give the equation or equation number from Tost et al., as done on 26107 line 5.

We have added the equation number (9).

A11) 26104 line 24 I think you mean "down to" or "as low as" 1 km.

Changed.

A12) 26109 line 20 "processed" should be "processes"

Changed.

A13) Fig. 5: What are the numbers in the upper right corner of each panel? Number of observations?

Correct. We have added a sentence to the figure description.