Authors' response (in blue) to the Reviewer #3's comments (in black):

The authors thank Reviewer #3 for his/her comments and suggestions that definitely improved the manuscript. Required changes and modifications have been introduced in the text of the revised version of the manuscript by using the Word Track Changes tools. The manuscript has been revised and restructured in order to present more clearly the results and to implement the changes after answering the reviewer's comments. Authors think that the way they are introduced in the new version of the manuscript will improve the reading and understanding.

In addition, the following general changes have been addressed throughout the manuscript:

- The title has been slightly modified in order to highlight the main issue of the work (Part 1), i.e. the shortwave dust direct radiative effect, that is: "Aerosol radiative impact during the summer 2019 heatwave produced partly by an inter-continental Saharan dust outbreak. Part 1. Shortwave dust direct radiative effect".
- 'J' in the dates have been replaced by 'June' for avoiding confusion.
- Figure 2 has been removed (also taking into account the reviewer #2's comments) for a more fluent reading in overall, since this figure doesn't provide any crucial additional information to the current modelling analysis performed (see Fig. 1 and previous Fig. 3). The following figures have been renumbered.
- Figures 3-5 (previous 4-6), and 7-9 (previous 8-10) have been improved. In particular, error bars have been included in Fig. 3 in order to show uncertainties.
- A new Table (Table 2 now) has been added. The rest of Tables have been renumbered.
- Symbols used for the single scattering albedo (SSA), the asymmetry factor (asyF) and the surface albedo (SA) have been replaced by SSA, asyF and SA, respectively, for avoiding confusion, as in the text as in the Figures.

Next, the authors respond to the particular comments of the reviewer #3.

- Reviewer 3

The authors present a case study on the use of ground-based measurements from various sensors (sunphotometers and lidars) and a radiative transfer model to calculate the direct radiative effect of fine and coarse mode dust particles, separately, over Barcelona, Spain and Leipzig, Germany. Overall, this is a nice paper that merits to be published in ACP after the authors address my comments and also the comments of the other reviewers. Below, please find my major and minor comments:

R3C1. English should be improved in the text. I believe the authors could do several improvements by going through the text several times.

Authors' response: The English has been revised in addition to the initial structure throughout all the manuscript.

R3C2. Line 26-27. Please rephrase this sentence. You might consider breaking it into two sentences.

Authors' response: This sentence together with the previous ones have been rephrased for clarity. Therefore, the text in the Abstract has been modified in the revised version of the manuscript, as follows:

Abstract, page 1, lines 26-30: "The dust produced a cooling effect on the surface with a mean daily DRE of -9.1 and -2.5 W m^{-2}, respectively, in Barcelona and Leipzig, but the Df/DD DRE ratio is larger for Leipzig (52%) than for Barcelona (37%). Cooling is also observed at the top-of-the-atmosphere (TOA), although less intense than on surface. However, the Df/DD DRE ratio at the TOA is even though higher (45% and 60%, respectively, in Barcelona and Leipzig) than on the surface."

R3C3. Line 34. Please replace concerning with another word (e.g. with important).
Authors' response: Done (line 38).

R3C4. Line 37. Relies instead of rely.
Authors' response: This sentence has been slightly modified: "This is mainly due to …" (lines 41-42).

R3C5. Line 50. Please give a reference here if there is any. Otherwise, rephrase.
Authors' response: The text has been modified in the revised version of the manuscript, including a reference, as follows:

Page 2, lines 53-56: "Therefore, the individual radiative estimate for both dust coarse and fine modes must be separately evaluated, and only a few works have addressed this issue. For instance, Sicard et al. (2014b) reported that dust coarse particles seem mainly to affect the LW radiation, being their fine mode mostly responsible of the SW radiative modulation."

Reference

Sicard, M., Bertolín, S., Muñoz, C., Rodríguez, A., Rocadenbosch, F., and Comerõn, A.: Separation of aerosol fine- and coarse mode radiative properties: Effect on the mineral dust longwave, direct radiative forcing, Geophys. Res. Lett., 41, 6978–6985, https://doi.org/10.1002/2014GL060946, 2014b.

R3C6. Line 52. Dust emissions are very uncertain and may be even higher (also much lower). See the paper of Huneeus et al. (2011). Quite a significant part of them is of anthropogenic origin (Ginoux et al., 2012).
Authors' response: We thank the reviewer's suggestions that definitely improve the paper. The text has been modified in the revised version of the manuscript, as follows:

Page 2, lines 57-65: "Mineral dust is the most abundant aerosol in the atmosphere; however, dust emissions are very difficult to predict. Despite emissions of 1000-3000 Tg yr^{-1} were estimated by global models (i.e., Zender et al., 2004), a later study carried out by means of a global dust model intercomparison (Huneeus et al., 2011) suggested that the emissions may range from 500 to 4000 Tg yr^{-1}. Globally the natural dust sources account for 75%, with the remaining 25% coming from anthropogenic (mainly from agricultural activities) origin (Ginoux et al., 2012). In particular, Saharan dust could represent half of the airborne abundance (Prospero 2002). According to Huneeus et al. (2011), North Africa deserts could emit between 400 to 2200 Tg yr^{-1} of dust particles, hence an 8% of that amount can be attributed to anthropogenic sources (Ginoux et al., 2012)."

In addition, the new references have been included in the reference list.

References

Ginoux, P., Prospero, J. M., Gill, T. E., Hsu, N. C., and Zhao, M.: Global-scale attribution of anthropogenic and natural dust sources and their emission rates based on MODIS Deep Blue aerosol products, Rev. Geophys., 50, RG3005, https://doi.org/10.1029/2012RG000388, 2012.

Huneeus, N., Schulz, M., Balkanski, Y., Griesfeller, J., Prospero, J., Kinne, S., Bauer, S., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Fillmore, D., Ghan, S., Ginoux, P., Grini, A., Horowitz, L., Koch, D., Krol, M. C., Landing,W., Liu, X., Mahowald, N., Miller, R., Morcrette, J.-J., Myhre, G., Penner, J., Perlwitz, J., Stier, P., Takemura, T., and Zender, C. S.: Global dust model intercomparison in AeroCom phase I, Atmos. Chem. Phys., 11, 7781–7816, https://doi.org/10.5194/acp-11-7781-2011, 2011.

Prospero, J. M., Ginoux, P., Torres, O., Nicholson, S. E., and Grill, T. E.: Environmental characterization of global sources of atmospheric soil dust identified with the Nimbus 7 Total Ozone Mapping Spectrometer (TOMS) absorbing aerosol product, Rev. Geophys., 40, 1002, doi:10.1029/2000RG000095, 2002.

Zender, C. S., Miller, R. L., and Tegen, I.: Quantifying mineral dust mass budgets: Terminology, constraints, and current estimates, Eos Trans. Am. Geophys. Union, 85, 509-512, https://doi.org/10.1029/2004EO480002, 2004.

R3C7. Line 57-59. You need a reference here. In a very recent study, Akritidis et al. (2020) present a very massive event of dust transport over Europe. Another study that could be also cited is Osborne et al. (2019).

Authors' response: We thanks the reviewer's suggestions. Both proposed references have been added to the text (also to the reference list), and also one more representing a generalized dust study over Europe (**page 2, line 70**). They are:

Akritidis, D., Katragkou, E., Georgoulias, A. K., Zanis, P., Kartsios, S., Flemming, J., Inness, A., Douros, J., and Eskes, H.: A complex aerosol transport event over Europe during the 2017 Storm Ophelia in CAMS forecast systems: analysis and evaluation, Atmos. Chem. Phys., 20, 13557–13578, https://doi.org/10.5194/acp-20-13557-2020, 2020.

Ansmann, A., Bösenberg, J., Chaikovsky, A., Comerón, A., Eckhardt, S., Eixmann, R., Freudenthaler, V., Ginoux, P., Komguem, L., Linné, H., Márquez, M. Á. L., Matthias, V., Mattis, I., Mitev, V., Müller, D., Music, S., Nickovic, S., Pelon, J., Sauvage, L., Sobolewsky, P., Srivastava, M. K., Stohl, A., Torres, O., Vaughan, G., Wandinger, U., and Wiegner, M.: Long-range transport of Saharan dust to northern Europe: The 11–16 October 2001 outbreak observed with EARLINET, J. Geophys. Res.-Atmos., 108, 4783, https://doi.org/10.1029/2003JD003757, 2003.

Osborne, M., Malavelle, F. F., Adam, M., Buxmann, J., Sugier, J., Marenco, F., and Haywood, J.: Saharan dust and biomass burning aerosols during ex-hurricane Ophelia:

observations from the new UK lidar and sun-photometer network, Atmos. Chem. Phys., 19, 3557–3578, https://doi.org/10.5194/acp-19-3557-2019, 2019.

R3C8. Line 96. Is this separation between coarse and fine mode aerosols or dust and no-dust? Is there a possibility of having other aerosol components into the coarse mode?
Authors' response: POLIPHON method can be used two ways by applying either the one-step (two-component separation) or the two-step (three-component separation) approach. The methodology is clearly explained in several works of Mamouri and Ansmann (2014, 2017) and Ansmann et al. (2019). In this work, we used the second (two-step) approach to separate the dust coarse (Dc), dust fine (Df) and non-dust (ND) components, by adapting the Mamouri and Ansmann's procedure to P-MPL data in particular (as described in Córdoba-Jabonero et al. (2018)). In the first step, the Dc component is separated from the fine mode (Df and ND). ND is assumed fine, as associated to background (continental, pollution) aerosols in both the urban (Barcelona and Leipzig) stations regarded in this work. In the second step, that fine mode is detached into Df and ND components, separately. The coarse mode is linked to the dust coarse (Dc) particles only. The predominance of dust coarse particles in the Saharan dust intrusions is primary, and a 'second' source of coarse particles can be considered insignificant in comparison. This statement is also confirmed by the particle linear depolarization ratio (PLDR) values found in this work, which are typical for dust. For more clarity, the text has been modified in the revised version of the manuscript, as follows:

Page 4, lines 114-120: "The algorithm is based on a two-step method for separating the three components of dusty mixtures. First, by using both the lidar-derived total particle backscatter coefficient (PBC), β_p, and the particle linear depolarization ratio (PLDR), δ_p, profiles, the coarse mode (predominantly, dust coarse particles, Dc), β_{Dc}, and the fine mode of the PBC are separated. Second, the fine mode of the PBC, which is composed of the dust fine particles (Df) and non-dust aerosols (ND), is separated in two more components, respectively β_{Df} and β_{ND}. The ND component is assumed to belong to the fine mode, associated to background (continental, pollution) aerosols in both the urban stations, BCN and LPZ, studied in this work."

References

Ansmann, A.; Mamouri, R.; Hofer, J.; Baars, H.; Althausen, D.; Abdullaev, S.F.: Dust Mass, Cloud Condensation Nuclei, and Ice-Nucleating Particle Profiling with Polarization Lidar: Updated POLIPHON Conversion Factors from Global AERONET Analysis, Atmos. Meas. Tech., 12, 4849-4865, https://doi.org/10.5194/amt-12-4849-2019, 2019.
Córdoba Jabonero, C.; Sicard, M.; Ansmann, A.; del Águila, A.; Baars, H.: Separation of the Optical and Mass Features of Particle Components in Different Aerosol Mixtures by using POLIPHON Retrievals in Synergy with Continuous Polarized Micro-Pulse Lidar (P-MPL) Measurements, Atmos. Meas. Tech., 11, 4775-4795, https://doi.org/10.5194/amt-11-4775-2018, 2018.
Mamouri, R.; Ansmann, A.: Fine and Coarse Dust Separation with Polarization Lidar, Atmos. Meas. Tech., 7, 3717–3735. https://doi.org/10.5194/amt-7-3717-2014, 2014.

Mamouri, R.; Ansmann, A.: Potential of Polarization/Raman Lidar to Separate Fine Dust, Coarse Dust, Maritime, and Anthropogenic Aerosol Profiles, Atmos. Meas. Tech., 10, 3403–3427. https://doi.org/10.5194/amt-10-3403-2017, 2017.

R3C9. Line 159. Please give the wavelengths at which the radiative transfer is implemented. Is there a specific spectral resolution of the model?
Authors' response: In the shortwave range, the model has no fixed, specific spectral resolution. It has several predefined resolutions in terms of wavenumber. For this study, the resolution was set to 100 cm^{-1} in the range 2500-14400 cm^{-1} (0.7 – 4.0 µm) and 400 cm^{-1} in the range 14400-50000 cm^{-1} (0.2 – 0.7 µm). The interval of the spectral range is set by the user (0.2 – 4.0 µm in our study. The resolution in terms of wave number has been added in the revised version of the manuscript (see), as follows:

Page 5, lines 175-176: "The solar spectral range was set from 0.2 to 4.0 µm (wave number resolution of 400 cm^{-1} from 0.2 to 0.7 µm and 100 cm^{-1} from 0.7 to 4.0 µm)."

R3C10. Line 203. Please define the term frequencies. How were they calculated?
Authors' response: The trajectory frequency ($F_{i,j}$) is the sum of the number of trajectories ($T_{i,j}$) that passed through each (i, j) grid cell (resolution: 1.0º x 1.0º) each 6 hours, divided by the total number (N) of trajectories analysed, that is,

$$F_{i,j} = 100 \sum T_{i,j}/N$$

In our case, N = 20.

However, Figure 2 has been removed in the revised version of the manuscript for the sake of a more fluent reading in overall, since this figure doesn't provide any additional crucial information to the current modelling analysis performed (see Fig. 1 and previous Fig. 3), and also following suggestions of the reviewer #2.

R3C11. Line 250. Remove "on".
Authors' response: The sentence has been slightly modified, as follows:

Page 11, lines 288-289: "On 28 and 29 June, the DD signature is also observed from around 4 km down to the surface; …"

R3C12. Personally, I do not like very much the use of J instead of January.
Authors' response: For avoiding confusion, 'J' has been replaced by 'June' in the whole manuscript, including figures and tables.

R3C13. As a general comment. The aging of dust is probably expected to modify the optical properties of dust. I would like to see a discussion on that in the paper.
Authors' response: Several aspects of the aging of dust have been put forward thanks to the dual-site analysis of the same dust outbreak. Some of them related to the mass loading, others to the optical properties (DOD). In all cases, the main consequence put forward is the gravitational settling of the largest particles. The observations used in our paper do not allow going more in details and quantifying other processes that occurred

during the aging, like e.g. nucleation, condensation, coagulation or deposition, and their impact on the optical properties.
Even from the dust radiative properties (Sect. 3.3.1), conclusions about the aging are not straightforward since the properties shown (see Fig. 6 in the revised version of the manuscript) are integrated in the column and cannot be attributed only to dust.
Several sentences have been added here and there in the revised manuscript suggesting the effect of the aging of dust on our results (not only on the optical properties):

Abstract, page 1, line 24: "Several aspects of the aging of dust are put forward."
Page 16, lines 472-474: "If the coarse mode in the column is formed exclusively of dust particles, it can be stated that the aging of dust has no effect on the absorption capabilities of the coarse mode."
Page 27, lines 764-765: "Indeed, this result reflects the aging of dust, and in particular the gravitational settling of Dc particles during their longer transport to LPZ."
Page 27, lines 774-777: "Concerning the relative mass incidence of each component, Dc particles were dominating (around 80%, in general) along the overall dusty period in both stations. However, a higher Df mass contribution with respect to the total dust mass loading was found in LPZ (13.5%) than in BCN (11%), reflecting again, through an increase of the fine mode mass contribution, the aging of the dust."
Page 27, lines 792-793: "The modification of the dust optical properties due to aging and its impact on the DRE is evidenced with the temporal dust evolution in BCN and with the comparison between BCN and LPZ dust scenarios."
Page 28, lines 804-805: "Along the dust 8-day event in BCN, the effect of dust aging is clearly visible on the Df/DD DRE ratio at the surface, which increased at a rate of +2.4%·day^{-1}, i.e., +16% between the first and the last day of the event."
Page 28, lines 829-831: "Second, and as a direct consequence of the dust aging, the mean Df/DD DRE ratio at the surface in LPZ was 52%, which is higher than in BCN (37%), likewise that observed at the TOA, where the Df/DD DRE ratio was 60% in LPZ and 45% in BCN."

R3C14. The use of two symbols for the single scattering albedo and asymmetry factor is confusing, i.e. AsyF and g.
Authors' response: We agree. The symbols have been removed and, if needed, replaced by SSA for the single scattering albedo, asyF for the asymmetry factor, and SA for the surface albedo.

R3C15. Please be more specific about the albedo data used. Are they part of the standard AERONET product?
Authors' response: The surface albedo (SA) was obtained from the AERONET data. This has been indicated in the text of the revised version of the manuscript, as follows:

Page 15, lines 412-413: "The SSA and asyF, as well as the surface albedo (SA), are available from AERONET database."

R3C16. It would be interesting to see a comparison of the radiative calculations with climatological values appearing in recent studies (e.g. Nabat et al., 2014 and Tsikerdekis et al., 2019). In this way, the reader will be able to evaluate the high or low instantaneous radiative effect values appearing in the paper.

Authors' response: First of all the authors thank the reviewer for pointing out these interesting papers.

In the original manuscript some comparisons of the instantaneous values (with Sicard et al., 2014a, https://doi.org/10.5194/acp-14-9213-2014) and of the daily averages (with Meloni et al., 2005, https://doi.org/10.1016/j.jqsrt.2004.08.035) were already made (Sect. 3.3.2). We have now also included a short comparison with Tsikerdekis et al. (2019) (also added to the reference list). Nabat et al. (2014) was not included in the comparison because they performed simulations over a shorter period of time (2003-2009) as well as 2 specific cases, and considered all aerosol types. The following text has been added in the revised version of the manuscript:

Page 18, lines 511-515: "Compared to climatological values, our findings are also in agreement with recent works reporting about the same region. For instance, Tsikerdekis et al. (2019) simulated with RegCM4 the dust shortwave direct radiative effect for a 10-year period (01 December 1999 to 30 November 2009) and found for the summer season values of -14.9 and -5.5 W m^{-2} over the Sahara region and for the Mediterranean Basin, respectively."

References

Meloni, D.; Di Sarra, A.; Di Iorio, T.; Fiocco, G.: Influence of the Vertical Profile of Saharan Dust on the Visible Direct Radiative Forcing, J Quant Spectrosc Radiat Transf., 93, 397-413, https://doi.org/10.1016/j.jqsrt.2004.08.035, 2005.

Nabat, P., Somot, S., Mallet, M., Sevault, F., Chiacchio, M., and Wild, M.: Direct and semi-direct aerosol radiative effect on the Mediterranean climate variability using a coupled regional climate system model, Clim. Dyn., 44, 1–29, https://doi.org/10.1007/s00382-014-2205-6, 2014.

Sicard, M.; Bertolín, S.; Mallet, M.; Dubuisson, P.; Comerón Tejero, A.: Estimation of Mineral Dust Long-Wave Radiative Forcing: Sensitivity Study to Particle Properties and Application to Real Cases in the Region of Barcelona, Atmos. Chem. Phys., 14, 9213-9231, https://doi.org/10.5194/acp-14-9213-2014, 2014a.

Tsikerdekis, A., Zanis, P., Georgoulias, A. K., Alexandri, G., Katragkou, E., Karacostas, T., and Solmon, F.: Direct and semi-direct radiative effect of North African dust in present and future regional climate simulations, Clim. Dynam., 53, 4311–4336, https://doi.org/10.1007/s00382-019-04788-z, 2019.

R3C17. Personally, I do not like some figures. E.g. Figure 2 or 5 might be polished a little bit. However, this is not a must.

Authors' response: We agree. Figure 2 has been removed for the sake of a more fluent reading in overall, since this figure doesn't provide any additional crucial information to the current modelling analysis performed (see Fig. 1 and previous Fig. 3). In addition, some data have been removed from the top panels in Figure 5, i.e., in particular, the daily-averaged/mean total mass loading, and keeping only that for the total dust. Therefore, Figures have been renumbered, and the text modified accordingly.

R3C18. Most probably the radiative effect of fine and coarse mode dust when added will not be equal to the total dust if a simulation is done for the total dust properties due to nonlinearities. This stems from the fact that dust particles are mingled and not separated in the atmosphere. I would appreciate a discussion on this from the authors.
Authors' response: This is totally true. As a matter of fact, the authors have demonstrated this statement in the longwave spectral range showing that, when fine and coarse mode properties are known independently, then DRE_{Dc} + DRE_{Df} is different from DRE_{DD} calculated with the total dust properties. See Sicard et al. (2014b, https://doi.org/10.1002/2014GL060946) for more details. Over 4 cases with AODs varying between 0.19 and 0.48 Sicard et al. (2014a) found that DRE_{DD} is reduced (in absolute value) by 10-20% compared to DRE_{Dc} + DRE_{Df}. For the shortwave spectral range, the difference is expected to be lesser since some parameters, e.g., the height of the aerosol layers, have a lesser influence on the estimation of the radiative effect compared to the longwave spectral range.
Anyway the direct radiative effects estimated in this work have been assessed independently with fine and coarse mode properties and are thus expected to be more "accurate" than the more classical approaches considering equivalent, total properties. In order to emphasize the benefit of having both fine and coarse properties vs. total properties the following text has been added in the conclusion section of the revised version of the manuscript:

Page 29, lines 863-866: "The study calls for a more generalized use of state-of-the-art algorithms, like POLIPHON, to independently retrieve aerosol properties for the fine and coarse modes. These retrievals are very valuable when used as input of radiative transfer models. Our findings clearly demonstrate that both fine and coarse modes are equally relevant for the estimation of SW direct radiative effects of long-range transported mineral dust."

References

Sicard, M.; Bertolín, S.; Mallet, M.; Dubuisson, P.; Comerón Tejero, A.: Estimation of Mineral Dust Long-Wave Radiative Forcing: Sensitivity Study to Particle Properties and Application to Real Cases in the Region of Barcelona, Atmos. Chem. Phys., 14, 9213-9231, https://doi.org/10.5194/acp-14-9213-2014, 2014a.
Sicard, M., Bertolín, S., Muñoz, C., Rodríguez, A., Rocadenbosch, F., and Comerón, A.: Separation of aerosol fine- and coarse mode radiative properties: Effect on the mineral dust longwave, direct radiative forcing, Geophys. Res. Lett., 41, 6978–6985, https://doi.org/10.1002/2014GL060946, 2014b.