Reviewer #4

Review of revised version of: "Implementation of aerosol-cloud interactions in the regional atmosphere-aerosol model COSMO-MUSCAT(5.0) and evaluation using satellite data"

The clarity and quantitative discussion of the results has improved significantly in the revised paper. In particular the addition of Fig. 5 has helped greatly in the discussion. Furthermore it further justifies focusing the detailed comparison on a single day of the simulated period. I can therefore now recommend the paper to be published at GMD following only minor technical/editorial comments.

Technical Comments:

1) P6L6: COSMO2M, COSMO2MR and COSMO-MUSCAT used here, but COSMO2MR has not yet been introduced. All acronyms are introduced again (in case of COSMO-MUSCAT and COSMO2M) on P6L18ff. Please adjust as necessary. **ANS:** All acronyms are revised in the manuscript.

2) P9L9: Please remove or reword first sentence. The aerosol-cloud-radiation coupling is already discussed in previous sections in context of cloud optical thickness. **ANS:** P9L9: Sentence is removed form the manuscript.

3) Fig. 5: Please relabel "overall simulations". Suggestions: either "15-24 Feb 2007", or "10 day period".

ANS: Overall simulation has been revised to 15-24 Feb 2007.

4) Fig7: I would suggest to reword the caption to something like: "Net shortwave and longwave radiative flux at the surface and top of atmosphere for COSMO2M (a-d) and COSMO2M (e-h). Differences in radiative fluxes between the two simulations are shown in panels (i-l). **ANS:** Revised as suggested.

Reviewer # 5

Reviewer Comments:

Title: Implementation of aerosol-cloud interaction in the regional atmosphere-aerosol model COSMO-MUSCAT and evaluation using satellite data Authors: S. Dipu, J. Quaas, R. Wolke, J. Stoll, A. Muhlbauer, M. Salzmann, B. Heinold, I. Tegen

The manuscript has substantially improved with respect to the first version. That not all changes (additions) to the text are marked in bold face makes the evaluation of the revised manuscript unnecessarily difficult. Some replies were given only as 'reply to reviewer' but should have entered the manuscript as well. Just one example to illustrate the point: while the 'reply to reviewer' now states that Figures 7 and 8 show a 24h average for February 17, this information is not given anywhere in the manuscript.

Evidence for the claimed superiority of COSMO-MUSCAT over COSMO2M is, however, still more on the qualitative than on the quantitative side. With comparatively little effort this could be further improved, I think, and I would encourage the authors to do so. What must be improved in any case is the language. In a number of places in the current manuscript it is not even clear what the authors want to say. Also, there is still an overall lack of precision (see minor pointsbelow).

The manuscript still requires major revisions to meet GMD standards.

Major points:

 $\overline{1)}$ The language remains a major issue that must be improved. In several places, it is not even clear what the authors want to say. I give only two examples.

ANS: We went through the entire document and clarified the formulations.

p.9, l.8: "Further, the satellite retrieval (mainly thin clouds) are affected by snow cover, which could be rather ignored."

ANS: p9,1.8 has been moved to section: "model evaluation method" and rephrased as, Since the analysis is carried out for winter, satellite retrievals can be affected by snow cover on the ground. However, the MODIS retrieval (Platnick et al., 2001) uses a combination of absorbing spectral channels for which the snow/ice albedo is relatively small which makes it suitable for retrieving cloud properties over snow.

p.9, l.20: "It is also noted that the cloud microphysics radiation coupling results in reduction in cloud optical properties, which would results more downward shortwave and upward longwave especially at the surface."

There are many more sentences that are equally unclear, but I do not consider it my task as a reviewer to list them all. If none of the authors has sufficient command of the English language they should seek assistance from a native speaker.

ANS: We carefully copyedited the text in this re-revision, and will also have the publisher help with it where necessary.

2) While improvements were made, the manuscript still lacks precision and relies more on qualitative than quantitative statements. For example, in the abstract it is said that the cloud effective radius shows an increasees of 1 to 4 micro meter and the cloud droplet number concentration is reduced by 100 to 200 cm-3. Where do we see that in the manuscript? I greatly appreciate the newly added pdfs (Figure 5). But if you want to make the point that COSMO-MUSCAT is closer to MODIS than COSMO2M, why not show also COSMO2M in that figure?

ANS1: The above sentence in the abstract has been modified to: The cloud effective radius shows an increase of 9.5%, and the cloud droplet number concentration reduced by 21.5%.

ANS2: COSMO2M is also included in Figure 5 and discussed in the text.

Another example concerns Figures 7 and 8, comparison of shortwave and longwave fluxes among the different models and with CERES. From looking at the figures you conclude that differences are neither large nor systematic. Equally based on just looking at the figures I would argue that panel 8e (CERES) is most similar to panel 7A (COSMO2M), and differs more strongly from panels 7e (COSMO2Mrad) and 8a (COSMO-MUSCAT). Why not remap all the data on the same grid and provide quantitative estimates (means, pattern correlations, etc) to decide the issue?

ANS: Figure 7 and 8 are modified, in the modified manuscript, net shortwave flux at the surface and net longwave flux at the TOP are compared, with different color bars. Also the fluxes are considered for 17 February 2007 and are daily averaged (0hrs to 23.00hrs). Figure R1 shows the spatial correlation between modeled and CERES flues.

Minor points:

p.4, l.11: What is the numerical value of mu?

ANS: Here, the value of μ is 2, which is now added to the manuscript as well.

p.5, l.9: What is TNO?

ANS: TNO is European emissions processing, and stands for Nederlandse Organisatie voor toegepast-natuurwetenschappelijk onderzoek (Netherlands Organisation for Applied Scientific Research). This is now added to the text.

p.6, l.3: What do you mean by a positional error due to mismatch between meteorological regimes?

ANS: We mean the error in location, rather than characteristics of an event in the forecast compared to reality.

p.6, l.25: You may want to add / state explicitly that COSMO-MUSCAT treats radiation in the same way as COSMO-2MR.

ANS: Revised as suggested.

p.7, l.9: Rainfall of 100 kg m³? Please clarify.

ANS: The unit of rainfall has been corrected to 100mm, which accumulated precipitation over 96 hrs.

p.7, l.22: You write that the cloud optical depth of the satellite data varies between 5 and 54. However, the pdf in Figure 5 shows values larger than 100. Please clarify.

ANS: The range of satellite data has been corrected to 5 to 100, althoug most of the values

are lies between 5 to 50.

p.7, l.24: You say that the satellite derived cloud optical depth and liquid water path are overestimated. Do you mean that the satellite overestimates these quantities systematically, for example with respect to surface based observations or reanalysis? If so, please give a reference.

ANS: In the case study, the domain averaged cloud optical depth, effective radius and LWP are 23.34, 11.30 μ m, 0.175 kg/m⁻², whereas the COSMO-MUSCAT derived values are 7.60, 9.93 μ m, and 0.056 kg/m², which illustrates the satellite derived cloud optical properties are overestimated (Page 8, 1.20-22.).

p.7, l.26: 'cloud droplet radius between 2 and 20 micro meter'. But the pdf in Figure 5 shows values up to 30 micro meter. Please clarify.

ANS: COSMO-MUSCAT derived cloud droplet radius varies between 3 and 16 μ m and MODIS effective radius ranges between 2 to 30 μ m, which is same as in Figure 5.

p.8, l.8: 'For cloud optical depth, the model overestimate low clouds (optical depth below 10)...' Do you mean optically thin clouds?

ANS: This sentence modified as: The cloud optical depth PDF shows that thin clouds (cloud optical depth < 10) in all model versions occur substantially more frequently than in the satellite retrievals, and thick clouds (cloud optical depth > 30), less frequently.

p.8, l.12: Why not also show COSMO2M in Figure 5? **ANS:** COSMO2M is also included in Figure 5

p.8, l.29: '...satellite derive Nd values are overestimated...' Can you give a reference? **ANS:** Zhang et al., 2012, Storelymo et al., 2009.

p.9, 1.5: 'However, model derived cloud optical properties are well correlate.' With what do they correlate?

ANS: Here, the model derived optical properties strongly correlated to MODIS level-2 products, despite the low magnitudes. This is now discussed in the section 3.2 of the manuscript.

Section 3.3: Looking at Figures 7 and 8 the agreement between CERES and the different models for the surface shortwave radiation seems better in the absence of the revised radiation scheme. Please comment.

ANS: Figure 7 and 8 are revised in the manuscript with net shortwave flux at the surface and net longwave flux at TOA, and are compared with CERES data.

p.10, l.14: "The satellite retrievals suggest the revised model version is more realistic in both quantities." I find it difficult to draw this conclusion based on Figure 4 (showing cloud effective radius, but only for MODIS and COSMO-MUSCAT) and Figure 6 (showing cloud droplet number, COSMO2M looking more similar (more red) to MODIS than COSMO-MUSCAT). Could you further corroborate your conclusion?

ANS: In the revised manuscript, MODIS cloud optical properties are under gone revised cloud screening based on the recommendations by Nakajima and King (1990) (cloud optical depth less than 5 and effective radius less than 2μ m are not considered), in which cloud optical depth, effective radius and LWP are more realistic with satellite observations. Based on the new analysis, the conclusion is also revised.

Conclusions: I think it would be worthwhile to state again that you consider only warm clouds. **ANS:** Revised as suggested

Figures 2 and 6: Does the 100% cloud cover from Figure 2 together with the close to zero cloud droplet number concentration in Figure 6 imply that the majority of clouds is ice (i.e. not warm / liquid clouds)?

ANS: Figure 2 is daily averaged product, where as Figure 6 is averaged for 8:14hrs of the same day. Additionally, N_d is in cloud which is independent of clod fraction and the re-gridding can results in more cloudy regions. Figure R2 shows comparison between ISCCP simulator derived cloud fraction and MODIS simulator derived cloud droplet number concentration.

References:

Nakajima, T. and M.D. King, (1990), Determination of the Optical Thickness and Effective Particle Radius of Clouds from Reflected Solar Radiation Measurements. Part I: Theory. J. Atmos. Sci., 47, 1878–1893, doi: 10.1175/1520-0469(1990)047j1878:DOTOTA¿2.0.CO;2.

Zhang, Y., P. Karamchandani, T. Glotfelty, D. G. Streets, G. Grell, A. Nenes, F. Yu, and R. Bennartz (2012), Development and initial application of the global-through-urban weather research and forecasting model with chemistry (GU-WRF/Chem), J. Geophys. Res., 117, D20206, doi:10.1029/2012JD017966.

Storelvmo, T., U. Lohmann, and R. Bennartz (2009), What governs the spread in short-wave forcings in the transient IPCC AR4 models? Geophys. Res. Lett., 36, L01806, doi:10.1029/2008GL036069.



Figure R1: Comparison between short wave and long wave fluxes at surface and top of the atmosphere with CERES satellite fluxes and correlation between satellite (CERES) and models (COSMO2MR and COSMO2M)



Figure R2: (a) Model derived cloud fraction (via ISCCP cloud fraction) and (b) cloud optical depth (via MODIS satellite simulator).

Reviewer # 6

Review of revised version of **Implementation of aerosol-cloud interactions** by S. Dipu Sudhakar et al.

The authors present the revised version of a numerical study on aerosol, clouds and radiation with mutual interactions and compare the results with saltellite derived data.

Although, the paper shows some improvement compared to the 1st version, it is still far from publication. Still, the presentation is partly vague, requires physical interpretation, lacks some corrections according to the suggestions in previous review, and requires a substantial improvement of the language. It is strongly recommended to follow all comments. Slanted fonts stand for citations from the 1st review.

Yet, before publication, I suggest another considerable revision.

Comments

• The following 2 questions have not been answered:

In midlatitude winter I expect that the ice phase plays an important role in the development of clouds and precipitation (Bergeron-Findeisen effect!), and you use the Seifert and Beheng (2006) scheme for mixed phase clouds. The paper, however, is devoted to the liquid phase alone.

Please discuss the effect of the modified treatment of drop nucleation on the ice phase properties, since a modification in one path of condensate formation is connected with an opposing trend in other path(s).

How do you determine the effective radius under cloud free conditions?

Do you use the scheme of Seifert and Beheng (2006) in its warm cloud version? If Yes, then please argue for the neglect of the ice phase in midlatitude winter. If No, then please explain the changes in the ice phase properties in the whole model domain when changing the nucleation treatment. See p.6 l15 'screened for liquid phase clouds only'. Next, concerning re. Do you assume a lower limit of re for cloud free conditions (see question above), as suggested in your response? If Yes, $r_{e,min} = 2\mu m$ (see p.6 l15), this - together with $\tau = 5$ - results in N_d = $5.4 \times 10^9 \text{ m}^{-3}$ (Eq. 9). Something goes wrong here......

ANS: In our models versions we have used the Seifert and Beheng (2006) scheme for mixed phase clouds, in which the ice-phase are also affected by the new droplet activation parameterisation, e.g. due to Bergeron-Findeisen process. However, we have used the Seifert and Beheng (2006) in warm cloud only. In the Seifert and Beheng scheme, the droplet nucleation scheme is parameterized as,

$$\frac{\partial N_{c}}{\partial t}\Big|_{nuc} = \begin{cases} C_{ccn} k S^{k-1} \frac{\partial S}{\partial z} w, & \text{if } S \ge 0, w \frac{\partial S}{\partial z} > 0, \\ & \text{and } S < S_{max}, \\ 0 & \text{else.} \end{cases}$$
(1)

and the ice nucleation rate is given by

$$\frac{\partial N_i}{\partial t}\Big|_{nuc} = \begin{cases} \frac{N_{IN}(S_i, T) - N_i}{\Delta t}, & \text{if } S_i \ge 0 \text{ and } N_i < N_{IN}(S_1, T) \\ 0 & \text{else.} \end{cases}$$
(2)

The modification is done only with the C_{ccn} of the equation 2, whereas N_{IN} is not coupled with prognostic aerosols. Even though, the C_{ccn} modification would modify the ice phase, the effect is very less compared to the liquid phase of the cloud. Figure R1, shows the ice cloud optical

properties for different model versions.

In the revised analysis, the effective radius are not diagnosed over cloud free regions.

Since the modification have done for the liquid phase of the clouds, for comparison, satellite cloud products are screened for the liquid phase clouds only.

Again, the screening of the satellite products has been corrected in the revised manuscript, in which the cloud optical properties are considered only if the cloud optical depth > 5 and effective radius > 2 μ m for a given pixel. This would eliminate high N_d values.

• Equations (3) - (5) as in the first review:

The reviewer is familiar with the calculation of the moments and related properties from the cloud drop size distribution $\phi(D)$. Then, (4) follows as,

$$\lambda = \left[\frac{\pi \rho_{\rm w} N \Gamma(\mu+4)}{6 \rho q_{\rm c} \Gamma(\mu+1)}\right]^{\frac{1}{3}}$$
(3)

with ρ air density, ρ_w bulk density of liquid water, q_c mass fraction of the liquid water, N number of drops per volume, as was already explained in the 1st review.

Please revise and check the relationship $\lambda(N, qc)$ used throughout the paper.

ANS: Equation 4 has been modified in the revised manuscript as

$$\lambda = \left[\frac{\pi \rho_{\rm w} \mathrm{N} \Gamma(\mu+4)}{6 \rho_{\rm qc} \Gamma(\mu+1)}\right]^{\frac{1}{3}} \tag{4}$$

• The following questions have not been answered:

Problem of avaraging.

7, Figs. 4, 6(new). Cloud water path is a property defined for the whole air column. Cloud effective radius, cloud droplet number concentration, and sulfate aerosol number concentration are defined locally, and for a grid point model the data are interpreted to be representative for the grid cell. For which level are the presented data relevant? If they are vertical averages, please discuss, how the vertical average is calculated, how cloud free layers are considered, how the result is to be interpreted, etc. This point is even more complicated for the local variable re, which depends nonlinearly on the local variables N and qc. Likewise, optical thickness is defined for a certain layer of thickness dz, maybe the layer where the respective re holds. The presented fields (do they hold for the whole column?) depend on the averaging method.

The same question arises for the daily averaging procedure and concerns also liquid water path. It concerns both, model and satellite data.

Please explain, and correct the discussion where necessary.

The reviewer is aware that you use the COSP satellite simulator. The question of averaging, however, is not answered, and the added text p. 5 l.27pp is not helpful in this context.

ANS: Cloud optical depth, effective radius and liquid water path are derived using the MODIS simulator which is included in the COSP satellite simulator. Figures 4 and 6 shows the COSP derived MODIS products and its estimations are discussed below, also included in the manuscript.

The MODIS satellite simulator uses profiles of particle size for liquid and ice and corresponding optical depths within each layer of sub-column as a function of model levels. Using the cloud overlap assumption, zero or one cloudiness in each sub-column is created in each level. The diagnostics are then integrated over the cloudy sub-columns to obtain in-cloud average cloud optical depth and liquid water path. In turn, cloud effective radius is sampled at the cloud tops, which is not a vertical integral. Further, the ISCCP simulator aggregates pixel scale cloud retrievals (fraction of the sub-column with $\tau \geq 0.3$) to estimate cloud fraction (more details: Pincus et al., 2012). Further, cloud droplet number concentration is estimated using equation 9.

To compare with MODIS satellite observation, cloud optical properties (optical depth, effective radius, and droplet number concentration) are averaged for the time 8.00 to 14.00 hrs, which is approximate MODIS Terra overpass time over the domain.

• As before: interpretation of Figs. 4, (new) 6.

Drop number concentration, liquid water content and path, optical thickness, and effective radius are interrelated, not independent of each other. The correlation may be positive or negative, see e.g., (5) states $\tau \propto LWP$ and $\tau \propto 1/r_e$, while Fig.4 suggests on first glance only the first relation. Please interpret the graphics in terms of these interrelations.

Again: For the discussion of the improvement of COSMO-MUSCAT to COSMO-2M it would be helpful to include the COSMO-2M-fields in Fig. 4 besides (or instead of) the difference fields.

ANS1: Interpretation of Figure 4 and 6 is given below.

From the above analysis, it can be inferred that COSMO-MUSCAT can be used as a tool for regional aerosol-cloud interaction estimates. The interactive aerosol results show an increase in the cloud droplet effective radius by 9.5% and a reduction in N_d by 21.5%. This indicates that the interactive aerosols in COSMO-MUSCAT model accounts for the increase in cloud droplet size by enhancing the cloud droplet growth and it accounts for lower N_d , which attributed to implicit aerosol cloud interactions in the model. Additionally, it reveals the importance of satellite simulators in weather forecast models, which is very efficient for validating model results with different satellite observations. Even though the degree of uncertainty can go together hand in hand with satellite observed and model derived cloud optical properties, the modeled cloud optical properties are in agreement with satellites data.

As for the influence of both LWP and r_e on cloud optical thickness: By construction, the dependencies as the reviewer suggests are relevant in both, observational and model datasets. In the model, we implemented the dependency of cloud optical thickness on r_e (please see Manuscript Section 3.3). In the satellite retrievals, LWP is not retrieved independently, but computed from cloud optical thickness and effective radius. However, the reviewer is of course correct in observing that the LWP variability substantially more impacts the cloud optical thickness variability than the variability in r_e .

ANS2: Authors think that rather than including COSMO-2M fields, the difference can give a quantitative information about the model modifications. In addition, for reviewers consideration COSMO-2M fields are included in the Figure R2.

• The following questions have not been answered:

The choice of the parameters C_{ccn} (p. 4 bottom) is a good general guess, however, not a universal constant. Did you do a similar run with modified C_{ccn} -values to check its influence - in opposition to the influence of the full interactive treatment with MUSCAT? COSMO-MUSCAT seems to result in much smoother distributions than COSMO-2M, in particular Fig. 5 (new: Fig. 6). Do you have an explanation?

ANS: Sensitivity experiments with different C_{ccn} values are already reported by Seifert et al.,

2012, which uses low $C_{ccn} = 100 \text{ cm}^{-3}$ and high $C_{ccn} = 3200 \text{ cm}^{-3}$ and it illustrates the influence of different C_{ccn} values. The main difference between COSMO-2M and COSMO-MUSCAT is to replace constant C_{ccn} value with gridded C_{ccn} proxies computed using Boucher and Lohmann parameterization. Hence, the smoother distribution can be explained by activation of more cloud droplets in the COSMO-MUSCAT simulations. Figure 6d is a representative of aerosols in the domain, and it can very with the model levels and time. So the smooth distribution can be due to the temporally and spatially varying aerosols, which results in more droplet nucleation.

• Please go through the whole paper carefully. Avoid repetitions, strengthen the physical interpretation, improve the verbal presentation, eliminate errors in grammer and spelling. **ANS:** We did go through the entire manuscript with careful copy-editing..

• p.7 l.9: wrong unit of rainfall amount.

ANS: Revised as 100mm, which is accumulated precipitation over 96 hrs.

• p.7 l.22, Fig. 4: You give the range of data for cloud optical depth with a minimum of 5 for both satellite and model data. This does not agree with the figures: Huge white areas occur, and white stand for data less than 5 according to legend. If on the other hand, you prescribe $\tau = 5$ as minimum (p.6 l.15), than refer to that chosen threshold. A similar problem concerns the effective radius (l. 26) with white standing for $r_e \ge 2 \mu m$, that is the mentioned minimum value.

ANS: Figure 4 is corrected, since the retrieval error is more for effective radius less than 2 μ m and cloud optical depth less than 5, the satellite retrievals are screened for above threshold values. This part is now corrected in the manuscript.

• p.8 l.7pp. Please clarify: The under-/overestimation refers to the frequency of the respective value of liquid water path or optical depth.

Does the model really overestimate 'low cloud' or does it overestimate the frequency of low optical depth cases?

Please clarify for all 3 properties.

ANS: Under/overestimation refers to the frequency of occurrence. This is now explained in the manuscript.

The statistical distribution of satellite and the model cloud microphysical properties are compared and evaluated in terms of probability density functions (PDFs). Figure 5 represents the probability density function of the spatiotemporal distribution of cloud optical depth, effective radius, and liquid water path, defined as the normalized count of occurrence per bin width of cloud optical property. The cloud optical depth PDF shows that thin clouds (cloud optical depth < 10) in all model versions occur substantially more frequently than in the satellite retrievals, and thick clouds (cloud optical depth > 30), less frequently. The modeled cloud effective radius PDFs is constrained to 3 and 16 μ m, where as the satellite retrievals shows a range of 4 to 30 μ m. A shift of the PDF is found in the COSMO-MUSCAT derived PDFs, which indicates the increased droplet size for the interactive Cccn. For liquid water path, modeled PDFs overestimates the clouds with low liquid water path and underestimates clouds with high water paths. The differences in PDFs largely follow what is found for the cloud optical depth, but model deficiencies compared to the satellite retrievals are substantially larger. The analysis also illustrates an increased in cloud optical PDF from COSMO-MUSCAT simulation. Certainly, the drop and preponderance of modeled cloud optical properties can be influenced by model tuning, an approach which, however, hasn't been performed yet for the

COSMO-MUSCAT model version.

• I appreciate the inclusion of fig. 5 for the different probability distribution functions. How do you define here the PDF? Unit?

The interpretation is given as a description of higher/lower PDF and the conclusion that the PDFs are similar for the single day and the period. Unfortunately, an interpretation of the differences/coincidences in the structure of the PDFs for the model and the satellite data is missing. It would be interesting to look for reasons of the shift in PDF for liquid water path, the more frequent occurrence of low τ in COSMO, and the preponderance of r_e around 10 μ m in COSMO, the peak in the MODIS-PDF for cloud optical depth between 150 and 200, the drop of COSMO-PDFs to 0 around LWP = 20m τ = 50, r_e = 30 μ m, and many more features. What are the PDFs in the COSMO-2M case? This may help the interpretation.

ANS: These plots represent the density of COD, CER and LWP, which means the normalized count of occurence per bin width of optical property. The density has a unit inverse to that of the bin width.

The shift in the PDF for MODIS cloud optical properties (especially LWP) can arise from the quality filtering of the data, also the log scale is responsible for large shit, which is revised in the manuscript.

The peak in the MODIS cloud optical depth around 150-200 can be due to high retrieval uncertainties towards high cloud optical depth, which is screened in the revised analysis.

the drop and preponderance of modeled cloud optical properties can be can be influenced by model tuning.

• p.8 l.19p. I do not see this.

p.8 l.20p: Something goes wrong with the sentence.

Please clarify.

ANS: p.8 l.19p. This sentence is removed from the manuscript

p.8 l.20p: The sentence is modified to : Likewise, the model derived N_d is also estimated using equation 9, which uses COSP (MODIS simulator) derived cloud optical depth and effective radius.

• p.7 l.20pp (new: p.8 12pp), Fig.4 g-i. You describe what is seen in the figures, but you do not give a physical interpretation. As suggested in the 1st review, the differences should be seen in relation to the signal, and then you find differences of 50% of the signal. If you use the full Seifert and Beheng schene, then the difference in LWP should be seen also in relation to the change in cloud ice concentrations (not only locally but in the whole domain). The sequel of e.g., red and blue bands over the Biscaya may be a phase shift.

p.8 l.19p. I cannot see the superiority of COSMO-MUSCAT from the presented material. p.8 l.20p. Sentence unclear. (i) Any explanation is missing. (ii) If you compare two models differring in 2 parameterizations, you cannot trace back the differences simply to the microphysics parameterization. Please clarify.

ANS: Physical interpretation: In COSMO-MUSCAT, the aerosol coupling leads to an increase in the cloud optical depth by 4.1%, the cloud effective radius by 9.5% and the cloud water path by 14.2%. This implies that gridded aerosols result in increaseing the cloud droplet size, optical depth and LWP. However, this would results in the reduction Cloud droplet number concentrations (N_d). This incicates that, as the drolpte size increase, N_d decreases and it would expline the implicit aerosol-cloud interaction in COSMO-MUSCAT.

Please see answer to the first comment above.

Figure R2 shows the MODIS simulator derived cloud optical properties for the COSMO2M Simulations (g-i). From this Figure, it is clear that, there is no phase shift over the Biscaya, However, it can be noticed that, there is an increase in the COSMO-MUSCAT derived cloud optical properties.

Figure R1 (ice optical properties) clearly indicates that, ice phase shows very low signal, which points the fact that, cloud microphysics modification has very little effect on ice cloud optical properties.

p.8 l.19p. I cannot see the superiority of COSMO-MUSCAT from the presented material: **ANS:** In COSMO-MUSCAT aerosol coupling leads to an increase in the cloud optical depth by 4.1%, the cloud effective radius by 9.5% and the cloud water path by 14.2% which explain the scope of aerosol-could interaction in regional modeling.

p.8 l.20p: The sentence modified to : Likewise, model derived N_d is also estimated using eqn 9, which uses COSP (MODIS simulator) derived cloud optical depth and effective radius.

• p.8 l.29-31. Under-/Overestimation - in comparison to the what?

l. 31
pp. '... explained by cloud microphysics modification.... MUSCAT-model.' I cannot see an explanation why $\rm N_d$ should be reduced in the MUSCAT-model. Please explain.

ANS : p.8 l.29-31.- On 17 February 2007, the domain averaged CDNC values are 150, 120, and 378 cm⁻³, respectively for COSMO2M, COMSO-MUSCAT and MODIS, which indicates an underestimation of model derived values (Figure 6a-c) as compared to MODIS. Further, from model inter-comparison (COSMO2M and COSMO-MUSCAT), it can be inferred that COMSO-MUSCAT derived CDNC is reduced by 21.5% (Figure 6a and 6b). This may be explained by implicit aerosol cloud interaction. From figure 5, it is also noticed that, there is an increase in cloud droplet size in COSOMO-MUSCAT, this would result in lower CDNC, which attribute to aerosol-cloud interaction in the COMOSO-MUSCAT model.

• See 1st review. Old: p.8 l.6, new: l.32 Fixed CCN= 300 cm^{-3} in COSMO-2M? This is in contradiction to Section 2.1, telling Nccn is given as function of S.

l. 32. Similar: 'constant cloud condensation nuclei profile'??

Please clarify.

You have answered the question in your response, but you did not clarify anything in the paper. Please precise your wording and distinguish clearly between the properties Nccn and Cccn as well as to talking about CCN (an abbreviation to shorten the text).

ANS: In COSMO-2M, N_{cn} (activated cloud droplets) is a function of S, whereas C_{ccn} kept constant. In the coupled model constant C_{ccn} is replaced by gridded C_{ccn} proxy from MUSCAT, which is a four dimensional variable.

This is included in the manuscript.

• As 1st review:

p.8 l. 9pp, new: p.8 l.34pp. The aerosol NUMBER (not 'mass') concentration is given in Fig. 6c. Could you please comment on the fact, that Nsulfate is so much larger than Nd for COSMO-MUSCAT? Is the result of Boucher and Lohmann (1995) transferrable to your model concept?

Please make clear which parameter you are talking about and give a precise explanation. **ANS:** p.8 l.34pp: It is revised to aerosol number concentration.

Figure 6d in the manuscript shows the spatial distribution of sulfate aerosol number concentration (aerosol number concentration proxy) below the convective cloud base (representative of aerosols in the model and it is also averaged for 8-14 hours on 17 February 2007), where high number concentrations are simulated over southeastern Europe. On contrast, N_d are smaller over the same region. This is because the Boucher and Lohmann (1995) parameterization models saturation of N_d over high aerosol or polluted regions (Penner et al., 2001) and the high pressure in this region results in trapping aerosol in the boundary layer.

• As 1st review: p.8 l.14pp, new: p.9 l6pp. Please revise the para.

'The model is unable to capture sub grid scale cloud patterns': A subgrid scale cloud cannot be captured by the microphysics parameterization of Seifert and Beheng (2006) or similar ones. You would need a different parameterization tool. '

You talk of the 'coarse resolution' of the model. I would not call a mesh size of 28 km 'coarse'. More important: Please tell the resolution of the satellite data when you compare the resolution.

If the snow cover can be ignored - how are the satellite retrievals affected by the snow cover? Please use a more precise wording.

ANS: In the revised manuscript, this part is revised and the revision is as follows.

From the above analysis, it can be inferred that COSMO-MUSCAT can be used as a tool for regional aerosol-cloud interaction estimates. Because the interactive aerosol coupling results show an increase in cloud droplet effective radius by 9.5% and a reduction in N_d by 21.5%. This indicates that the interactive aerosols in COSMO-MUSCAT model accounts for the increase in cloud droplet size by enhancing the cloud droplet growth and it accounts for lower N_d , which attributed to implicit aerosol cloud interactions in the model. Additionally, it reveals the importance of satellite simulators in weather forecast models, which is very efficient for validating model results with different satellite observations. Even though the degree of uncertainty can go together hand in hand with satellite observed and model derived cloud optical properties, the modeled cloud optical properties are in agreement with satellites data.

The effect of snow cover on cloud retrials is included in the model evaluation methods, which is follows,

Since the analysis is carried out for winter, satellite retrievals can be affected by snow cover on the ground. However, the MODIS retrieval (Platnick et al., 2001) uses a combination of absorbing spectral channels for which the snow/ice albedo is relatively small which makes it suitable for retrieving cloud properties over snow.

• As 1st review: p. 8, new: p.9, Section 3.3. l. 25 new: l. 19. Fig. 7. The colorbars are differently scaled. Sometimes this is straightforeward, but sometimes, however, confusing. Please unify the scaling insofar as to use the same scaling at least for SFC and TOA net down SWR. Same for Fig. 7.

Fig. 7 a-d contains is repetition of Fig. 6 e-h. Use the difference fields COSMO2M rad minus CERES instead.

Are figs. 7 i and k really different?

Once again: Please interpret the radiative flux differences also in terms of the cloud properties. **ANS:** Section 3.3 is revised in the manuscript. In the revised version, we have used net shortwave at the surface and net long wave at the top of the atmosphere (TOA), with different color bars. Also the fluxes are considered for 17 February 2007 and are daily averaged (0hrs to 23.00hrs).

ANS: In the revised manuscript, Figure 7 is moved to Figure 9, which shows the comparison

between the COSMO-MUSCAT and CERES derived fluxes (net downward shortwave flux at the surface and net downward longwave flux at the top of the atmosphere (TOA)). Figure 7i and k are revised in the manuscript, which are not similar.

• p. 10

Now you have a contradiction in conclusions 1 and 2. Conclusion 1 - 'modification has only a minor effect'. Conclusion 2 - COSMO MUSCAT shows an improvement in the cloud microphysical properties.

Please clarify.

What is the outcome of the PDF analysis?

From the 1st review for Conclusion 1: If you refer to the model runs COSMO-2M and COSMO-MUSCAT, please say so. Then, this statement does not agree with p.7 l. 20-29 (new: p.8 l.12-21). Please clarify.

Conclusion 3. You can find differences in the model runs with and without the effect on radiation. How do you know that the new approach gives results closer to reality? **ANS:** Conclusions have been modified:

A case study has been carried out to compare the model output with observations. The incorporated COSP satellite simulator serves as a link between model and satellite comparisons. Despite the resolution, COSP derived ISCCP cloud fraction shows similar spatial Further, MODIS level-2 cloud optical products such as cloud pattern and magnitude. optical depth, effective radius, and liquid water path are compared. The COSMO-MUSCAT derived cloud optical properties show a similar spatial distribution compared to the MODIS observation. In COSMO-MUSCAT, the cloud optical depth has been increased by 4.1%, cloud droplet effective has been increased by 9.5%, and liquid water path has been increased by 14.2% in comparison to CSOMO2M. In turn, the cloud droplet number concentration estimated from COSMO-MUSCAT model shows a reduction of 21.5% compared to the COSMO2M model. Furthermore, considerable changes in the radiation budget have been found. This analysis indicates that the coupled model (COSMO-MUSCAT) with interactive aerosol treatment results in an increase in cloud droplet size and reduction in cloud droplet number concentration by activation and growth of droplets, which illustrates implicit aerosol-cloud interactions. Also, the cloud properties in COSMO-MUSCAT agree reasonably well with observations, so that it can be used for regional aerosol-cloud interaction studies.



Figure R1: COSMO-MUSCAT cloud ice optical properties on 17 February 2007.



Figure R2: Cloud optical properties on 17 February 2007, top panel: MODIS level 2 products, bottom panel: COSMO-MUSCAT simulated cloud products, and bootom panel: COSMO2M derived cloud products. All model products are averaged between 08:00 to 14:00 hrs, which is the MODIS aqua overpass time ove the domain.

Implementation of aerosol-cloud interactions in the regional atmosphere-aerosol model COSMO-MUSCAT(5.0) and evaluation using satellite data

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Abstract. The regional atmospheric model Consortium for Small Scale Modeling (COSMO) coupled to the MultiScale Chemistry Aerosol Transport model (MUSCAT) , is extended in this work to represent aerosol-cloud interactions. Previously, only one-way interactions (scavenging of aerosol and in-cloud chemistry) and aerosol-radiation interactions were included in this model. The new version allows for a microphysical aerosol effect on clouds. For this, we use the optional two-moment cloud

- 5 microphysical scheme in COSMO and the online-computed aerosol information for cloud condensation nuclei (CCN) concentrations, replacing the constant CCN concentration profile. In the radiation scheme, we implement have implemented a droplet-size-dependent cloud optical depth, allowing now for aerosol-cloud-radiation interactions. In order to evaluate the model To evaluate the models with satellite data, the Cloud Feedback Model Inter-comparison Project Observational Simulator Package (COSP) has been implemented. A case study has been carried out to understand the effects of the modifications, in which where
- 10 the modified modeling system was is applied over the European domain with a horizontal resolution of $0.25^{\circ} \times 0.25^{\circ}$. Further, to To reduce the complexity in aerosol cloud interaction interactions only warm-phase clouds are considered. It is We found that the online coupled aerosol introduces significant changes for some cloud microphysical properties. The cloud effective radius shows an increase of 1 to 4 μ m9.5%, and the cloud droplet number concentration is reduced by 100 to 200 cm⁻³. The microphysics modifications have a smaller effect on other parameters such as optical depth, cloud water content, and cloud
- 15 fraction. <u>21.5%</u>.

1 Introduction

The quantification of aerosol cloud interactions in models continues to be a challenge (*IPCC*, 2013). Estimates of effective radiative forcing and assessments of the radiative effects due to aerosol cloud interactions to a large extent rely on numerical modeling. A large effort has been made to represent such effects in general circulation models (GCMGCMs) (*Penner et al.*,

20 2006; Quaas et al., 2009; Ghan et al., 2016). However, GCMs do not resolve the processes relevant for cloud dynamics well. Improved process understanding for Improving the understanding of processes of aerosol-cloud interactions thus largely relies on simulations with cloud-resolving models and large-eddy simulations (LES) (*Ackerman et al.*, 2000, 2004; *Xue et al.*, 2006; *Sandu et al.*, 2008; *Seifert et al.*, 2015; *Berner et al.*, 2013). However, LES often focus on case studies and use idealised idealized boundary conditions and also an idealised-idealized representation of the aerosol. This leads to uncertainties in particular specifically because, when analyzing cloud systems, or cloud regimes, rather than individual clouds, aerosol-cloud-

- 5 precipitation interaction processes often are buffered (*Stevens and Feingold*, 2009). Regional climate modeling is a powerful tool to overcome these limitations of small-domain , idealised LES. Much idealized LES, and much higher resolutions are possible than for GCMs. Compared to LES that only simulate simulates individual cloud systems, regional climate models are able to simulate the feedbacks between clouds and aspects of the large-scale circulation and its variability are simulated by regional climate models. Even though regional models do not describe part of the large scale feedbacks.
- 10 it provides large-scale feedbacks, may be considered a good optimal compromise (*Bangert et al.*, 2011; *Van den Heever and Cotton*, 2007; *Chapman et al.*, 2009; *Forkel et al.*, 2015; *Yang et al.*, 2012).

A still often applied cloud microphysics parameterization in numerical weather prediction is a bulk, one-moment scheme (*Kessler*, 1969; *Lin et al.*, 1983), which uses the specific mass-masses for different hydrometeor species as prognostic variables. However, it cannot treat aerosol cloud interaction because interactions because it calculates only one moment of the size distri-

- 15 butionis calculated, do, and does not carry information about size or number concentration of cloud droplets. In contrast, bin microphysical schemes numerically resolve the size spectrum and are thus able to predict the spatio-temporal spatiotemporal behavior of a number of size categories for each hydrometeor type explicitly (*Khain et al.*, 2000; *Simmel et al.*, 2015). However, this approach is numerically very expensive especially when applied for regional atmospheric models. As a compromise between these two approaches, two-moment microphysical schemes are able to can predict the number concentration of the
- 20 liquid and ice hydrometeors, in addition to mass variables (*Cotton et al.*, 1986; *Meyers et al.*, 1997; *Seifert and Beheng*, 2006). Furthermore, numerous studies have shown that using two-moment schemes is a promising avenue to be used in future operational forecast models (*Reisner et al.*, 1998; *Tao et al.*, 2003; *Seifert and Beheng*, 2006) and is also computationally efficient.

At present, several weather prediction and global models have applied apply with two-moment cloud microphysical schemes.

- 25 For example, the Weather Research and Forecasting model (WRF) model is available with different types of two-moment microphysical schemes (*Thompson et al.*, 2008; *Morrison et al.*, 2008; *Lim et al.*, 2010). *Morrison et al.* (2009) demonstrated the showed that using a two-moment scheme in the WRF model produced more trailing stratiform precipitation in an idealized two-dimensional squall case with WRF model, which is consistent with surface which is more consistent with observations. In another study, *Li et al.* (2008) investigated the effect of aerosol on cloud microphysical processes with a two-moment
- 30 microphysical scheme in WRF model. Also, *Lim et al.* (2010) have included the a prognostic equation for cloud water and cloud condensation nuclei (CCN) number concentration number concentration (C_{ccn}), which could reduce the uncertainty to investigate in investigating the aerosol effect on cloud properties and the precipitation process in WRF model. Furthermore *Weverberg et al.* (2014) discuss the comparison between *Seifert et al.* (2012) and *Weverberg et al.* (2014) compared the operational one-moment and two-moment microphysical schemes in the Consortium for Small Scale Modeling atmospheric model (COSMO).
- 35 Further, other some groups previously implemented aerosol-cloud interactions in COSMO, albeit with a different aerosol

scheme (*Bangert et al.*, 2011; *Zubler et al.*, 2011; *Possner et al.*, 2015) and very few are coupled to the radiation scheme(*Seifert et al.*, 2012). *Seifert et al.* (2012) reported a strong positive bias while comparing 2-m temperature in <u>Seifert et al.</u> (2012) compared the operational one-moment scheme with cloud radiation coupled microphysics scheme to a two-moment scheme, which indicates that radiative aerosol induced effects are more relevant compared to precipitation. They found that aerosol perturbation have

- 5 significant effect on radiation and near surface temperature, rather than the resulting surface precipitation.
 - In this paper, we discuss the improved cloud microphysics parameterization in the COSMO model (*Doms et al.*, 1999), via the online-coupled aerosol model, MUlti-Scale Chemistry-Aerosol Transport (MUSCAT; (*Wolke et al.*, 2004, 2012))(MUSCAT, *Wolke et al.*) The two-moment cloud microphysical scheme in the COSMO model (*Seifert and Beheng*, 2006) uses fixed profiles of CCN concentrations C_{cen} . Rather than this simplification, here we use CCN concentrations C_{cen} predicted on the basis of the simu-
- 10 lated aerosol from the MUSCAT module. This will enable the COSMO model to have temporally and spatially varying $\frac{\text{CCN}}{\text{concentrations}}$ at each grid point, which are fully consistent with the cloud and precipitation fields, as well as with dynamics (e.g. scavenging is taken into account, as is vertical transport) to represent aerosol cloud interactions. In two further steps, (i) the radiation scheme is slightly revised to take into account consider the cloud droplet size information (so far considered constant even when applying the two-moment cloud microphysical scheme), and (ii) a diagnostic tool, the Cloud Feedback
- 15 Model Intercomparison Project Observational Simulator Package (*Bodas-Salcedo et al.*, 2011, 2008; *Nam and Quaas*, 2012) is implemented that allows for a consistent evaluation using satellite observations. The paper is organized as follows; section 2 gives a brief introduction to the coupled model systems, datasystem COSMO-MUSCAT, data, and methodology. The comparison between the improved two-moment cloud microphysical parameterization with the available two-moment scheme making use of the COSP satellite simulator is discussed in section 3. Finally, concluding remarks are given found in section 4.

20 2 Data and Methodology

2.1 The COSMO-MUSCAT model and revised cloud activation

The non-hydrostatic three-dimensional model, COSMO, which was originally developed for limited-area operational predictions (Doms et al., 1999; Steppeler et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 1999; Steppeler et al., 1999; Steppeler et al., 1999; Steppeler et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 1999; Steppeler et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 1999; Steppeler et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 1999; Steppeler et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 1999; Steppeler et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 1999; Steppeler et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 1999; Steppeler et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 2003) numerical weather predictions (NWP), is used in this study (Doms et al., 2003) num

- 25 vice (Deutscher Wetterdienst, DWD) (Baldauf et al., 2011)(Deutscher Wetterdienst, DWD, Baldauf et al., 2011). In this study, we have used COSMO version 5.0, which is initialized and forced by reanalyzed data provided by the global meteorological model GME (Global Model of the Earth) of DWD, which is a hydrostatic weather prediction model (Majewski et al., 2002). GME operates on an icosahedral hexagonal icosahedral-hexagonal grid having a horizontal resolution of approximately 40 km and vertical resolution of 40 layers up to 10 hPa. The COSMO model is initialized with the interpo-
- 30 lated GME initial state and nested within GME with hourly updates of lateral boundary values. In this study, the COSMO model has been configured in a non-convection non-convection permitting mode with a uniform horizontal grid resolution of 0.25° (≈28 km). The two-moment scheme in COSMO model consists of cloud microphysics scheme in the COSMO model (*Seifert and Beheng*, 2006) distinguishes between five hydrometeors classes, namely cloud droplets, rain, ice crystals, snow,

and graupel. Processes in the warm (liquid) phase considered by this scheme include the nucleation of cloud droplets, autoconversion of cloud droplets to form rain, accretion, and self-collection of water rain droplets. The formulations have been derived by *Seifert and Beheng* (2001) from the theoretical formulation of *Beheng and Doms* (1986). However, the radiation scheme does not yet make use of the additional information about cloud particle sizes provided by the two-moment microphysics. It

5 uses the *Ritter and Geleyn* (1992) parameterization for the cloud optical properties in radiation scheme. According to *Ritter* and Geleyn (1992), the cloud optical properties were are approximated by the relation between specific liquid water content q_c , and cloud effective radius r_e of cloud drop size distribution, thus cloud. Thus cloud optical depth τ_c is expressed as,

$$\tau_c = (c_1 + \frac{c_2}{r_e})q_c dz \tag{1}$$

where dz is layer thickness, and c_1 and c_2 are constants. Similarly, the effective radius r_e is related to specific cloud water 10 content and is approximated as,

$$r_e = c_3 + c_4 q_c \tag{2}$$

where c_3 and c_4 are constants (*Ritter and Geleyn*, 1992). In order to take into account of in to account the two-moment microphysics scheme, the simulated variable cloud droplet size, the cloud optical properties in radiation scheme have been microphysics in radiation scheme the cloud optical properties have to be modified. The cloud effective radius r_e is derived by di-

15 viding the third and second moment of the size distribution (*Martin et al.*, 1994) (*Martin et al.*, 1994; *Morrison et al.*, 2008) which, after rearranging, yields,

$$r_e = \frac{\Gamma(\mu+4)}{2\lambda\Gamma(\mu+3)} \tag{3}$$

where μ is the spectral shape parameter , (here, $\mu = 2$), Γ is the gamma distribution function and λ is the slope parameter, which is given by

$$20 \quad \lambda = \left[\frac{\pi \rho N_d \Gamma(\mu+4)}{6q_c \Gamma(\mu+1)} \frac{\pi \rho_w N_d \Gamma(\mu+4)}{6\rho q_c \Gamma(\mu+1)}\right]^{\frac{1}{3}} \tag{4}$$

where ρ is the density of the air, $\rho_w = 1000 \text{ kg m}^{-3}$ is the bulk density of liquid water, N_d is the droplet number concentration, and q_c is the specific water content. The corresponding cloud optical depth is given by

$$\tau_c = \frac{3\rho q_c dz}{2\rho_w r_e} \tag{5}$$

where, dz is the layer thickness, $\rho_w = 1000 \text{ kg m}^{-3}$ the is the bulk density of liquid water and the droplet size spectrum is considered vertically constant in the grid layer.

25

The online coupled model system COSMO-MUSCAT (*Wolke et al.*, 2012; *Renner and Wolke*, 2010; *Wolke et al.*, 2004) (*Wolke et al.*, 2010) used for prognostic cloud condensation nuclei in the cloud microphysics parameterization in COSMO model. The chemistry/aerosol transport model , MUSCAT treats atmospheric transport as well as chemical transformationreactions, with the Regional Atmospheric Chemistry Mechanism (RACM) (*Stockwell et al.*, 1997)(RACM, *Stockwell et al.*, 1997). In MUSCAT, all meteorological fields are given with respect to the uniform horizontal meteorological grid from the online coupled COSMO2M (COSMO with two-moment scheme) model, whereas the aerosol information is fed back to the COSMO2M model from MUS-CAT. In the previous setting, the interactions only considered the radiative effects of aerosols (scattering and absorption of solar radiation), as well as the scavenging of aerosol and in-cloud aerosol chemistry. A diagram illustrating the COSMO-MUSCAT

5 modeling set up is shown in Figure 1. In COSMO model, the the COSMO model with two-moment approach, the nucleation of cloud droplets has been treated explicitly and the aerosol activation parameterization is based on empirical activation spectra, which is in the form of power lawrelation,

$$N_{ccn} = C_{ccn} S^k, S in \%$$
⁽⁶⁾

where S is supersaturation, C_{ccn} = 1.26×10⁹m⁻³, and k = 0.308 for continental condition conditions or C_{ccn} = 1.0×10⁸m⁻³
and k = 0.462 for maritime condition (*Khain et al.*, 2001). conditions (*Khain et al.*, 2001). *Seifert et al.* (2012) investigated the influence of substantially perturbing C_{ccn} from 100 to 3200 cm⁻³ (see above for a brief discussion of this paper). Accordingly, the grid scale explicit nucleation rate is calculated from the time derivative of the activation relation (*Seifert and Beheng*, 2006),

$$\frac{\partial N_c}{\partial t}\bigg|_{nuc} = \begin{cases} C_{ccn}kS^{k-1}\frac{\partial S}{\partial z}w, & \text{if } S \ge 0, w\frac{\partial S}{\partial z} > 0, \\ & \text{and } S < S_{max}, \\ 0 & \text{else} \end{cases}$$
(7)

- 15 The above parameterization scheme uses constant C_{ccn} concentrations in accordance with different atmospheric conditions. Also, and S_{max} varies with atmospheric conditions (maritime in maritime conditions, C_{ccn} assumes that at $S_{max} = 1.1\%$, all C_{ccn} are already activated). In the above equation, nucleation is explicitly depends on grid scale supersaturation in combination with saturation adjustment assumed in the cloud scheme, which has limitations . This has been overcome by applying an operator splitting method to treat process numerically (Seifert and Beheng, 2006). As an initial step, a coupled model simulation
- 20 is carried out by setting $S_{max} = 2.0\%$, which is the optimum the condition for intermediate aerosols in COSMO model. FurtherIn a second step, we have used simulated sulfate (SO_4) aerosol mass concentration information from MUSCAT model (the MUSCAT model. The emission inventory in the MUSCAT model is proved by TNO for provided by the TNO (European emissions processing, and stands for Nederlandse Organisatie voor toegepast-natuurwetenschappelijk onderzoek (Netherlandse Organisation for Applied Scientific Research)) for the Air Quality Model Evaluation International Initiative (AQMEII) project
- 25 (Pouliot et al., 2012)) to derive C_{ccn} concentration proxy is derived using the following empirical relation (Boucher and Lohmann, 1995),

$$C_{ccn} = 10^{\underline{2.21+0.41log(mSO_4)2.21+0.41\log(mSO_4)}}_{\underline{}}$$
(8)

where mSO_4 is the sulfate aerosol mass concentration in $\mu gm^{-3} \mu gm^{-3}$. The constant C_{ccn} in the equation (7) is replaced by the spatially and temporally varying C_{ccn} values, derived from equation (8), using the sulfate aerosol mass concentra-

30 tion from the MUSCAT module. Even though, this empirical relationship that This empirical relationship which links sulfate aerosol mass concentration to C_{ccn} are widely used is subject to substantial uncertainty. Representing sulfate aerosol as a surrogate for all aerosols is probably too simple simplistic to capture the complexity of the whole activation process. Future work will introduce a more complex aerosol-cloud coupling, taking into account also other aerosol compounds. The Seifert and Beheng (2006) cloud microphysical scheme considers both phases. Also mixed-phase clouds are affected by the revision of the CCN parameterisation, e.g. via the Bergeron-Findeisen process. Nevertheless, the current analysis focuses on

5 the liquid phase only, investigations on mixed and ice-phase clouds are left for future research.

2.2 Model evaluation method

Satellite retrievals have been used to evaluate the performance of the numerous GCMs and NWP models (*Quaas et al.*, 2004, 2009; *Zhang* However, a meaningful evaluation of modeling with satellite observations is challenging because of the difference in the model variables and the satellite retrievals. To address this problem, the integrated satellite simulator COSP (CFMIP Observational

- Simulator Package, *Bodas-Salcedo et al.*, 2011) has been developed within the framework of the Cloud Feedback Model Intercomparison Project (CFMIP). The COSP satellite simulator produces model diagnostics, which are fully consistent to with satellite products such as , the International Satellite Cloud Climatology Project (ISCCP; *Rossow and Schiffer*, 1999), MODerate Resolution Imaging Spectroradiometer (MODIS; *Platnick et al.*, 2003; *Pincus et al.*, 2012), Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO; *Chepfer et al.*, 2010) and the CloudSat cloud radar (*Marchand et*
- 15 al., 2009). To produce similar output to satellite data, COSP requires the grid mean vertical profile of temperature, humidity, hydrometer mixing rationatios, cloud optical thickness and emissivity, the surface temperature and emissivity from the model. It produces the output comparable with satellite data in three steps. First, it addresses the mismatch between model and satellite pixel resolution by generating sub-columns using model information about subgrid-scale variability e.g. from the assumption on vertical overlap of fractional cloudiness, second, second vertical profiles of individual sub-columns are passed to
- 20 each instruments and finally instrument simulator, and third the COSP statistic module gathers the output from all instruments (*Bodas-Salcedo et al.*, 2011). Since COSP is running online with COSMO model, it is able to produce outputsimilar to model simulation (in every hour). The positional errors due to mismatch between meteorological regimes in the observation and models is not considered in COSP satellite simulator. This tool instrument simulators (*Bodas-Salcedo et al.*, 2011). COSP is implemented online in the COSMO model with hourly output. While using COSP facilitates a more consistent comparison
- 25 between model output and satellite data, differences between the model simulation and the satellite can for example still arise due to displacements in simulated storm tracks. COSP has previously been used with COSMO by *Muhlbauer et al.* (2014, 2015). The output diagnostics include a variety of cloud properties, which enables consistent inter-model and facilitate consistent model-to-observation comparisons . In spite of COSP satellite products, CERES Clouds and the Earth's Radiant Energy System, *Loeb et al.* (2012)satellite observations are also used for model evaluations. One should keep in mind that
- 30 the satellite products, just like models, are prone to biases. Comparisons of satellite retrievals with in-situ measurements have shown overestimation. Nonetheless, spatial correlation of the cloud structures are well represented(*Noble and Hudson*, 2015; *Min et al.*, 20 In the next step, we evaluate the model results in terms of cloud optical and microphysical properties with MODIS as well as consistent inter-model comparisons.

In the next section, we evaluate the model results (derived using MODIS and the ISCCP satellite simulators) with MODIS level-2 data sets. In all model versions (COSMO2M, COSMO2MR and COSMO-MUSCAT), we make use of the MODIS simulator diagnostics. The different swath data sets and ISCCP satellite observations, in terms of cloud optical and microphysical properties (cloud optical depth, effective radius, liquid water path and cloud fraction). The MODIS satellite simulator uses

5 profiles of particle size for liquid and ice and corresponding optical depths within each layer of sub-column as a function of model levels. Using the cloud overlap assumption, zero or one cloudiness in each sub-column is created in each level. The diagnostics are then integrated over the cloudy sub-columns to obtain in-cloud average cloud optical depth and liquid water path. In turn, cloud effective radius is sampled at the cloud tops, which is not a vertical integral. Further, the ISCCP simulator aggregates pixel scale cloud retrievals (fraction of the sub-column with $\tau \ge 0.3$) to estimate cloud fraction (more details: *Pincus et al.*, 2012)

10

The precision of weather forcasts for longer times is inherently limited by the non-linear nature of the problem. As forecast progresses, the uncertainty in weather prediction also increases. In turn, the earliest forecast time-steps are still substantially affected by the initialization. Hence, we have considered the third day of the simulation for evaluating the model with satellite observations. The synoptic condition which is discussed in the next section. To compare with model simulations, different

- 15 swath data-sets of MODIS level-2 on 17 February 2007 (day time daytime overpass only) are combined and gridded to the model domain . To reduce the uncertainty in cloud phase, MODIS (Terra) and model outputs are averaged between 8.00-14.00 UTC (corresponding approximately to the MODIS-Terra overpass time over the domain). Also, MODIS level-2 products and model simulations are screened for liquid phase clouds onlybecause in COSMO-MUSCAT only cloud microphysics for liquid clouds was modified. Additionally, MODIS cloud optical depth and effective radius are applied with threshold values of 5 and
- 20 2μm (Sourdeval et al., 2016; Zhang et al., 2012a). Further, the COSP-diagnosed Since the analysis is carried out for winter, satellite retrievals can be affected by snow cover on the ground. However, the MODIS retrieval (*Platnick et al.*, 2001) uses a combination of absorbing spectral channels for which the snow/ice albedo is relatively small which makes it suitable for retrieving cloud properties over snow. Furthermore, the COSP diagnosed model clouds are compared to ISCCP daily cloud products. To compare with ISCCP satellite retrievals, model results For that, modeled ISCCP cloud products are re-gridded
- 25 from 28 km to 280 km resolution, (ISCCP satellite resolution) using a grid interpolation method and model outputs are daily averaged. Besides these satellite observations, Clouds and the Earth's Radiant Energy System, (CERES, *Loeb et al.*, 2012) satellite observations are also used for model evaluation, which are daily products (*Kato et al.*, 2003). One should keep in mind that the satellite products, just like models, are prone to biases. Nonetheless, the spatial correlation of the cloud structures is well represented (*Noble and Hudson*, 2015; *Min et al.*, 2012).

30 3 Results for a case study

The simulations are carried out for a time period of 10 days (15 - 25 February 2007). The weather is evidently a complex processes which exhibits lots of variations. As forecast time progress the uncertainty in weather prediction also increases.

Hence, we have considered third day of the simulation for validating model and satellite simulators. Moreover, the synoptic conditions are favorable for comparisons, which discussed in the next section.

2.0.1 Numerical Simulations

To isolate and analyse analyze the effects of the modifications, three different simulations were carried out, (a) model modifications,

- 5 we have performed three different model simulations with the same interpolated GME initial conditions for the time period of 10 days (15 24 February 2007). They are (a) a standard COSMO two-moment simulation with fixed C_{ccn} (COSMO2M), with fixed CCN (3.0×10⁸m⁻³) (COSMO2M), (b) a COSMO two-moment simulation with radiation coupled to cloud microphysics (COSMO2MR), with fixed CCN (3.0×10⁸m⁻³), which uses equation which uses equations 3 to 5 in radiation scheme the radiation scheme (here also C_{ccn} is kept fixed as in COSMO2M), and (c) coupled a coupled COSMO-MUSCAT simulation,
- 10 i.e. using interactive rather than prescribed $\frac{\text{CCN}}{\text{concentrations}} C_{ccn}$ and treating radiation in the same way as COSMO2MR (COSMO-MUSCAT). In most of the discussion we have used simulations (a) and (c). In all three model versions (COSMO2M, COSMO2MR, and COSMO-MUSCAT), we make use of the COSP diagnostics for the MODIS and ISCCP satellite simulators.

3 Results for a case study

15 3.1 Synoptic situation

The simulation starts on 15 February and ends on 25-24 February 2007. At the beginning of the simulation (00:00 UTC), the meteorological condition is dominant by low pressure system over dominated by a low-pressure system over the north Atlantic and high pressure systems over the a high-pressure system over land. The 2-m temperature still shows shows a temperature gradient with a warm ocean and a cool continent, mostly in the northeastern part of the domain. The winds are mostly strong

- 20 southwesterly over the Atlantic and northerly and northwesterly in the southern region as well (Figure S1). Since the case study has been conducted for 17 February, the model derived key meteorological parameters -at 12:00 UTC are illustrated in Figure 2. On February 17, the low pressure system has been low-pressure system has moved to the French Atlantic coast and a cyclonic circulation has been setup set up over the region. FurtherFurthermore, a strong high pressure is clearly seen over northeastern Europe. The 2-m temperature shows that -prominent winter synoptic condition conditions still exist in the northern part with a
- warm oceanic region (Atlantic) and cold northeastern part. The southern region has a maximum temperature of $20^{\circ}C$, whereas the northeastern continental region experiences a minimum temperature of $-20^{\circ}C$. The cyclonic circulation drives the airmass from the oceanic region and results in the formation of clouds along the eyelonic circulation frontal systems. Besides, the airmass from the high pressure results in cloud free region in the middle high pressure in the Eastern part of the domain results in a cloud-free region due to subsidence. However, most of the domain is cloud covered with cloud fraction close to 100%.
- 30 Further, the total amount of rainfall Furthermore, rainfall around 100 mm (accumulated precipitation over 96 h) on 17 February is observed along with the cyclonic circulation and the south eastern part of Europe, with highest value over south of the low

pressure system, which is 100 kgm³. The . The modeled convective cloud bases are observed located between 500 to 4000 m over the domain.

3.2 Evaluation with satellite data

The model derived cloud fraction is daily averaged to (0:00 to 24:00 UTC) to illustrate the comparison between the model
(COSP) and ISCCP satellite retrievals (Figure 3). The observed cloud fraction shows more cloud free cloud-free regions compared to the model simulations. This may arise due to the coarse (280 km resolution) resolution of the satellite observation or poor parameterization of clouds in the model. Nevertheless, it is evident that Nevertheless, the model derived cloud fraction is in broad agreement with ISCCP satellite retrievals, allowing now for a more detailed analysis of the cloud microphysical properties with a fine resolution which is the center of this study. Further, flux comparison-Furthermore, a comparison of radiative fluxes with CERES (Clouds and the Earth's Radiant Energy System) satellite products are is discussed in section 3.3.

- Figure 4 shows the comparison between MODIS observed and model simulated (averaged between 8.00 -14.00 UTC, COSPretrieved (Figure 4a-c) and COSMO-MUSCAT simulated (Figure 4d-f) cloud optical depth, cloud droplet effective radius, and cloud liquid water path, respectively. In general, we find . From the figure, it can be noticed that the simulated cloud optical depth exhibits a spatial pattern similar to the observations, with a albeit higher magnitude in the MODIS level-2
- 15 retrievals (Figure 4a and d). In satellite the satellite retrievals, it varies between 5 to 54 and in 100 and in the model between 5 to 45, with maximum values observed over similar geographical regions. However, the satellite derived satellite-derived cloud optical depth and liquid water path are overestimated while comparing-larger in comparison with model (COSMO-MUSCAT) outputs. Although the The model derived cloud effective radius is underestimated compared to MODIS data, both exhibit exhibits both a similar spatial pattern and magnitude compared to that of the MODIS satellite retrievals (Figure 4b and e). The
- 20 model modeled cloud droplet effective radius varies between 3 to 1416μ m, whereas it is in the range between 2 to 2030μ m in the satellite retrievals. The spatial pattern clearly indicates that, satellite derived cloud effective radius is overestimated, which may be due to the horizontal heterogeneity and it is specially visible in marine stratocumulus. Also, note that MODIS possibly overestimate cloud droplet effective radius (*Min et al.*, 2012; *Noble and Hudson*, 2015). The effect of marine stratocumulus is also visible in the case of observed MODIS cloud optical depth and cloud water path. Similar to cloud optical depth, eloud
- 25 liquid water path also exhibit exhibits comparable spatial patterns for both, model and observations. Its simulated magnitude also is in broad agreement with the satellite retrievals, with an underestimation in the model mainly over central eastern Europe and over the Atlantic coast. The cloud water path in both cases ranges between about 0.025 and Whereas the modeled liquid water path varies between 0.025 and 0.425 kgm^{-2} , and in the satellite observation it varies between 0.25 and 1.0 kgm^{-2} . The white regions region (missing values) in satellite retrievals can be explained by the very strict quality-filtering of the
- 30 MODIS cloud products. The domain averaged cloud optical depth, effective radius and liquid water path are 23.34, 11.30 μm , 0.175 kgm⁻², whereas the COSMO-MUSCAT derived values are 7.60, 9.93 μm , and 0.056 kgm⁻², which illustrates an underestimation of all simulated quantities compared to the satellite derived cloud optical properties. The above cloud optical properties are calculated using equations 3 and 5 in the models, although their correlations are valid only for that particular model layer/levels.

Even though, spatial distribution of satellite and model cloud microphysical propertiescan be compared and validated, absolute comparison like spatial correlation and area mean can add uncertainty to the analysis. To overcome this, we have evaluated the statistical representation of In the following, the statistical distribution of satellite and the model cloud micro-physical properties as are compared and evaluated in terms of probability density functions (PDFs)corresponding to model

- 5 (COSMO-MUSCAT) and satellite, which can account for different resolution of model and satellite instruments (Figure 5). The solid lines in . Figure 5 indicates, PDF for 17 February and the dashed line indicates for entire simulation period (15-24 February 20017). Figure 5 indicates that, in represents the probability density function of the spatiotemporal distribution of cloud optical depth, effective radius, and liquid water path, the model shows an overestimation in the lower range (below 0.08 kgm⁻²) and underestimation above 0.08 kgm⁻², which is same for both cases (17 February and overall). For defined as
- 10 the normalized count of occurrence per bin width of cloud optical property. The cloud optical depth , the model overestimate low clouds (optical depth below PDF shows that thin clouds (cloud optical depth < 10) and underestimate high clouds (above 20). In the case of in all model versions occur substantially more frequently than in the satellite retrievals, and thick clouds (cloud optical depth > 30), less frequently. The modeled cloud effective radius , overestimation is between 6 and 12PDFs is constrained to 3 and 16 μ mand underestimation above 12, where as the satellite retrievals shows a range of 4 to 30 μ m, which
- 15 is same for both cases. This clearly indicates that, 17 February can be a representative for the entire simulation to compare with satellite observations. A shift of the PDF is found in the COSMO-MUSCAT derived PDFs, which indicates the increased droplet size for the interactive C_{ecp} . For liquid water path, modeled PDFs overestimates the clouds with low liquid water path and underestimates clouds with high water paths. The differences in PDFs largely follow what is found for the cloud optical depth, but model deficiencies compared to the satellite retrievals are substantially larger. The analysis also illustrates
- 20 an increased in cloud optical PDF from COSMO-MUSCAT simulation. Certainly, the drop and preponderance of modeled cloud optical properties can be influenced by model tuning, an approach which, however, hasn't been performed yet for the COSMO-MUSCAT model version.

The outcome of eloud microphysics modification is the cloud microphysics modifications can be analyzed by considering the difference between the two simulations (COSMO-MUSCAT and COSMO2M), which is shown in Figure 4g, h, and i4g-i.
The version considering the interactive aerosol number concentration (COSMO-MUSCAT) exhibits an increase in the cloud droplet effective radius by a range of 1-4 μm throughout the domain with an overall increase of 9.5%, although a slight reduction can be noticed in a few areas. This indicates the impact of the activation and growth of the sulphate aerosol from MUSCAT model. In the case of For cloud optical depth and eloud liquid water path, both generally show an increases despite

of increases despite the reduction in a few areas. The revised parameterization in the coupled model has made modification in modified the spatial distribution of the cloud optical depth in the range of \pm 15 and the liquid water exhibits a variation in the range of \pm 0.12 kgm⁻². For the cloud droplet effective radius, the revised model version (COSMO-MUSCAT) represented the retrieved distribution better compared to other two variables. Additionally, analyzing the difference between COSMO2MR with COSMO2M accounts for the cloud mircophysics modification in COSMO-MUSCAT modelHowever, the domain averaged cloud optical depth and liquid water path has been increased by 4.1% and 14.2%, which is also observed in PDF analysis. Cloud The cloud droplet number concentration N_d can be used as a diagnostic for aerosol cloud interaction. From satellite observation, observations it can be expressed in terms of cloud optical depth τ_c and effective radius r_e (Quaas et al., 2006), which is given by,

$$N_d = \alpha \tau_c^{0.5} r_e^{-2.5} \tag{9}$$

- 5 where $\alpha = 1.37 \times 10^{-5} m^{-0.5}$. Uncertainty in deriving Likewise, the model derived N_d can arise from satellite droplet effective radius. In order to compare the model simulation with satellite observations, we have used above equation to compute model N_d , as the COSP simulator can provide is also estimated using equation 9, which uses COSP (MODIS simulator) derived cloud optical depth and effective radiussimilar to MODIS satellite. Figure 6 shows N_d comparison between models and satellites. From the figure it is noticed that, model derived N_d values are underestimated (a-c) shows the comparison between modeled
- 10 (CSOMO2M and COSMO-MUSCAT) and observed N_d . On 17 February 2007, the domain-averaged N_d values are 153, 120, and 378 cm⁻³ for COSMO2M, COMSO-MUSCAT and MODIS, which indicates an underestimation of model derived values (Figure 6a-c) compared to MODIS - Again, in comparison with (*Zhang et al.*, 2012b; *Storelvmo et al.*, 2009). The inter-model comparison (COSMO2M , an underestimation is noticed in and COSMO-MUSCAT) reveals that COMSO-MUSCAT derived N_d , which is in the order of 100 to 200 cm⁻³. This underestimation can be explained by cloud microphysics modification
- 15 in coupled model. In the basic version of the COSMO2M, the CCN is fixed as 300 cm^{-3} (for intermediate aerosol types), whereas the coupled model uses gridded CCN (Cloud Condensation Nuclei) information from the MUSCAT model. Figure shows a decrease of 21.5%. Figure 6d shows the vertically and daily averaged sulfate aerosol number concentration, which varies between 20 to 300 cm^{-3} . From figure 6d, maximum aerosol mass concentration is observed over south eastern shows the spatial distribution of sulfate aerosol number concentration (aerosol number concentration proxy) below the convective cloud
- 20 base (representative of aerosols in the model and it is also averaged for 8-14 hours on 17 February 2007), where high number concentrations are simulated over southeastern Europe. On the contrarycontrast, N_d shows less are smaller over the same region. This is because *Boucher and Lohmann* (1995) parameterization shows the *Boucher and Lohmann* (1995) parameterization models saturation of N_d over high aerosol or polluted regions (*Penner et al.*, 2001) and the high pressure in this region result results in trapping aerosol below in the boundary layer. Further, it may be difficult to correlate the spatial patterns of aerosol
- 25 number concentration and cloud droplet number concentration because the droplet activation also controlled by several other meteorological properties, such as vertical velocity, microphysical links. However, model derived cloud optical properties strongly correlated. From the above analysis, it can be inferred that COSMO-MUSCAT can be used as a tool for regional aerosol-cloud interaction estimates. The interactive aerosol coupling results show an increase in cloud droplet effective radius by 9.5% and a reduction in N_d by 21.5%.
- 30 While comparing modified two-moment scheme results with MODIS level-2 satellite products, the model shows more cloud free (clear) grid points. This indicates that model is unable to capture the sub grid scale cloud patterns accurately (?), which may be due to the coarse resolution (0.25°) of the model. Further, the satellite retrieval (mainly thin clouds) are affected by snow cover, which could be rather ignored. The COSP satellite simulator derives the cloud information using specific cloud water content, ice content and snow content from cloud microphysical scheme. Additionally, the model simulations via COSP

is able to reproduce spatial patterns similar/comparable to that of satellite observations regardless of overestimation of satellite retrievals (MODIS), which are reported in previous studies.

3.3 Impact on radiative balance

In addition, we have also implemented aerosol-cloud-radiation interactions in the COSMO model, by revising the radiation scheme in order to make use of a droplet-size-dependent cloud optical depth. Incorporating aerosol-cloud-radiation interactions in the model results in a causes significant change in the radiation fluxes. The analysis reveals an increase in shortwave wave flux distribution, which is in the order of 10 to 40 Wm^{-2} . Figure 7 shows the spatial distribution of net downward shortwave flux at the surfaceand 2 to 20 Wm^{-2} , net downward longwave flux at top of the atmosphere . In turn, the long wave flux distribution shows (TOA), and the corresponding difference between COSMO2M and COSMO2MR simulations (with fixed

- 10 C_{ccn}). Similar to the above analysis, we also compare fluxes for 17 February 2007, and are daily averaged. The figure shows that in the radiation modified version (COSMO2MR) there is an increase in the net downward shortwave flux at the surface. Likewise, an overall reduction in the range of -2 is observed in the net downward longwave flux at the TOA despite the increase in the northeast part of the domain. The net downward shortwave radiation at the surface shows an increase of about 10 to 60 Wm^{-2} and the net downward longwave flux shows a decrease of 10 to $-20 Wm^{-2}$ at the surface and top of the atmosphere.
- 15 An exception with some increase (20 to $2040 Wm^{-2}$) is noted top of the atmosphere in the northern part of the domain (Figure 7). It is also noted that the cloud microphysics radiation coupling results in reduction in cloud optical properties, which would results more downward shortwave. This illustrates the reduction of cloud cover in the COSMO2MR simulations, which implies that reduced cloud cover results in more shortwave radiation reaching the surface and less longwave radiation reflected back to TOA. This can be inferred by considering the cloud optical depth, and liquid water path difference between the COSMO2M
- 20 and upward longwave especially at the surface. Further, the effect of aerosol-cloud-radiation interaction can be seen to larger extent over ocean than over land, especially for surface net downward short wave and long wave fluxes. The boundary effect in the difference can be ignored, which arise due to different physics in COSMO and GME. In comparison with CERES Clouds and the Earth's Radiant Energy System, *Loeb et al.* (2012)satellite observations, the COSMO2MR simulations, which is also daily averaged (Figure 8). From the figure, the regions with reduced cloud optical depth and liquid water path are correlated
- 25 with increased net shortwave flux at the surface and decreased net longwave flux at the TOA. To illustrate the combined effect of revised radiation scheme and interactive aerosols, COSMO-MUSCAT derived radiative fluxes are compared with CERES satellite observations (*Kato et al.*, 2003; *Loeb et al.*, 2012). For comparison, we have considered computed CERES fluxes (derived based on state and composition of the atmosphere, surface, and the incoming solar radiation) with spatial resolution $1^{\circ} \times 1^{\circ}$ and care must be taken while interpreting the results. Also, during winter the uncertainty in CERES flux
- 30 observation are slightly higher (*Guo et al.*, 2007). The spatial pattern and the magnitude of model simulations simulated fluxes are comparable with satellite observations, however the differences are neither systematic nor large (Figure 8). Further, during winter the uncertainty in CERES flux observation are little higher (*Guo et al.*, 2007). The difference observed in cloud optical properties (Figure 4) can also be attributed from impact of radiative balancein which the surface net downward shortwave flux ranges between 20 to 26 Wm^{-2} and TOA net downward longwave flux varies between -290 to -140 Wm^{-2} (Figure 9).

Additionally, correlations between satellites and models (COSMO2M and COMO-MUSCAT, model outputs are re-gridded to satellite resolution) are illustrated in Figure 9 c and d. The models modifications (revised radiation scheme and interactive aerosols) result in an increase in the correlation coefficient from 0.61 (COSMO2M) to 0.84 (COSMO-MUSCAT) in the case of net shortwave flux at the surface, whereas the modifiations do not have much effect on the longwave flux.

5 4 Conclusions

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This paper discusses the modification of the *Seifert and Beheng* (2006) two-moment scheme in COSMO model. This has been done with <u>aerosol information from the</u> online-coupled MUSCAT model<u>aerosol information</u>, which allows for a microphysical aerosol effect on clouds. It has been achieved by replacing the constant cloud condensation nuclei profile in the COSMO two-moment scheme with gridded aerosol information derived from online-coupled MUSCAT model, using the *Boucher and*

- 10 Lohmann (1995) parameterization, which takes sulfate aerosol as a proxy for all aerosols. In addition, the radiation scheme was is revised to a droplet-size-dependent cloud optical depth, allowing now for aerosol-cloud-radiation interactions. In order to facilitate an evaluation using satellite retrievals, the COSP satellite simulator has been incorporated into the modeling system, which runs online with in the model. The model results are evaluated with satellite observations from the ISCCP, MODIS, and CERES projects and instruments, respectively. Since the cloud microphysics modification has been done for
- 15 cloud droplet nucleation, the analysis are restricted to the liquid part of clouds in the model and MODIS level-2 cloud products are screened for liquid phase cloud products. Although the two-moment cloud microphysics and radiation scheme in COSMO model has been modified, the model did was not re-tuned to get reasonable 2m temperature or precipitation. The conclusions are summarized below.

1. The modified two-moment scheme results have been compared with two-moment version of COSMO model. In terms of the cloud distributions, this modification has only a minor effect.

2.-A case study has been carried out to compare the model output with observations. Daily averaged cloud optical depth, droplet-The incorporated COSP satellite simulator serves as a link between model and satellite comparisons. Despite the resolution, COSP derived ISCCP cloud fraction shows similar spatial pattern and magnitude. Further, MODIS level-2 cloud optical products such as cloud optical depth, effective radius, and liquid water path are compared with MODIS level-2 products.

- 25 The interactive treatment of acorosls in COSMO-MUSCAT simulations show an improvement in the cloud microphysical properties. Further, the PDF analysis has contributed to a quantitative comparison of model reuslts with satellite observations. The cloud effective radius exhibits an increase and The COSMO-MUSCAT derived cloud optical properties show a similar spatial distribution compared to the MODIS observation. In COSMO-MUSCAT, the cloud optical depth has been increased by 4.1%, cloud droplet effective has been increased by 9.5%, and liquid water path has been increased by 14.2% in comparison
- 30 to CSOMO2M. In turn, the cloud droplet number concentration estimated from COSMO-MUSCAT model shows a reduction in the modified simulation. This is due to the reduced CCN number concentrations from the MUSCAT model. The satellite retrievals suggest the revised model version is more realistic in both quantities.

3. The representation of cloud microphysical properties in the radiation scheme has been revised in order to digest the additional information about cloud particle sizes the two-moment microphysics scheme offers. Again of 21.5% compared to the COSMO2M model. Furthermore, considerable changes in terms of the radiation budget were also found. The new approach now, however, allows to explicitly take into account the radiative effects of have been found. This analysis indicates that

5 the coupled model (COSMO-MUSCAT) with interactive aerosol treatment results in an increase in cloud droplet size and reduction in cloud droplet number concentration by activation and growth of droplets, which illustrates implicit aerosol-cloud interactions. Also, the cloud properties in COSMO-MUSCAT agree reasonably well with observations, so that it can be used for regional aerosol-cloud interaction studies.

In As a next step, further improvement in the two-moment scheme will be carried out through the use of the newly included aerosol model M7 (*Vignati et al.*, 2004) framework in the MUSCAT model, which is able to provide aerosol number concentration information to the COSMO two-moment scheme by replacing *Boucher and Lohmann* (1995) parameterization. This can result in more precise cloud droplet activation parameterization, involving different aerosol species as CCN*C*_{ccn}, and thus improving the cloud droplet number calculation of microphysical aerosol effect on clouds (*Lohmann et al.*, 2007). Also, the role of aerosols on ice nucleation will be addressed.

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Code and data availability

The COSMO-MUSCAT(5.0) model is freely available under public license policy. The source code, external parameters and documentation can be obtained through Ralf Wolke (wolke@tropos.de).

Acknowledgements. This work was supported by an ERC starting grant "QUAERERE" (GA no 306284). We acknowledge the development work by the COSMO consortium. We thank Axel Seifert for and anonymous reviewers for their valuable suggestions.

References

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- Ackerman, A. S., O. B. Toon, D. E. Stevens, A. J. Heymsfield, V. Ramanathan, and E. J. Welton: Reduction of tropical cloudiness by soot, *Science*, 288, 1042-1047, doi:10.1126/science.288.5468.1042, 2000.
- Ackerman, A. S., M. P. Kirkpatrick, D. E. Stevens, and O. B. Toon: The impact of humidity above stratiform clouds on indirect aerosol climate forcing, *Nature*, 432, 1014-1017, doi:10.1038/nature03174, 2004.
- Baldauf, M., A. Seifert, J. Förstner, D. Majewski, M. Raschendorfer, T. Reinhardt: Operational convective-scale numerical weather prediction with the COSMO Model: Description and sensitivities, *Mon. Wea. Rev., 139*, 3887-3905, doi: 10.1175/MWR-D-10-05013.1, 2011.

- Beheng, K. D., G. Doms: A general formulation of collection rates of clouds and raindrops using the kinetic equation and comparison with parameterizations, *Contrib. Atmos. Phys.* 59, 66-84, 1986.
 - Berner, A. H., Bretherton, C. S., Wood, R., and Muhlbauer, A.: Marine boundary layer cloud regimes and POC formation in a CRM coupled to a bulk aerosol scheme, *Atmos. Chem. Phys.*, *13*, 12549-12572, doi:10.5194/acp-13-12549-2013, 2013.
- 15 Bodas-Salcedo, A. A., M. J. Webb, S. S. Bony, H. H. Chepfer, J. L. Dufresne, S. A. Klein, Y. Y. Zhang, R. R. Marchand, J. M. Haynes, R. R. Pincus, and V. O. John: COSP: Satellite simulator software for model assessment, *Bull. Amer. Meteor. Soc.*, 92, 1023-1043, doi: 10.1175/2011BAMS2856.1, 2011.
 - Bodas-Salcedo, A., M. J. Webb, M. E. Brooks, M. A. Ringer, K. D. William, S. F. Milton, and D. R. Wilson: Evaluating cloud systems in the Met Office global forecast model using simulated CloudSat radar reflectivities, *J. Geophys. Res.*, 113, D00A13, doi:10.1029/2007JD009620, 2008.
 - Boucher, O., and U. Lohmann: The sulfate-CCN-cloud albedo effect: A sensitivity study with two general circulation models, *Tellus*, 47 Ser. *B*, 281-300, 1995.
 - Brunke, M. A., S. P. de Szoeke, P. Zuidema, and X. Zeng: A comparison of ship and satellite measurements of cloud properties with global climate model simulations in the southeast Pacific stratus deck, *Atmos. Chem. Phys.*, *10*, 6527-6536, doi:10.5194/acp-10-6527-2010, 2010.
- 25 Chepfer, H., S. Bony, D. Winker, G. Cesana, J. L. Dufresne, P. Minnis, C. J. Stubenrauch, and S. Zeng: The GCM oriented CALIPSO cloud product (CALIPSO-GOCCP), J. Geophys. Res., 115, D00H16, doi:10.1029/2009JD012251, 2010.
 - Chapman, E. G. and Gustafson Jr., W. I. and Easter, R. C. and Barnard, J. C. and Ghan, S. J. and Pekour, M. S. and Fast, J. D.:, Coupling aerosol-cloud-radiative processes in the WRF-Chem model: Investigating the radiative impact of elevated point sources, *Atmos. Chem. Phys.*, *9*, 945-964, doi:10.5194/acp-9-945-2009, 2009.
- 30 Cherian, R., C. Venkataraman, S. Ramachandran, J. Quaas, and S. Kedia: Examination of aerosol distributions and radiative effects over the Bay of Bengal and the Arabian Sea region during ICARB using satellite data and a general circulation model, *Atmos. Chem. Phys.*, 12, 1287-1305, doi:10.5194/acp-12-1287-2012, 2012.

Cotton, W.R., G. P. Tripoli, R. M. Rauber, E. A. Mulvihill: Numerical simulation of the effects of varying ice crystal nucleation rates and aggregation processes on orographic snowfall, *J. Clim. Appl. Meteorol.*, 25, 1658-1680, 1986.

35 Doms, G. and U. Schättler: The Nonhydrostatic Limited-Area Model LM (Lokal-Modell) of DWD: Part I: *ScientificDocumentation (Version LM-F90 1.35), Deutscher Wetterdienst*, Offenbach, 1999.

Bangert, M., Kottmeier, C., Vogel, B., and Vogel, H.: Regional scale effects of the aerosol cloud interaction simulated with an online coupled comprehensive chemistry model, *Atmos. Chem. Phys.*, *11*, 4411-4423, doi:10.5194/acp-11-4411-2011, 2011.

- Forkel R., A. Balzarini, R. Baró, R. Bianconi, G. Curci, P. Jiménez-Guerrero, M. Hirtl, L. Honzak, C. Lorenz, Ulas Im, J. L. Pérez, G. Pirovano, R. S. José, P. Tuccella, J. Werhahn, R. Žabkar: Analysis of the WRF-Chem contributions to AQMEII phase2 with respect to aerosol radiative feedbacks on meteorology and pollutant distributions, *Atmos. Environ.*, 115, 630-645, http://dx.doi.org/10.1016/j.atmosenv.2014.10.056, 2015.
- 5 Ghan, S., M. Wang, S. Zhang, S. Ferrachat, A. Gettelman, J. Griesfeller, Z. Kipling, U. Lohmann, H. Morrison, D. Neubauer, D. G. Partridge, P. Stier, T. Takemura, H. Wang, and K. Zhang: Challenges in constraining anthropogenic aerosol effects on cloud radiative forcing using present-day spatiotemporal variability, *PNAS 2016 113 (21)*, 5804-5811, doi:10.1073/pnas.1514036113, 2016.
- IPCC, 2013: Climate change 2013: The physical science basis. Contribution of working group I to the fifth assessment report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y.
- 10 Xia, V. Bex and P.M. Midgley (eds.)], Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 1535 pp, doi:10.1017/CBO9781107415324.

Otkin, J.A. and T.J. Greenwald: Comparison of WRF model simulated and MODIS derived cloud data, *Mon. Wea. Rev., 136*, 1957-1970, doi:10.1175/2007MWR2293.1, 2008.

Guo, S., L. Henry, and M. Murray: Surface-Absorbed and Top-of-Atmosphere Radiation Fluxes for the Mackenzie River Basin from Satellite

- 15 Observations and a Regional Climate Model and an Evaluation of the Model, *Canadian Meteorological & Oceanographic Society, v. 45, no. 3*, p. 129-139, 2007.
 - Kato, S. and N. G. Loeb: Twilight irradiance reflected by the Earth estimated from Clouds and the Earth's Radiant Energy System (CERES) measurements, *J. Climate*, *16*, 2646-2650, doi: 10.1175/1520-0442(2003)016<2646:TIRBTE>2.0.CO;2, 2003.

Kessler, E.: On the distribution and continuity of water substance in atmospheric circulations, Atmos. Res., 38, 109-145, doi:10.1016/0169-

20 8095(94)00090-Z, 1969.

35

Khain, A., M. Ovtchinnikov, M. Pinsky, A. Pokrovsky, H. Krugliak: Notes on the state-of-the-art numerical modeling of cloud microphysics, *Atmos. Res.*, 55, 159-224, http://doi.org/10.1016/S0169-8095(00)00064-8, 2000.

Khain A. P., D. Rosenfeld, A. Pokrovsky: Simulating convective clouds with sustained supercooled liquid water down to -37.5° using a spectral microphysics model, *Geophys. Res. Lett.* 28, 3887-3890, doi:10.1029/2000GL012662, 2001.

25 Li, G., Y. Wang, and R. Zhang: Implementation of a two-moment bulk microphysics scheme to the WRF model to investigate aerosol-cloud interaction, J. Geophys. Res., 113, D15211, doi:10.1029/2007JD009361, 2008.

Lim, K. Sunny, S. Hong: Development of an effective double-moment cloud microphysics scheme with prognostic cloud condensation nuclei (CCN) for weather and climate models, *Mon. Wea. Rev.*, 138, 1587-1612, doi: 10.1175/2009MWR2968.1, 2010.

Lin, Y.-L., R. D. Farley, H. Orville: Bulk parameterization of the snow field in a cloud model, J. Clim. Appl. Meteorol., 22, 1065-1092, doi:

30 10.1175/1520-0450(1983)022<1065:BPOTSF>2.0.CO;2, 1983.

Loeb, N. G., J. M. Lyman, G. C. Johnson, R. P. Allan, D. R. Doelling, T. Wong, J. Soden, and G. L. Stephens: Observed changes in topof-the-atmosphere radiation and upper-ocean heating consistent within uncertainty, *Nature Geosci.* 5, 110-113, doi:10.1038/ngeo1375, 2012.

Majewski, D., D. Liermann, P. Prohl, B. Ritter, M. Buchhold, T. Hanisch, G. Paul, W. Wergen, and J. Baumgardner: The operational global Icosahedral-Hexagonal Gridpoint Model GME: description and high-resolution tests, *J. Atmos. Sci.*, *139*, 319-338, 2002.

Lohmann, U., Stier, P., Hoose, C., Ferrachat, S., Kloster, S., Roeckner, E., and Zhang, J.: Cloud microphysics and aerosol indirect effects in the global climate model ECHAM5-HAM, *Atmos. Chem. Phys.*, *7*, 3425-3446, doi:10.5194/acp-7-3425-2007, 2007.

- Marchand, R., J. Haynes, G. G. Mace, T. Ackerman, and G. Stephens: A comparison of simulated cloud radar output from the multiscale modeling framework global climate model with CloudSat cloud radar observations, *J. Geophys. Res.*, 114, D00A20, doi:10.1029/2008JD009790, 2009.
- Martin, G. M., D. W. Johnson, and A. A. Spice: The measurement and parameterization of effective radius of droplets in the warm stratocumulus clouds, *J. Atmos., Sci., 51*, 1823-1842, doi: 10.1175/1520-0469(1994)051<1823:TMAPOE>2.0.CO;2, 1994.
- Meyers, M. P., R. L. Walko, J. Y. Harrington, W. R. Cotton: New RAMS cloud microphysics parameterization: Part II: The two-moment scheme, *Atmos. Res.* 45, 3-39, http://dx.doi.org/10.1016/S0169-8095(97)00018-5, 1997.
- Min, Q., Joseph, E., Lin, Y., Min, L., Yin, B., Daum, P. H., Kleinman, L. I., Wang, J., and Lee, Y.-N.: Comparison of MODIS cloud microphysical properties with in-situ measurements over the Southeast Pacific, *Atmos. Chem. Phys.*, 12, 11261-11273, doi:10.5194/acp-

10 12-11261-2012, 2012.

- Morrison, H., A. Gettelman: A new two-moment bulk stratiform cloud microphysics scheme in the community atmosphere model, Version 3 (CAM3). Part I: Description and numerical tests, *J. Climate*, *21*, 3642-3659, doi: 10.1175/2008JCLI2105.1, 2008.
- Morrison, H., G. Thompson, V. Tatarskii: Impact of cloud microphysics on the development of trailing stratiform precipitation in a simulated squall line: Comparison of one and two-moment schemes, *Mon. Wea. Rev., 137*, 991-1007, 2009.
- 15 Muhlbauer, A., T. P. Ackerman, R. P. Lawson, S. Xie, and Y. Zhang: Evaluation of cloud-resolving model simulations of midlatitude cirrus with ARM and A-train observations, J. Geophys. Res. Atmos., 120, 6597-6618. doi: 10.1002/2014JD022570, 2015.

Muhlbauer, A., E. Berry, J. M. Comstock, and G. G. Mace: Perturbed physics ensemble simulations of cirrus on the cloud system-resolving scale, J. Geophys. Res. Atmos., 119, 4709-4735, doi:10.1002/2013JD020709, 2014.

- 20 the ECHAM5 general circulation model using CALIPSO and CloudSat satellite data, J. Adv. Model. Earth Syst., 6, 300-314, doi:10.1002/2013MS000277, 2014.
 - Nam, C., and J. Quaas: Evaluation of clouds and precipitation in the ECHAM5 general circulation model using CALIPSO and CloudSat , J. Clim., 25, 4975-4992, doi:10.1175/JCLI-D-11-00347.1 2012.

Noble, S. R., and J. G. Hudson: MODIS comparisons with northeastern Pacific in situ stratocumulus microphysics, J. Geophys. Res. Atmos.,

- 25 *120*, 8332-8344, doi:10.1002/2014JD022785, 2015.
 - Penner, J. E., Andreae, M., Annegarn, H., Barrie, L., Feichter, J., Hegg, D., Jayaraman, Leaitch, R., Murphy, D., Nganga, J., and Pitari, G.: Aerosols, their Direct and Indirect Effects, in: Climate Change 2001: The Scientific Basis, Contribution of working group I to the Third Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Houghton, J. T., Ding, Y., Griggs, D. J., Noguer, M., Van der Linden, P. J., Dai, X., Maskell, K., and Johnson, C. A., *Cambridge Univ. Press, New York*, 881, 2001.
- 30 Penner, J. E. and Quaas, J. and Storelvmo, T. and Takemura, T. and Boucher, O. and Guo, H. and Kirkevåg, A. and Kristjánsson, J. E. and Seland, Ø.: Model intercomparison of indirect aerosol effects, *Atmos. Chem. Phy.*, *6*, *11*, 3391-3405, 2006.
 - Pincus, R., S. Platnick, S. A. Ackerman, R. S. Hemler, and R. J. P. Hofmann:, Reconciling simulated and observed views of clouds: MODIS, ISCCP, and the limits of instrument simulators, *J. Climate*, 25, 4699-4720. doi:10.1175/JCLI-D-11-00267.1, 2012.

Platnick, S., King, M. D., Ackerman, S. A., Menzel, W. P., Baum, B. A., Riedi, J. C., and Frey, R. A.: The MODIS cloud products: Algorithms

- and examples from Terra, *IEEE Trans. Geosci. Remot. Sens.*, 41, 459-473, 2003.
- Platnick, S., J. Y. Li, M. D. King, H. Gerber, and P. V. Hobbs: A solar reflectance method for retrieving the optical thickness and droplet size of liquid water clouds over snow and ice surfaces, *J. Geophys. Res.*, *106*(*D14*), 15185–15199, doi:10.1029/2000JD900441, 2001.

Nam, C., J. Quaas, R. Neggers, C. Siegenthaler-Le Drian, and F. Isotta: Evaluation of boundary layer cloud parameterizations in

- Pouliot, G., Pierce, T., Denier van der Gon, H., Schaap, M., Moran, M., Nopmongcol, U.: Comparing emissions inventories and model-ready emissions datasets between Europe and North America for the AQMEII project, *Atmos. Environ.*, 53, 4-14, http://doi.org/10.1016/j.atmosenv.2011.12.041, 2012.
- Possner, A., Zubler, E., Lohmann, U., and Schär, C.: Real-case simulations of aerosol-cloud interactions in ship tracks over the Bay of Biscay,
 Atmos. Chem. Phys., *15*, 2185-2201, doi:10.5194/acp-15-2185-2015, 2015.
- Quaas, J., Y. Ming, S. Menon, T. Takemura, M. Wang, J. Penner, A. Gettelman, U. Lohmann, N. Bellouin, O. Boucher, A. M. Sayer, G. E. Thomas, A. McComiskey, G. Feingold, C. Hoose, J. E. Kristjánsson, X. Liu, Y. Balkanski, L. J. Donner, P. A. Ginoux, P. Stier, B. Grandey, J. Feichter, I. Sednev, S. E. Bauer, D. Koch, R. G. Grainger, A. Kirkevág, T. Iversen, Ø. Seland, R. Easter, S. J. Ghan, P. J. Rasch, H. Morrison, J. -F. Lamarque, M. J. Iacono, S. Kinne, and M. Schulz: Aerosol indirect effects general circulation model intercomparison
- 10 and evaluation with satellite data, *Atmos. Chem. Phys.*, *9*, 8697-8717, doi:10.5194/acp-9-8697-2009, 2009.
 - Quaas, J., O. Boucher, and F. -M. Bréon: Aerosol indirect effects in POLDER satellite data and in the Laboratoire de Météorologie Dynamique-Zoom (LMDZ) general circulation model, J. Geophys. Res., 109, D08205, doi:10.1029/2003JD004317, 2004.
 - Quaas, J., O. Boucher, and U. Lohmann: Constraining the total aerosol indirect effect in the LMDZ and ECHAM4 GCMs using MODIS satellite data, *Atmos. Chem. Phys.*, *6*, 947-955, doi:10.5194/acp-6-947-2006, 2006.
- 15 Reisner, J., R. M. Rasmussen, and R. T. Bruintjes: Explicit forecasting of supercooled liquid water in winter storms using the MM5 mesoscale model, Q. J. R. Meteorol. Soc. 124, 1071-1107, doi:10.1002/qj.49712454804, 1998.
 - Renner, E., R. Wolke: Modelling the formation and atmospheric transport of secondary inorganic aerosols with special attention to regions with high ammonia emissions, *Atmos. Environ.*, 44, 1904-1912, http://doi.org/10.1016/j.atmosenv.2010.02.018, 2010.
- Ritter, B., and J. Geleyn: A comprehensive radiation scheme for numerical weather prediction models with potential applications in climate
 simulations, *Mon. Wea. Rev.*, *120*, 303-325, doi: 10.1175/1520-0493(1992)120<0303:ACRSFN>2.0.CO;2, 1992.
- Rossow, W. B., and R. A. Schiffer: Advances in understanding clouds from ISCCP, *Bull. Amer. Meteorol. Soc.*, 80, 2261-2288, doi:10.1175/1520-0477(1999)080<2261:AIUCFI>2.0.CO;2, 1999.
 - Sandu, I., J. L. Brenguier, O. Geoffroy, O. Thouron and V. Masson: Aerosol impacts on the diurnal cycle of marine stratocumulus, *J. Atmos. Sci.*, *65*, 2705-2718, doi: 10.1175/2008JAS2451.1, 2008.
- 25 Seifert, A., T. Heus, R. Pincus, and B. Stevens: Large-eddy simulation of the transient and near-equilibrium behavior of precipitating shallow convection, J. Adv. Model. Earth Syst., 7, 1918-1937, doi:10.1002/2015MS000489, 2015.

Seifert, A., C. Köhler, and K. D. Beheng: Aerosol-cloud-precipitation effects over Germany as simulated by a convective-scale numerical weather prediction model, *Atmos. Chem. Phys.*, 12, 709-725, doi:10.5194/acp-12-709-2012, 2012.

- Seifert, A. and K. D. Beheng: A two-moment cloud microphysics parameterization for mixed- phase clouds. Part 1: Model description,
 Meteorol. Atmos. Phys., 92, 45-66, doi:10.1007/s00703-005-0112-4, 2006.
- Seifert, A. and K. D. Beheng: A double-moment parameterization for simulating autoconversion, accretion and selfcollection, *Atmos. Res.*, 59-60, 265-281, http://doi.org/10.1016/S0169-8095(01)00126-0, 2001.
 - Simmel, M., Bühl, J., Ansmann, A., and Tegen, I.: Ice phase in altocumulus clouds over Leipzig: remote sensing observations and detailed modeling, *Atmos. Chem. Phys.*, *15*, 10453-10470, doi:10.5194/acp-15-10453-2015, 2015.
- 35 Steppeler, J., G. Doms, U. Schüttler, H. W. Bitzer, A. Gassmann, U. Damrath, Gregoric: Meso-gamma scale forecasts using the nonhydrostatic model LM, *Met. Atmos. Phys.*, 82, 75-96, doi:10.1007/s00703-001-0592-9, 2003.
 - Stevens, B., and G. Feingold: Untangling aerosol effects on clouds and precipitation in a buffered system, *Nature*, 7264, 607-613, doi:10.1038/nature08281, 2009.

- Stockwell, W.R., F. Kirchner, M. Kuhn, and S. Seefeld: A new mechanism for regional atmospheric chemistry modeling, J. Geophys. Res., 102(D22), 25847-25879, doi:10.1029/97JD00849, 1997.
- Storelvmo, T., U. Lohmann, and R. Bennartz: What governs the spread in shortwave forcings in the transient IPCC AR4 models?, *Geophys. Res. Lett.*, *36*, L01806, doi:10.1029/2008GL036069, 2009.
- 5 Sourdeval, O., C.-Labonnote, L., Baran, A. J., Mülmenstädt, J. and Brogniez, G.: A methodology for simultaneous retrieval of ice and liquid water cloud properties. Part 2: Near-global retrievals and evaluation against A-Train products. *Q.J.R. Meteorol. Soc.*, *142*, 3063-3081. doi:10.1002/qj.2889, 2016.
 - Tao, W.-K., J. Simpson, D. Baker, S. Braun, M. -D. Chou, B. Ferrier, D. Johnson, A. Khain, S. Lang, B. Lynn, C. -L. Shie, D. Starr, C. -H. Sui, Y. Wang and P. Wetzel: Microphysics, radiation and surface processes in the Goddard Cumulus Ensemble (GCE) model, *Meteor. and Atmos. Phys.*, 82, 97-137, doi:10.1007/s00703-001-0594-7, 2003.
- Thompson, G., P. R. Field, R. M. Rasmussen, W. D. Hall: Explicit forecasts of winter precipitation using an improved bulk microphysics scheme. part II: Implementation of a new snow parameterization *Mon. Wea. Rev., 136*, 5095-5115, doi: 10.1175/2008MWR2387.1, 2008.

10

- Vignati, E., J. Wilson and P. Stier: M7: An efficient size-resolved aerosol microphysics module for large-scale aerosol transport models, J. Geophys. Res. 109, D22202, doi:10.1029/2003JD004485, 2004.
- 15 Van den Heever, Susan C. and Cotton, William R.: Urban aerosol impacts on downwind convective storms, J. of Appl. Meteor. Climatol., 46, 828-850, doi: 10.1175/JAM2492.1, 2007.
 - Weverberg Van, K., E. Goudenhoofdt, U. Blahak, E. Brisson, M. Demuzere, P. Marbaix, J. -P. van Ypersele: Comparison of one-moment and two-moment bulk microphysics for high-resolution climate simulations of intense precipitation, *Atmos. Res.*, 147-148, 145-161, http://doi.org/10.1016/j.atmosres.2014.05.012, 2014.
- 20 Wolke, R., W. Schröder, R. Schrödner, E. Renner: Influence of grid resolution and meteorological forcing on simulated European air quality: a sensitivity study with the modeling system COSMO-MUSCAT, *Atmos. Environ.*, 53, 110-130, http://doi.org/10.1016/j.atmosenv.2012.02.085, 2012.
 - Wolke, R., O. Knoth, O. Hellmuth, W. Schröder and E. Renner: The parallel model system LM-MUSCAT for chemistry-transport simulations: Coupling scheme, parallelization and application, in: G. R. Joubert, W. E. Nagel, F. J. Peters, and W. V. Walter, Eds.,
- 25 Parallel Computing: Software Technology, Algorithms, Architectures, and Applications, Elsevier, Amsterdam, The Netherlands, 363-370, doi:10.1016/S0927-5452(04)80048-0, 2004.
 - Xue, H., and G. Feingold: Large eddy simulations of trade-wind cumuli:Investigation of aerosol indirect effects, J. Atmos. Sci., 63, 1605-1622, doi:10.1175/JAS3706.1, 2006.
 - Yang, Q. and Gustafson Jr., W. I. and Fast, J. D. and Wang, H. and Easter, R. C. and Wang, M. and Ghan, S. J. and Berg, L. K. and Leung, L.
- 30 R. and Morrison, H.: Impact of natural and anthropogenic aerosols on stratocumulus and precipitation in the Southeast Pacific: a regional modelling study using WRF-Chem, *Atmos. Chem. Phys.*, 11, 8777-8796, doi:10.5194/acp-12-8777-2012, 2012.
 - Zhang, M. H., et al.: Comparing clouds and their seasonal variations in 10 atmospheric general circulation models with satellite measurements, *J. Geophys. Res.*, *110*, D15S02, doi:10.1029/2004JD005021, 2005.
 - Zhang, Z., A. S. Ackerman, G. Feingold, S. Platnick, R. Pincus, and H. Xue: Effects of cloud horizontal inhomogeneity and drizzle
- 35 on remote sensing of cloud droplet effective radius: Case studies based on large-eddy simulations, J. Geophys. Res., 117, D19208, doi:10.1029/2012JD017655, 2012. 2012a.

Zhang, Y., P. Karamchandani, T. Glotfelty, D. G. Streets, G. Grell, A. Nenes, F. Yu, and R. Bennartz, Development and initial application of the global-through-urban weather research and forecasting model with chemistry (GU-WRF/Chem), J. Geophys. Res., 117, D20206, doi:10.1029/2012JD017966, 2012. 2012b.

Zubler, E. M., D. Folini, U. Lohmann, D. Lüthi, A. Mühlbauer, S. Pousse-Nottelmann, C. Schär, and M. Wild: Implementation and evaluation of aerosol and cloud microphysics in a regional climate model, *J. Geophys. Res.*, *116*, D02211, doi:10.1029/2010JD014572, 2011.

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Figure 1. COSMO-MUSCAT modeling system. Lefthand_Left hand side, setup of COSMO modeling system system with GME inputand Righthand, Right hand side: MUSCAT modeling system with land use and emissions.



Figure 2. Model synoptic conditions for 17 February 2007 at $\frac{0012:00hrs00UTC}{0012:00hrs00UTC}$, (a) Surface pressure in contours and 2 meter m temperature in closed contours (°C) as colour shading, (b) 500 mb wind vector vectors and total cloud area fraction as colour shading.



Figure 3. (a) Satellite and (b) model (COSMO-MUSCAT) derived ISCCP cloud fraction, for 17 February 2007 (daily averaged).



Figure 4. MODIS Level-2 (a) cloud optical depth, (b) cloud effective radius, (c) cloud water path, COSMO-MUSCAT derived (day-time averaged between 8.00-14.00 UTC, approximate MODIS-Terra overpass time over the domain) (d) cloud optical depth, (e) cloud effective radius, (f) cloud water path, and difference between COSMO-MUSCAT and COSMO-2M COSMO2M simulations (g,h,i), for 17 February 2007.



Figure 5. Probability density functions (PDF) of cloud optical depth, cloud effective radius, <u>cloud Liquid</u> water path from COSMO-MUSCAT (green), <u>COSMO2M</u> (red) and MODIS Level-2 products (greenblue), for 17 February 2007 (solid line) and for <u>entire-10 day period</u> (15-24 February 2007) simulation (dashed line).



Figure 6. Day time averaged cloud Cloud droplet number concentration (averaged between 8.00 -14.00 UTC, MODIS-Terra overpass time over the domain) for (a) COSMO-2M, (b) COSMO-MUSCAT, (c)MODIS level-2, and (d) Sulfate aerosol number concentration below the convective cloud base from MUSCAT model, for 17 February 2007.



Figure 7. Comparison and difference between short wave and long wave radiation fluxes surface and top of the atmosphere, and it is difference between two simulation (COSMO-2MR COSMO2MR radiation coupled minus COSMO-2MCOSMO2M).



Figure 8. Daily averaged cloud optical depth and liquid water path difference between COSMO2MR and COSMO2M on 17 February 2007.



Figure S1: Model synoptic conditions for 15 February 2007 at 00:00hrs, (aCERES) Surface pressure in contours and 2 meter temperature in closed contours, models (b) 500 mb wind vector COSMO2MR and total cloud area fractionCOSMO2M).

Figure S1: Model synoptic conditions for 15 February 2007 at 00:00hrs, (aCERES) Surface pressure in contours and 2 meter temperature in closed contours, models (b) 500 mb wind vector COSMO2MR and total cloud area fraction COSMO2M).

Figure 9. Comparison between short wave and long wave fluxes at surface and top of the atmosphere with CERES satellite fluxes (top panel: model COSMO-MUSCAT, bottom Panel: and correlation between satellite).

Figure S1: Model synoptic conditions for 15 February 2007 at 00:00hrs, (aCERES) Surface pressure in contours and 2 meter temperature in closed contours, models (b) 500 mb wind vector COSMO2MR and total cloud area fractionCOSMO2M).



Figure 10. Figure S1: Model synoptic conditions for 15 February 2007 at 00:00 UTC, (a) Surface pressure in contours and 2m (°C) as colour shading, (b) 500 mb wind vectors and total cloud area fraction as colour shading.