Reply to Referee 1

We thank the reviewer for the careful and thorough evaluation of the manuscript and helpful suggestions to improve the manuscript.

(*) *This paper focuses on the impact of mineral dust on cloud formation. Given that, a more sophisticated cloud droplet activation parameterization (than one used) is needed to represent these interactions. Kumar et al (2009a;2011a) suggested that adsorption activation theory (AT) better represents fresh dust-water interactions than Köhler theory (KT), as the dependence of critical supersaturation with particle dry diameter is closer to observations. The CCN activity of wet processed dust though, is more consistent with KT at small particle sizes and with AT at larger particles. To address this gap, the unified CCN activity framework can be used, developed by Kumar et al. (2011b), that accounts for concurrent effects of solute and adsorption to describe the CCN activity of aged or hygroscopic dusts. Karydis et al. (2011) used this framework to provide a first assessment of the contribution of insoluble dust to global CCN and cloud droplet number concentration (CDNC). Bangert et al. (2011), using this framework, also investigated the impact of mineral dust particles on clouds, radiation and atmospheric state during a strong Saharan dust event over Europe in May 2008. Overall, considering the hydrophilicity from adsorption and hygroscopicity from solute is required to comprehensively capture the dust-warm cloud interactions. I suggest to add a similar discussion in the manuscript and also comment on the simplification of i) using the KT for describing CCN activity of particles that contain mineral dust and ii) assuming that the bulk aerosol hygroscopicity is the mean value over all bulk aerosol components.*

(*) *8. Page 32368 line 8: The most appropriate theory for describing the CCN activity of mineral dust is the FHH-adsorption theory (Kumar et al., 2009b), which uses the AFHH and BFHH parameter to express its hygroscopicity. In case of using the Khler theory though, the use of a hygroscopicity parameter between 0.005 and 0.05 for freshly emitted mineral dust seems more realistic according to Kumar et al. (2011a; Figure 2).*

– We have added a discussion as recommended. Please find the new discussion below. We have further added references to the introduction referring to AT.

In our model setup, we calculate cloud droplet activation following Koehler theory (KT). However, Kumar et al. (2009) and Kumar et al. (2011) suggest that adsorption activation theory (AT) better represents fresh dust-water interactions than KT. The CCN activity of wet processed dust though, is more consistent with KT at smaller particle sizes and with AT at larger particle sizes. Therefore, Kumar et al. (2011) developed a unified CCN activity framework (UAF). However, this framework is difficult to incorporate in numerical models using internally mixed aerosol particles. The implementation of externally mixed dust in addition to different dust age classes would be required to properly make use of the UAF, as for differently aged dust either adsorption growth or equilibrium water uptake would be calculated.
Bangert et al. (2011) use the UAF in one of their sensitivity simulations investigating the impact of dust on cloud formation over central Europe, assuming that all dust particles are coated by ammonium sulfate. Thus they describe the activation of dust more accurately than we do. However, they cannot consider changes in the dust hygroscop-

icity during transport time, which is the basis for our sensitivity calculations. For their study, focusing on the impact of dust on cloud formation over Central Europe, this is not crucial because it can be assumed that all long-distance transported dust has undergone chemical processing and should be to some extent associated with soluble compounds. As our case study focuses on a region closer to the dust source, we consider it necessary to explicitly account for changes in the bulk hygroscopicity and composition and using a less sophisticated CCN activation formulation rather than assuming internally mixed aerosols. Nevertheless, Bangert et al. (2011) obtain very similar results from their study using slightly different parameterisations and a different model (COSMO-ART) than we do. In both studies, the effect of dust on warm cloud formation is very small.

(*) *Specific Comments:*

(*) *1. Page 32364 line 4: Replace days with cases. The second episode lasts for two days and not one.*
– We have changed this sentence.

(*) *2. Page 32364 line 23: The start of the sentence is confusing. The authors may need to rephrase, i.e. In addition aged, chemically altered dust particles can act: : :*
– We have rephrased the sentence:
In addition, aged dust particles that have undergone chemical processing can act as giant cloud condensation nuclei (GCCN) which can enhance rain formation as they efficiently collect moisture and grow at the expense of smaller CCN (e.g. Levin et al., 2005).

(*) *3. Page 32366 line 5-6: Mineral dust is hydrophilic and not hydrophobic as mentioned in the text (Sorjamaa and Laaksonen, 2007; Gustafsson et al., 2005; Hatch et al., 2008; Vlasenko et al., 2005). In addition, Kumar et al. (2009a, 2011a) suggest that even fresh unprocessed mineral dust can also affect warm clouds by acting as CCN. Please comment on these as well.*

– We have added references to the introduction related to fresh dust, its hygroscopicity and abilities to act as CCN.
The reviewer states that mineral dust is hydrophilic, supported by several references. We have checked these references (and others) and found no experimental evidence for this. Even though several references indicate the possibility of dust being hydrophilic, e.g. based on theoretical considerations, most of the literature converges on fresh dust being hydrophobic. It is nevertheless possible that some fresh dust is affected by the deposition of soluble material or rapidly accumulates acids from the ambient atmosphere. It is conceivable that minor amounts of soluble material in dust may suffice to allow the particles to act as CCN.

(*) *4. Page 32366 line 20-21: Other studies though suggest that dust particles with a soluble coating can also maintain their activity as IN (Levin et al., 2005). Please add this (or similar) reference too.*
– We have added this reference.

(*) *5. Page 32366 line 24-26: Tropical Atlantic Ocean is also often influenced from dust particles originating from Northern and Central Africa (Karyampudi and Carlson,*

1988;Karyampudi et al., 1999;Chiapello et al., 2005;Kallos et al., 2006)
– We have added these references to the introduction.

(*) *6. Page 32367 line 21: Replace parametrisation with parameterization. This error is repeated several times in the manuscript.*
– We have changed 'parametrisation' to 'parameterisation'. We have written the manuscript in British English.

(*) *7. Page 32368 line 6: Which is the size range of the four largest size classes? Several studies have reported mineral dust size distributions including modes with median diameter as low as 160 nm (DAlmeida, 1987;Chou et al., 2008).*
– The lower diameter of the smallest size class to which dust is emitted is 312nm and the upper limit of the largest size class is 10000nm.

(*) *9. Page 32368 line 26: The 12h case is neither in Table 1 nor discussed later in the manuscript.*
– We excluded this case study from the final manuscript and decided to only discussed the extreme scenarios τ_1 and τ_{48}. We erased the 12h case from the model setup description.

(*) *10. Page 32369 line 15: Replace case study days with case studies*
(*) *11. Page 32370 line 1: Replace summer/autumn with autumn*
– We have replaced both phrases.

(*) *12. Page 32370 line 11: Please refer the number of the figure in the supplement. This omission is repeated several times in the manuscript.*
– We have added the number of the figure from the supplement to which we refer to.

(*) *13. Page 32371 line 23: Please use either spacial or spatial in the entire manuscript.*
– We have replaced 'spacial' with 'spatial'

(*) *14. Page 32372 line 9: Please include a brief description of the NOfeedb scenario in model setup section.*
– We have added a brief description of scenario NOfeedb to the 'model setup' section.

(*) *15. Page 32375 line 16: Replace range of with in the range of*
– We have replace 'range' with 'in the range'.

(*) *16. Page 32375 line 28: It is not apparent from figure 4 that precipitation is initiated earlier when dust is present. The authors should explain this more. They can i.e. place an arrow or a box in the figure to help the reader focus on the area that this shift is more apparent.*
– 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)'

(*) *17. Page 32376 line 4-6: Some readers may confuse and refer to Figure 4 for this sentence. I suggest either to include a figure with the 24h accumulated precipitation (it may be interested) or to add (not shown) at the end of the sentence.*
– The 24h accumulated precipitation is shown in figure 2 in the supplement. We have added a reference to this figure in the text.

(*) *18. Page 32376 line 12: Replace summer with autumn*
– We have replaced 'summer' with autumn.

(*) *19. Page 32376 line 24-27: As mentioned in the general comment, other weaknesses of the model setup are i) the use of KT for describing CCN activity of particles that contain mineral dust and ii) the assumption that the bulk aerosol hygroscopicity is the mean value over all bulk aerosol components. Please also comment on them.*
– As stated above we have added a section to the 'Discussion' where we discuss the potential impact of using KT versus AT.
- We do not necessarily see the usage of the bulk aerosol hygroscopicity as a disadvantage of our model setup.
Over northern Africa the coarse mode hygroscopicity is very low, as the aerosol composition is mainly dust. The further north the dust plume is advected over the Mediterranean (e.g. in the September case study) the more the hygroscopicity increases due to the increasing sea salt fraction in the coarse mode. As dust and sea salt particles can mix, increasing the hygroscopicity of a former pure dust particle the usage of the bulk hygroscopicity is even an advantage of the simulation setup as the uptake of other acids into the coarse mode is not very efficient.
However, we consider the change of hygroscopicity within the dust plume (due to mixing with sea salt) as most likely overestimated. Therefor we performed the series of sensitivity simulations allowing only a fraction of coarse mode particles to contribute to the CCN population. In that case, also only a 'fractional' bulk hygroscopicity is used (please refer also to our reply to reviewer 2, (2)). In case that only a very small dust fraction is allowed to act as CCN, the bulk hygroscopicity is thus mainly equivalent to the hygroscopicity of sea salt (for the coarse mode). In case that almost all dust is allowed to act as CCN the bulk hygroscopicity is reduced, however it is still high enough to allow the coarse mode particles to get activated - at least in the regions of interest in the afternoon/evening hours, i.e. in regions where the dust concentrations were high and cloud formation was facilitated.
From our point of view, one of the largest uncertainties is our poor understanding of the timescales of the chemical surface processing of dust and the related changes in the dust hygroscopicity. If laboratory and field data would give better evidences on dust aging, we could better constrain the (bulk) hygroscopicity in our model simulations.

(*) *20. Page 32377 line 27: Replace case study days with case studies*
– We have replaced 'case study days' with 'case studies'.

(*) *21. Figures: Please improve the quality of the figures. For instance, the numbers*

and letters on the axes are not very clear.
– We have enlarged the axes labels in the figures.

References

Bangert, M., Nenes, A., Vogel, B., Vogel, H., Barahona, D., Karydis, V., Kumar, P., Kottmeier, C., and Blahak, U.: Saharan dust event impacts on cloud formation and radiation over Western Europe,, Atmos. Chem. Phys. Discuss., 11, 31 937–31 982, 2011.

Kumar, P., Nenes, A., and Sokolik, I.: Importance of adsorption for CCN activity and hygroscopic properties of mineral dust aerosol, Geophys. Res. Lett., 36, L24804, doi:10.1029/2009GL040 827, 2009.

Kumar, P., Sokolik, I., and Nenes, A.: Cloud condensation nuclei activity and droplet activation kinetics of wet processed regional dust samples and minerals, Atmos. Chem. Phys., 11, 8661–8676, 2011.