Response to **Anonymous Referee #2** concerning the paper "Dust size parameterization in RegCM4: Impact on aerosol burden and radiative forcing" (http://www.atmos-chem-phys-discuss.net/acp-2016-434).

November 30, 2016 Dear editorial and respected reviewer,

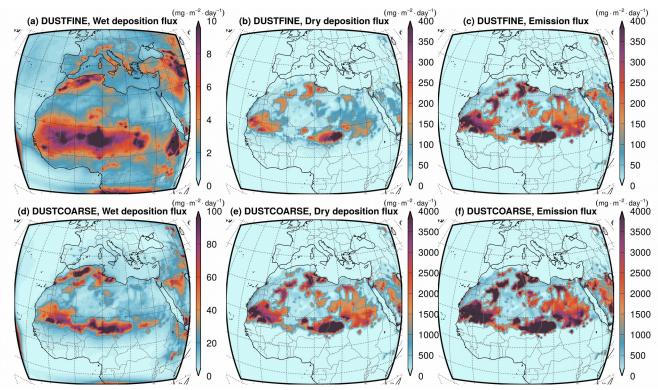
Thank you very much for reviewing our manuscript and providing us with such a constructive feedback. We believe that your comments helped us highlight some critical aspects of the paper and add important content which elevates the quality of our work. All the short comments and typographical errors were corrected in the new version of the paper, while comments that needed more explanation and additional material are discussed below. The structure of the responses includes (1) comments from Referee, (2) a detailed response (3) changes implemented in the new version of the paper. Quotes from the initial version of the paper are highlighted with pale red along with the corresponding page and lines. Quotes from the new version of the paper are highlighted with pale blue along with the corresponding page and lines. The specific phrases changed-modified are underlined for convenience. In cases where the quotes are discussed but not changed are highlighted with grey. I sincerely hope you will be fully satisfied with all the changes we have made.

Best Regards, Athanasios Tsikerdekis

1. The paper does not present much about emission which is heavily affected by size distribution. Can you show more about dust emission, e.g., spatial distributions and size distributions?

This is an important issue that you have raised and we sought to address in the paper. We agree it adds value and it will additionally help the readers understand the work better. The Fine (DUST01+DUST02) and Coarse (DUST03+DUST04) emission tendencies (kg kg⁻¹ s⁻¹) are shown in Figure 5 and Figure 6 as monthly values averaged over a subregion for the DUST4 experiment. But of course the spatial distribution of emission flux (mg m⁻² day⁻¹) might be helpful for the reader so we are adding the following figures (emission, dry deposition, wet deposition) as supplementary material. Note that the spatial distribution of emission and deposition fluxes are almost identical between the fine and the coarse dust particles, although the coarse particles deposition and emission fluxes are ten times greater. Also, the following comment was added in the main paper.

P12, L4-5: The spatial distribution of wet deposition, dry deposition and surface emission fluxes is depicted for the fine and coarse particles in FigureS 3.



FigureS 3: Wet deposition, dry deposition and surface emission fluxes of fine (a, b, c) and coarse (d, e, f) dust particles in DUST4 experiment for the period December 2006 to November 2014.

2. P1, L20-21 and P3, L18-19: Please consider removing "minimize the error". More size bins shall provide more information but it does not guarantee if it is more realistic than less bin numbers. Both 12-bin and 4-bin approaches are globally homogeneous and both have large uncertainties. This study shows that 12-bin results some changes in dust loading (3-4%) and DOD (10%). Since the model sensitivity is small or moderate it needs statistical significance test to prove the improvement in 12-bin approach. Emission, DOD, and/or RF from the new model need to be compared with observations to show how much bias is reduced compared to the old model.

As you correctly point out, the phrase "minimize the error" is too generic and can be misinterpreted as "minimizing the bias in comparison to observations", which is not always the case. Therefore, we have removed and rephrased it respectively.

- P1, L19-21: Increasing the number of transported dust size bins theoretically improves the representation of the physical properties of dust particles within the same size bin. Thus, more size bins minimize the error and improve the simulation of atmospheric processes.
- P1, L20-22: Increasing the number of transported dust size bins theoretically improves the representation of the physical properties of dust particles within the same size bin. Thus, more size bins minimize the error and improve the simulation of atmospheric processes.
- P3, L18-19: The greater number of dust size bins <u>minimizes model error</u> especially for particle dry deposition and thus allows to more accurately simulate both the atmospheric dust burden and the interaction with radiation (Foret et al., 2006; Menut et al., 2007).
- P3, L22-23: The gGreater number of dust size bins minimizes model error especially for improves particle dry deposition and thus allows to more accurately simulate both the atmospheric dust burden

More size bins provide more information than less bin numbers. Furthermore, the isolog partitioning method is arbitrary and mathematical, with no connection to the physical world processes. Isogradient partitioning method of the bins takes into account the deposition velocity of dust particles according to their size. Considering both the number and the partitioning method we are highlighting that the 12bin-isogradient is actually more "realistic" than the 4bin-isolog method (closer to reality in terms of the physical processes that take place in the atmosphere). Of course this improvement does not guarantee that is going to fix the biases that originate from other processes as already stated in the paper:

Pg 14, lines 30 - Pg 15, lines 5: Previous studies (Foret et al., 2006; Menut et al., 2007) revealed that the new approach in dust size bin partitioning and number (which is also adopted in our DUST12 experiment) more realistically simulates the transport and dry deposition of the dust size bins. This does not imply that the simulated biases will be reduced for all regions in the DUST12 experiment. The major factor that controls and potentially creates the first order biases in dust models is the balance between the emission and sedimentations terms (e.g. Figure 5, Figure 6). As discussed in Section 3.1, some positive DOD biases (e.g. Eastern Sahara) might be due to an underestimation of the outflow of dust or a local overestimation of the emission flux from surface wind velocity errors. Therefore, although the new dust size parameterization theoretically improves dry deposition, it does not necessarily regulate or improve the biases that originate from other processes.

Thank you for highlighting the statistical significance issue. We have calculated the statistical significance of the differences using the monthly data for each grid point for the DOD, dust column burden and radiative forcing using the two-tailed paired t-test. In all variables and almost in all grid points the differences are statistically significant at the 95% confidence level (p.value < 0.05). We have also updated the text in the new version of the paper to highlight this issue. The plots shown below were not updated in the paper since we believe that they do not offer additional information to the reader; the vast majority of the grid points show differences that are statistically significant (shaded areas in the differences), hence we are just mentioning this in the text.

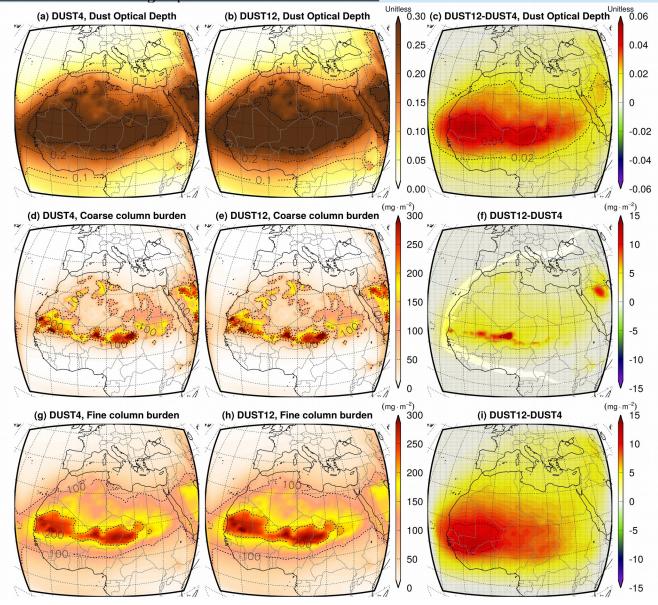
Pg 14, lines 13-20: The DOD percent increase is between 10.4% and 13% for all the subregions. Furthermore, there is a distinctive increase by 0.04 with the 12-bin model over the Sahara desert and especially along the Sahel region where the DOD values are higher (Figure 11c). In comparison with the DUST4 simulation the DUST12 simulation increases the deposition lifetime (column burden/total deposition flux) by 3.5 hours and 2 minutes for fine and coarse particles respectively. Consequently, that increases the dust column burden of fine (+4%) and coarse (+3%) particles (Figure 11f,i). The changes in the fine particles correlates better with the changes in DOD, because dust extinction coefficient is much higher for fine particles (<2.5μm) (Figure 1). Over the Middle East and the northern part of the Arabian Peninsula we observe a distinct increase on the coarse dust column burden by 10mg•m-2.

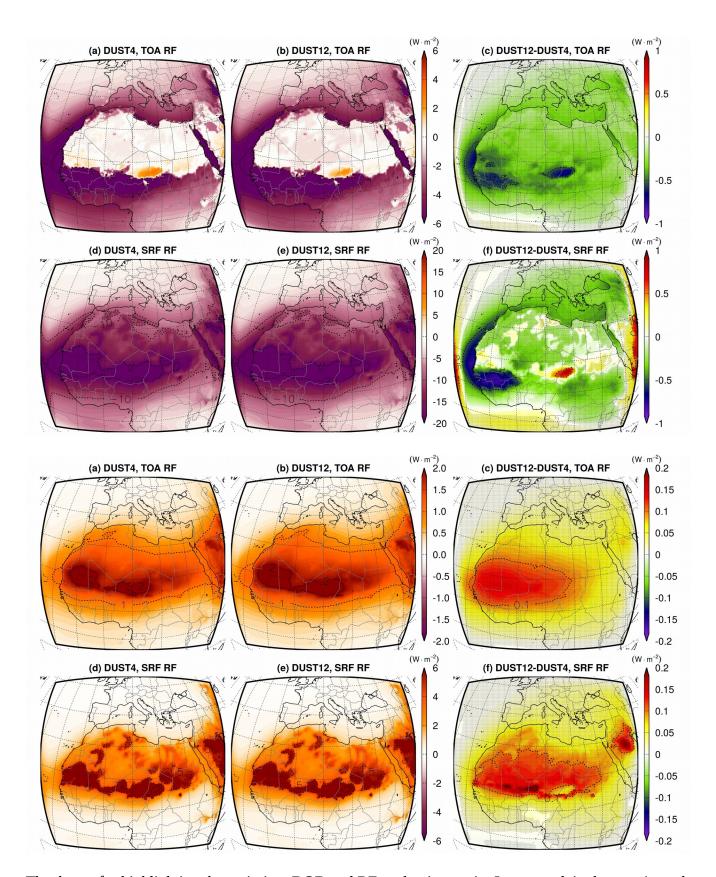
Pg 15, lines 17-26: The DOD percent increase is between 10.4% and 13% for all the subregions. Furthermore, there is a distinctive increase by 0.04 with the 12-bin model over the Sahara desert and especially along the Sahel region where the DOD values are higher (Figure 11c). In comparison with the DUST4 simulation the DUST12 simulation increases the deposition lifetime (column burden/total deposition flux) by 3.5 hours and 2 minutes for fine and coarse particles respectively. Consequently, that increases the dust column burden of fine (+4%) and coarse (+3%) particles (Figure 11f,i). The changes in the fine particles correlates better with the changes in DOD, because dust extinction coefficient is much higher for fine particles ($<2.5\mu$ m) (Figure 1). Over the Middle East and the northern part of the Arabian Peninsula we observe a distinct increase on the coarse dust column burden by

10mg•m-2. The differences of DOD and column burden between the two experiments, calculated from the monthly data for each grid, are statistically significant at the 95% confidence level according to a two-tailed paired t.test for almost all the grid points of the simulated domain.

Pg 16, lines 11-13: Similarly, at the surface the positive radiative forcing rise by 0.08W·m-2 (3.0%) and 0.09W·m-2 (2.7%) in eastern and western Sahara and 0.9W·m-2 (6.3%), 0.08W·m-2 (4.8%) and 0.9W·m-2 (6.5%) in western, central and eastern Mediterranean (Figure 14f).

Pg 17, lines 16-20: Similarly, at the surface the positive radiative forcing rise by 0.08W·m-2 (3.0%) and 0.09W·m-2 (2.7%) in eastern and western Sahara and 0.9W·m-2 (6.3%), 0.08W·m-2 (4.8%) and 0.9W·m-2 (6.5%) in western, central and eastern Mediterranean (Figure 14f). The shortwave and longwave radiative forcing differences between the two experiment, calculated from the monthly data for each grid, are statistically significant at the 95% confidence level according to a two-tailed paired t.test for almost all the grid points of the simulated domain.





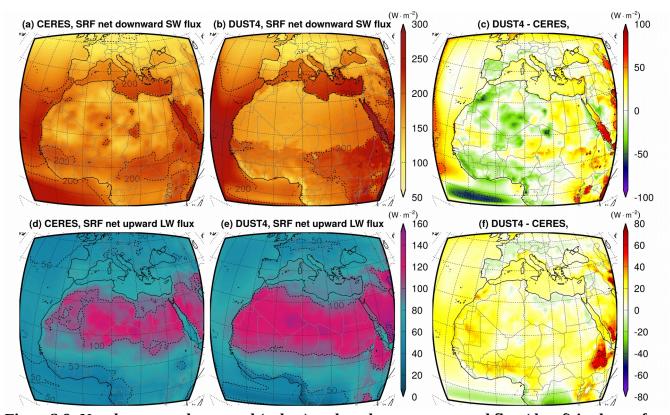
Thank you for highlighting the emission, DOD and RF evaluation again. Let us explain the case in each one of them below. Dust emission is identical between the two experiments (DUST4, DUST12) and this was done on purpose (as already stated in Section 2.5 Experimental set-up) in order to compare the

size bin changes in the two experiments. The two experiments have the same meteorological fields, the same wind fields, thus the same emission fluxes. A direct evaluation of the emission fluxes with observations is not available at this spatial scale, thus we evaluate and discuss emission flux indirectly by using the surface wind speed from the ERA-interim in the Section "3.1 Evaluation".

Dust Optical Depth (DOD) is already being evaluated for both experiments with the state-of-the-art as of right now dust product LIVAS (FigureS 5). FigureS 5 was added due to the comments that we have received in the 1st review phase. As already discussed in the paper (Section "3.2 Comparison of 4-bin and 12-bin experiments"), the reduction of bias in comparison to observation doesn't necessarily mean that we are improving the physics in the model. DUST4 experiment exhibits lower bias in comparison to DUST12 experiment in most regions since conflicting processes that are overestimated-underestimated in the model (e.g. wind fields, precipitation, surface characteristics) might give a DOD closer to the observational data, but for the wrong reasons.

To our knowledge there are no observational data for Dust Radiative Forcing (DRF). But evaluating DOD is the next best thing, since DOD is the extinction of radiation by the dust particles in the atmosphere. Of course since DRF is also a function of the simulated SW and LW radiation, we have added as supplementary material the following plots which evaluate the net ShortWave (SW) and LongWave (LW) in the SuRFce (SRF) using the CERES measurements. We also added the following phrase at the end of the Section "3.1 Evaluation".

P15, L7-9: The model was evaluated also in terms of the surface net downward shortwave and net upward longwave radiation flux against CERES satellite measurements (FigureS 8). In the shortwave spectrum radiation flux bias ranges between -10Wm⁻² and 10Wm⁻² above the desert and the Mediterranean, while in the longwave the differences are mostly positive (~10Wm⁻²).



FigureS 8: Net shortwave downward (a, b, c) and net longwave upward flux (d, e, f) in the surface

3. P5, L20: Please clarify what is a new dust scheme. Size distribution only or new scheme?

Thank you for pointing this out. We have rephrased the following sentence.

P5, L19-20: Following the methodology of (Foret et al., 2006), we have implemented a <u>new dust scheme</u> that resolves twelve size bins instead of four.

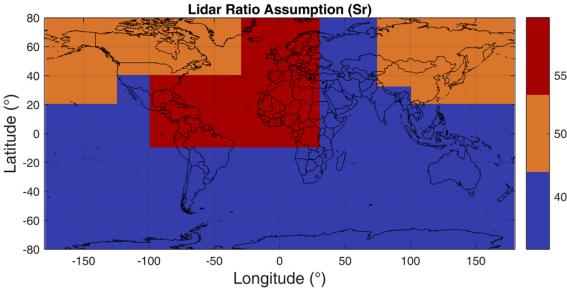
P5, L25-27: Following the methodology of (Foret et al., 2006), we have implemented a <u>new dust size</u> <u>discretization scheme</u> that resolves twelve <u>dust transport</u> size bins instead of four.

4. P6, L31: Please provide the old and new lidar ratio values.

Thank you for highlighting this issue, I think is of great importance therefore we have added the Lidar Ratio (LR) values that was used prior in the CALIPSO measurements and the regional specific new LR values used in the LIVAS product in the last sentence. Plus in the supplementary we have added the following plot to show the spatial distribution of LR assumption.

P6, L31 - P7, L4: The LIVAS extinction dust product is corrected for the Lidar Ratio (LR) based on multi-year measurements performed by the ground-based lidar stations of the EARLINET lidar network (https://www.earlinet.org). The LR of dust particles depends on their refractive index and may vary for aerosols of the same type. The refractive index values rely upon the composition of dust and most importantly on the relative proportion of clay-sized mineral illite in dust (Schuster et al., 2012). Thus, regions with different physiochemical dust characteristics leads to different LR values. The 0.3.1 version of LIVAS separates the globe into three regions, specified based on known dust sources and loadings with specific physio-chemical composition and LR for each region.

P7, L4-L12: The LIVAS extinction dust product is corrected for the Lidar Ratio (LR) based on multi-year measurements performed by the ground-based lidar stations of the EARLINET lidar network (https://www.earlinet.org) and intensive campaigns in different dust regions arround the globe (Wandinger et al., 2010; Hänel et al., 2012; Baars et al., 2016). The LR of dust particles depends on their refractive index and may vary for aerosols of the same type. The refractive index values rely upon the composition of dust and most importantly on the relative proportion of clay-sized mineral illite in dust (Schuster et al., 2012). Thus, regions with different physiochemical dust characteristics leads to different LR values. The 0.3.1 version of LIVAS separates the globe into three regions, specified based on known dust sources and loadings with specific physio-chemical composition and LR for each region. The globally LR value 40sr used in CALIPSO retrieval algorithm (Omar et al., 2009) was replaced with region specific LRs of 40sr, 50sr and 55sr (FigureS 2).



FigureS 2: The region specific Lidar Ratio assumption used in LIVAS.

5. P7, L8-11: The sentence is unclear, since the study domain is over land. I believe MODIS Deep Blue AOD is well validated with AERONET data over the source region (please check with the NASA Deep Blue website and other documentations).

We agree with the reviewer and indeed we believe the new changes help situate the case better. The retrieval accuracy and the spatial coverage of the MODIS Deep Blue AOD has been improved in the latest version C6 in comparison to the C5 version. Future studies will determine the correlation between MODIS Deep Blue (C6) AOD and the LIVAS DOD. Therefore, we are highlighting in the text the version of MODIS (C5) evaluated with LIVAS and the need for future comparison with the new MODIS Deep Blue (C6) AOD. We feel the need again to note that the LIVAS post-processing analysis provides the DOD (and not AOD like MODIS) which can be directly used to evaluate the DOD of the model in this paper.

P7, L5-13: LIVAS has been evaluated against AERONET stations globally by Amiridis et al. (2015). The results show that the aerosol optical depth differences are between ±0.1 in most cases. Over the southwestern Sahara desert, LIVAS underestimates the AERONET AOD by -0.1, and this bias may be related with the dust underestimation of CALIPSO found in previous studies (Amiridis et al., 2013; Schuster et al., 2012; Tesche et al., 2013; Wandinger et al., 2010). Amiridis et al. (2013) showed that LIVAS correlates well with the Dark Target MODIS retrieval over sea, yet the correlation between MODIS Deep Blue and LIVAS over Sahara are weak (results not shown). This could be attributed to the Deep Blue MODIS retrieval, which uses passive remote sensors for dust aerosol optical depth that take into account numerous assumptions (e.g. high reflectivity). Thus, LIVAS is a more reliable product over the deserts with higher accuracy than the products coming from passive remote sensing techniques.

P7, L13-L21: LIVAS has been evaluated against AERONET stations globally by Amiridis et al. (2015). The results show that the aerosol optical depth differences are between ±0.1 in most cases. Over the southwestern Sahara desert, LIVAS underestimates the AERONET AOD by -0.1, and this bias may be related with the dust underestimation of CALIPSO found in previous studies (Amiridis et al., 2013; Schuster et al., 2012; Tesche et al., 2013; Wandinger et al., 2010). Amiridis et al. (2013) showed that

LIVAS correlates well with the Dark Target MODIS retrieval over sea, yet the correlation between MODIS Deep Blue (version C5) and LIVAS over Sahara are weak (results not shown). The new version of MODIS Deep Blue (C6) improved its retrieval accuracy and spatial coverage globally (Sayer et al., 2014, 2015) and the Mediterranean region (Georgoulias et al., 2016a) in comparison to AERONET. Thus, further research is needed to determine the correlation between LIVAS DOD and MODIS Deep Blue (C6) AOD. This could be attributed to the Deep Blue MODIS retrieval, which uses passive remote sensors for dust aerosol optical depth that take into account numerous assumptions (e.g. high reflectivity). Thus, LIVAS is a more reliable product over the deserts with higher accuracy than the products coming from passive remote sensing techniques.

6. P8, L15: Please be specific the size is radius or diameter.

Thank you for pointing out this clarification. It is diameter.

P8, L15-16: Using a simple one-dimensional box model they simulated an experiment with a detailed particles size distribution that used 1000 size bins within the range of 0.001-100µm.

P8, L24-25: Using a simple one-dimensional box model they simulated an experiment with a detailed particles size distribution that used 1000 size bins within the range of 0.001-100µm diameter.

7. P9, L5 and Figure 2: About one half of the Sahel is desert and the other half is non source according to the map. Semi-arid source is only small fraction. Bodele is in Sahel. It is unclear what are the characteristics of the Sahel in this study. The seasonality of wind, precipitation, loading, and DOD over the Sahel is mixed with Sahara and Savanna.

We agree with the reviewer. The rough delimitation of the box-shaped subregions may affect the levels (but as it was found not the seasonality) of the dust column burden, DOD, wind velocity and precipitation annual cycles (for example DOD of Sahel). Therefore, we have re-organized all the subregions making them more representative for dust fluxes and concentration as well as for the meteorological variables.

The new delimitation of each region is shown in the following plot. Sahel was delimited using the CRU precipitation for the period 2001-2014. The selected grid cells for the Sahel receive annual precipitation between 100mm and 600mm (Ali and Lebel, 2009; Nicholson, 2013) and now Bodele Depression belongs to the Eastern Sahara subregion. Eastern and Western Sahara were selected according to the desert and semi-desert landuse assigned by the model. Their southern border was masked by the Sahel grid points. The three Mediterranean subregions now contain only non-desert grid points, while their western, northern and eastern boundaries were kept the same. Due to these changes some plots were updated accordingly in the final manuscript: Figure 4, Figure 5, Figure 6, Figure 8, Figure 9, Figure 10, Figure 12 as well as Figure S4 and Figure S5 (which are FigureS7 and FigureS9 in the new version of the paper). The following paragraph was added in the Section "2.5 Experimental set-up" to explain the delimitation of each region.

P9, L15-22: The simulated domain was separated into six distinct subregions: Sahel, Eastern Sahara (ESah), Western Sahara (WSah), Eastern Mediterranean (EMed), Central Mediterranean (CMed) and Western Mediterranean (WMed) (Figure 2b). Sahel was delimited using CRU precipitation for the period 2001-2014. The selected grid cells receive annual precipitation between 100mm and 600mm (Ali and Lebel, 2009; Nicholson, 2013) and are located in the southern border of Sahara. Eastern and

Western Sahara were selected according to the desert and semi-desert landuse assigned by the model. Their southern borders were masked by the Sahel grid points. The three Mediterranean subregions contain only non-desert grid points and are separated according to their DOD seasonality (Israelevich et al., 2012).

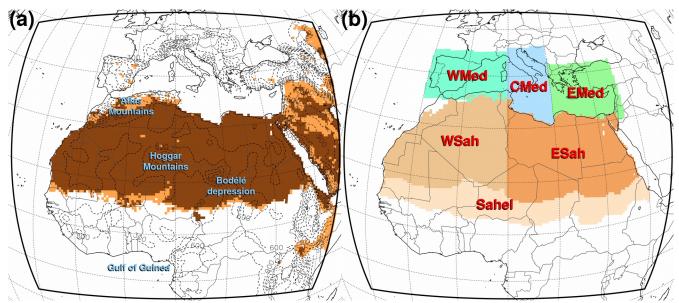


Figure 2: (a) The desert (dark brown) and semi desert (light brown) grid cells assign by the model along with the simulated topography used on RegCM4 in meters. The black box depicts the simulated domain. (b) The distinct six subregions used in the analysis (details in text).

8. P13, L34-P14, L5: The sentences do not belong to the results. Please consider to relocate or remove.

Thank you for raising this issue and we can understand your concern. The structure of our paper is consisted by a combined Results+Discussion Sections (3.1, 3.2, 3.3). Thus, we have changed the title of the Section "3 Results" into "3 Results and Discussion". We believe that this structure greatly helps the flow of the paper without making it too long. Thus, we believe that the following lines belong to the Results and Discussion section, since they are concluded from the analysis of Figure 10 (Dust Extinction evaluation). Prior to this section we are explaining what processes govern dust vertical distribution in each altitude and in (P13, L34-P14, L5) we are suggesting possible reason for an overestimation in the mid Troposphere by the model.

P13, L34-P14, L5: Considering the overestimated DEX profile in this altitude range it suggests that the cumulus convection activity/convective transport mechanism is overactive in Tiedtke scheme (Tiedtke, 1989) or another negative sign process (e.g. sedimentation) is not properly represented by the model. Discrepancies in the vertical distribution of dust can also be misinterpreted from possible local emission errors, the lack of simulated vertical levels or wet deposition biases. Further research is needed in this regard to reduce the mean vertical distribution bias of dust in the model.

9. P16, L1-2: Unclear sentence. Please consider to remove it.

Indeed, the following sentence was too generic, therefore we have removed it. Thank you for indicating

this.

P16, L1-2: A fact that should be taking into account by future researches that study the same region. A fact that should be taking into account by future researches that study the same region.

10. P16, L14-30: Again, it does not belong to result section.

Thank you for brining this up, though let me elaborate on it. As already noted in comment 8, we have written our Section "3 Results and Discussion", since it helps the reading flow. Thus, the following paragraph belongs to the resul-discussiont section since we are presenting and discussing results between four 1-yearly experiments in order to more accurately validate our radiative forcing results in the longwave spectrum. All the plots presented (FigureS 8 and FigureS 9) are analyzing the radiative forcing of dust in the longwave spectrum, thus we have put them in the section "3.3 Radiative Forcing".

P16, L14-30: However, the current treatment of the optical properties of dust in the longwave spectrum of CCM3 scheme is limited and does not account for specific absorption coefficient for each dust size bin. Furthermore, the CCM3 longwave bands are concentrated on the absorption of H₂O and CO₂ and they do not integrate in detail the absorbing aerosol component part of the longwave. Therefore, we have conducted two similar dust experiments for June 2008 using the radiation transfer scheme RRTM, which is known for its detailed longwave calculation. We note again that the current version of RRTM (+McICA) produces a random generated noise on radiation fields, therefore the dust emission fluxes are not identical between the DUST4 and DUST12 experiments. Our preliminary simulations using the RRTM scheme (FigureS 8), have shown notably lower longwave dust RF over the desert in comparison to CCM3 (FigureS 7) for the same period and roughly the same DOD levels and spatial patterns. More specifically, the DOD and the longwave radiative forcing averaged above the Sahara for the CCM3_DUST4 experiment is DOD=0.31, SRF=3.5W·m-2 and TOA=2.1W·m-2. While the related values for the RRTM_DUST4 experiment is DOD=0.29, SRF=1.9W·m-2 and TOA=0.6W·m-2. Although DOD increases in the 12 bin experiment in both radiation schemes (FigureS 7c, FigureS 8c), the longwave RF changes display a striking difference. With the CCM3 radiation scheme, the longwave radiation linearly increases with higher DOD and dust burden values (FigureS 7f and i). In comparison, the RRTM scheme uses specific absorption coefficient for each dust size bin, taking into account the fine/coarse dust burden changes. Thus is exhibits local increases or decreases of the longwave RF (FigureS 8f and i), according to the changes of fine to coarse dust burden. Overall the total longwave RF increase in RRTM is smaller compared to CCM3 when spatially averaged over the Sahara desert.

References Added.

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Hänel, A., Baars, H., Althausen, D., Ansmann, A., Engelmann, R. and Sun, J. Y.: One-year aerosol profiling with EUCAARI Raman lidar at Shangdianzi GAW station: Beijing plume and seasonal variations, J. Geophys. Res. Atmos., 117(D13), n/a–n/a, doi:10.1029/2012JD017577, 2012.

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Sayer, A. M., Hsu, N. C., Bettenhausen, C., Jeong, M.-J. and Meister, G.: Effect of MODIS Terra radiometric calibration improvements on Collection 6 Deep Blue aerosol products: Validation and Terra/Aqua consistency, J. Geophys. Res. Atmos., 120(23), 12,157–12,174, doi:10.1002/2015JD023878, 2015.

Sayer, A. M., Munchak, L. A., Hsu, N. C., Levy, R. C., Bettenhausen, C. and Jeong, M.-J.: MODIS Collection 6 aerosol products: Comparison between Aqua's e-Deep Blue, Dark Target, and "merged" data sets, and usage recommendations, J. Geophys. Res. Atmos., 119(24), 13,965–13,989, doi:10.1002/2014JD022453, 2014.

Acknowledgements added.

Pg 19, lines 22-31: This work is supported by the project GEO-CRADLE (Coordinating and integRating state-of-the-art Earth Observation Activities in the regions of North Africa, Middle East, and Balkans and Developing Links with GEO related initiatives towards GEOSS), Grant Agreement No. 690133, funded under European Union Horizon 2020 Programme - Topic: SC5-18b-2015, Integrating North African, Middle East and Balkan Earth Observation capacities in GEOSS. We would like also to acknowledge the support for international research staff exchange by REQUA (Regional climate-air quality interactions) project (FP7-PEOPLE-2013-IRSES - Marie Curie Action, PIRSES -GA -2013 -612671) and ACTRIS-2 project (Grand Agreement No. 654109, funded under European Union's Horizon 2020 programme). CALIPSO data were provided by NASA. LIVAS team thanks the

ICARE Data and Services Center (http://www.icare.univ-lille1.fr/) for providing access to CALIPSO data used for the production of LIVAS dataset.

Response to **Anonymous Referee #3** concerning the paper "Dust size parameterization in RegCM4: Impact on aerosol burden and radiative forcing" (http://www.atmos-chem-phys-discuss.net/acp-2016-434).

November 30, 2016 Dear editorial and respected reviewer,

Thank you very much for reviewing our manuscript and providing us with such a constructive feedback. We believe that your comments helped us highlight some critical aspects of the paper and add important content which elevates the quality of our work. All the short comments and typographical errors were corrected in the new version of the paper, while comments that needed more explanation and additional material are discussed below. The structure of the responses includes (1) comments from Referee, (2) a detailed response (3) changes implemented in the new version of the paper. Quotes from the initial version of the paper are highlighted with pale red along with the corresponding page and lines. Quotes from the new version of the paper are highlighted with pale blue along with the corresponding page and lines. The specific phrases changed-modified are underlined for convenience. In cases where the quotes are discussed but not changed are highlighted with grey. I sincerely hope you will be fully satisfied with all the changes we have made.

Best Regards, Athanasios Tsikerdekis

1) I suggest to join all three datasets in the validation part. It makes the reader easier to see the difference between the two aerosol size bin discretization schemes in the view of the reference observational data. Of course, this holds only for those quantities, where observations are available.

We agree with the reviewer and therefore Figure 12 was updated and now includes the LIVAS DOD annual cycles along with the DUST4 and DUST12 experiments. Future readers can now directly compare DOD of LIVAS with the two experiments in the dust size bin discretization Section of the paper "3.2 Comparison of 4-bin and 12-bin experiments". Although since the DUST12 experiment is not discussed in the Section "3.1 Evaluation", we believe it would be confusing to include it in prior plots. Also, the following phrase was updated:

P 14, L 27: Figure 12 shows the annual cycle of DOD for the DUST4 and DUST12 experiments. P 16, L 1: Figure 12 shows the annual cycle of DOD for LIVAS, DUST4 and DUST12 experiments.

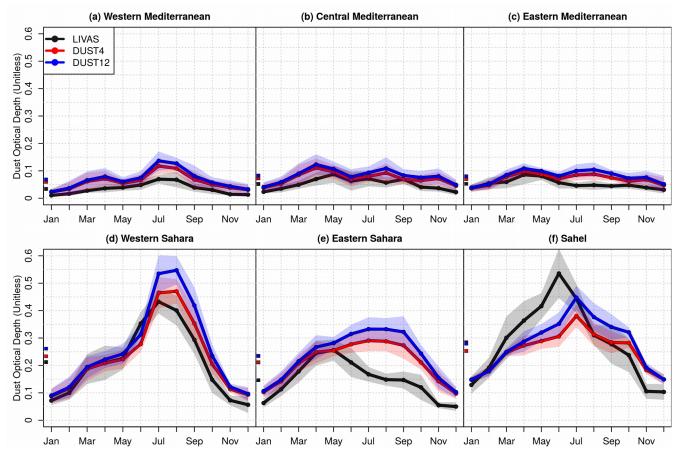


Figure 12. Dust optical depth annual cycle of LIVAS, DUST4 and DUST12 experiments for the period December 2006 to November 2014.

2) The presented changes caused by the introduction of new aerosol size bin scheme are rather small. I agree with the other reviewer, that this requires a statistical significance test of the differences.

Thank you for highlighting the statistical significance issue. We have calculated the statistical significance of the differences using the monthly data for each grid point for the DOD, dust column burden and radiative forcing using the two-tailed paired t-test. In all variables and almost in all grid points the differences are statistically significant at the 95% confidence level (p.value < 0.05). We have also updated the text in the new version of the paper to highlight this issue. The plots shown below were not updated in the paper since we believe that they do not offer additional information to the reader; the vast majority of the grid points show differences that are statistically significant (shaded areas in the differences), hence we are just mentioning this in the text.

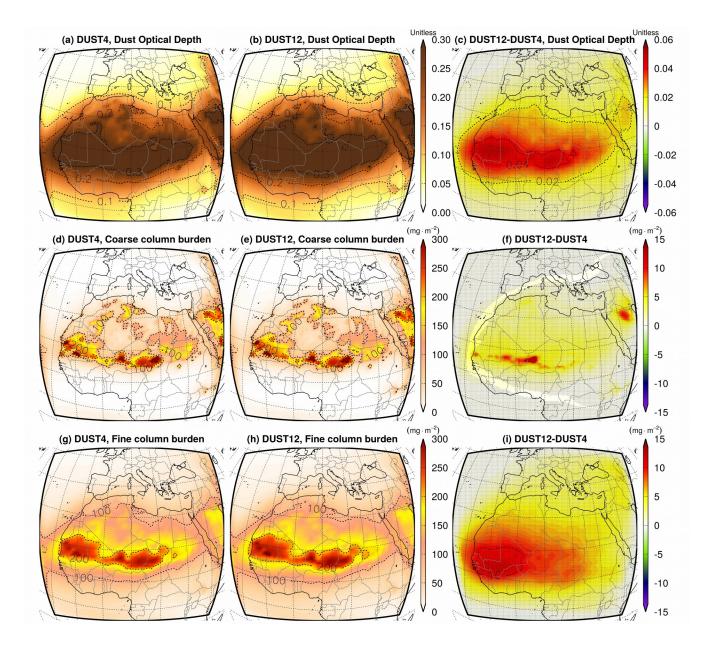
Pg 14, lines 13-20: The DOD percent increase is between 10.4% and 13% for all the subregions. Furthermore, there is a distinctive increase by 0.04 with the 12-bin model over the Sahara desert and especially along the Sahel region where the DOD values are higher (Figure 11c). In comparison with the DUST4 simulation the DUST12 simulation increases the deposition lifetime (column burden/total deposition flux) by 3.5 hours and 2 minutes for fine and coarse particles respectively. Consequently, that increases the dust column burden of fine (+4%) and coarse (+3%) particles (Figure 11f,i). The changes in the fine particles correlates better with the changes in DOD, because dust extinction coefficient is much higher for fine particles ($<2.5\mu$ m) (Figure 1). Over the Middle East and the northern

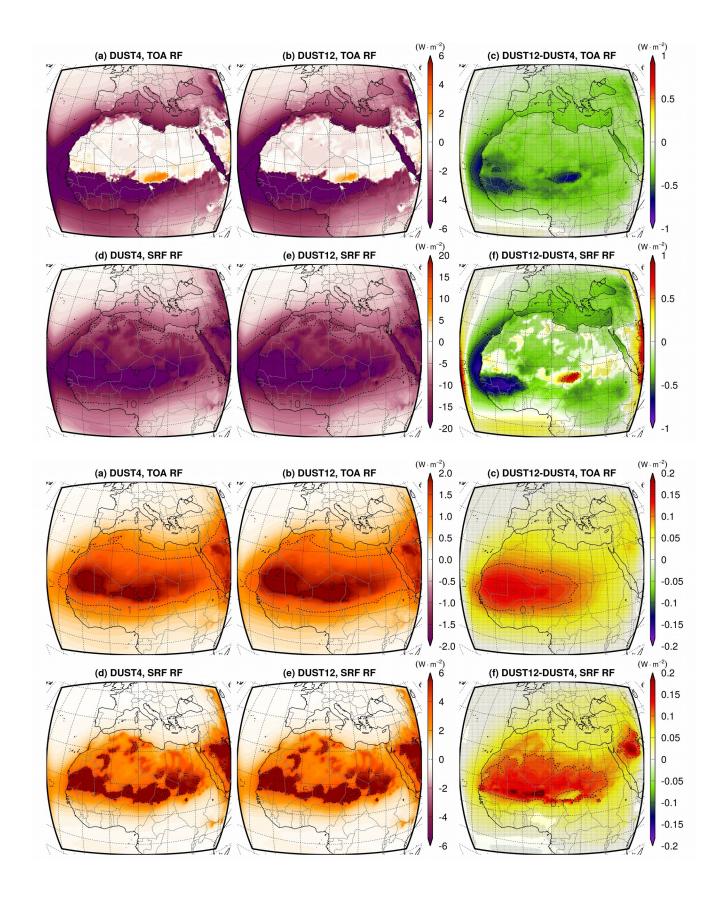
part of the Arabian Peninsula we observe a distinct increase on the coarse dust column burden by 10mg•m-2.

Pg 15, lines 17-26: The DOD percent increase is between 10.4% and 13% for all the subregions. Furthermore, there is a distinctive increase by 0.04 with the 12-bin model over the Sahara desert and especially along the Sahel region where the DOD values are higher (Figure 11c). In comparison with the DUST4 simulation the DUST12 simulation increases the deposition lifetime (column burden/total deposition flux) by 3.5 hours and 2 minutes for fine and coarse particles respectively. Consequently, that increases the dust column burden of fine (+4%) and coarse (+3%) particles (Figure 11f,i). The changes in the fine particles correlates better with the changes in DOD, because dust extinction coefficient is much higher for fine particles (<2.5μm) (Figure 1). Over the Middle East and the northern part of the Arabian Peninsula we observe a distinct increase on the coarse dust column burden by 10mg•m-2. The differences of DOD and column burden between the two experiment, calculated from the monthly data for each grid, are statistically significant at the 95% confidence level according to a two-tailed paired t.test for almost all the grid points of the simulated domain.

Pg 16, lines 11-13: Similarly, at the surface the positive radiative forcing rise by 0.08W·m-2 (3.0%) and 0.09W·m-2 (2.7%) in eastern and western Sahara and 0.9W·m-2 (6.3%), 0.08W·m-2 (4.8%) and 0.9W·m-2 (6.5%) in western, central and eastern Mediterranean (Figure 14f).

Pg 17, lines 16-20: Similarly, at the surface the positive radiative forcing rise by 0.08W·m-2 (3.0%) and 0.09W·m-2 (2.7%) in eastern and western Sahara and 0.9W·m-2 (6.3%), 0.08W·m-2 (4.8%) and 0.9W·m-2 (6.5%) in western, central and eastern Mediterranean (Figure 14f). The shortwave and longwave radiative forcing differences between the two experiment, calculated from the monthly data for each grid, are statistically significant at the 95% confidence level according to a two-tailed paired t.test for almost all the grid points of the simulated domain.





3) Pg 3, lines 15-16: This statement is true, however please add 1-2 sentences on how this number affects the modeling of aerosols, at least in theory.

Indeed, adding a discussion at this point makes the initial statement more robust. Therefore, we have explained briefly why this is the case as it was suggested.

Pg 3, lines 15-16: An important component that affects the transport and the radiative properties of dust in climate modelling is the number of transport dust size bins.

Pg 3, lines 15-20: An important component that affects the transport and the radiative properties of dust in climate modelling is the number of transport dust size bins. Small dust particles, due to their weight, can travel over long distances and can efficiently reflect/backscatter the incoming shortwave solar radiation, while larger particles, with shorter atmospheric life, can effectively absorb and re-emit in the longwave spectrum. Thus, both the partitioning and the number of dust transport bins, used in atmospheric models, should carefully distinguish dust particles with contrasting radiative properties and transport characteristics.

4) Pg 5, lines 25-26: So aerosols cannot move from one bin to another meaning that there is no aerosol fragmentation or aerosol coagulation?

Yes. The current state of the model assumes that dust particles retain their size and they do not fragment into smaller particles during transport. Thus remaining in the same bin throughout their atmospheric life.

Pg 5, lines 25-26: Each transported bin is considered as a distinct tracer, which assumes that there is no mixing between the dust size bins.

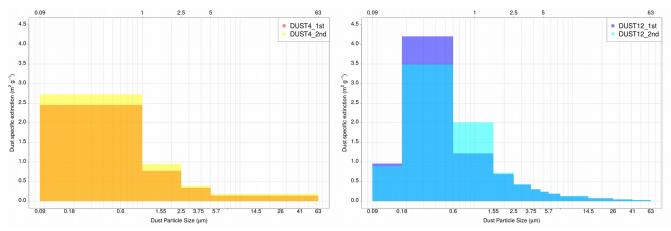
5) Pg 6, lines 5-7: I would be interesting to compare the presented optical properties with those obtained by using only one effective diameter – i.e. the one that describes the bin (the center of it).

Thank you for giving us the opportunity to discuss a more technical aspect of our work. The bin specific extinction coefficient is displayed below for 4 and 12 bins (left and right respectively). The 1^{st} method uses as effective particle radius the mean diameter of each size bin in order to calculate the extinction coefficient, while the 2^{nd} method calculates the extinction coefficient for multiple radii within the range of each size bin and average them in the end. The 2^{nd} method is numerically more accurate and thus it was adopted in our paper. The highest differences between the two methods are observed in fine particles ($<2.5\mu m$) where the optical properties change rapidly with particle size.

These informations could be valuable for future reader we have added the following plot as supplementary material and cited it also in the main text:

Pg 6, lines 4-7: The differences for all the optical parameters are relatively small, because the calculations were performed for multiple effective particle radii within the range of each size bin and averaged in the end, instead of using the mean effective radius of each size bin. Using this method the optical properties between the two experiments are almost identical.

Pg 6, lines 10-13: The differences for all the optical parameters are relatively small, because the calculations were performed for multiple effective particle radii within the range of each size bin and averaged in the end, instead of using the mean effective radius of each size bin (FigureS 1). Using this method the optical properties between the two experiments are almost identical.



FigureS 1: Dust bin specific coefficient for 4 and 12 dust size bins. The first 1st method uses as effective particle radius the mean diameter of each size bin in order to calculate the extinction coefficient, while the 2nd method calculates the extinction coefficient for multiple radii within the range of each size bin and average them in the end.

6) Pg 17, line 2: ..on emissions? I missed something in the manuscript? At this point, I would recommend to present an emission figure to gain some idea about its spatial distribution.

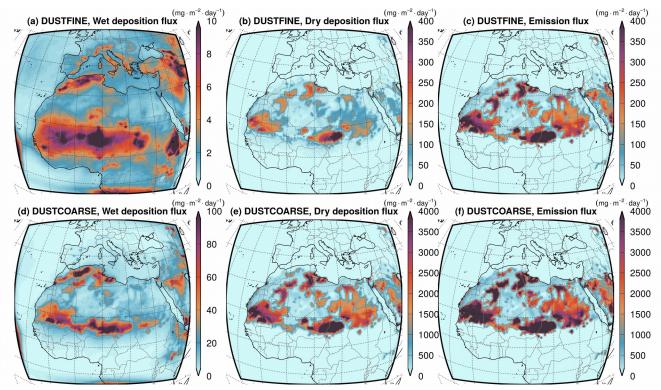
Thank you for pointing this out. Emission is incorrectly referred at this point. The two experiments have the same emission fluxes, since we are testing only the binning effect on DOD, dust column burden and RF. Therefore, we have removed emission and added DOD in the sentence.

Pg 17, Line 1-2: In the present study, we investigate the role of the modelled particle size distribution on the emission, total column burden and radiative effects of dust in a regional climate model.

Pg 18, Line 5-6: In the present study, we investigate the role of the modelled particle size distribution on DOD, the emission, total dust column burden and radiative effects forcing of dust in a regional climate model.

Although since the spatial distribution of emission and deposition of dust might be interesting for future readers we have included the following plot as supplementary material and cite it in the main paper:

Pg 12, lines 4-5: The spatial distribution of wet deposition, dry deposition and surface emission fluxes is depicted for the fine and coarse particles in FigureS 3.



FigureS 3: Wet deposition, dry deposition and surface emission fluxes of fine (a, b, c) and coarse (d, e, f) dust particles in DUST4 experiment for the period December 2006 to November 2014.

7) Pg 18, line 14: The use of word 'underestimates' is not correct, as we have no comparison of RF figures with observation: there is no information on the reference values of the RF, so it can possibly be that the 4 bin approach is closer to the reality.

As correctly indicated the word "underestimates" may be misleading for the reader, therefore we have rephrased the sentence in order to be more comprehensive.

Pg 18, line 12-15: Overall, this study highlights that the radiative differences between the two dust size bin treatments are relatively small. The simulated shortwave radiative forcing by the 4-bin isolog method is to some extent numerically efficient and acceptable. Nevertheless, our work emphasize that the simplified representation of the 4-bin approach underestimates the direct radiative forcing, a fact that should be taking into account by future researches that study the same region.

Pg 19, line 16-19: Overall, this study highlights that the DOD, dust column burden and radiative forcing differences between the two dust size bin treatments are relatively small. The 12-bin isogradient method represents more realistically the physical processes such as deposition and optical properties of dust, nevertheless the 4-bin isolog method is numerically efficient and can be useful for long term regional climate simulations.

Acknowledgements added.

Pg 19, lines 22-31: This work is supported by the project GEO-CRADLE (Coordinating and integRating state-of-the-art Earth Observation Activities in the regions of North Africa, Middle East,

and Balkans and Developing Links with GEO related initiatives towards GEOSS), Grant Agreement No. 690133, funded under European Union Horizon 2020 Programme - Topic: SC5-18b-2015, Integrating North African, Middle East and Balkan Earth Observation capacities in GEOSS. We would like also to acknowledge the support for international research staff exchange by REQUA (Regional climate-air quality interactions) project (FP7-PEOPLE-2013-IRSES - Marie Curie Action, PIRSES -GA -2013 -612671) and ACTRIS-2 project (Grand Agreement No. 654109, funded under European Union's Horizon 2020 programme). CALIPSO data were provided by NASA. LIVAS team thanks the ICARE Data and Services Center (http://www.icare.univ-lille1.fr/) for providing access to CALIPSO data used for the production of LIVAS dataset.

Dust size parameterization in RegCM4: Impact on aerosol burden and radiative forcing

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Abstract. We investigate the sensitivity of aerosol representation in the regional climate model RegCM4 for of two dust parameterizations of the regional climate model RegCM4-for the period 2007-2014 over the Sahara and the Mediterranean. We apply two discretization methods of the dust size distribution keeping the total mass constant: 1) the default RegCM4 4bin approach, where the size range of each bin is calculated using an equal, logarithmic separation of the total size range of dust, using the diameter of dust particles and 2) a newly implemented 12-bin approach with each bin defined according to an isogradient method where the size ranges are dependent on the dry deposition velocity of dust particles. Increasing the number of transported dust size bins theoretically improves the representation of the physical properties of dust particles within the same size bin. Thus, more size bins minimize the error and improve the simulation of atmospheric processes. The radiative effects of dust over the area are discussed and evaluated with the CALIPSO Dust Optical Depth (DOD). This study is among the first studies evaluating the vertical profile of simulated dust with a pure dust product. Reanalysis winds from ERA-interim and the total precipitation flux from the observational gridded database CRU are used to evaluate and explain the discrepancies between model and observations. The new dust binning approach increases the dust column burden by 4% and 3% for fine and coarse particles respectively, which increases DOD by 10% over the desert and the Mediterranean. Consequently, negative shortwave RF is enhanced by more than 10% at the top of the atmosphere and by 1% to 5% on the surface. Positive longwave RF locally increases by more than 0.1 W m⁻² in a large portion of the Sahara desert, the northern part of the Arabian peninsula and the Middle East. The 4-bin isolog method is to some extent numerically efficient, nevertheless our work highlights that the simplified representation of the 4-bin approach underestimates the dust optical depth and the radiative forcing, a fact that should be taking-taken into account by future researches that study the same region.

1 Introduction

Aerosols affect the energy budget of the planet through numerous feedback mechanisms. The direct aerosol climatic impact of -0.27W·m-2 and the indirect effect of -0.55W·m-2 was estimated in the most recent IPCC report, yet the forcing uncertainty remains the highest among all the other factors (Boucher et al., 2013). Aerosols strongly affect Earth's climate by scattering and absorbing the incoming solar radiation (direct effect), burn low clouds through longwave heating (semi-direct effect) or by producing brighter clouds (first indirect effect) with longer cloud lifetime (second indirect effect) (Bangert et al., 2012; Hansen et al., 1997; Karydis et al., 2011; Lohmann and Feichter, 2001; Nabat et al., 2014; Ramanathan et al., 2001; Tegen, 2003). Small particles can effectively scatter/reflect the incoming shortwave radiation while large particles can more effectively absorb and re-emit on the longwave part of the spectrum (Liao and Seinfeld, 1998). Globally, dust is considered to have a slightly negative radiative forcing -0.1W·m-2 (-0.3W·m-2 to 0.1W·m-2) on climate (Boucher et al., 2013). A recent detailed evaluation of RegCM4-CCM3 using satellite-based observations over the European domain highlighted that surface solar radiation bias can be attributed to an overestimation or underestimation of counteracting parameters, with cloud fractional cover, cloud optical thickness and aerosol optical depth being the most important (Alexandri et al., 2015). Since airborne dust play a fairly significant role on the total aerosol optical depth, it is important to understand and improve the probable causes of dust optical depth biases.

Dust production begins when surface wind or wind at a certain height exceeds a threshold. In most studies, the threshold friction velocity is determined as the minimal wind velocity that is capable of sustaining particle movement (Iversen and White, 1982). Practical threshold wind velocity is depended on soil particle size, inter-particle cohesion forces and surface roughness features. Assuming a constant particle density, soil particle size affects their weight. Thus, large particles require higher surface friction velocity to initiate dust movement. On the other hand the inter-particle forces (van der Waals, Capillary, Coulomb forces) acting upon small dust species, that depends on soil moisture and chemical composition, are higher compared to the larger particles and difficult to estimate (Shao and Lu, 2000). Due to the opposite dependency of particles weight and inter-particle cohesion forces size particles with diameter 60µm was estimated to have the minimum wind erosion threshold (Knippertz and Stuut, 2014). Particles of this size have a higher probability to be emitted first and trigger the next stages of dust emission (saltation and disaggregation).

Mineral dust is produced mainly in deserts. Its production depends on surface wind, precipitation and changes in the vegetation cover (Tegen et al., 2000). The Sahara desert is the most important source of aeolian soil dust on the planet (Prospero et al., 2002). Dust from the Sahara is transported northward in Europe and Mediterranean on episodic events quite frequently (11.4 year-1) (Gkikas et al., 2013). The dust burden in the Mediterranean region differs spatially and seasonally with the Eastern Mediterranean experiencing high dust load in spring and the Western Mediterranean reaching its peak in the summer and autumn (Moulin et al., 1998). The central Mediterranean can be considered a transition region where dust transport episodes can occur throughout spring to autumn (Israelevich et al., 2012). Depending on season and area of the Mediterranean, dust can originate from different places in the Sahara desert. Possible dust pathways have been identified for

each season using the TOMS AI satellite data (Israelevich, 2003). In spring for example, dust follows a long trajectory from the Bodélé depression to southern Algeria and then moves northward across the coast of northern Africa to reach Eastern Mediterranean. This long path affects the size of soil particles that reach the Mediterranean in spring in comparison with other seasons. Since dust particles have to travel for such a long distance, dry deposition through gravitational settling forces coarse particles to deposit. Thus, the size of dust particles across Mediterranean in the spring (1.5µm) is half compared to the particles during summer and autumn (3µm), which follow a shorter trajectory (Israelevich, 2003).

The vertical dust distribution has remained uncertain in the past decade, because satellites measurement provided mainly the columnar aerosol optical depth and the stationary observations were spatially sparse. Modelling studies have shown different vertical dust distribution between the Mediterranean, the Atlantic and the Sahara desert (Alpert et al., 2004), while after the launch of the CALIPSO satellite in 2006, our understanding on aerosol vertical distribution increased rapidly (Winker et al., 2009). The particle size distribution of dust changes over height with finer particles reaching higher in the atmosphere and therefore having a higher probability to be transported further because wind speed is also increasing with height. Considering that the fine particles scatter and reflect efficiently the incoming solar radiation, we can grasp the importance of the vertical dust distribution on the shortwave radiative forcing.

An important component that affects the transport and the radiative properties of dust in climate modelling is the number of transport dust size bins. Small dust particles, due to their weight, can travel over long distances and can efficiently reflect/backscatter the incoming shortwave solar radiation, while larger particles, with shorter atmospheric life, can effectively absorb and re-emit in the longwave spectrum. Thus, both the partitioning and the number of dust transport bins, used in atmospheric models, should carefully distinguish dust particles with contrasting radiative properties and transport characteristics. The number of dust bin and the size range resolved in global climate models varies between models, with most using 1-6 dust bins spanning a size range of 0.01-25µm (e.g. Kinne 2003; Huneeus et al. 2011 and reference there in). The gGreater number of dust size bins improves minimizes model error especially for particle dry deposition and thus allows to more accurately simulate both the atmospheric dust burden and the interaction with radiation (Foret et al., 2006; Menut et al., 2007). However, the high computational requirements of global climate models demands as few as possible dust size aggregates and consequently a low size range of aerosol species. Regional climate models with smaller domains of interest usually simulate between 4 to 12 dust size bins (e.g. Alexandri et al., 2015; Basart et al., 2012; Giorgi et al., 2012; Nabat et al., 2012; Solmon et al., 2008; Spyrou et al., 2013; Zakey et al., 2006), while dust transport models have conducted simulation experiments with up to 40 dust bins (Menut et al., 2007).

Dust particle size measurements in the Sahara desert from the Fennee 2011 aircraft campaign show that the dust coarse mode volume median diameter ranges between 5.8 and 45.3 μ m (Ryder et al., 2013). Moving away from the dust source regions the mean/median size of dust particles drops dramatically. In the Mediterranean the mean diameter of dust particles ranges between 2-30 μ m (Goudie and Middleton 2001 and reference therein). It is evident that particles with diameter >25 μ m possess a major role in the burden of dust close and in some cases away from the dust sources. Thus, Foret et al. (2006)

proposed that dust size for the transported bins should range from $0.09\mu m$ to $63\mu m$, considering both the total number and the mass distribution of soil particles.

This study presents new dust features in RegCM4, highlights the sensitivity of dust emission and deposition processes with various meteorological schemes, and analyzes the effect of size bin distribution on dust burden and radiative forcing. In Section 2 we describe the regional climate model RegCM4 and the gridded observational data LIVAS, ERA-interim and CRU. We discuss previous studies on dust size parameterization and explain how the present work expands these studies further. In Section 3 we evaluate the regional climate model RegCM4 using the dust product LIVAS, where high resolution vertical aerosol extinction coefficient measurements were made available and new techniques for the discrimination of pure dust were developed (Amiridis et al., 2013). We also highlight the importance of the partitioning method and the number of dust size bins by comparing two simulations in terms of the dust column burden and dust optical depth. Finally, we discuss the effect of the two binning methods on the dust radiative forcing both in the short and the longwave spectrum.

2 Material and Methods

2.1 RegCM4

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The Regional Climate Model (RegCM) is a space limited numerical model developed at the National Center of Atmospheric Research (NCAR) and the Abdus Salam International Center for Theoretical Physics (ICTP). Notably, it was the first area limited model used for long-term climate simulations (Giorgi and Anyah, 2012). Here we employ the RegCM4, which is described in detail in Giorgi et al. (2012). The hydrostatic core of the model, which is based on the fifth version of the Mesoscale Model (MM5) (Grell et al., 1994), restricts the minimum horizontal resolution of the model to 10km. Thus, the regional scale convective precipitation on RegCM4 is resolved through various convective scheme parameterizations. The vertical computation of the atmosphere is applied on sigma levels. Land-Atmosphere interactions are analyzed with the Biosphere-Atmosphere Transfer Scheme (BATS) (Dickinson et al., 1993), while there is a recently implemented alternative option to use the Community Land Model (CLM4.5) (Oleson et al., 2013). The radiation transfer scheme used in RegCM4 is based on the NCAR Community Climate Model version 3 (CCM3) (Kiehl et al., 1996) with the additional option of the correlated-k Rapid Radiation Transfer Model (RRTM) (Mlawer and Clough, 1997; Mlawer et al., 1997) to improve the model representation of the radiation processes improve radiation processes in the longwave spectrum (Iacono et al., 2000).

The chemical part of the model contains gas phase chemistry (Shalaby et al., 2012; Steiner et al., 2014) as well as natural and anthropogenic aerosols (Solmon et al., 2006; Zakey et al., 2006, 2008). Most of the natural aerosols, such as dust and sea salt, are driven by RegCM4 meteorologyproduced internally (online) in RegCM4 using the meteorological fields, while all the anthropogenic aerosols, organic carbon, black carbon and pollen require emission datasets.

The dust emission scheme is activated in a grid cell when the friction velocity, resolved as a function of RegCM4 simulated wind speed and surface roughness, is higher than the minimum friction velocity threshold. Dust saltation flux is calculated

following Marticorena and Bergametti (1995) and Zakey et al. (2006), while the calculation of dust aerosol vertical flux from the saltation flux follows Laurent et al. (2008).

Particle size distribution (PSD) of the emitted dust and it's relation to surface wind conditions can be modelled using either Alfaro and Gomes (2001) or Kok theory (2011a). The first theory, based on wind tunnel experiments, demonstrated that increasing wind speed close to the surface increases the portion of fine dust particles emitted (Alfaro et al., 1997). Based on these results (Alfaro and Gomes, 2001) suggested that the saltator kinetic energy increases with wind speed, thus the impact produces more fine particles and affects the PSD of dust. On the other hand Kok (2011a) represented the impact of saltator on the surface to act as the fragmentation of brittle materials and suggested that the saltator impact speed does not depend on wind friction speed (Kok, 2011b). Using observational data he established a theoretical expression of particle size distribution independent of surface wind speed. Kok PSD theory shows a significant improvement of the simulated RegCM4 dust optical depth over the European and Northern African domain (Nabat et al., 2012), thus we use this parameterization in these simulations.

Surface roughness and soil moisture, which are essential for the calculation of threshold friction velocity and saltation fluxes, are provided by the surface scheme BATS (Zakey et al., 2006). The soil aggregate distribution, is used to determine threshold velocity and the saltation flux, differs from Zakey et al. (2006) and is taken from Menut et al. (2013), based on the FAO texture class and spatial distribution. Emission processes take place only over desert or semi-desert grid cells. RegCM4 also accounts for possible subgrid partial desert cover emissions on grid cells dominated by different types of soil and soil texture.

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Following the calculation of the dust mass emission fluxes, the tracer transport equation is applied for each transported bin (Solmon et al., 2006). The equation includes transport of the tracers through resolvable winds, horizontal and vertical turbulent diffusion as well as vertical transport due to cumulus convection. The default number of dust size bins that are resolved for transportation in RegCM4 are four (Zakey et al., 2006) with the selection of each size bin defined according to the diameter of dust particles. The size range of each bin is calculated using an equal, logarithmic separation of the total size range of dust, using the diameter of dust particles. This method creates some biases that depend on how representative the selected size bins are compared to the actual dust particle size distribution in each bin. Following the methodology of (Foret et al., 2006), we have implemented a new dust size discretization scheme that resolves twelve dust transport size bins instead of four. The specification of each bin is linked to the gradient of dry deposition velocity as a function of particle diameter. When the change rate of dry deposition velocity to particle size is high, more bins are created on that specified size range, while the opposite happens when the rate changes are slower. The improved partitioning of dust size bins as well as the higher number of bins theoretically enhances the representation of physical removal processes during transport, the estimation of the effective radius of dust particles and the calculation of dust optical properties. Each transported bin is considered as a distinct tracer, which assumes that there is no mixing between the dust size bins.

ground) and turbulent transfer as well as impaction, interception and particle rebound (Zhang, 2001). Wet deposition processes include a size dependent washout scavenging parameterization (Gong, 2003; Seinfeld and Pandis, 1998) and occurs for a small fraction of dust (10%) considered as soluble.

The optical properties of dust are pre-calculated for every size bin and each spectral band of the radiation scheme in use (CCM3 or RRTM) using Mie theory. Due to the high non-linearity of the Mie calculation, we calculated optical properties for each bin and each spectral band, based on a weighted average on size and wavelength assuming that the sub-size distribution follows Kok (2011a), consistent with the emission size distribution. We ensure that the 12 and 4 bins options cover the exact same total size range and that the integrated optical properties over the whole distribution are conserved regardless of the number of bins. Figure 1 depicts the dust specific extinction coefficient, single scattering albedo and asymmetry parameter for both binning options. The differences for all the optical parameters are relatively small, because the calculations were performed for multiple effective particle radii within the range of each size bin and averaged in the end, instead of using the mean effective radius of each size bin (FigureS 1). Using this method the optical properties between the two experiments are almost identical.

In our simulation, we use the default radiation scheme CCM3 to retain the same meteorological fields between the two binning experiments. In the current version of RegCM4, the RRTM scheme calculates cloud-radiation interaction using a Monte Carlo Integration of the Independent Column Approximation (McICA) (Pincus, 2003). The McICA method, although it provides a more detailed and complicated perspective on the horizontal and vertical radiative structure of the clouds, it introduces a random generated noise even when simulating the exact same experiment. In his work, Pincus (2003) accurately highlights that this random generated noise can be significantly reduced to zero with enough ensemble members. But sensitivity tests with RegCM4 show that the changes in the radiative effect between the two dust bin partitioning methods are already small, thus the results can be significantly altered even with the small perturbation contained in an ensemble mean. A downside of the CCM3 scheme is the simplistic representation of the longwave spectrum as far as the dust particles. In Section 3.3 we discuss the relevant impact of the binning methods in the longwave radiative spectrum between the two radiative transfer schemes.

25 **2.2 LIVAS**

The "Lidar climatology of Vertical Aerosol Structure for space-based lidar simulation studies" (LIVAS; Amiridis et al., 2015), is a 3-dimensional global climatic dataset derived from CALIPSO measurements and funded by the European Space Agency (ESA). CALIPSO obtains high resolution profiles of the attenuated backscatter of aerosols and clouds at 532nm and 1064nm, and retrieves aerosol optical properties below optically thin clouds, in clear skies and above clouds (Winker et al., 2009). Thus, in regions with frequent optically thick clouds, aerosol measurements are limited. In regions with an absence of clouds like the Sahara desert, measurements of aerosols species (most likely dust) are available more frequently.

LIVAS utilizes Level 2 (Version 3) product of CALIOP measurements (Amiridis et al., 2015). The CALIPSO Level 2 database determines the vertical location of cloud/aerosol layers (Vaughan et al., 2009), distinguishes the clouds from the

aerosol layers (Liu et al., 2009), identifies aerosol layers into six subcategories (dust, marine, smoke, polluted dust, polluted continental and clean continental; Omar et al. 2009) and calculates the AOD for each selected layer (Young and Vaughan, 2009).

The LIVAS extinction dust product is corrected for the Lidar Ratio (LR) based on multi-year measurements performed by the ground-based lidar stations of the EARLINET lidar network (https://www.earlinet.org). and intensive campaigns in different dust regions arround the globe (Wandinger et al., 2010; Hänel et al., 2012; Baars et al., 2016). The LR of dust particles depends on their refractive index and may vary for aerosols of the same type. The refractive index values rely upon the composition of dust and most importantly on the relative proportion of clay-sized mineral illite in dust (Schuster et al., 2012). Thus, regions with different physiochemical dust characteristics leads to different LR values. The 0.3.1 version of LIVAS separates the globe into three regions, specified based on known dust sources and loadings with specific physiochemical composition and LR for each region. The globally LR value 40sr used in CALIPSO retrieval algorithm (Omar et al., 2009) was replaced with region specific LRs of 40sr, 50sr and 55sr (FigureS 2).

LIVAS has been evaluated against AERONET stations globally by Amiridis et al. (2015). The results show that the aerosol optical depth differences are between ±0.1 in most cases. Over the southwestern Sahara desert, LIVAS underestimates the AERONET AOD by -0.1, and this bias may be related with the dust underestimation of CALIPSO found in previous studies (Amiridis et al., 2013; Schuster et al., 2012; Tesche et al., 2013; Wandinger et al., 2010). Amiridis et al. (2013) showed that LIVAS correlates well with the Dark Target MODIS retrieval over sea, yet the correlation between MODIS Deep Blue (C5 version) and LIVAS over Sahara are weak (results not shown). The new version of MODIS Deep Blue (C6) improved its retrieval accuracy and spatial coverage globally (Sayer et al., 2014, 2015) and the Mediterranean region (Georgoulias et al., 2016a) in comparison to AERONET. Thus, further research is needed to determine the correlation between LIVAS DOD and MODIS Deep Blue (C6) AOD. This could be attributed to the Deep Blue MODIS retrieval, which uses passive remote sensors for dust aerosol optical depth that take into account numerous assumptions (e.g. high reflectivity). Thus, LIVAS is a more reliable product over the deserts with higher accuracy than the products coming from passive remote sensing techniques. Furthermore, Georgoulias et al. (2016b, submitted in ACP) showed that LIVAS correlates well with the high resolution TERRA MODIS and MACC dust optical depth over land covered regions in Eastern Mediterranean.

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Monthly mean LIVAS data is available at a 1°x1° horizontal resolution with a vertical resolution ranging from 60 m (-0.5km and 21km) to 180 m (above 21km) (Amiridis et al., 2015). We concentrate on the troposphere where the vertical distribution of layers is constant. In our work we use the specialized LIVAS pure dust product which includes the extinction coefficient of pure dust calculated from the dust percentage of "dust" and "polluted dust" aerosol subcategories of CALIPSO (Amiridis et al., 2013). Since CALIPSO is a non-geostationary satellite, the obtained mean monthly profiles are highly dependent on the date and time the measurements were taken. Thus, a spatiotemporal mask was produced according to the exact flight track of CALIPSO and applied to the nearest RegCM timestep prior to evaluation.

2.3 Meteorological Data

ERA-interim is a state-of-the-art global atmospheric reanalysis product developed by the European Centre for Medium-Range Weather Forecasts (ECMWF) (Dee et al., 2011). The data assimilation system uses various types of observational data from ground-based stations, radiosondes, ships and satellites. Data availability starts from 1979 and it is extended forward in near-real time. Its assimilation scheme uses a 12 hour cycle, where observational data are combined with forecast information from the previous timestep in order to construct the global atmospheric conditions. ERA-interim reanalysis database is exceptionally useful over isolated areas like the Sahara desert, where meteorological measurements are limited. Wind velocity and direction, associated with dust emission and transport, were used to evaluate RegCM4 wind fields. ERA-interim is used also as boundary conditions in several regional climate models (e.g. RegCM4) for hindcast simulations.

Climate Research Unit (CRU) data is a gridded global climate database of monthly meteorological measurement from ground-based stations (Harris et al., 2014). The dataset includes surface measurements of six meteorological variables, notably precipitation and temperature. Stations are interpolated into a 0.5x0.5 grid that covers all the land surface of the planet (except Antarctica). Data availability peaks between the period 1950-2000 and drops dramatically in the last decade. During the period 2007-2014 and around the area of Sahara desert, more than 600 monthly measurements of precipitation are included in the database from various stations. However, we note that most of the continual measurements occur at stations close to the border of the desert, and station coverage is generally limited over the Sahara.

2.4 Dust particles size discretization

Dust tracers in climate models are usually represented by describing the size distribution with a specified number of bins defined according to the dust particle diameter (D). Increasing the number of transported dust size bins improves the representation of the physical properties and behaviorbehaviour of dust particles within same bin. However, the computational cost, especially in climate studies, generally limits the number of bins used to describe (dust). The transported size bins must be separated in an affordable number of bins and with a method that minimizes the numerical inaccuracies due to dust bin partitioning.

The isolog method, which partitions the dust bins in equal range of log D, is frequently used to specify the range of the dust size bins in climate models (Huneeus et al., 2011 and reference therein). An alternative partitioning method was introduced in (Foret et al., 2006), where they divide the total size range in each bin according to the changes of dry deposition velocity with size. Using a simple one-dimensional box model they simulated an experiment with a detailed particles size distribution that used 1000 size bins within the range of 0.001-100µm diameter. They used this experiment as a reference to evaluate the differences between the isolog and isogradient method. The error ratio between the total dust particle mass (or number) simulated with the given particle size bin scheme (isolog or isogradient) and bin number (4-30bins), to the total dust particle mass (or number) simulated with the reference size distribution (1000bins), showed that for a given number of bins, isogradient scheme scored much better in terms of total dust mass, but isolog scheme produced somewhat better results in

term of total dust particle number. The isogradient method yielded errors lower than 2% with regard to the total particle number and the representation of aerosol optical depth when using more than 8 dust bins (Foret et al., 2006).

Following the above results Menut et al. 2007 implemented the isogradient scheme into the 4D transport model CHIMERE-DUST and did several nested simulations over the Sahara region. To exclude any biases introduced by the emission, deposition and dynamics of the model they evaluated their results using a reference simulation that resolved 40 size bins. They concluded that the isogradient scheme reduced the errors by a factor of 2 compared to the isolog method and that 6 size bins are not sufficient to reproduce the correct concentration levels of dust even with the methodology proposed by Foret et al. 2006.

Both of these studies have shown that using a higher number of bins and the use of isogradient scheme improves the validity of the simulated results. However, they evaluate only the effect of dust bin partitioning on the size distribution dust emissions, either with a simple one-box model (Foret et al., 2006) or a three-dimensional dust transport model CHIMERE-DUST (Menut et al., 2007). In this manuscript, we take this research further by comparing the method and number of dust size bins with in RegCM4 results, and evaluate using observational data, account for biases introduced by the balance of production and loss processes, the calculate of optical properties of dust and evaluate these results with satellite products.

5 2.5 Experimental set-up

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The key parameters of the RegCM4 simulations are presented in the Table 1. The simulation domain includes the Sahara desert and the largest part of the Arabian Peninsula (Figure 2), which captures the two main sources of dust in the Earth (Tegen, 2003). To reduce the contribution of dust from outside the domain requiring chemical lateral boundary conditions, the domain extends south of the Sahara desert. The simulated domain was separated into six distinct subregions Sahel, Eastern Sahara (ESah). Western Sahara (WSah), Eastern Mediterranean (EMed), Central Mediterranean (CMed) and Western Mediterranean (WMed) (Figure 2b). Sahel was delimited using CRU precipitation for the period 2001-2014. The selected grid cells receive annual precipitation between 100mm and 600mm (Ali and Lebel, 2009; Nicholson, 2013) and are located in the southern border of Sahara. Eastern and Western Sahara were selected according to the desert and semi-desert landuse assigned by the model. Their southern borders were masked by the Sahel grid points. The three Mediterranean subregions contain only non-desert grid points and are separated according to their DOD seasonality (Israelevich et al., 2012). Two 8-yearly simulations (4 dust size bins and 12 dust size bins) were performed from September 2006 to November 2014, excluding in our analysis the initial 3 months as a spin-up time. The 4 dust bin experiment (DUST4) implements the isolog approach for the partitioning of the dust size bins, while the 12 dust bin experiment (DUST12) uses the isogradient method. Both experiments use the Kok (2011a) dust particle size distribution theory.

To ensure that the two experiments are identical in every way except the dust bin number and partitioning method, we have both distributions span the exact same total size range and conserve the integrated optical properties (over the whole distribution) whether we consider 4 or 12 bins. Furthermore, there is no interaction between the dust particles and the radiation fields (e.g. no direct aerosol feedback on climate) to ensure that there are no meteorologically driven changes

between the two simulations. The radiative forcing of dust is calculated using separate radiation call during the simulation. Consequently, the meteorology of the two experiments is identical.

The initial and boundary conditions of the mean monthly dust burden were acquired from a global simulation (CAM + EC-EARTH), for the decades 2000-2009 and 2010-2019. Therefore, a constant static map was used as boundary conditions for each month. A certain degree of error is expected at the boundaries of our domain that will not capture individual dust events or strong interannual variability. However, our analysis shows that the boundary forcing in the examined sub-regions is negligible in the dust annual cycle column burden of fine and coarse particles (Section 3.1). Because the aerosol lateral boundary conditions used a 4-bin dust model, we modified the boundary conditions for the 12 bin experiment. The new bins were defined according to the ratio between the 4 and 12 bin size range and size limits. Multiplying this ratio by each month and atmospheric level we have created the necessary global boundary condition for the 12 bin dust scheme. This method assumes that the number of particles is homogeneously distributed in each bin.

The dust size range of the boundary conditions dataset spans $0.01\mu\text{m}$ - $20\mu\text{m}$. Therefore, coarse dust size bins in both DUST4 and DUST12 scheme (e.g. that exceed the upper limit of the $20\mu\text{m}$) are not influenced by boundary conditions. This does not introduce a significant bias in our results since large particles (> $20\mu\text{m}$) have a short atmospheric lifetime and the main dust sources from outside the domain are located far from our simulation boundaries.

Table 1. Simulation options implemented in RegCM4.

Grid Dimensions	140 x 160, 18 sigma levels
Horizontal Resolution	50km
Meteorological Boundary Conditions	ERA-Interim (Dee et al., 2011)
Surface Model	BATS (Dickinson et al., 1993)
Chemical Boundary Conditions	CAM + EC-EARTH
Cumulus Convection Scheme	Tiedtke (Tiedtke, 1989)
Transfer Radiation Scheme	CCM3 (Kiehl et al., 1996)
Moisture Scheme	SUBEX (Pal et al., 2000)
Planetary Boundary Layer Scheme	Modified Holtslag (Holtslag et al., 1990)
Dust Tracers	DUST4 (4bins, isolog)
	DUST12 (12bins, isogradient; Foret et al., 2006)
Dust Size Particle Distribution	Kok (Kok, 2011a)

3 Results and Discussion

3.1 Evaluation

The 4 bin simulation (DUST4) was evaluated against the climatological LIVAS Dust Optical Depth (DOD) (Figure 3). The high dust belt (DOD > 0.2) between 15°-25°N in RegCM4 generally matches the observed DOD of LIVAS with some spatial inconsistencies. We discuss these differences for key regions (Figure 2) according to the mean bias, the lower (LCI) and upper (UCI) 95% confidence intervals of the mean and the percent bias (Pbias). RegCM4 underestimates DOD in a zone south of the Sahel around to 10°N by about 0.05. Over the eastern Sahara (ESah), RegCM4 overestimates the climatological DOD by 0.083 (LCI:0.078, UCI:0.088, PBias:63.1%), mainly in Chad and expanding to Libya, Egypt and northern Sudan. In western Sahara (WSah) where the DOD is higher, the model mean overestimation is 0.043 (LCI:0.039, UCI:0.047, PBias:26.2%). Over the Mediterranean all the grid points show a weak overestimation of DOD which for the three Mediterranean regions combined is 0.027 (LCI:0.026, UCI:0.029, PBias:54.7%).

Figure 4 depicts the seasonal cycle of DOD in LIVAS observations and the DUST4 simulation for the six subregions depicted in Figure 2. In the Mediterranean the observed monthly mean DOD values are typically less than 0.1, with the RegCM4 maximum values reaching 0.15 in some months (Figure 4a-c). Although there is an almost constant overestimation of DOD for all Mediterranean regions, the observed annual cycle maximum and minimum values are simulated adequately and we note that generally, the monthly DOD values are relatively low. In the western Mediterranean DOD peaks in summer (Figure 4a) while the eastern Mediterranean peak is in spring (Figure 4c). Generally, the RegCM4 DUST4 simulation overestimates DOD with a secondary annual maximum in spring and summer over the western and eastern Mediterranean,

respectively. The central Mediterranean is the transition area that receives dust throughout the year from transport paths that affect both Eastern and Western Mediterranean (Israelevich et al., 2012). Therefore, it exhibits a broad maximum that peaks mainly in spring and remains until late summer (Figure 4b).

The spatial distribution of wet deposition, dry deposition and surface emission fluxes is depicted for the fine and coarse particles in Figure 8. Over desert and semi-desert areas, DOD values are higher and show greater intra-annual variability and amplitude in comparison to the Mediterranean subregions (Figure 4d-f). Western Sahara DOD shows a strong summer maximum of 0.35 that is seven times higher than winter values (Figure 4d). RegCM4 simulates accurately the mean monthly values in the first six months and overestimates in the summer and autumn by 0.03-0.07. In the eastern Sahara, there is a significant overestimation of DOD during summer and autumn where the positive bias of the model is more than 0.1 in most months (Figure 4e). The Sahel annual cycle is affected by the southward transport of dust from Sahara and the seasonal movement of the Inter Tropical Convergence Zone (ITCZ) (Ridley et al., 2012; Rodríguez et al., 2015). The local emission sources are activated mainly during winter (dry season) when soil moisture is low but DOD peak in summer (Figure 4f), indicating that the dust annual cycle is strongly affected by the inflow of dust from the Sahara into the Sahel. RegCM4 underestimates DOD during August-January and overestimates it during March-June (Figure 4f).

To understand the simulated processes of dust production-destruction-transport in the model and explain the discrepancies identified in the DOD annual cycle, we investigate the annual cycle of dust column burden for coarse-silt size (Dd > 2.5µm, Figure 5) and fine-clay size (Dd < 2.5µm, Figure 6) dust particles alongside the production/destruction column tendencies of dust. Fine particles dominate the annual cycle of the total column burden and control its seasonal variability, whereas coarse particles display a weak seasonal variability and its intra-annual amplitude is generally negligible in comparison to fine particles. The major tendencies that controls the dust column burden in both fine and coarse particles is emission and sedimentation. Additionally, vertical turbulence, horizontal and vertical advection plays a considerable role in the desert regions for fine particles. The fine column burden annual cycle is anti-correlated with the absolute values of production/destruction tendencies in the desert, e.g., during the warm season the absolute values of tendencies tends to be small and fine dust column burden high, with the opposite in the cold season. In most regions, fine and coarse particles peak during the same month, enhancing the total column burden seasonal variability (e.g. western Sahara, Sahel; Figure 5e-f, Figure 6e-f). However, in the eastern Sahara (Figure 5d and Figure 6d), fine and coarse annual cycles follow a completely different pattern. Coarse particles peak in spring while fine particles exhibit a broad spring-summer maximum that peaks during late summer. This indicates that fine particles are responsible for the overestimation of DOD in the Eastern Sahara as their burden and the size specific extinction coefficient (Figure 1) is higher in comparison to the coarse dust particles.

Over the Eastern and Western Sahara both emission and sedimentation fluxes display a distinct spring maximum in March (Figure 5d,e and Figure 6d,e), that does not explain the annual cycles of dust column burden. The ratio of emission to dry deposition flux is connected to the dust column burden, rather than the absolute values of emission and dry deposition flux. Values greater than 1 demonstrate periods where the emission dominates in comparison to dry deposition rate, while values smaller than 1 depicts the dominance of deposition. Coarse particles, due to their low atmospheric lifetime, are deposited

close to their source region and shortly after emission, therefore exhibiting a very low seasonal variability. Fine particles on the other hand remain in the atmosphere for longer and can travel in long distances, affecting substantially the emission to dry deposition ratio.

In the Western Sahara the fine particle emission to deposition ratio for the full simulation period is 1.46, which indicates that the model emits 46% more fine particles compared to its deposition. The coarse particle ratio is close to 1. In Eastern Sahara the emission to deposition ratio is much higher than in the Western Sahara, with values of 1.80 and 1.31 for fine and coarse particles, respectively. The strong and almost constant annual NNE and NNW winds in the region transports dust away from the source region increasing the dust outflow of the Eastern Sahara (Figure 7). Thus, deposition in the Eastern Sahara decreases and the emission to deposition ratio rises. Furthermore, the fine particle emission to deposition ratio steadily increases from March to August, leading to an accumulation of fine dust particles in the atmosphere that increases the column burden and the DOD. In the Sahel throughout the year, emissions are two times greater than the deposition for fine particles, with a mean ratio 2.26. As discussed later, RegCM4 exhibits some biases in the total precipitation annual cycle that influences this ratio.

The column burden of dust can be affected by numerous meteorological variables that may alter the emission, deposition or the circulation and transport of dust. Thus, we have evaluated these variables with the observational gridded and reanalysis data to explain the spatial biases observed in the DOD. We chose the ERA-Interim for the evaluation of wind fields (surface, 925hPa and 850hPa) in order to have large spatial coverage and a long-term, continuous availability of data. The surface wind velocity in RegCM, that constitutes the main driver of dust production through erosion, is comparable to ERA-Interim (FigureS 14). We observe some positive and negative biases over the desert that generally are less than 1 m s-1. Although the mean wind velocity is linked with dust emission, wind gustiness and dust emission fluxes exhibit higher temporal and spatial correlation (Engelstaedter and Washington, 2007). A wind gust is defined as a sudden increase of wind speed that last less than 20 seconds. In our simulations, wind speed is calculated within the internal timestep (120 seconds) of our model and wind gust is considered as the maximum value of wind velocity within the last output timestep (6 hours). Typically, this calculation method is applied in most climate models and reanalysis products with different internal and output timesteps. Therefore, it is difficult to draw conclusions when comparing the wind gust in RegCM and ERA-interim, because the wind gust was calculated in different time frames. Alternatively we calculated the mean field of the 6hourly timesteps that exceeded the 0.9 percentile value of wind velocity, averaged over the three desert subregions (Eastern Sahara, Western Sahara, Sahel) (Figure \$\frac{25}{25}\$). The 0.9 percentile value of the averaged wind over the desert is 4.93m/s-1 and 5.92m/s-1 for ERA-interim and RegCM respectively. RegCM clearly overestimates locally high wind velocity over the desert by 1-3m s-1, which indicates that the model potentially overestimates dust emission flux.

In Egypt and northern Sudan, the strong surface NNE winds depicted in ERA-interim is underestimated by RegCM, which may lower the simulated production of dust in the Eastern Sahara. But considering that the same wind pattern continues at both 925hPa and 850hPa and that the larger amount of dust is concentrated on these layers (Figure 7), it can also decrease its southward transport and the outflow of dust in Eastern Sahara. In Figure 8, the annual cycle of the meridional wind

component confirms that during the summer (June-September) the southward wind is underestimated by the model by more than 1m·s-1. Thus, a high load of dust remains stationary for longer period over Eastern Sahara that likely increases the modelled column burden and the DOD (Figure 4e).

Away from the Sahara region and especially in semi-arid environments, like the Sahel, precipitation can affect both emission and wet deposition processes. According to CRU database RegCM overestimates precipitation in the Sahel by 10-20 mm month-1 (FigureS 36) which increases wet deposition as well as soil moisture and vegetation (Engelstaedter et al., 2006). Consequently, more dust is deposited and less is emitted. According to the annual cycle of total precipitation (Figure 9) the overestimation of precipitation during April, May and June probably contributes into the underestimation of DOD (Figure 4f). The same process prevents the southward transport of dust to lower latitudes and is partly responsible for the strong underestimation of DOD close to the Gulf of Guinea.

The vertical distribution of dust is evaluated for the six subregions for vertical tendencies (advection, convective transport, vertical turbulence and sedimentation) in Figure 10. The first 200m of Dust Extinction (DEX) values were excluded from the analysis due to measurement restriction of LIVAS. The tendencies are illustrated in the plot as percentages for the layers 0.2-5km and 5-10km. DEX is overestimated by RegCM in all subregions in the middle and upper Troposphere. More than 95% of dust is located in the first 5km from the surface according to DEX measurements from LIVAS. However, RegCM places more dust at higher altitudes, with only 80-90% located between 0 to 5km. In Western Sahara and Sahel, DEX is underestimated by RegCM until the height of 5km (Figure 10d and 10f) and above that height there is a constant positive bias that decreases with altitude. The large observed DEX values between 3km and 5km over the Sahel indicates the high amount of fine dust particles that reach higher altitudes as compared to the Eastern and Western Sahara. In the Eastern Sahara the DEX is overestimated between 3-13km. At the western, central and eastern Mediterranean regions, the DEX is overestimated above about 2-3km and underestimated below this height (Figure 10a-c).

In the model, the vertical transport of dust is controlled by four main processes: sedimentation, vertical turbulence, vertical advection and convective transport. The sedimentation tendency is always negative and equally important in all heights, whereas vertical turbulence plays a major role in the first couple kilometers of the atmosphere and vertical advection and convective transport, prevail in higher altitudes. As shown in Figure 10, in first 5km of the atmosphere sedimentation exhibit negative values (contribution >-40%), which removes dust from the atmosphere. On the other hand, vertical turbulence is the main force that raises dust upwards (contribution >50%). The planetary boundary layer (PBL) scheme based on Holtslag et al. (1990) produces high vertical turbulences in the first couple of kilometers in the atmosphere, and this may contribute to the underestimation or overestimation of the modelled DEX profile as compared to LIVAS measurements. We have simulated 2008 with the alternative PBL scheme option in RegCM4 based on Bretherton et al. (2004), which suggests an improvement as far as the vertical distribution of dust in the PBL, although column DOD and DEX bias increases, especially over Eastern Sahara (FigureS 47). The vertical advection and more importantly convective transport between 5 and 10km show positive values on average, which transports dust upward. Over the Mediterranean region vertical advection (>20%) and convective transport (>30%) contribution is equally important (Figure 10a-c), while in Sahara desert and Sahel

convective transport is the most significant factor (>65%)(Figure 10d-f). Considering the overestimated DEX profile in this altitude range it suggests that the cumulus convection activity/convective transport mechanism is overactive in Tiedtke scheme (Tiedtke, 1989) or another negative sign process (e.g. sedimentation) is not properly represented by the model. Discrepancies in the vertical distribution of dust can also be misinterpreted from possible local emission errors, the lack of simulated vertical levels or wet deposition biases. Further research is needed in this regard to reduce the mean vertical distribution bias of dust in the model.

The model was evaluated also in terms of the surface net downward shortwave and net upward longwave radiation flux against CERES satellite measurements (FigureS 8). In the shortwave spectrum radiation flux bias ranges between -10Wm-2 and 10Wm-2 above the desert and the Mediterranean, while in the longwave the differences are mostly positive (~10Wm-2).

10 3.2 Comparison of 4-bin and 12-bin experiments

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The optical properties for the 4 and 12 dust size bins simulations were calculated using a Mie scattering code for each CCM3 radiation band. As noted above, the meteorological forcing between the two models is the same, therefore the emission fluxes which depends on the model's resolvable winds remains the same. Thus, the changes in column burden between the two experiments can only emerge from the new dust size discretization that theoretically improves dust transport and dry deposition processes, and the changes in DOD can only be attributed in changes related to transport and deposition.

Figure 11 compares the DOD coarse and fine column burden between the DUST4 and DUST12 for the full simulation period. The DOD percent increase is between 10.4% and 13% for all the subregions. Furthermore, there is a distinctive increase by 0.04 with the 12-bin model over the Sahara desert and especially along the Sahel region where the DOD values are higher (Figure 11c). In comparison with the DUST4 simulation the DUST12 simulation increases the deposition lifetime (column burden/total deposition flux) by 3.5 hours and 2 minutes for fine and coarse particles respectively. Consequently, that increases the dust column burden of fine (+4%) and coarse (+3%) particles (Figure 11f,i). The changes in the fine particles correlates better with the changes in DOD, because dust extinction coefficient is much higher for fine particles (<2.5μm) (Figure 1). Over the Middle East and the northern part of the Arabian Peninsula we observe a distinct increase on the coarse dust column burden by 10mg·m-2. The differences of DOD and column burden between the two experiments, calculated from the monthly data for each grid, are statistically significant at the 95% confidence level according to a two-tailed paired t.test for almost all the grid points of the simulated domain.

The dust bin resolution also depends on the emission size distribution considered for the dust. In our study we use the Kok (2011a) dust Particles Size Distribution (PSD), which as already mentioned improves the DOD and the seasonality over the Sahara region (Nabat et al., 2012). But Kok (2011a) PSD drops very fast for large and small dust particles. Other typical dust size distributions (e.g. Alfaro et al. 1998; Zender et al. 2003) do not show such a sharp drop when increasing or decreasing the dust particle diameter (Kok, 2011a), thus they could be much more sensitivity to the binning partitioning method and number which will generate bigger changes in dust column burden and DOD.

Figure 12 shows the annual cycle of DOD for LIVAS, the DUST4 and DUST12 experiments. The monthly differences of the two experiments against LIVAS can be found in the supplementary FigureS 59. In all regions, the DUST12 experiment increases the DOD in comparison to DUST4. This is attributed to the increase of lifetime in fine and coarse dust particles in DUST12 with respect to DUST4 as also discussed earlier. Previous studies (Foret et al., 2006; Menut et al., 2007) revealed that the new approach in dust size bin partitioning and number (which is also adopted in our DUST12 experiment) more realistically simulates the transport and dry deposition of the dust size bins. This does not imply that the simulated biases will be reduced for all regions in the DUST12 experiment. The major factor that controls and potentially creates the first order biases in dust models is the balance between the emission and sedimentations terms (e.g. Figure 5, Figure 6). As discussed in Section 3.1, some positive DOD biases (e.g. Eastern Sahara) might be due to an underestimation of the outflow of dust or a local overestimation of the emission flux from surface wind velocity errors. Therefore, although the new dust size parameterization theoretically improves dry deposition, it does not necessarily regulate or improve the biases that originate from other processes.

3.3 Radiative Forcing

Dust particles can interact both with the shortwave and the longwave radiation, creating a dimming or a heating radiative effect on climate (Liao and Seinfeld, 1998). Some of these processes are illustrated for the shortwave spectrum in FigureS 610. According to the model the radiation forcing over the Sahel and the North Atlantic ocean is great than -5W·m-2 (Figure 13a,b). The albedo on the desert areas is already very high and in most cases, surface albedo does not change from the suspended dust. However, close to the high emission dust source of Bodélé depression we can observe positive RF values. In the model dust aloft decreases the already high albedo of the desert and creates a positive RF at the TOA. In the Bodélé depression, sediments were deposited during the Holocene in the bed of the Megachad paleolake and the large diatomite sediments formed there have high albedo values that are visible in satellite images (Bristow et al., 2010). It is interesting that RegCM4 simulates a positive radiative response caused by the combination of high surface albedo values and high emission fluxes on that area, and the change to DUST12 decreases this positive response (Figure 13c). The DUST12 experiment enhances the negative radiative forcing in the central and eastern Mediterranean by -0.24W·m-2 (10.5%) and in eastern by -0.18W·m-2 (8.7%). The highest absolute changes are located over the Sahel where the negative radiative forcing strengthens by -0.41W·m-2 (12.1%).

The radiative process that backscatters and reflects the incoming solar radiation is the dominant global mechanisms that affects climate. As shown in Figure 13d and Figure 13e, dust prevents more than 20W·m-2 of incoming solar radiation from reaching the surface on the desert and 5-10W·m-2 on the Mediterranean. The highest absolute differences between the two experiments are located on the western part of Africa between 15°-20°N, where the highest fine dust burden differences between DUST4 and DUST12 were noted (Figure 11i). These negative changes are located in the region with the most persistent dust concentrations (Figure 11g,h) and changes in the dust size bins might have a bigger effect over the long range Trans-Atlantic transport than local forcing. It is worth discussing the source of the surface positive radiative forcing

differences between the DUST12-DUST4 experiment (Figure 13f), which is related to the TOA discussion. The positive RF at the TOA over the Bodélé depression is reduced in the DUST12 experiment, allowing less downward shortwave radiation to reach the surface. Consequently, there is less available downward radiation to be scattered and reflected on these areas, and the negative radiative forcing in DUST12 is smaller than DUST4. As a result the difference (DUST12-DUST4) is positive. The simulated shortwave radiative forcing by the 4-bin isolog method is to some extent numerically efficient and acceptable. Nevertheless, our work emphasize that the simplified representation of the 4-bin approach underestimates the direct radiative forcing of dust by 13.7% (-0.29 W·m-2) and the SW SRF radiative forcing by 1.8% (-0.23 W·m-2). A fact that should be taking into account by future researches that study the same region.

On the longwave spectrum, the RF is always positive. Coarse dust particles in the atmosphere absorb the upward longwave Earth's radiation and remit it either towards the surface or upwards to space. A part of the Earth's longwave radiation is absorbed by dust and limits the portion of upward longwave radiation that reaches the TOA, making the RF positive (Figure 14a,b). A fraction of the absorbed longwave radiation is remitted back towards the Earth and increases the downward longwave radiation on the surface, creating a positive RF (Figure 14d,e). In both cases the DUST12 experiment enhances the positive RF by 0.1W·m-2 in a large portion over the Sahara desert, the northern part of the Arabian peninsula and the middle east (Figure 14c,f). More specifically at the TOA the positive radiative forcing enhances over the eastern and western Sahara by 0.08W·m-2 (6.9%) and 0.07W·m-2 (5.9%) while in western, central and eastern Mediterranean increases by 0.02W·m-2 (7.8%), 0.03W·m-2 (7.8%) and 0.04W·m-2 (8.3%) (Figure 14c). Similarly, at the surface the positive radiative forcing rise by 0.08W·m-2 (3.0%) and 0.09W·m-2 (2.7%) in eastern and western Sahara and 0.9W·m-2 (6.3%), 0.08W·m-2 (4.8%) and 0.9W·m-2 (6.5%) in western, central and eastern Mediterranean (Figure 14f). The shortwave and longwave radiative forcing differences between the two experiment, calculated from the monthly data for each grid, are statistically significant at the 95% confidence level according to a two-tailed paired t test for almost all the grid points of the simulated domain.

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However, the current treatment of the optical properties of dust in the longwave spectrum of CCM3 scheme is limited and does not account for specific absorption coefficient for each dust size bin. Furthermore, the CCM3 longwave bands are concentrated on the absorption of H₂O and CO₂ and they do not integrate in detail the absorbing aerosol component part of the longwave. Therefore, we have conducted two similar dust experiments for June 2008 using the radiation transfer scheme RRTM, which is known for its detailed longwave calculation. We note again that the current version of RRTM (+McICA) produces a random generated noise on radiation fields, therefore the dust emission fluxes are not identical between the DUST4 and DUST12 experiments. Our preliminary simulations using the RRTM scheme (FigureS §12), have shown notably lower longwave dust RF over the desert in comparison to CCM3 (FigureS 711) for the same period and roughly the same DOD levels and spatial patterns. More specifically, the DOD and the longwave radiative forcing averaged above the Sahara for the CCM3_DUST4 experiment is DOD=0.31, SRF=3.5W·m-2 and TOA=2.1W·m-2. While the related values for the RRTM_DUST4 experiment is DOD=0.29, SRF=1.9W·m-2 and TOA=0.6W·m-2. Although DOD increases in the 12 bin experiment in both radiation schemes (FigureS 7e11c, FigureS 8e12c), the longwave RF changes display a striking difference. With the CCM3 radiation scheme, the longwave radiation linearly increases with higher DOD and dust burden

values (FigureS 7f-11f and i). In comparison, the RRTM scheme uses specific absorption coefficient for each dust size bin, taking into account the fine/coarse dust burden changes. Thus it exhibits local increases or decreases of the longwave RF (FigureS 8f-12f and i), according to the changes of fine to coarse dust burden. Overall the total longwave RF increase in RRTM is smaller compared to CCM3 when spatially averaged over the Sahara desert.

5 4 Conclusions

In the present study, we investigate the role of the modelled particle size distribution on <u>DOD</u>, the emission, total column burden and radiative <u>effects forcing</u> of dust in a regional climate model. We evaluate the regional climate model RegCM4 dust optical depth and dust extinction with the LIVAS dust product to understand potential model biases and link these with size-dependent emission and atmospheric processes. Generally, RegCM4 overestimates dust optical depth over important source regions such as the eastern and western Sahara by 0.083 (Pbias:63.1%) and 0.043 (PBias:26.2%) respectively and in Mediterranean by 0.027 (PBias:54.7%).

The dust optical depth annual cycle of LIVAS correlates with RegCM4 adequately in most regions. In the western Mediterranean, RegCM shows an annual cycle with a summer maximum similarly to LIVAS and in eastern Mediterranean, RegCM captures the spring maximum observed in LIVAS but additionally it illustrates a secondary maximum in summer. In the western Sahara, the RegCM DOD annual cycle correlates with LIVAS, although there is a constant overestimation during summer and autumn while in the eastern Sahara RegCM shows a broad spring-summer maximum while the observations show a clear spring maximum. In the Sahel, the model captures the observed DOD summer maximum.

We then evaluate the modelled emission and sedimentation terms, as these are the most important factors for the production and deposition of dust. Additionally, vertical turbulence, horizontal and vertical advection pose a considerable negative tendency for the fine dust particles ($Dd < 2.5\mu m$). According to the annual cycle of the meridional wind component in ERA-interim, the model underestimates the southward wind in eastern Sahara and probably decreases the outflow of dust in the area. Furthermore, the 0.9 percentile value of the averaged wind over the desert is 4.93 m·s-1 and 5.92 m·s-1 for RegCM and ERA-interim respectively, which indicates that the model potentially overestimates dust emission flux. At the Sahel during April, May and June the total precipitation is overestimated by the model in comparison with the CRU database. Thus, wet deposition is enhanced and emission flux is decreased since the ground has higher moisture. Overall, this process-level analysis suggests that meteorological drivers such as wind and precipitation may explain some model biases with observations.

These changes in the physical processes then link to the radiative properties that affect climate. We evaluate the Dust Extinction (DEX) from the model with LIVAS observations, and find the RegCM overestimates DEX in all subregions in the middle and upper troposphere, that can be either attributed to an overestimation of dust convective transport or to an underestimation of some other deposition process (e.g. sedimentation). In the Western Sahara and Sahel, DEX is underestimated by RegCM below 5km, suggesting that the emissions and deposition processes are not well-balanced in the

model. Our one year sensitivity study showed that the PBL mixing scheme (e.g., changing from Holtslag et al. (1990) to Bretherton et al. (2004)) can potentially improve the dust vertical distribution in the boundary layer by reducing overactive mixing.

The effect of the two different approaches on the number and the partitioning method of dust size bins was investigated in the model with two experiments: a 4 size bin simulation based on the isolog approach (DUST4) and a 12 size bin simulation based on the isogradient approach (DUST12). The DUST12 experiment increases the deposition lifetime by 3.5 hours and 2 minutes for fine and coarse particles respectively in comparison to DUST4. Consequently, the dust column burden increases by 4% (fine) and 3% (coarse) that boosts DOD by approximately 10% over the desert and the Mediterranean.

The negative surface radiative forcing of DUST12 experiment in the shortwave spectrum is regionally enhanced by -0.5W·m-2 while the top of the atmosphere radiative forcing is intensified by almost -1W·m-2 (10%). The negative changes in the surface radiative forcing are concentrated in the western part of Africa and extend over the Eastern Atlantic, where the near-constant annual dust plume is located. On the longwave spectrum DUST12 experiment enhances the positive RF at the surface and TOA by 0.1W·m-2 in a large portion of the Sahara desert, the northern part of the Arabian Peninsula and the middle east. Although the radiation transfer scheme RRTM, known for its detailed integration on the longwave spectrum, revealed that these longwave RF changes can be also locally negative and therefore smaller when spatially averaged over the Sahara desert.

Overall, this study highlights that the radiative differences between the two dust size bin treatments are relatively small. The simulated shortwave radiative forcing by the 4 bin isolog method is to some extent numerically efficient and acceptable. Nevertheless, our work emphasize that the simplified representation of the 4 bin approach underestimates the direct radiative forcing, a fact that should be taking into account by future researches that study the same region. Overall, this study highlights that the DOD, dust column burden and radiative forcing differences between the two dust size bin treatments are relatively small. The 12-bin isogradient method represents more realistically the physical processes such as deposition and optical properties of dust, nevertheless the 4-bin isolog method is numerically efficient and can be useful for long term regional climate simulations. We have to note that other typical size distribution (e.g. Alfaro et al. 1998; Zender et al. 2003) could be much more sensitivity to the binning partitioning method and number which will generate bigger changes in dust column burden DOD and RF.

Acknowledgements

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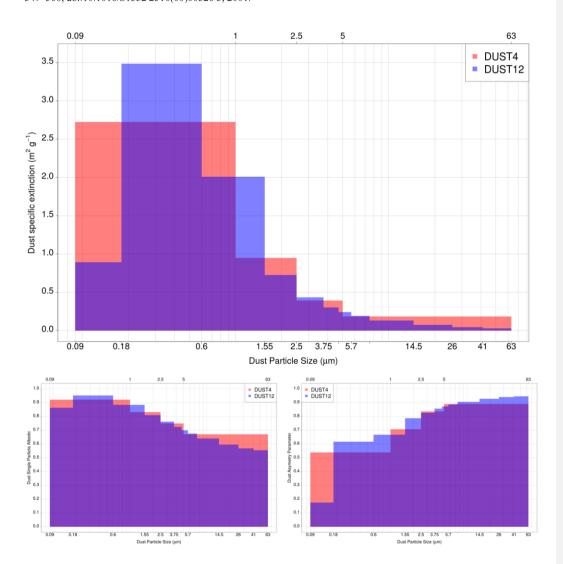
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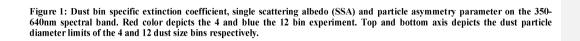
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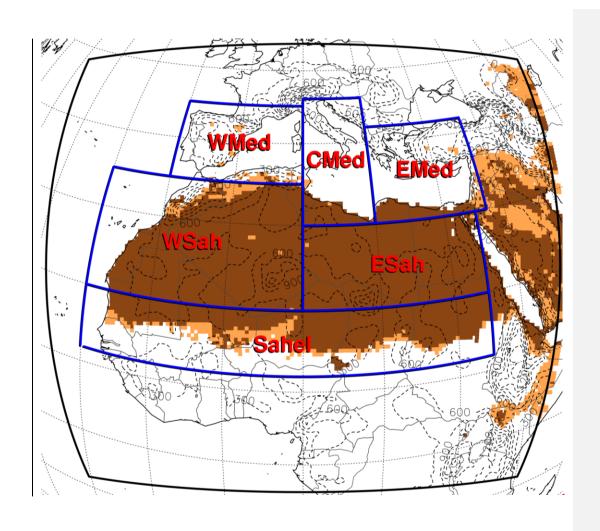
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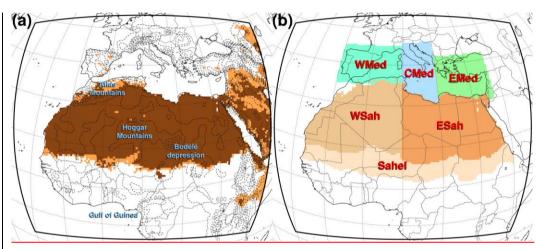


Figure 2: RegCM4 simulated domain (black box) and the subregions selected for the analysis (blueboxes). Background colors depict the desert (dark brown) and semi-desert (light brown) grid cells assign by the model. Dashed lines illustrates the simulated topography used on RegCM4 in meters.

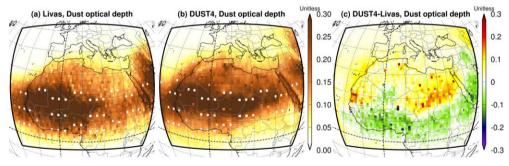
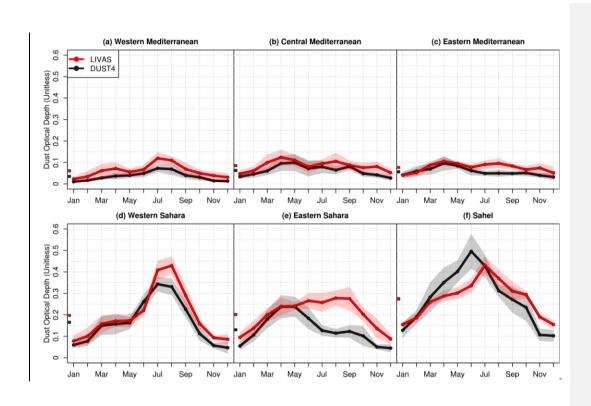


Figure 3: Dust optical depth of the DUST4 experiment and the LIVAS dust product for the period January 2007 to November 2014.



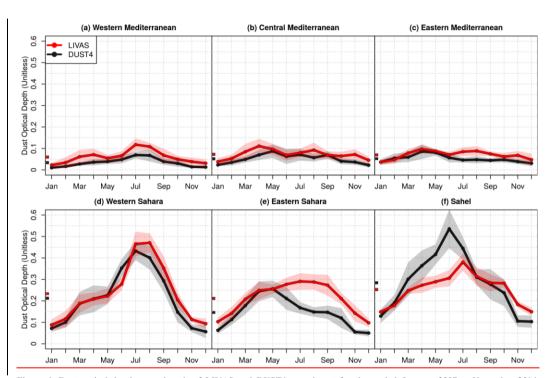
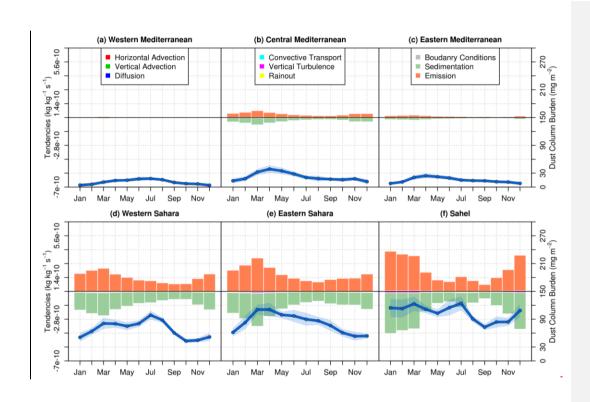


Figure 4: Dust optical depth annual cycle of LIVAS and DUST4 experiment for the period January 2007 to November 2014. Shaded areas display the 95% confidence interval of the mean.



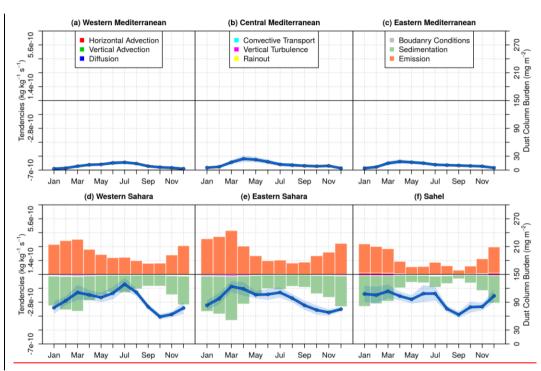
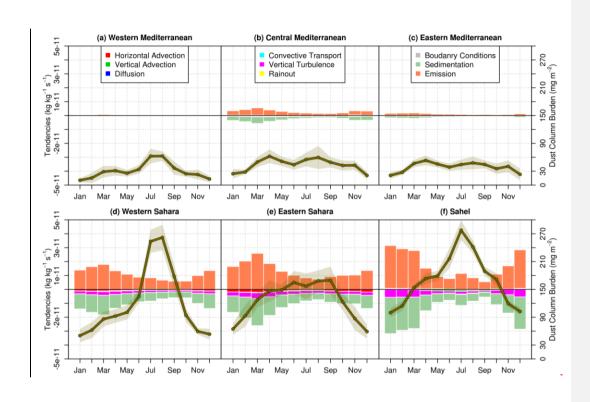


Figure 5: Coarse dust particle column burden of the DUST4 experiment (lines) alongside with the production/destruction column tendencies (bars) averaged for the period December 2006 to November 2014. Shaded areas display the 95% confidence interval of the mean.



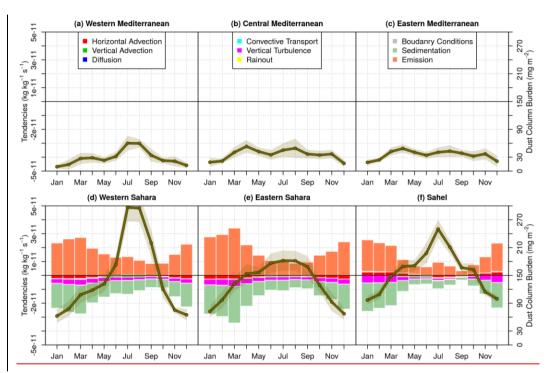


Figure 6: Fine dust particles column burden of the DUST4 experiment (lines) alongside with production/destruction column tendencies (bars) averaged for the period December 2006 to November 2014. Shaded areas display the 95% confidence interval of the mean.

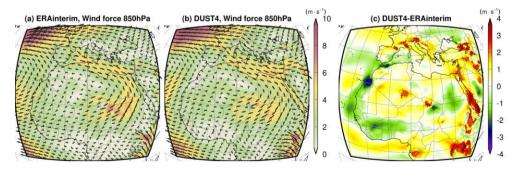


Figure 7: Wind velocity at 850hPa of the DUST4 experiment against the reanalysis ERA-interim for the period December 2006 to November 2014.

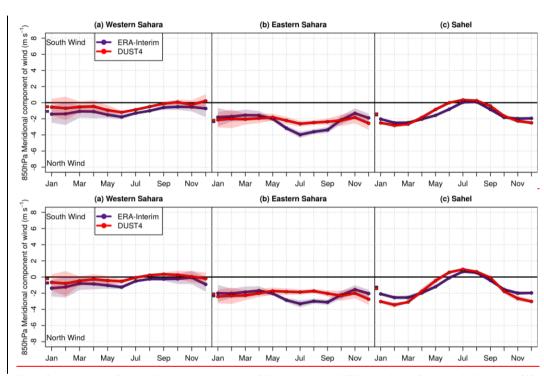
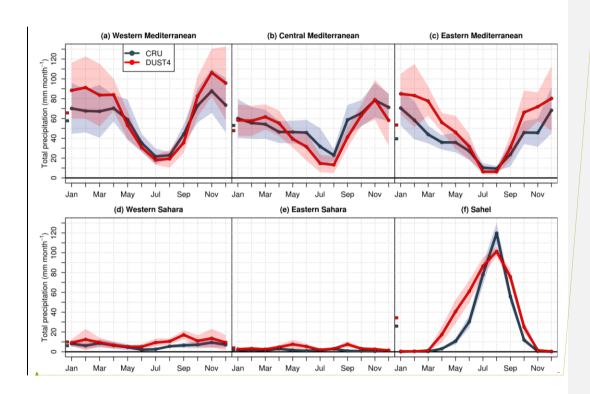


Figure 8: Annual cycle of the meridional wind component of ERA-interim and DUST4 experiment for the period December 2006 to November 2014. Positive and negative values indicate south and north wind respectively. Shaded areas display the 95% confidence interval of the mean.



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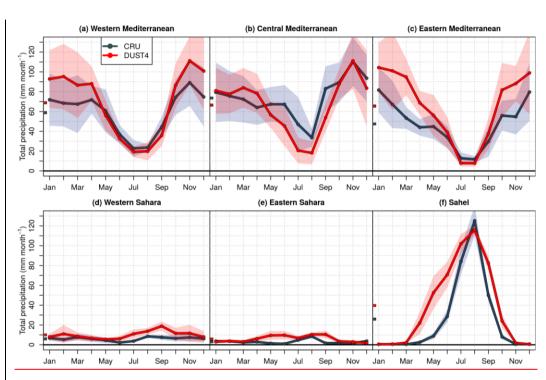
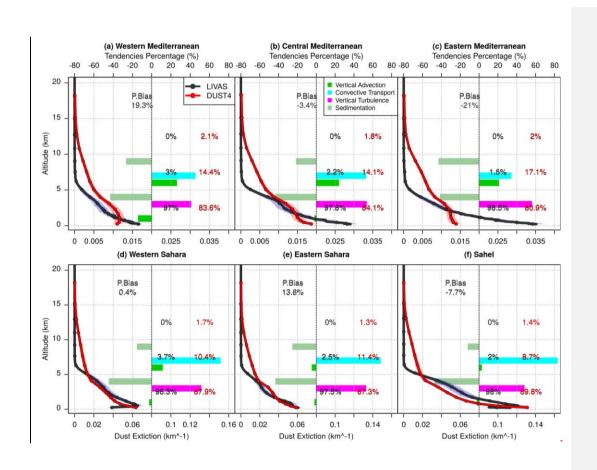


Figure 9: Total precipitation annual cycle of the ERA-interim and DUST4 experiment for the period December 2006 to November 2014.



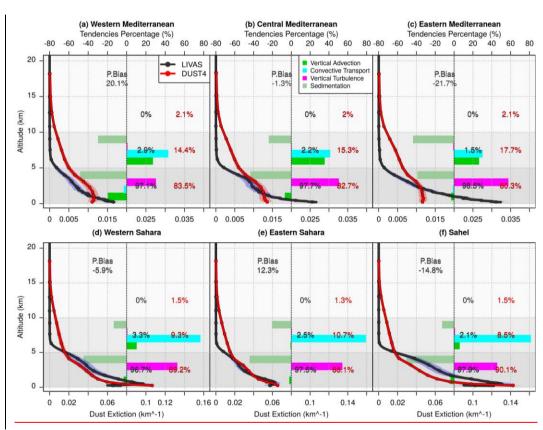


Figure 10: Dust extinction profiles of LIVAS and DUST4 experiment. The percent bias (P.Bias) of dust extinction located between 0.2-5km, 5-10km and 10-20km for each subregion is illustrated on the plots. The bars depict the percent of the vertical tendencies for 0.2-5km and 5-10km.

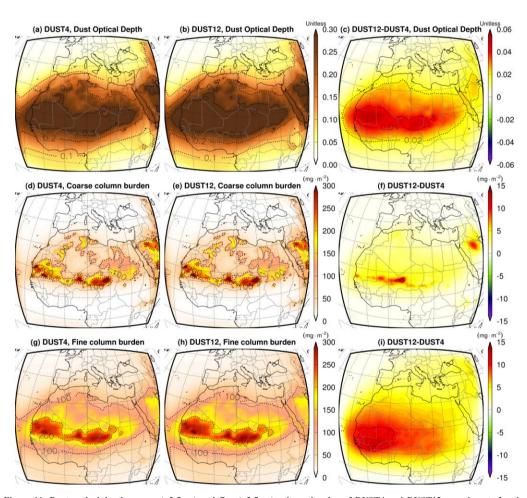
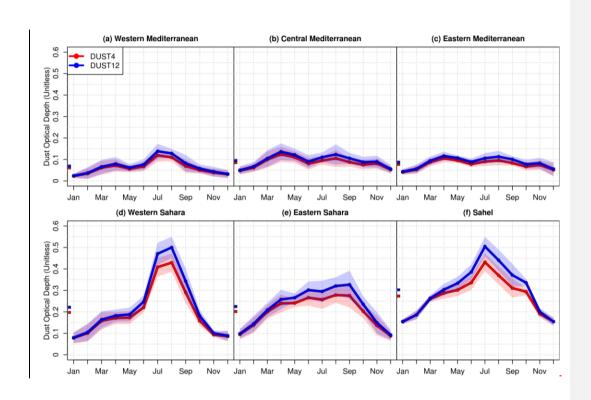


Figure 11: Dust optical depth, coarse (>2.5 μ m) and fine (<2.5 μ m) column burden of DUST4 and DUST12 experiments for the period December 2006 to November 2014.



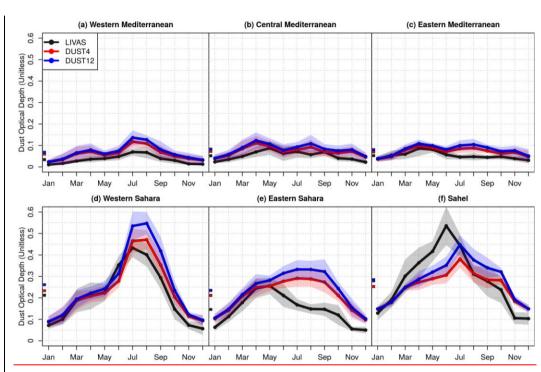


Figure 12: Annual cycle of dust optical depth of <u>LIVAS</u>, <u>DUST4</u> and <u>DUST12</u> and <u>DUST4</u> experiments for the period December 2006 to November 2014.

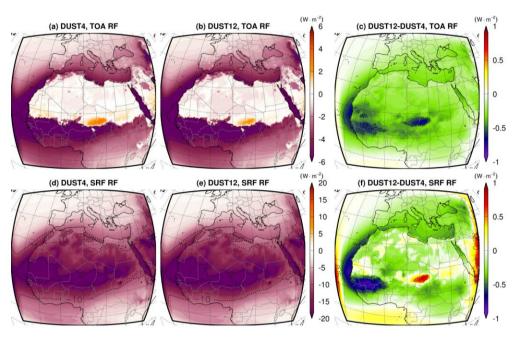


Figure 13: Top of the atmosphere (top) and surface (bottom) radiative forcing on the shortwave spectrum for the period December 2006 to November 2014.

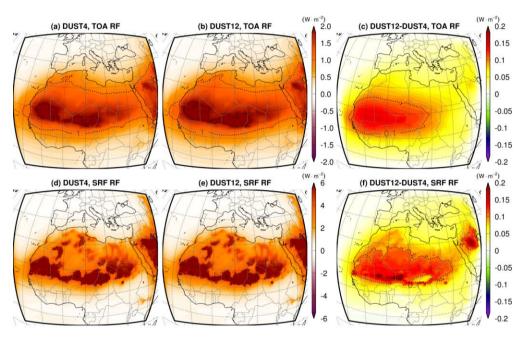


Figure 14: Top of the atmosphere (top) and surface (bottom) radiative forcing on the longwave spectrum for the period December 2006 to November 2014.

Dust size parameterization in RegCM4: Impact on aerosol burden and radiative forcing

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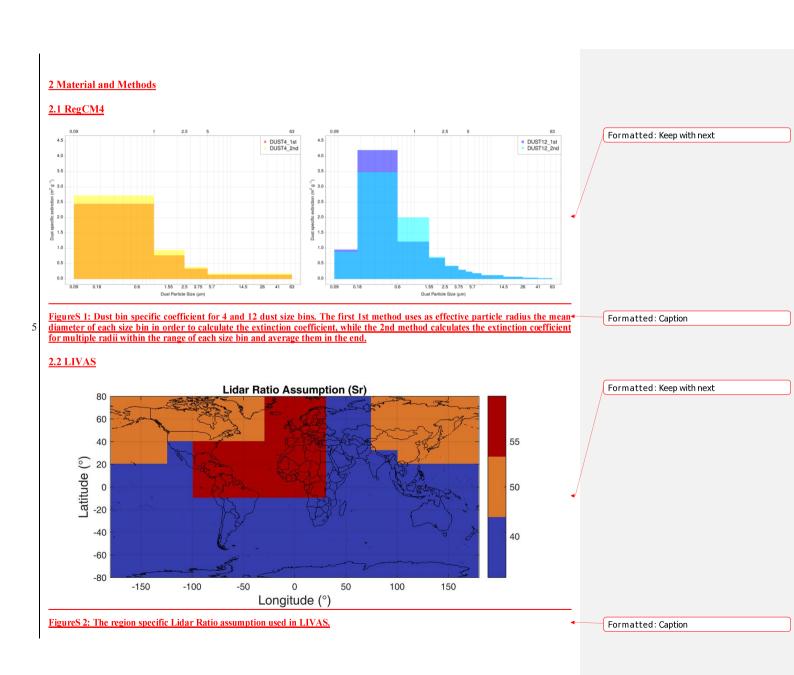
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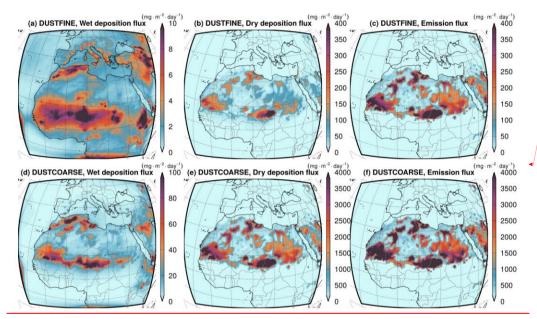
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15 This is the electronic supplement of the article "Dust size parameterization in RegCM4: Impact on aerosol burden and radiative forcing" submitted to Atmos. Chem. Phys. Diss. (2016)

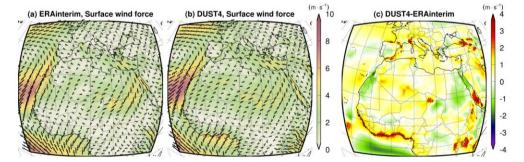


3 Results and Discussion

3.1 Evaluation



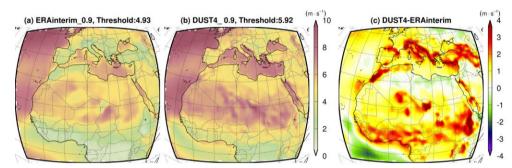
FigureS 3: Wet deposition, dry deposition and surface emission fluxes of fine (a, b, c) and coarse (d, e, f) dust particles in DUST4* experiment for the period December 2006 to November 2014.



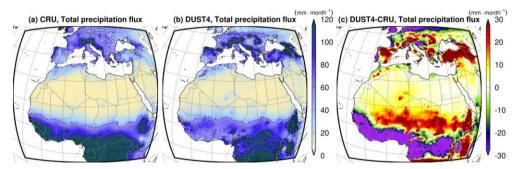
| FigureS 41: Wind velocity at the surface of DUST4 experiment against the reanalysis ERA-interim for the period December 2006 to November 2014.

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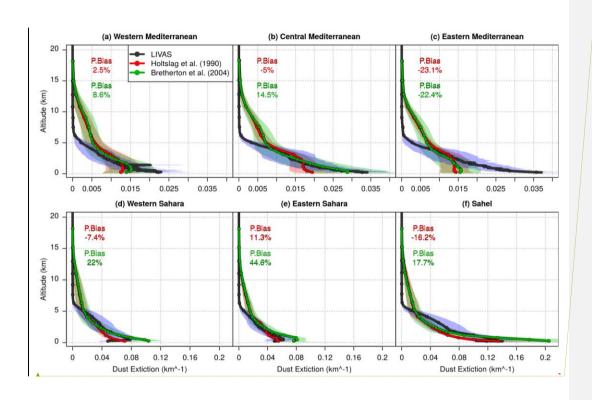
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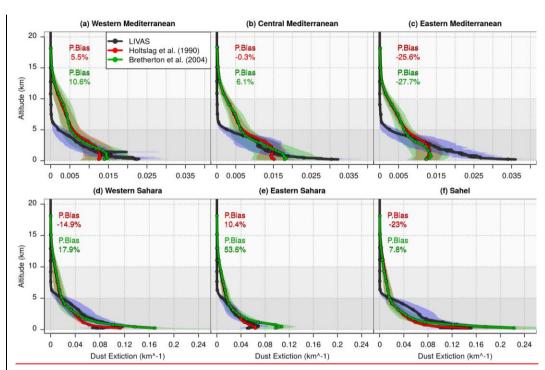
FigureS 52: Wind velocity at the surface of the 0.9 percentile over the desert for the period December 2006 to November 2014.



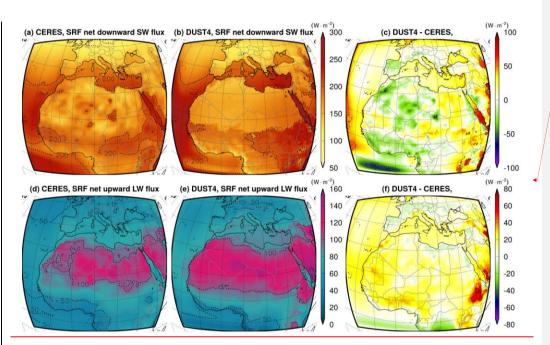
| FigureS 63: Total precipitation flux of DUST4 experiment against the reanalysis ERA-interim for the period December 2006 to November 2014.



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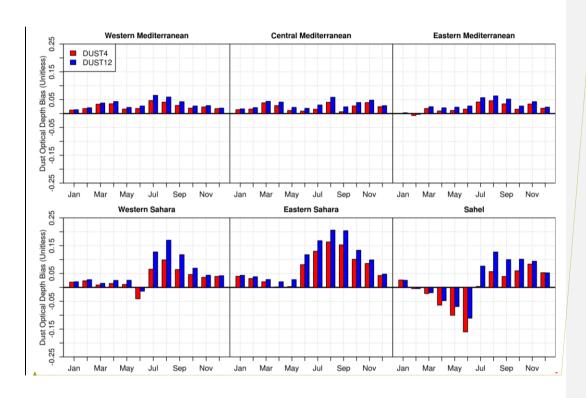
FigureS 74: Dust extinction profiles of LIVAS and DUST4 using Holtslag et al. (1990) and Bretherton et al. (2004) PBL schemes for 2008. P.Bias indicates the column percentage bias of DEX.



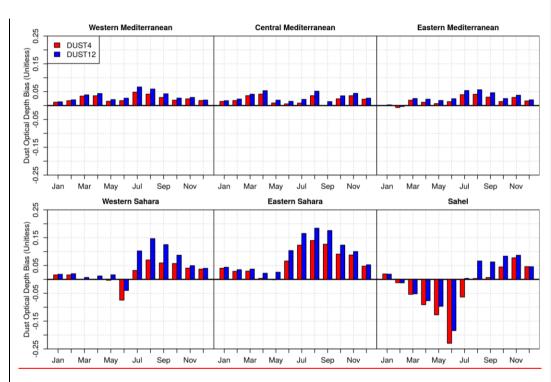
FigureS 8: Net shortwave downward (a, b, c) and net longwave upward flux (d, e, f) in the surface of CERES and DUST4 experiment for the period December 2006 to November 2014.

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3.2 Comparison of 4-bin and 12-bin experiments

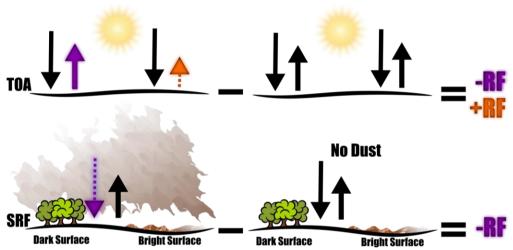


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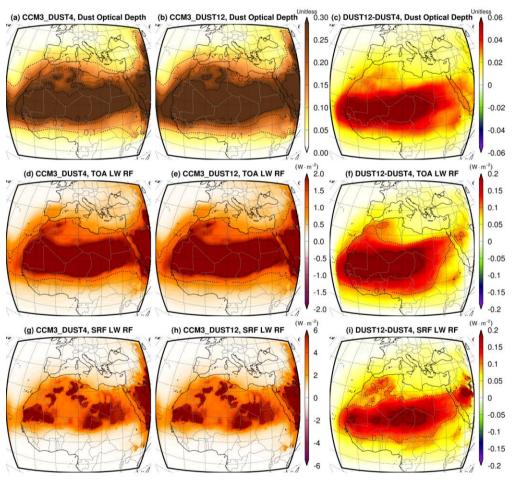


FigureS <u>95</u>: Monthly biases of DUST4 and DUST12 experiment in comparison to the LIVAS dust product for the period December 2006 to November 2014.

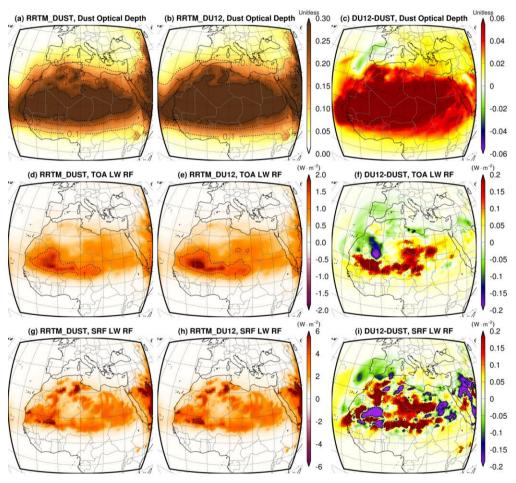
3.3 Radiative Forcing



FigureS 106: Illustration of shortwave radiative forcing of dust at the surface and at the top of the atmosphere. Left and right panel depicts a dust loaded and a dust free atmosphere respectively. Arrows indicate the downward and upward radiation. The purple and the orange color correspond to a negative and positive radiative effect of dust particles respectively.



| FigureS 117: Dust optical depth, top of the atmosphere (TOA) and surface (SRF) radiative forcing on the longwave spectrum using the Community Climate Model 3 (CCM3) radiation transfer scheme for June 2008.



FigureS 128: Dust optical depth, top of the atmosphere (TOA) and surface (SRF) radiative forcing on the longwave spectrum using as radiation transfer scheme the Rapid Radiation Transfer Model (RRTM) for June 2008.