Reply to Referee 2

We thank the reviewer for the comments and suggestions to improve the manuscript. We think that most of the critique of our work is related to misunderstandings of the capabilities of the model we are using and we hope that we can clarify all issues which were raised in the reviewers comment.

(1) *At the end of the introduction it would be nice to have a list of the questions that the paper wants to address and why. Why have you chosen these episodes? What is the main purpose of the work?*

– We have rephrased the last paragraph of the introduction where we describe the objective of our work.

'The objective of our study is to consider the impact of dust as a CCN in regions which are often affected by dust such as outflow regions of the Sahara. In this study we focus on the Eastern Mediterranean, an area often affected by dust events. We have selected two case studies where cyclonic systems lead to the formation of deep clouds and significant precipitation over coastal areas as well as over the ocean, and where at the same time a dust plume originating in Northern Africa crossed the Mediterranean and strongly influenced aerosol concentrations and potentially clouds and precipitation.'

(2) *The paper suffers from a detailed description of the method that was applied to simulate the aerosol cloud interaction. Although several references are given that might describe the details the authors should address in the model description at least the following questions. Which kind of cloud scheme is used? Is it a bulk scheme? Is it a hybrid scheme? Are size distributions of the hydrometeors taken into account? Which processes (e.g. heterogeneous vs. homogeneous freezing) are taken into account? How is the radiative effect of the clouds that depends on cloud droplet and ice crystal numbers and sizes distributions treated? As this is a model study some equations my help to explain what is really used and in which way the interaction between the aerosol particles and the cloud quantities is accounted for. This is necessary, to understand the complexity of the model system and how it differs from other models used in previous and future studies. It is also not clear how the change in the soluble fraction is treated within the simulation. Please explain that in more detail; again maybe an equation would help to elucidate this.*

We have extended the model description.
As WRF-chem is a widely used model in the community which has been documented well by the model developers in numerous publications, we do not consider it appropriate to describe every part of the model in great detail in our manuscript. We list all processes which are related to clouds and which are included in the model setup and have included the appropriate references.
Cloud microphysics:
We used a two moment bulk microphysics scheme with five different hydrometeor classes (cloud water, rain water, ice, graupel, snow). The size distribution of each hydrometeor class is prescribed and fix during the simulation time. The implementation of the microphysics scheme into the WRF-chem code follows strictly the theoretical framework described by Lin et al. (1983). All processed included in this microphysics scheme are

listed in Fig.1, Lin et al. (1983) (e.g. homogeneous freezing, depositional growth of cloud ice).
The coupling of the microphysics scheme with the aerosol scheme in the model (prognostic treatment of cloud droplet number concentrations) is described by Chapman et al. (2009).
The treatment of aerosol indirect effects follows the work by Abdul-Razzak and Ghan (2002) and its implementation into WRF-chem is described by Chapman et al. (2009), section 2 and references therein (e.g. Ghan et al. (2001), section 3.2)
The treatment of aerosol direct effects is described by Fast et al. (2006), section 2.
We have added a parameterisation to the code treating the number of activated ice nuclei diagnostically, based on DeMott et al. (2010).
The only other change to the default code version of WRF-chem is the following (which we explain now in more detail in the manuscript):

The following equations describe the calculation of the maximum supersaturation following Abdul-Razzak and Ghan (2002).
S_{max}: maximum supersaturation, equation (1) from Abdul-Razzak and Ghan (2002):

$$S_{max} = \frac{S_m}{\left[f(\sigma)\left(\frac{\zeta}{\eta}\right)^{3/2} + g(\sigma)\left(\frac{S_m^2}{\eta+3\zeta}\right)\right]} \tag{1}$$

With:

$$S_m = \frac{2}{\sqrt{(B)}}\left(\frac{A}{3a_m}\right)^{3/2} \tag{2}$$

$$\zeta = \frac{2A}{3}\left(\frac{\alpha V}{G}\right)^{1/2} \tag{3}$$

$$\eta = \frac{(\alpha V)^{3/2}}{2\pi\rho_w\gamma\mathbf{N}} \tag{4}$$

(equations (4-6) from Abdul-Razzak and Ghan (2002); A: surface tension, B: hygroscopicity, V: updraft velocity, G: diffusion of heat and moisture to the particle, ρ_w: density of water, γ, α: coefficients in the supersaturation balance equation)

N : the total number of aerosol particles in each section (as MOSAIC uses a sectional representation of the aerosol size distribution).
As explained in the model description and as explained in detail by Zaveri et al. (2008) and Fast et al. (2006) the aerosol module currently only treats one type of aerosol and consequently bulk properties are used for describing the aerosol number, volume and mass required for calculating aerosol activation.

$\Longrightarrow$ We now assume that **N** is not the total number of aerosols in one section, but only a fraction of the total number of aerosol particles (i.e. the fraction we allow to get activated, we assume that a fraction of the coarse mode particles consists of insoluble dust which is not allowed to become activated).

$$N_{insol} = (N \cdot f_{dust})exp\left(\frac{-T}{\tau}\right) \tag{5}$$

$$N = N_{other} + (N_{dust} - N_{insol}) \tag{6}$$

f_{dust} : dust (number) fraction
N_{dust}: Number of dust particles
N_{other}: Number of particles with a chemical composition other than dust
T: Time since model start
τ : Half-lifetime of insoluble dust fraction $\longrightarrow$ this half-lifetime is varied in the different sensitivity scenarios.

(3) *The section 6 (Discussion and Conclusions) should be completely rewritten. It should really concentrate on the discussion and the conclusions based on the results that were achieved. I cannot see why the weakness of neglecting the GCCN is placed in this section. This should be moved to the model description. As the authors neglect the GCCN I cannot see the basis to speculate about their potential roles. This again should be moved to the section where a detailed model description including the advantages and disadvantages of the applied methods is given. The effect on temperature is discussed for the first time in the conclusion section which makes no sense.*

We think that the critique on section 6 is mainly related to a misunderstanding of the following statement given by us and we hope to clarify the confusion about the capabilities and limitations of the model system we used:
We wrote:
'A weakness of the model set up is that we did not explicitly account for dust as GCCN leading to larger cloud droplets which may increases the rate of precipitation formation due to efficient collection/coalescence of smaller droplets, as this is only possible with a prognostic cloud droplet size distribution. Thus in our model setup an increased number of CCN cloud droplets leads to the opposite effect. More cloud droplets compete for the same amount of moisture leading to smaller autoconversion rates, thus delaying precipitation formation.'

As the overall scientific objective of this manuscript is the investigation of the impact of dust on cloud formation we certainly include dust (and dust acting as CCN) in the model setup.
The section cited above refers to the general weakness of using a bulk microphysics scheme where the collision and coalescence of cloud droplets to form raindrops is parametrised (equation 50 in Lin et al. (1983)). This parameterisation only depends on the number concentration of cloud droplets and a fixed cloud droplet size distribution is assumed. Thus, if more cloud droplets are activated (considering dust as CCN vs not considering dust as CCN), those cloud droplets compete for the same amount of available water. This effect finally leads to smaller autoconversion rates and thus a delay in precipitation initiation.
In reality the collection/coalescence of small droplets by large droplets leads to faster autoconversion rates from cloud to larger rain droplets. However, to consider this process accurately in a numerical model, the cloud droplet size distribution would be needed to be treated prognostically, a process not included in any commonly used microphysics scheme on regional and global scales. This process could be however, explicitly treated when using a spectral (or bin) microphysics scheme. Those schemes are however, computationally very expensive and are not applied in a model setup including full chemistry and on scales such as we use it.

We have slightly rephrased the above cited sections in the manuscript:

"A weakness of the model set up, using a bulk microphysics scheme, is that we cannot explicitly account for dust as GCCN leading to larger cloud droplets which may increase the rate of precipitation formation due to efficient collection/coalescence of smaller droplets, as this is only possible with a prognostic cloud droplet size distribution. Thus in our model setup an increased number of CCN and cloud droplets leads to the opposite effect. More cloud droplets compete for the same amount of moisture leading to smaller autoconversion rates, thus delaying precipitation formation. A model setup treating the cloud droplet size distribution prognostically would however require the application of a spectral microphysics scheme which is computationally very expensive and has to our knowledge not yet been used in a model in combination with a state of the art aerosol scheme and explicitly calculating chemistry and meteorology online."

(*) *Specific comments*

(4) *Page 32370 line 23: The statement about the quality of the agreement of the simulated and the observed precipitation is very optimistic and by no means documented. A simple scatter plot would show this. On the other hand, can we expect a good agreement?*

With our study we do not intend to discuss the impact of dust on the rainfall prediction skills of the model. The overall objective of our work is to investigate the potentially different evolution of cloud formation processes under different aerosol conditions. The comparison with observations shown in the supplement only shows that all relevant model parameters especially simulated aerosol concentrations are realistic. Further the plots give additional information about the meteorological conditions on the case study days and the comparison shows that the model simulates clouds in the right region and the amount of precipitation is neither unrealistically low or high at any place for which measurements are available.

(5) *Page 32371 line 5: What means in all cases where dust is allowed to act as CCN*

In our reference simulation NOdust, we have switched the emission of dust particles off. Thus in this scenario dust is not present in the model and is therefore not available to act as CCN. In all other scenarios dust is emitted and is fractionally allowed to act as CCN as a consequence of chemical aging .

(6) *line 6: The slightly northward extension of the cloudy area is not visible in Figures 2a-c. Please check.*

Without including dust, the northern part of the cloud (the area we refer to corresponds to the area marked with a white circle in Fig.1, indicated in the text) only consists of a very few cloud droplets (purple area in Fig. 2a) whereas the cloud droplet number concentration is significantly enhanced when dust acts additionally as CCN (Fig. 2 b,c). As the northward extension of the cloudy area is not significant we wrote 'slightly northward'. We rephrased this sentence to:
'In all cases where dust is allowed to act as CCN, particles are activated at the southern edge of the dust plume leading to a slightly, though not significant northward extension of the cloudy area compared to the NOdust scenario (Fig. 2ac).

(7) *line 7: Please explain the meaning of the total cloud droplet number in this region is 166391 cm-3 and the corresponding numbers for the other cases. Please check the units.*

The number gives the vertically and horizontally summed number of cloud droplets in the area between 29° to 31°E and 33.5° to 35.4°N.
We have changed the units of the above given numbers in the revised version of the manuscript and we now give the mean value (over the are between 29° to 31°E and 33.5° to 35.4°N) of the total column burden of the cloud droplet number concentration (#/m^{-2})

(8) *lines 23-25: Where can I see that the clouds are thin in scenario τ_{48}.*
You can see that clouds are thinner as the vertically integrated number of cloud droplets is smaller in scenario τ_1 than in scenario τ_{48} (e.g. the region between 30.-30.6° and 33.9-35°N, marked now as area (1), which is pinkish in scenario τ_1 and greenish in scenario τ_{48}).

(9) *Section 4.1.2: This section contains numerous findings which are not documented in corresponding figure(s). At lease it is not clear which figure the authors are referring to. Please add figures that document your findings (if necessary in the supplement).*

We wrote:
' The differences in the domain-integral precipitation is less than 5% between scenario NOdust and scenario τ_1 as well as between scenario NOdust and NOfeedb. '
We have added a figure to the supplement showing the domain-integral precipitation and we marked the area we refer to in figure 2.
– We have added a reference to figure 2 (d-f) after the following sentence:
'However, a small shift of the rain maximum can be seen with increased precipitation over land by introducing hygroscopic dust. Since we did not perform ensemble simulations it is not possible to determine the level of significance of this shift.'
– We have added another figure to the supplement showing the difference in hourly accumulated precipitation between the scenario including dust (τ_1) and scenario NOdust and we refer to this figure after the following sentence:
'In addition the rain maximum over the sea is shifted slightly further South in scenarios τ_1 and τ_{48} compared to scenario NOdust (between 15:00 and 18:00 UTC) as dust particles have highest concentrations there and subsequently act as additional CCN and IN (Fig. 6, supplement).'

(10) *Section 4.1.3: From the material presented it is very difficult and almost impossible to follow the description of the results. Maybe it would help to indicate the areas where averages were taken and to add some tables. This procedure would also be very helpful for the following sections.*

– As mentioned above, we have marked the regions we refer to in the figures.

(11) *Figure 5 is very confusing as long as the figure caption does not explain all the information that is depicted. Which scenario is presented in Figure 5?*

– Both plots in figure 5 refer to scenario τ and we have added this information to the figure caption.

(*) *Section 5.1: It is hard to follow a lot of the findings based on the figures that are presented.*
(12) *Page 32375: line 8. How can you discuss the role of GCCN if you have neglected them in your model setup? To avoid confusion, the authors should use the term GCCN in a consistent way throughout the text. Sometimes GCCN refers in general to CCN originating from dust [e.g. in the abstract or Line 8 page 32375] and sometimes it is used to refer to large particles that impact the colloidal stability of the cloud, which is the common definition.*

– Please refer also to our reply at point (3).
Throughout the text we refer to large CCN by using the term 'GCCN'. Generally, this expression is not restricted to dust. Large Sea Salt particles are also addressed when speaking of Giant Cloud condensation nucleii.
As discussed above (point (3)), the treatment of dust as CCN should also lead to changes in the cloud droplet size distribution, an effect which cannot be taken into account with a model setup such as we use.

(13) *Page 32376: line 1: how can you determine a time shift of 30 min from figures 4a,b.*

– We have added another figure to the supplement, showing the 10 minute accumulated precipitation at the time of rainfall initiation in scenarios NOdust and τ_1. We have further rephrased this sentence and write now more precisely:
'With dust being present:
Precipitation is initiated slightly later along the northern Israel coast (north of 32.8°N) compared to scenario NOdust. However, precipitation along the shoreline between 31.6°N and 32.8°N is initiated slightly earlier (≈ 30 min, Fig. 7, supplement)'

(14) *line 15: You are talking about the coagulation of dust particles and sea salt. Is that process treated in your model? Do you have indication that this process is significant in reality? Which coagulation kernel did you use?*

(i) Coagulation is treated in the model and follows the approach by:
Jacobson m. et al. (1994) modeling coagulation among particles of different composition and size. atmos. environ. 28a, 1327 - 1338.
Jacobson m. z. (1999) fundamentals of atmospheric modeling. cambridge university press, new york, 656 pp.
(ii) The chemical composition of aerosol particles collected during the MEIDEX campaign (on 21 January, 2003) indicates that up to 33% of the coarse mode aerosol are internally mixed particles containing dust and sea salt. (Levin et al., 2005).'
(iii) Apparently the sentence you refer to lead to a misunderstanding and we rephrased it in the revised version of the manuscript:
We wrote:
'However, the dust transport time over the sea is much shorter on 28 January. Thus the time for dust to coagulate (we rephrased 'to coagulate' with 'to mix') with sea salt is much shorter. In addition, part of the dust along the coast of Israel does not pass over the Mediterranean hence cannot mix with sea salt.'

However, in the interpretation of the simulated data, coagulation is meant differently and we refer to the description of the treatment of aerosols given in the model description: 'Since in the current version of WRF-chem/MOSAIC only one type of aerosol is considered and aerosols are internally mixed in each bin, the bulk aerosol hygroscopicity used in the aerosol activation scheme is a mean value over all bulk components. Thus as soon as the dust plume crosses the Mediterranean the aerosol composition changes from almost pure dust in the coarse mode to an approximately equal mass ratio of sea salt (having a high hygroscopicity) and dust (having a low hygroscopicity) quickly leading to a bulk hygroscopicity for coarse mode particles which is high enough to activate particles into cloud droplets. Thus the effect of coating and/or mixing of dust with sea salt is to some extent (indirectly) accounted for in the model.'

(15) *Figure 3: Which scenario is shown? What means mixing ratios of integrated total condensate in g k-1? Please explain.*

Figure 3 refers to scenario τ_1 and we have added this information to the figure caption.
'Total condensate' means the sum of the mixing ratios of all five hydrometeor classes included in the microphysics scheme (cloud water, rain water, ice, snow, graupel). As this figure mainly shows the time evolution of the frontal system and the dust plume we show the vertically summed total condensate. In the revised version of the manuscript we have changed the units of the figure and now show the column burden of the total condensate (in g/m^2).
Showing cloud quantities at only one level might give a wrong impression of the overall conditions.

(16) *Figure 5: Which variables are shown in this figure? Which scenario*

Both plots in figure 5 refer to scenario τ_1 and we have added this information to the figure caption.
Both plots in figure 5 show the coarse mode bulk hygroscopicity (water attraction potential of a particle, Zaveri et al. (2008), Fast et al. (2006)) which is equal to the unweighted arithmetic mean of the bulk hygroscopicity of the aerosol bins 5-8.

References

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