Reviewer 2:

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.

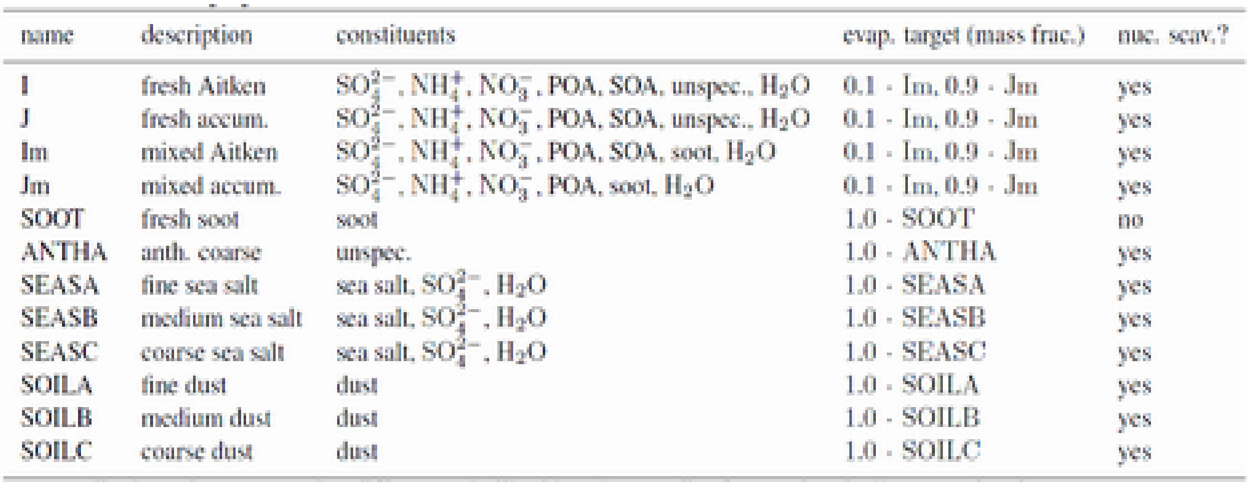
We are thankful for the reviewers efforts and comments on our manuscript.

Specific comments:

2 Methods
2.1 Modeling system

B1) 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.

The following table has been added, and is referenced in sections 2.1 and 2.3.2:



name	description	constituents	evap. target (mass frac.)	nuc. scav.?
I	fresh Aitken	SO_4^{2-}, NH_4^+, NO_3^-, POA, SOA, unspec., H_2O	0.1 · Im, 0.9 · Jm	yes
J	fresh accum.	SO_4^{2-}, NH_4^+, NO_3^-, POA, SOA, unspec., H_2O	0.1 · Im, 0.9 · Jm	yes
Im	mixed Aitken	SO_4^{2-}, NH_4^+, NO_3^-, POA, SOA, soot, H_2O	0.1 · Im, 0.9 · Jm	yes
Jm	mixed accum.	SO_4^{2-}, NH_4^+, NO_3^-, POA, soot, H_2O	0.1 · Im, 0.9 · Jm	yes
SOOT	fresh soot	soot	1.0 · SOOT	no
ANTHA	anth. coarse	unspec.	1.0 · ANTHA	yes
SEASA	fine sea salt	sea salt, SO_4^{2-}, H_2O	1.0 · SEASA	yes
SEASB	medium sea salt	sea salt, SO_4^{2-}, H_2O	1.0 · SEASB	yes
SEASC	coarse sea salt	sea salt, SO_4^{2-}, H_2O	1.0 · SEASC	yes
SOILA	fine dust	dust	1.0 · SOILA	yes
SOILB	medium dust	dust	1.0 · SOILB	yes
SOILC	coarse dust	dust	1.0 · SOILC	yes

2.2 Scavenging and aqueous-phase chemistry scheme

B2) 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.

We agree with the reviewer that this is a limitation of the modal scheme. We think that this has been addressed sufficiently in the last paragraph of section 2.3.2. Further it should become clear from the table added (question B1) which target modes are used upon evaporation.

2.3 Coupling and extension

B3) 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.

We did do tests with an unmodified version of SCAV. This was of interest in the development of a “diagnostic” formulation (i.e. without cloud / rain water composition as transported variables), as both reviewers have asked about.

There are however a number of problems with the original formulation in case it is used in the regional model context such as overestimated cloud uptake / evaporation rates, which did not allow us so far to create a diagnostic version producing correct results. Further work is needed to create such a version.

2.3.1 COSMO operational cloud and precipitation microphysics

B4) 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?

In the version presented here we do not consider cloud ice or snow chemical composition as prognostic quantities. Hence there is no target reservoir for scavenged chemical components for these hydrometeor types. As a consequence, the fraction of cloud liquid water / rain water that is converted to cloud ice and snow is considered to be additional evaporation for the chemical constituents in the droplet, that is, the additional fraction of constituents are released in the same way as if the droplet would be evaporating. It is therefore mass conserving, although it does not consider snow / cloud ice.

2.3.2 Cloud uptake and release of aerosols

B5) 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?).

All modes and constituents are subject to impaction scavenging by rain droplets, as well as Brownian motion scavenging by cloud droplets. All modes except the fresh soot modes take part in nucleation scavenging. We have added an overview table (B1) to describe modes, constituents, evaporation targets and participiation in nuc. scavenging. We have further added a sentence in section 2.3.2 (see reply A3).

B6) 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)?

Not very much. See reply B8.

B7) 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?

COSMO-ART carries both mass (contributions of each constituent) and number concentrations as prognostic quantities for each mode. The standard deviation of each mode is fixed. Upon evaporation, released masses and numbers are simply added to the prognostic quantities. The mass to be released is the known scavenged mass for each component, plus - in case e.g. of SO4 - in-cloud converted contributions, scaled by f_evap, the calculated evaporated fraction. Number concentrations added are the number of cloud droplets evaporated (N_0 * f_evap for cloud droplets, N_r * f_evap for rain drops).

Mean mode diameter and further properties of the mode are then diagnosed from known mass and number densities, the fixed mode standard deviation and the known densities of each constituent. See also our reply to reviewer 1 (A3) for updates to the manuscript.

B8) 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?

In the current version the cloud droplet number density is a fixed quantity (200 cm^{-3}). We do not consider an indirect effect of aerosols on clouds. As described in section 2.3.2 the number of cloud droplets evaporating scales with the available cloud liquid water content (cLWC)- the lower the cLWC the more droplets are evaporating, i.e. we assume that under high cLWC only few droplets evaporate completely but mass is released into the gas-phase primarily through shrinking of droplets. At lower cLWC more particles evaporate until the cloud completely evaporates. Then, all droplets (i.e. 200 cm^{-3}) evaporate.
We conducted a sensitivity simulation with N_0=400cm^{-3}, which show that there is negligible sensitivity on this parameter regarding the change in size distribution. A sentence has been added at the end of section 4.4:

"A sensitivity simulation where the maximum CDNC used in activation and evaporation calculations has been doubled (N_0 = 400 cm^{-3}) showed no noticeable effect on the changes in size distributions."

B9) 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.

We agree the sentence could be misleading. We intend to state that this is how we formulate "cloud processing" of aerosols in the model. The sentence has been rephrased and now reads:

"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..."

2.3.3 Precipitation uptake and release of aerosols

B10) 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?

see our reply to question B4 above.

B11) 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)?

Receiving modes and evaporation mechanisms are the same for cloud and rain droplet evaporation.

3 Idealized 2D simulation

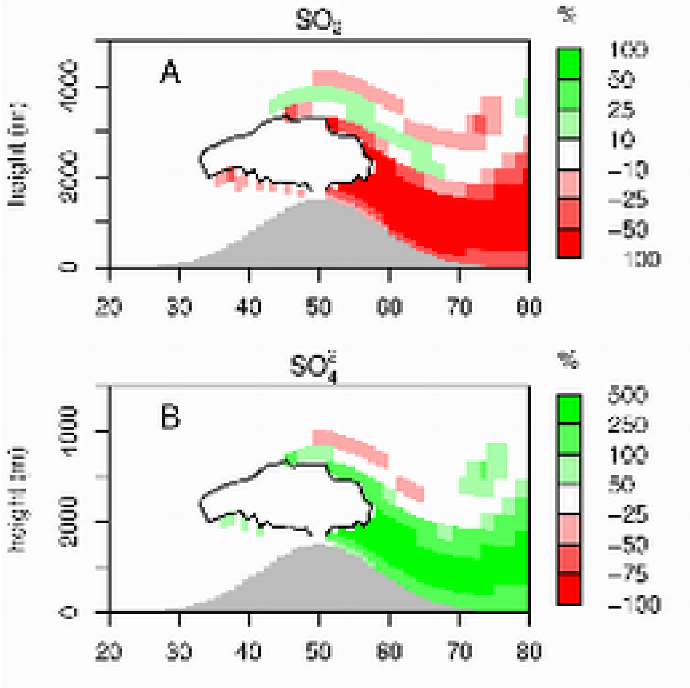
B12) With an idealised case like this, one can expect a more quantitative evaluation of the system, e.g., whether there is a mass closure on the depleted SO2 and the increased aerosol sulfate at the wake of the cloud (Figure 2) and similarly the loss in Aitken mode sulfate and the gain in accumulation 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).

We made a mistake in the way the sulfate increase had been calculated, as it was based on time averaged fields including the spin up time which did not allow the system to reach steady state. Therefore, no mass closure was achieved. We know that the scheme itself is mass conserving (see below).

The 0.55 ug/m3 increase in SO42- given in the text had been calculated based on the (indeed) oxidant limited case, and is revised in the updated manuscript (see answer to next question). With the non-limited simulations we find (updated excerpt from section 3):

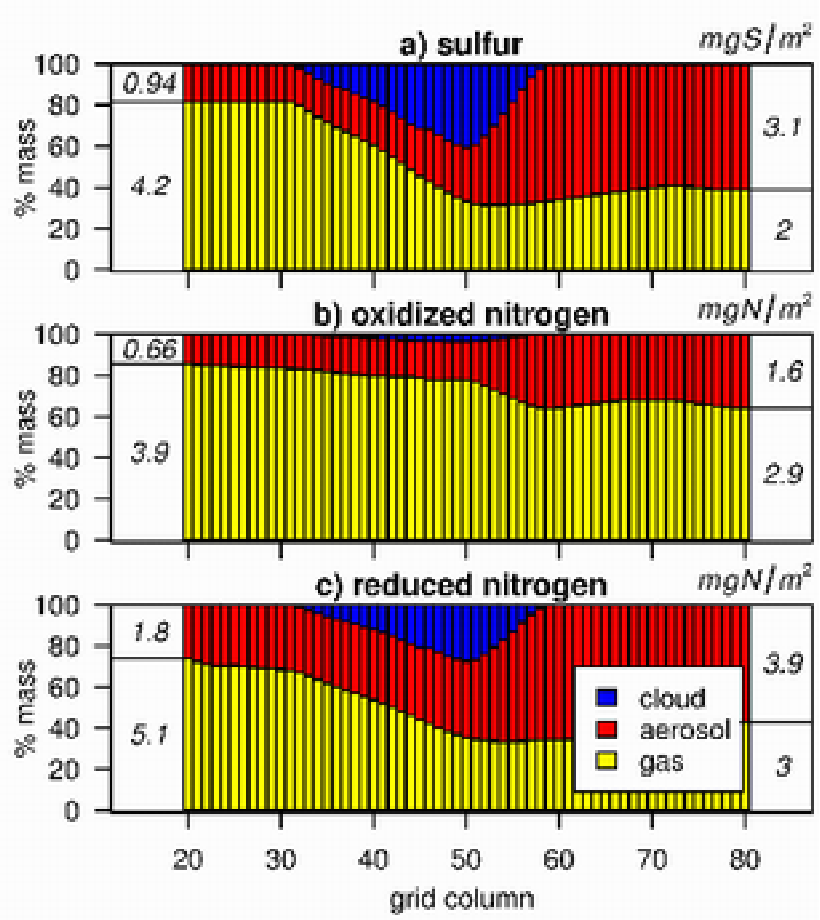
"Figure 2 shows how SO2 is efficiently scavenged after the air has passed the cloud, and that in turn total aerosol sulfate mass has increased by up to 500 %. The time an air parcel spends in
the cloud in this setup is about 450 s (cloud with approx. 9 km diameter, 20 m s^{-1} wind speed), and the maximum absolute difference in SO_4^{2-} in the wake of the cloud is +3.2 µg m^{-3}. This means that with an initial SO2 concentration of about 1 µgm-3 we generated approximately 7.1 ng m^{-3} of SO_4^{2-} per second due to aqueous-phase chemistry."

The updated figure 2 now looks as follows:



This large increase in in-cloud produced SO_4^{2-} had the consequence that Figure 3 (cloud processing through shifting of mass from smaller to larger mode) now shows an increase in SO_4^{2-} in both modes. This is the result of a dominating formation process over the reshuffling of existing mass between modes. As Figure 3 did not provide much further information we decided to remove it, as it would only lead to confusion of the reader. The sentence “Furtheron, Fig. 3 shows that mass released from the cloud...” was deleted.

As already stated in the response to reviewer 1 we ensured that the scavenging scheme itself is mass-conserving and we confirmed this with box-model/single column simulations (not shown). Figure 2 shows percentage changes and is intended to emphasize the relative importance of the effects, while the text should give a more quantitative description.
We refer the reviewer also to the updated Figure 4 (now Figure 3), in which a mass closure is now shown:



The updated figure caption reads:

"Partitioning of different groups of compounds between phases during passage of the cloud (between grid columns 30 and 60 of the idealized simulation). Shown are grid column totals. The three phases considered are gas (g), aerosol (a), and cloud liquid water (l). The compound groups are a) sulfur: SO2 (g), H2SO4 (g), SO42- (s), SO2 (l), H2SO4 (l), HSO4- (l), SO42- (l), HSO3- (l), SO32- (l). b) oxidized nitrogen: NO (g), NO2 (g), HNO3 (g), N2O5 (g), NO3 (g), NO3- (s), HNO3 (l), NO3- (l). c) reduced nitrogen: NH3 (g), NH4+ (s), NH3 (l), NH4+ (l). Numbers to the left (before cloud passage) and right (after) show column mass totals of the gas and aerosol reservoirs."

The paragraph in section 3 referring to this figure now reads:

"Figure 3 shows the column-total partitioning of mass between gas, aerosol and cloud phase for sulfur and nitrogen compounds during passage of the cloud. Wet deposition was negligibly small for mass calculations in this idealized simulation. Note that only about one third of the column is actually exposed to the cloud. Both, total sulfur and oxidized nitrogen show

increases in aerosol mass by 230% (+2.16 mg(S) m^{-2}) and 140% (+0.94 mg(N) m^{-2}) column total,respectively. Reduced nitrogen shows an increase of 120% (+2.1 mg(N) m^{-2}) after cloud passage, as a result of its thermodynamic equilibrium with sulfate and nitrate.
The idealized simulations provide useful...”

B13) 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)?

We thank the reviewer for this valuable comment. The case was indeed oxidant-limited and we now find much stronger increases in sulfate concentrations.
Figure 3 a) showed a 12% increase in column-total SO_4 aerosols. We have improved the idealized setup (see response to reviewer 1), especially through increases in H_2O_2 and NH_3 concentrations, and we now find much stronger increases in sulfate concentrations. The text in section 3 has been extended to reflect this

“It is calculated as the mean of all 12:00UTC profiles at Payerne during the 20 days of simulation of the “autumn” case in Knote et al. (2011). The profile was modified by increases in the concentrations of H_2O_2 (factor of 5) and NH_3 (factor of 2.5) to ensure that no oxidant or counter-ion limitation occurs. Two simulations have been made, …”

and Figure 4 has been updated (see above).

4 Application to a real case
4.1 Comparison against long-term station measurements

B14) 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.

The results of our previous model evaluation study (Knote et al., GMD, 2011) indicate that at most sites where speciated (AMS) aerosol data is available sulfate is only a minor mass contributor. Even if sulfate would increase several times, $PM_{2.5}$ would still be dominated by organics and nitrate. As can be seen in Figure 9 in Knote et al., GMD, 2011, only at Helsinki and Hyytiälä (both stations in Finnland) there is a share of sulfate of more than 25% of total aerosol mass (as seen by the AMS, see Knote et al., GMD, 2011 for a discussion of this restriction). Further, HNO_3 is efficiently washed out which leads to a compensating reduction in nitrate aerosols.
We did look at changes in modelled chemical components and found that there are indeed compensating effects. This is also mentioned in the text (section 4.3).

4.3 Improvements in aerosol chemical composition

B15) 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.

We disagree. The reviewer is right in that nitrate mass decreased. However, looking at the changes in absolute numbers it can be seen clearly that absolute sulfate mass of $PM_{1\text{-}NR}$ increased. To make this point clearer we added a table showing the absolute amounts of the different components:

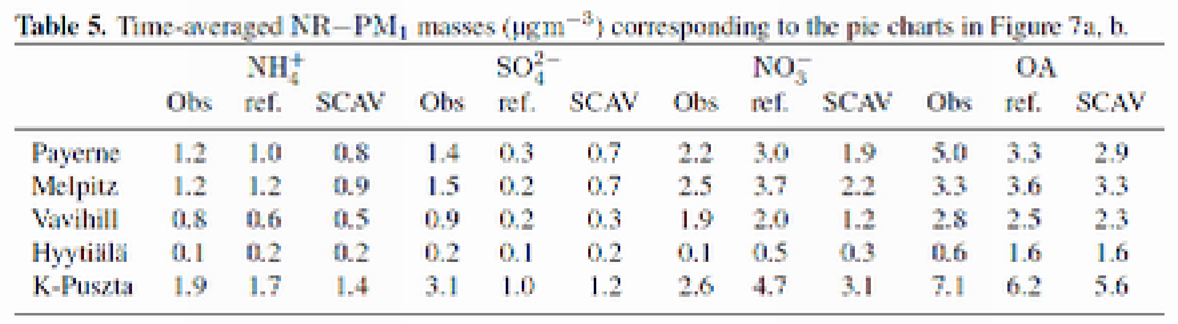


Table 5. Time-averaged $NR-PM_1$ masses ($\mu g m^{-3}$) corresponding to the pie charts in Figure 7a, b.

	NH_4^+			SO_4^{2-}			NO_3^-			OA		
	Obs	ref.	SCAV	Obs	ref.	SCAV	Obs	ref.	SCAV	Obs	ref.	SCAV
Payerne	1.2	1.0	0.8	1.4	0.3	0.7	2.2	3.0	1.9	5.0	3.3	2.9
Melpitz	1.2	1.2	0.9	1.5	0.2	0.7	2.5	3.7	2.2	3.3	3.6	3.3
Vavihill	0.8	0.6	0.5	0.9	0.2	0.3	1.9	2.0	1.2	2.8	2.5	2.3
Hyytiälä	0.1	0.2	0.2	0.2	0.1	0.2	0.1	0.5	0.3	0.6	1.6	1.6
K-Puszta	1.9	1.7	1.4	3.1	1.0	1.2	2.6	4.7	3.1	7.1	6.2	5.6

We have also rephrased the paragraph describing the results of the AMS comparison (as we included an updated SOA formation scheme in the meantime and hence some of the conclusions regarding OA underestimation were outdated, see response A8). The paragraph now reads:

"Figure 7a, b and Table 5 show that the inclusion of SCAV has considerable impact on the chemical composition.
The overestimation of nitrate compounds found in previous simulations is reduced, which is mostly due to the efficient washout of the HNO_3 precursor. Some discrepancy remains, like for example a tendency of the model to retain too much nitrate in the particle phase during daytime (e.g. in Payerne and Melpitz in Fig. 7a or K-Puszta in Fig. 7b). Sulfate aerosol mass increases substantially and now compares better to measured contributions, although at all stations the simulated values are still below the measurements. Together with the overestimation of SO_2, this points to a still too slow oxidation of SO_2 in the aqueous-phase possibly due to the lack of an explicit representation of several potentially relevant ions (Mn, Fe, Ca, K) that affect pH in COSMO-ART and missing minor oxidation pathways (transition metals, formaldehyde, dichloride ions, Jacobson, 2005). Biases in cloud pH would also influence the efficiency of oxdiation by O_3 (Kreidenweis et al., 2003).
Organic aerosol mass was not changed substantially due to the new coupling. Note the significantly improved agreement with observed values compared to what was presented in Knote et al. (2011). This is the result of a change from the use of the SORGAM module (Schell

et al., 2001) to describe the formation of secondary organic aerosols (SOA) to a volatility basis set approach (Athanasopoulou et al., 2013).
There is evidence that SOA can be formed in the aqueous-phase, and that these contributions can be substantial (Turpin et al., 2000; Lim et al., 2010; Ervens et al., 2011). As we focused on sulfate, we did not include any of these formation pathways so far.
Overall the relative contributions of the different components to ...”

B16) 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.

The new table 5 (see B15) shows that the inclusion of SCAV has positive effects on both SO_4 and NO_3. NH_4 (in equilibrium with NO_3 and SO_4) was reduced since the decrease in NO_3 is stronger than the increase in SO_4, which somewhat worsened the comparison with AMS. Effects (even though they are minor) are also observed for organic aerosols - at one station (Melpitz) results got better, at one worse (Payerne). That means that SCAV affects all components measured by the AMS, which usually represent the majority of particle mass below 1um.
NR-PM1 is defined in section 4.3. We have changed Figure 4 (now Figure 3) and removed the use of abbreviations NR and NO for reduced and oxidized nitrogen, respectively,so that this should not be confusing anymore.

B17) 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.

We agree with the reviewer that this might be the case. It is however, as the reviewer also notes, difficult to untangle what the real reason for this discrepancy is. We have added 2 sentences in this paragraph to state that. It now reads

“Overall the relative contributions of the different components to NR-PM1 show better agreement with AMS measurements due to the inclusion of SCAV, though total mass of NR-PM1 is slightly reduced. It is possible that this indicates missing sources for submicron aerosols in the model like wrong emission size distributions, or underestimated new particle formation. Further, aerosol growth due to cloud processing could be overestimated. To compare with AMS measurements, the AMS transmission function needs to be applied to the modeled aerosol mass size distribution to accurately represent the particle size range ``visible'' for the AMS.”

4.4 Effects on aerosol size distribution

B18) Are there any size distribution measurements available for comparison with the modelled aerosol size distribution?

We already performed a comparison of number concentration size distribution measurements in Knote et al., GMD, 2011. There, we used the first available homogenized dataset made during the EUCAARI project. However, the authors of this dataset caution that data quality is strongly degraded outside of a range of 20 - 200 nm (particle diameter). In the current study we focus on changes between <1 um (AMS) and 2.5 um - a size range for which this dataset is not well suited.

4.5 Wet deposition

B19) 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?

The model contains the major inorganic species (ammonium-sulfate, ammonium-nitrate) and its aqueous-phase products (see Table 2, 3). CO_2 is fixed. Table 1 describes the species that can partition into the aqueous-phase from the gas-phase and participate in pH calculation. While HCl and HBr are listed in Table 1 and 2 (because the aq.-phase chemistry mechanism includes them), no values are provided because COSMO-ART presently does not consider halogen chemistry. Further ions that may influence pH like e.g. transition metals are not included as no information is provided from COSMO-ART. The chemical system is solved each timestep and pH itself is then calculated from the estimated H+ ion concentration (pH:=-log10([H+]).

B20) How does the modelled precipitation compare to the observed?

COSMO itself is operationally used by several European weather services, and our model configuration is very similar to the operational setup. We can hence imply some initial trust in the meteorological quality of our simulations. We further compared against SYNOP stations in Knote et al., GMD, 2011. To ensure that precipitation is of good quality at the deposition stations we used in comparison in this work, we have added a plot to Figure 9 (now Figure 8), in which we compare against the reported precipitation values (daily accumulated values). Interestingly we find that the model underestimates the accumulated precipitation, which coincides with underestimations in wet deposition amount. We have updated the paragraph accordingly:

“... There is reasonable agreement with precipitation pH, with a mean value of 5.4 for measured and 6.0 for modelled values. Most of the comparisons of wet deposition with single station measurements lie within a factor of 5. Deposited nitrate and ammonia mass generally correspond to measurements within a factor of 2. Only sulfate mass shows a stronger underestimation, which reflects the still too low sulfate aerosols concentrations.

We find an average underprediction of accumulated precipitation by the model by a factor of 2. Hence we conclude that the underprediction of wet deposited mass of nitrate and ammonium, and part of the underestimation of sulfate wet deposition, is likely reasoned by insufficiencies of the meteorological simulation. The stochastic nature of precipitation contributes to the strong scatter of the comparison for such a short period (20 days) (Fig. 8 A) - and would arguably be reduced when simulating a longer time period. Also, very low precipitation intensities (drizzle) may not be registered by the instrumentation as precipitation event due to the measurement principle, which requires the opening of a lid (Aas et al., 1996). One possibility to circumvent the problem of statistical undersampling which does not involve simulating longer periods is to calculate the mean deposition (as arithmetic mean over all stations, black circles in Fig. 8), which can serve as a proxy for total wet deposition in Europe. There, SO$_{2-}$ is underestimated by a factor of 2–3. Model and measurements show better agreement for NO$_{-3}$ and NH$_{+4}$, where the underestimations are below a factor of 2. Results shown in Gong et al. (2006) for a comparable modeling system indicated similar underpredictions."

4.6 Contribution of evaporating rain to aerosol cloud processing

B21) 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.

We have removed the whole paragraph, as the investigation of the contributions of rain to evaporation require more model development and additional simulations. Also, the sentences referring to results from this section in the abstract and the conclusions have been removed.

B22) It may be more instructive to show the sensitivity on modelled vertical profiles with vs. without rain evaporation.

Section 4.6 has been removed. We agree with the reviewer that this would have been the more instructive way to do it.

5 Discussion

5.1 Limitations of the current implementation

B23) 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).

We tried to remedy this lack by including a sensitivity study on the assumption of maximum CDNC allowed. This allows to address both uptake and release. See answers above on the results.

B24) Just to add, the scavenging and processing by ice crystals and snow flakes can be important for lower troposphere, during winter season for example.

We agree, sentence added:

"… For studies of the lower troposphere this is, however, of much lesser importance and arguably negligible. It can become important, however, if a winter period is simulated. In the light of the additional computational …"

5.2 Comparison with other model systems

B25) Need to include references to the various models mentioned here.

We have added the appropriate references to AURAMS, CMAQ, WRF/Chem, and GATOR.

B26) I very much agree with the last statement made in this section, but this work does not demonstrate the benefit of this feature.

The last statement made in section 5.2 is "Only few models consider transport of in-cloud substances, and the fact that the chemical composition of rain droplets is a prognostic quantity in our approach is to our knowledge a unique feature".

We agree that no direct comparison has been made with a diagnostic formulation (see replies to several comments above the technical reasons why this did not happen). Further work is needed to show this, but was out of scope of this work, which focussed on the inclusion of a wet scavenging and aq-phase scheme at all, not a method comparison.