Reviewer's report on the manuscript by Knote and Brunner (2012) "An advanced scheme for wet scavenging and liquid-phase chemistry in a regional online-coupled chemistry transport model", acpd-12-26099-2012

This manuscript reports an effort in extending the capability of an existing on-line chemistry transport model COSMO-ART to include more comprehensive representation of wet scavenging and aqueous-phase processes. The methodology used in coupling with an existing wet scavenging and aqueous-phase chemistry module (SCAV) is unique, in that it is one of very few chemical transport models to include chemical tracers in cloud and rain water as prognostic (advected) variables to be consistent with the microphysics scheme in the meteorological model (COSMO in this case). The coupled COSMO-ART with SCAV was tested in a 2-D idealised setup and then further evaluated for a regional simulation over Europe for a selected time period. The authors showed that the coupled system is able to address some of the previous model deficiencies with the improved representation of wet scavenging processes and the inclusion of aqueous-phase chemistry. While I was impressed by the originality and sophistication in the coupling technique developed in this work, I was somewhat disappointed that the authors did not address through this study the impact of such careful coupling as opposed to the more commonly adopted approach of simulating cloud life cycle at each model time step in most of the contemporary chemical transport models. Nevertheless I do think that this work presents a valuable contribution to regional air quality modelling community. The following are my specific comments and some suggestions to improve the manuscript.

Specific comments:

2   Methods

  2.1 Modeling system

It would help to list the various aerosol type/modes represented in the COSMO-ART model (in a table for example) and indicate the ones participating in (or affected by) cloud processing in this work.

  2.2 Scavenging and aqueous-phase chemistry scheme

On distributing cloud processed aerosols upon cloud droplet evaporation, what is the largest soluble mode specifically in this case? Significant growth of aerosol particle as a result of cloud processing is due to the in-cloud aqueous-phase production (of sulphate primarily) rather than the scavenging of multiple interstitial aerosols by a single cloud droplet. It should perhaps be acknowledged that this simplified treatment of cloud processing (i.e., distributing the droplet-borne aerosols to the largest soluble mode upon evaporation) is in part due to the limitation of modal representation.

  2.3 Coupling and extension

On the coupling and extension, was there a test done with the original SCAV? As many regional CTMs do still use similar methodology as the original SCAV (i.e., treating a cloud life cycle at each model time step), it would have been very valuable to show the impact on the model results due to the different coupling schemes.

2.3.1 COSMO operational cloud and precipitation microphysics

If equation (1) is also applied to cloud chemical components, it would require tracer components to be carried as prognostic variables for all the hydrometeor types (i.e., cloud water, cloud ice, rain water, and snow) in order to ensure mass conservation. Is this the case?

2.3.2 Cloud uptake and release of aerosols

Need to clarify which of the various aerosol modes are considered for cloud uptake (nucleation and impact scavenging; e.g., all fresh modes, all mixed modes; what about dust, sea salt, and soot?).

How sensitive is the model results to this selection of the upper limit on the cloud water content (or the droplet number density assumed – we know that cloud droplet number density can be significantly greater than 200 cm^{-3})?

On evaporation, are both number and mass imposed on the distribution of the released aerosols to the receiving modes? How is this done (mathematically) in this implementation, given the lognormal distribution? Does COSMO-ART carry aerosol number densities (of the various modes) as prognostic (advected) variables?

What's the sensitivity to the assumed cloud droplet number density (and hence the number of evaporated droplets) on the distribution of released aerosols and on the resultant aerosol size distribution?

The statement made at the beginning of the last paragraph of this section ("This treatment of evaporation … what is often termed 'cloud processing' of aerosols") is somewhat a misinterpretation of cloud processing.

2.3.3 Precipitation uptake and release of aerosols

Again, since it is implied that equation (6) is applied to chemical tracers in rain, are tracers in snow and ice also include as prognostic variables?

On the release of aerosols due to precipitation evaporation, similar to the question with regard to cloud droplet evaporation above, how are the number and mass mapped onto the receiving modes (and what are the receiving modes in this case)?

3 Idealized 2D simulation

With an idealised case like this, one can expect a more quantitative evaluation of the system, e.g., whether there is a mass closure on the depleted SO2 and the increased aerosol sulfate at the wake of the cloud (Figure 2) and similarly the loss in Aitken mode sulfate and the gain in accumulation mode (Figure 3). It is not clear from the discussion on Figure 2 and 3 whether these changes are consistent (quantitatively). For example, for a 50% decrease in SO2 (based on a 5 ug/m3 pre-cloud concentration) one would expect a much greater increase in aerosol sulfate than the 0.55 ug/m3 quoted (the number should be closer to 3.75 ug/m3, based on conservation of S, when no precipitation removal is involved).

There is also very little evidence of the enhanced sulphur oxidation in cloud from Figure 4A. Is this an oxidant-limited case (e.g., lack of H2O2)?

4   Application to a real case

    4.1 Comparison against long-term station measurements

It is somewhat surprising to see very little difference it made in PM2.5 at "rural" sites given the significant decrease in SO2 at these sites. Although the speciated measurements may not be available at these long-term sites, it would still be worthwhile to look at the modelled speciated components (of the relevant modes) to help with the interpretation of the results.

    4.3 Improvements in aerosol chemical composition

Why only focus on the fractional contributions, when different components may be influenced by different sources and processes? For example, for all three sites shown on Figure 7a, the increased fractional contribution of sulfate to the total PM1 (the simulation with SCAV vs. the base-case) is largely due to the reduction in nitrate rather than the actual increase in sulfate due to the coupling with SCAV. So to state (2nd paragraph of this section) "Sulfate aerosol mass increases substantially and now compares quite well to measured contribution" is somewhat misleading.

Last paragraph of this section: Again to say that the inclusion of SCAV has improved the overall relative contributions of the different components to NR-PM1 comparing to AMS measurement is not as meaningful when the impact is mostly on a single component. BTW, should define "NR-PM1" here (I take "NR" here refers to non-refractory here?) particularly when "NR" refers to something else, i.e., reduced nitrogen, earlier in the manuscript.

While it is reasonable to expect that the modelled PM1 may be reduced from the additional cloud processing (i.e., effectively moving some of the aerosol particles from Aitken mode to accumulation mode), it does show that the extended model significantly under-predicted submicron aerosol concentration at most of the sites shown in Figure 7 as compared to AMS measurements. This could indicate that the model is missing sources for submicron aerosols and/or the growth of aerosol particles due to cloud processing

may be over-represented by the extended model. The new AMS instrument being developed to measure the full size range of PM2.5 will not make the model deficiency in modelling submicron aerosols go away.

4.4 Effects on aerosol size distribution

Are there any size distribution measurements available for comparison with the modelled aerosol size distribution?

4.5 Wet deposition

How was the precipitation pH computed for model? Does the model include the full set of ions and base cations that contributes to the measured precipitation pH?

How does the modelled precipitation compare to the observed?

4.6 Contribution of evaporating rain to aerosol cloud processing

First paragraph of this section: The argument presented here is not very clear and may need to be rephrased – not sure what the authors really mean by "a diagnostic precipitation treatment" and "a diagnostic treatment of wet scavenging by precipitation". Further more the proposed argument is not tested or evaluated in this study. The tracer release due to rain evaporation can be parameterized even when rain water and tracers in rain water are not treated as prognostic variables.

It may be more instructive to show the sensitivity on modelled vertical profiles with vs. without rain evaporation.

5 Discussion

5.1 Limitations of the current implementation

There is a lack of assessing the impact on model results with regard to the assumptions made in the coupling scheme (related to both the uptake and release of aerosols, for example).

Just to add, the scavenging and processing by ice crystals and snow flakes can be important for lower troposphere, during winter season for example.

5.2 Comparison with other model systems

Need to include references to the various models mentioned here.

I very much agree with the last statement made in this section, but this work does not demonstrate the benefit of this feature.