

Author's Response to the Referee Comments

Manuel Gutleben, Silke Groß, Christian Heske and Martin Wirth

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The authors would like to thank the referees very much for carefully reading the submitted manuscript and for their helpful and very valuable suggestions and feedbacks. In the following, all comments and questions will be addressed and answered. The comments are repeated and a direct response is given below. In addition, changes in the manuscript are highlighted in the appended marked-up manuscript version using blue (additions) and red (removals) colors.

Reply to Comments of Referee #1 on 9 March 2022

Major Issues

(I) The biggest issue to me is that the authors seem to draw quite general conclusions from what really is a case study of an individual event. I'd therefore urge them to not overinterpret the findings and clearly state that these measurements - unique as they are - mark a limited sample that does not allow for drawing more general conclusions. This should also be expressed by re-categorization of the manuscript type as Measurement Report and a change of title to, e.g. Airborne lidar observations of a case of wintertime Saharan dust transport towards the Caribbean during EUREC⁴A.

Thank you for this very valuable comment. We agree, that this study draws conclusions from measurements during only one research campaign. It was already explicitly stated in the title of the submitted manuscript, that the paper is based on measurements during one campaign, namely EUREC⁴A. Nevertheless, we agree that it is appropriate to change the title of the manuscript to: 'Wintertime Saharan dust transport towards the Caribbean - an airborne lidar case study during EUREC⁴A', to point out that the paper is indeed a case study. Moreover, we point it out once more in the conclusion of the study. However, since we observed for the first time that enhanced concentrations of water vapor are also advected during winter-time dust transport, we follow the suggestion of Referee #2 not to change the manuscript type to a measurement report.

(II) Figure 1 should be omitted. The same is shown in a much better way in Figure 2.

Thank you for this suggestion. Please refer to the reply to comment (1) by Referee #2.

(III) The methods section (maybe better data and methods?) should also include the auxiliary data use in your work, i.e. MODIS, HYSPLIT, etc. Lines 173 to 193 should be moved to that section and expanded towards a discussion of typical values.

We followed your suggestion and moved the introduction to the aerosol separation technique to Chapter 2. We also changed the title of the chapter to 'Data and Methods'. HYSPLIT and MODIS are introduced in Chapter 'Data and Methods' in the revised manuscript. You can find all changes in the attached marked-up manuscript version.

(IV) Figures 3 and 4 should be split into three figures each and placed at positions in the text so that the reader doesn't have to go back and forth to follow their discussion. As is, the panels in Figure 3 are too small. The last sentence in the caption of Figure 3 should be moved to the methodology section. A statement regarding the grey shaded areas should be mentioned.

Thank you for this valuable comment. We split up Figure 3 and Figure 4 in the revised manuscript such that the reader does not have to jump back and forth. This also increases the size of the panels in Figure 3. Furthermore, we followed your suggestion and specified that the median profiles of $R_{||}$ and S refer to the grey shaded regions in the respective right panels.

5

Minor Issues

(1) line 15: is there an estimate of the dust contribution based on measurements?

We are not aware of any based estimate of annual North African dust aerosol emission, which is solely based on measurements. Hence, we prefer to stick to the estimate by Huneeus et al. (2011) which is based in a comparison of 15 global aerosol models.

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(2) line 21: transport instead of transportation

We corrected that.

(3) line 24: the Intertropical Convergence Zone is generally referred to as ITCZ

We changed the abbreviation in the revised manuscript.

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(4) line 44: cloud condensation nuclei

The mistake is corrected in the revised manuscript.

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(5) line 50: this region instead of these regions

We corrected that.

(6) line 53: What plumes?

The mineral dust plumes mentioned in the sentences before. For a better understanding we changed the sentence to: "This unique data set now enables a detailed investigation of macrophysical properties of the observed long-range-transported dust plumes and the state of the atmosphere."

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(7) line 70: enable a characterization of winter-time dust transport: please clearly state that you are discussing just three research flights within four days and that those flights are likely to cover the same dust event. In that context, your aim of characterizing wintertime dust transport is quite overstates what is possible with your data set.

We pointed that out by revising the paragraph to: "Collected airborne lidar data sets during measurement flights on these days enable a characterization of long-range-transported African dust plumes during EUREC⁴A, although the flights tracks have not been specifically designed for dust observations. As dust aerosols could only be measured in this 4-day period, it is likely that the observed aerosol originated from the very same African dust outbreak."

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(8) Figure 2 and related text: Please: at what wavelength of AOD measurement. Caption: one station is marked by one dot.

The wavelength of MODIS AOD is 550 nm. We added that to the figure caption and the related text. The dots mark the Grantley Adams International Airport on Barbados - this is already denoted in the caption of the figure.

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(9) lines 93 - 96: You could drop the index 532 after clearly stating that all measurements have been performed at that wavelength.

We followed your suggestion and dropped the index in the revised manuscript for simplicity.

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(10) line 98: Please clarify for the non-experts that DIAL gives the water vapour profiles and that HSRL gives the aerosol profiles.

We added a corresponding clarification.

50

(11) line 114: add reference to 10.1029/2009JD011862 and 10.1111/j.1600-0889.2011.00548.x regarding the use of lidar measurements to characterize aerosol mixtures

We added the citations and modified the references in the revised manuscript.

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(12) line 143 and 145: AT these altitudes

We corrected the mistakes.

(13) Figure 5: please add the abbreviations for the different aerosol types (MA, DU, BB) in line 3 of the caption when marking their colour in the plot.

We added the abbreviations to the figure captions.

(14) line 211: dominates the aerosol mixture?

We followed your suggestion and changed the phrase.

(15) Figure 6: Is it possible to apply the information from Figure 5 to this plot to get more quantitative results rather than the coarse ellipses?

In this Figure, we explicitly wanted to separate the variables δ_p and S with regard to r_m from each other to give a better overview on the distribution of water vapor. This is why we used ellipses to roughly outline the regions of the two mixed regimes (BB-DU and MA-DU). We discussed how we could use the information of Figure 5 in the histograms shown in Figure 6, but thought it would be best to stick to the figures in the submitted manuscript. However, we decided to change the transparency of the ellipses, in a way that they don't appear too dominant.

Reply to Comments of Referee #2 on 11 March 2022

General comments

(I) First, I would like to underline that I consider the title to be appropriate and also, I like the presentation of the results in Fig.3. This way it is more visible the variability of the measurements during the three flights.

Thank for this comment. As the results of the study are based on measurements during one research campaign we agree with Referee #1 to rename the manuscript in a way that the reader immediately recognizes that this paper is on a lidar case study. Therefore, we changed the title accordingly (see response to comment (I) of Referee #1). We also followed the suggestion of Referee #1 to split up Figure 3 and Figure 4 such that the reader does not have to jump back and forth in the manuscript (see response to comment (IV) of Referee #1).

This also increases the size of the panels in Figure 3.

(II) I agree with TROPOS team (comment by Albert Ansmann): The lidar scientists will use your measurements in follow-on papers, so please consider TROPOS suggestions. Furthermore, previous campaigns should be mentioned and also references to their publications.

Thank you for your feedback. For our response to this comment, please refer to the response to Albert Ansmann later in this document.

Minor technical suggestions /corrections

(1) Figure 1 should be moved in section 2.1 (after it is mentioned in the text). I think it should be kept in the manuscript; it gives the exact overlaying of the flights. Table 1 should be moved in section 3.1 (after it is mentioned in the text)

Thanks for these comments. Even though we placed Figure 1 and Table 1 after their introduction in the text (in the \LaTeX source code), \LaTeX automatically put them to the top of the pages. However, we managed to modify the template to put them to the bottom of the respective pages (after they have been introduced in the main body) in the revised manuscript. We followed your suggestion and kept Figure 1 in the revised manuscript as it gives a nice overview of all EUREC⁴A flight tracks.

(2) Please specify in the caption what is the grey shaded areas on the right panels of Fig.3.

Thank you for this valuable comment. We specified that the median profiles of $R_{||}$ and S refer to the grey shaded regions in the respective right panels.

Reply to Comments by Albert Ansmann on 11 March 2022

(I) By reading the introduction (lines 30-36), the reader may get the impression, the authors introduce a new aspect: Winter transport of polluted dust over the remote tropical Atlantic towards, e.g., Barbados or even South America (Amazonia). But this impression needs to be avoided. We at TROPOS (partly together with Munich University, Wiegner, Gross, Freudenthaler) did so much work already in this field (since the SAMUM 2008 and later on in the framework the SALTRACE activities in 2013-2014) that needs to be mentioned:

Ansmann et al., Dust and smoke transport from Africa to South America: lidar profiling over Cape Verde and the Amazon rainforest, *Geophys. Res. Lett.*, 36, L11802, doi:10.1029/2009GL037923, 2009.

Baars et al., Further evidence for significant smoke transport from Africa to Amazonia, *Geophys. Res. Lett.*, 382, L20802, doi:10.1029/2011GL049200, 2011.

Rittmeister et al., Profiling of Saharan dust from the Caribbean to western Africa – Part 1: Layering structures and optical properties from shipborne polarization/Raman lidar observations, *Atmos. Chem. Phys.*, 17, doi.org/10.5194/acp-17-12963-2017, 2017.

Ansmann et al., Profiling of Saharan dust from the Caribbean to western Africa – Part 2: Shipborne lidar measurements versus forecasts, *Atmos. Chem. Phys.*, 17, 14987–15006, <https://doi.org/10.5194/acp-17-14987-2017>, 2017.

Haarig et al., ACP, 2019 (in the references)

Haarig et al., ACP, 2017, <https://doi.org/10.5194/acp-17-14199-2017> on dry sea salt depolarization should also be mentioned as a source for uncertainties in the depolarization observations close to Barbados.

You may now realize why I personally was motivated to write this comment!

Dear Albert,

thank you very much for this very valuable and important comment. It was never our intention to publish a paper in which we describe transatlantic winter-time transport of Saharan dust for the first time. Our aim was to investigate whether winter-time dust transport is also coming along with enhanced water vapor concentrations compared to the dry free troposphere (like already observed for summertime transport). This is why we did not focus on former studies on wintertime transport without focus on water vapor. However, we completely agree that those studies have to be mentioned in the manuscript. Otherwise the reader of the paper would get a false impression of the topic. Of course we included the list of papers that you have mentioned in the revised manuscript as they preceded our study. You can find the reworked parts in the appended marked-up manuscript version.

(II) We have a severe problem with the PURE SMOKE particle linear depolarization ratio (PLDR) of 0.14 at 532nm in the troposphere! This has never been observed, except for the upper dry troposphere (for cases in which the smoke particles were unable to age quickly..., so that the irregular, fractal-like structures remained for a long time and caused enhanced PLDR values of up to 0.2, Burton et al.). However, in the lower and middle troposphere such enhanced PLDR values for pure smoke have never been observed. Extreme values may be here, 0.07 (Falcon observations during LACE98, and Falcon observation presented by Dahlkoetter et al., 2014, in the upper troposphere). But usually the smoke PLDR values are <0.05. This is the reason that one is able to properly separate smoke and dust contributions to lidar backscatter coefficients (Tesche et al., JGR2009, Tesche et al., Tellus2011). It is general accepted that aged biomass burning smoke particles at heights in the lower to middle troposphere cause PLDRs of <0.05. See Haarig et al., ACP 2018, on smoke in the troposphere and stratosphere... As long as you cannot demonstrate by lidar observations (or by a proper reference) that the PURE smoke PLDR is about 0.14 one should avoid to mention that. To our opinion, such a statement is not acceptable and even 'dangerous' because lidar scientists may use that in follow-on papers! All in all: Nice work!

Thank you for this very helpful and valuable comment. You are absolutely right, that the depolarization ratios used for outlining the region of pure biomass burning aerosols in the submitted manuscript (i.e. Figure 6) are referring to values for non-aged and irregularly shaped biomass-burning aerosols. Of course we would have not observed such an aerosol species upstream the island of Barbados. During EUREC⁴A we never observed pure

biomass burning aerosol. We always observed aerosol mixtures of biomass burning aerosol and mineral dust. The optical properties of these aerosol mixtures are also in good agreement with those observed during SAMUM-2 at the beginning of transatlantic transport (Teschke et al. 2009, in the references). Furthermore, we rectified this mistake and changed the assumed particle linear depolarization ratio at 532 nm as well as the lidar ratio of biomass burning aerosol to values published by Haarig et al. 2018 for the lidar-based aerosol separation. This shifts the reference lines in Figure 6, but has no further impact on our results.

Wintertime Saharan dust transport towards the Caribbean :- an airborne lidar **observations** case study during EUREC⁴A

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Abstract. Wintertime Saharan dust plumes in the vicinity of Barbados are investigated by means of airborne lidar measurements. The measurements were conducted in the framework of the EUREC⁴A field experiment (Elucidating the Role of Cloud-Circulation Coupling in Climate) upstream the Caribbean island in January/February 2020. The combination of the water vapor differential absorption and high spectral resolution lidar techniques together with dropsonde measurements aboard the German HALO (High Altitude and Long-Range) research aircraft enable a detailed vertical and horizontal characterization of the measured dust plumes. In contrast to summertime dust transport, mineral dust aerosols were transported at lower altitudes and were always located below 3.5 km. Calculated backward trajectories affirm that the dust-laden layers have been transported in nearly constant low-level altitude over the North Atlantic Ocean. Only mixtures of dust-particles with other aerosol species, i.e. biomass burning aerosol from fires in West Africa and marine aerosol, were detected by the lidar. No pure mineral dust regimes were observed. Additionally, all the dust-laden airmasses that were observed during EUREC⁴A came along with enhanced water vapor concentrations compared to the free atmosphere above. Such enhancements have already been observed during summertime and were found to have a great impact on radiative transfer and atmospheric stability.

1 Introduction

Mineral dust aerosol is known to be a major contributor to the Earth's aerosol mass burden (Cakmur et al., 2006) and is estimated to contribute between 25 % and 30 % to the total aerosol optical depth (Tegen et al., 1997; Kinne et al., 2006). The greatest source region of mineral dust is the Saharan desert and its arid surrounding landscapes. From a comparison of 15 global aerosol models, Huneus et al. (2011) derived annual dust emissions from North Africa that range from 400 to 2200 Tg a⁻¹. This makes up roughly 50 % of the total global annual dust emission (1000 to 4000 Tg a⁻¹).

Once injected into the atmosphere, Saharan dust particles can be transported far away from their origin. While only 15 % of the total emitted dust load is transported north- and eastwards towards the Mediterranean and the Middle East (e.g. Shao et al., 2011), almost 85 % of the dust burden gets carried south- and westwards over the Atlantic Ocean. Transatlantic **transportation** transport routes vary throughout the year and depend on the large-scale synoptic situation (e.g. Ben-Ami et al., 2009; Tsamalis et al., 2013). During the winter months the Harmattan carries Saharan dust particles at low levels (<3.5 km) southwestwards towards the Gulf of Guinea (Schepanski et al., 2009). Due to the southernmost position of Intertropical Convergence Zone

25 (~~ITCITCZ~~) during this season, dust particles are then frequently transported towards the South American continent and the Amazon region (e.g. Yu et al., 2015). During boreal summer, however, strong solar insolation over the Sahara causes a deep boundary layer (e.g. Messenger et al., 2009) which can reach up to 6 km altitude (≈ 500 hPa). This allows dust particles to be mixed upward to higher altitudes where they get caught by the trade winds. Due to the northward shift of the ~~ITCITCZ~~ during boreal summer, they then get carried westwards as far as the Caribbean as well as Central and North America along a more northerly transportation route (e.g. Prospero and Carlson, 1972; Prospero et al., 2010).

Summertime dust advection can frequently be observed in the Caribbean. During that time of the year the dust particles are usually advected in elevated and decoupled layers - so called Saharan air layers (SALs; Prospero and Carlson, 1972; Carlson and Prospero, 1972; Prospero et al., 2021 Prospero and Carlson, 1972; Carlson and Prospero, 1972). Wintertime dust plumes, however, reach the Caribbean less frequently and only in connection with favorable synoptic situations. ~~They-~~

Transatlantic Saharan dust transport has been studied during several previous research campaigns, e.g. the Saharan Mineral Dust Experiments SAMUM 1 and 2 (Ansmann et al., 2011), the closure experiment SALTRACE (Saharan Aerosol Long-range Transport and Aerosol-Cloud-Interaction Experiment, Weinzierl et al., 2017) or the Next-Generation Aircraft Remote Sensing for Validation Studies II (Stevens et al., 2019). While observations during SAMUM 1 and 2 predominantly focused on the beginning of transatlantic dust transport near the North African coast (Ansmann et al., 2011; Groß et al., 2011, 2013), long-range-transported Saharan dust has been studied during SALTRACE and NARVAL-II on airborne (Gutleben et al., 2019a, b, 2020), shipborne (Rittmeister et al., 2017; Ansmann et al., 2017) and ground-based platforms (Groß et al., 2015; Haarig et al., 2017). In contrast to dust transport in elevated SALs during boreal summer, it was found that dust particles are mostly transported at lower-levels-and-low atmospheric levels during wintertime. It was also observed that they are sometimes mixed with other aerosol types like biomass burning aerosols or marine aerosols in the marine boundary layer (MBL; Chiapello et al., 1995; Ben-Ami et al., 2009; Groß et al., 2011 e.g. Ansmann et al., 2009; Baars et al., 2011; Groß et al., 2011; Haarig et al., 2019).

~~Recent studies have shown,~~ In recent studies it was also shown that summertime SALs may come along with enhanced concentrations of water vapor compared to the surrounding dry free troposphere (Gutleben et al., 2019b, 2020; Ryder, 2021). It was shown that not the aerosol but the water vapor inside the SALs is the dominating driver for net radiative heating during transatlantic dust transport. In this way, water vapor associated with dust layers has the potential to modify the atmospheric stability and consequently to impact the evolution as well as macrophysical properties of shallow marine trade wind clouds.

In addition to that, the transportation of dust particles at low atmospheric levels within/atop of the MBL during wintertime implies aerosol radiative effects. The particles may not only have a direct effect on the evolution of shallow marine clouds via absorption and scattering, but also have an indirect effect as ~~dust particles-aerosols~~ can act as cloud condensation nuclei and ice nucleating particles (Karydis et al., 2011; Bègue et al., 2015; DeMott et al., 2015; Boose et al., 2016). ~~However, before an investigation of these effects can be performed, wintertime transatlantic dust transport towards the Caribbean has to be investigated in detail, which is the focus of this study.~~

60 Up to now, the concurrence of transatlantic Saharan dust and water vapor transport during boreal winter has never been studied. In early 2020, extensive wintertime mineral dust plumes could be observed upstream the Caribbean island of Barbados during the EUREC⁴A field experiment (Elucidating the Role of Cloud-Circulation Coupling in Climate; Stevens et al., 2021). Several research flights with the High Altitude and Long Range Research Aircraft (HALO; Krautstrunk and Giez, 2012) were carried out over ~~these regions~~ this region and an extensive lidar and dropsonde data set was collected. This unique data set now
65 enables a detailed investigation of ~~the plumes~~ macrophysical properties of the observed long-range-transported dust plumes and the state of the atmosphere.

This paper is structured as follows. In section 2 an overview of the EUREC⁴A project as well as ~~of the performed research flights is given. Moreover,~~ the capabilities and features of the airborne lidar system WALES (Water Vapor Lidar Experiment in Space; Wirth et al., 2009) are introduced. Moreover, an explanation of how lidar data can be used to separate the contributions
70 of individual aerosol species in an aerosol mixture is outlined. A detailed analysis of the observed Saharan dust events by means of airborne lidar and dropsonde measurements is presented in section 3. Finally, in section 4, the results are discussed and the study is concluded.

2 Data and Methods

2.1 HALO research flights during EUREC⁴A

75 The EUREC⁴A field campaign aimed at investigating the driving factors for the evolution of trade-wind cumulus clouds in the winter trades (Stevens et al., 2021). As one of several employed research platforms during EUREC⁴A, the German high-flying research aircraft HALO conducted remote sensing measurements east of the Caribbean island of Barbados (Konow et al., 2021). In the period from 18 January to 18 February 2020, the modified Gulfstream G550 research aircraft performed a total of 15 scientific flights (13 local flights from and to Barbados and two transfer flights from and to Germany). An overview of
80 the HALO flight tracks is shown in Figure 1. The circular flight patterns of the research flights were flown for dropsonde-based analyses of the respective prevailing large-scale divergences (Bony and Stevens, 2019).

On three measurement days, Saharan mineral dust ~~plumes were was~~ advected to the research area, i.e. on 30 Jan 2020 during HALO-0130, on 31 Jan 2020 during HALO-0131 and on 2 Feb 2020 during HALO-0202. Collected airborne lidar data sets during measurement flights on these days enable a characterization of ~~winter-time dust transport~~ long-range-transported
85 African dust plumes during EUREC⁴A, although the flights tracks have not been specifically designed for dust observations. As dust aerosols could only be measured in this 4-day period, it is likely that the observed aerosol originated from the very same African dust outbreak. The enhanced total column aerosol optical depth (AOD) at 550 nm over the dust-affected research areas was ~~also~~ captured by MODIS (Moderate-resolution Imaging Spectroradiometer) and is shown in Figure 2. MODIS is a spaceborne imaging sensor which operates at 36 spectral bands from 0.4 μm to 14.4 μm wavelength. While on 2 February the
90 AOD in the research area ranged from 0.2 to 0.4, it was higher during the two other days and took maximum values greater than 0.6. The African origin of the aerosol plumes is also verified by calculated backward ~~trajectories~~ (-) trajectories using the

Hybrid Single Particle Lagrangian Integrated Trajectory model (HYSPLIT; Stein et al., 2015) [with NCEP GDAS \(National Centers for Environmental Prediction Global Data Assimilation System\) input data.](#)

In total, approximately 18.5 hours of lidar data could be collected during the three flights. Additionally, a total of 164
95 dropsondes were launched (Vaisala RD-41; George et al., 2021; Vaisala, 2020). 157 of them worked properly and collected thermodynamic data from aircraft to ground level.

2.2 The WALES lidar instrument aboard HALO

During EUREC⁴A the WALES lidar instrument (Wirth et al., 2009) was operated aboard HALO. It is an airborne ~~water-vapor~~
differential absorption lidar (DIAL) system [for profiling water vapor mass mixing ratios \(\$r_m\$ \)](#) with depolarization and high
100 spectral resolution lidar (~~HSRL; Esselborn et al., 2008~~) ~~capabilities~~ [capabilities \(HSRL; Esselborn et al., 2008\) for vertically resolved measurements of aerosol optical properties.](#)

The DIAL module operates at four wavelengths (three online and one offline wavelength) around the H₂O- absorption band at 935 nm. ~~This setup allows for measurements of water-vapor mass mixing ratios (r_m) that cover the whole extent from aircraft to ground level.~~ Due to its high pulse-repetition rate of 0.01 s between online and offline pulses, it enables
105 horizontally and vertically highly resolved measurements of r_m with relative uncertainties of less than 5 % (Kiemle et al., 2008). To obtain the high pulse repetition rate, two Q-switched Nd:YAG (Neodymium-doped Yttrium Aluminum Garnet; ~~Nd:Y₃Al₅O₁₂~~ [Nd:Y₃Al₅O₁₂](#)) ring lasers, that generate pulses at 1064 nm wavelength, are operated temporally interleaved. The required frequencies for DIAL measurements are generated via frequency-doubling of parts of the emitted pulses and subsequent pumping of injection-seeded optical parametric oscillators (Mahnke et al., 2007).

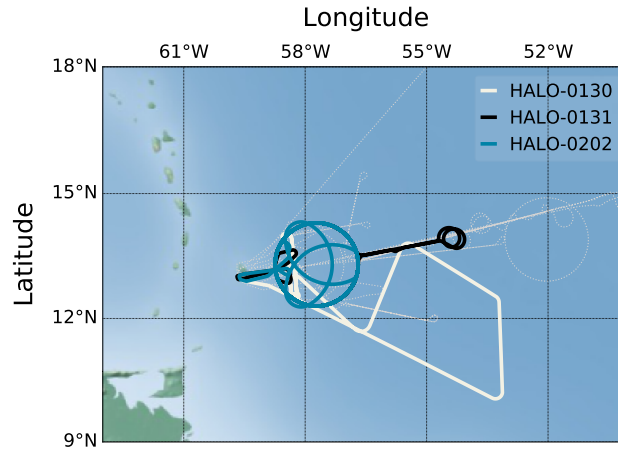


Figure 1. The HALO flight tracks during EUREC⁴A. Flights over long-range-transported Saharan mineral dust are colored. The thin dashed white lines indicate the remaining dust-free research flight-tracks.

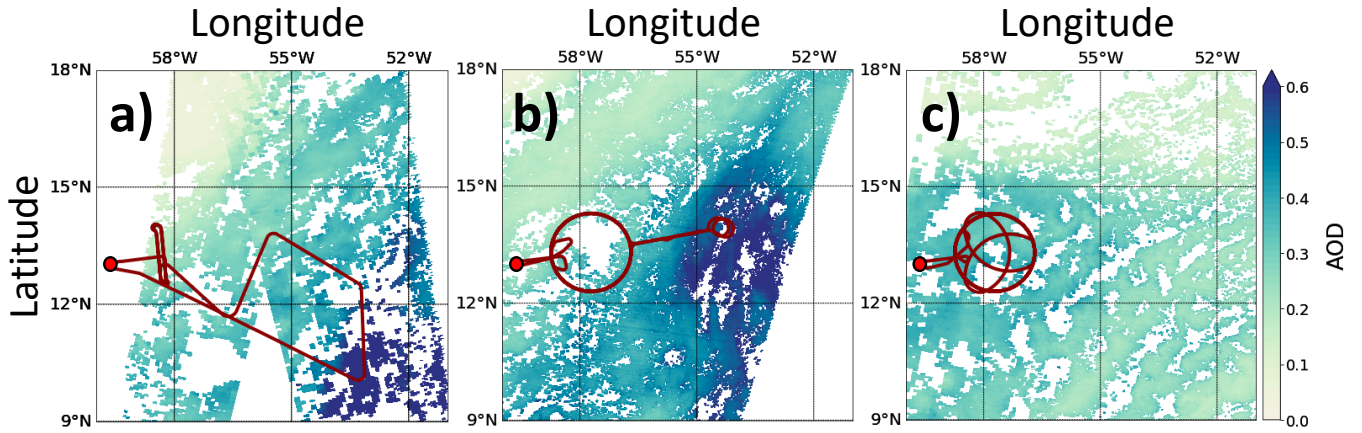


Figure 2. The HALO flight-tracks of the three research flights over long-range-transported Saharan dust aerosol on 30 Jan 2020 (a), 31 Jan 2020 (b) and 2 Feb 2020 (c) atop of MODIS (Moderate-resolution Imaging Spectroradiometer) satellite imagery of aerosol optical depth (AOD) at 550 nm. All MODIS images were taken between 14 and 18 UTC. The Grantley Adams International Airport on Barbados is marked with red dots.

110 In addition to the DIAL capability, the integrated HSRL-module and depolarization sensitive channels allow for highly re-
 solved measurements of particle linear depolarization ratios ($\delta_{p(532)}\delta_p$), backscatter ratios ($R_{532} = 1 + \beta_{p(532)}/\beta_{m(532)}$; with
 $\beta_{p(532)}$ and $\beta_{m(532)}$ $R = 1 + \beta_p/\beta_m$; with β_p and β_m being the particle backscatter and molecular backscatter coefficients),
 particle extinction coefficients ($\alpha_{p(532)}\alpha_p$) as well as the lidar ratios (S) - all at 532 nm wavelength. Relative uncertainties in
 measurements of $\delta_{p(532)}$, R_{532} and $\alpha_{p(532)}$ δ_p , R and α_p are 10 % to 16 %, 5 % and 10 % to 20 %, respectively (Esselborn et al.,
 115 2008).

WALES data is spatially and temporally averaged to improve the signal to noise ratio. At typical aircraft speed of 200 m s⁻¹,
 the horizontal resolutions amount to 3000 m (DIAL) and 200 m (HSRL). The vertical resolution is 15 m from and measurements
 are taken from aircraft to ground level. Since measurements of the WALES DIAL are masked at altitudes <200 m (contamination
 from surface echoes), no water vapor measurements near ground level are available.

120 2.3 Lidar-based aerosol separation

For an in-depth investigation of the predominating aerosol mixtures during EUREC⁴A (i.e. possible mixtures of transported
 mineral dust with marine aerosol or biomass burning aerosol), the well-established lidar-based aerosol-separation technique for
 two-component aerosol mixtures shown by Tesche et al. (2009) and Groß et al. (2011) is applied. It allows for the calculation
 of the contribution (in percent) of mineral dust to both the measured particle linear depolarization ratio δ_p and the measured
 125 lidar ratio S .

The percentage that Saharan mineral dust is contributing to a measured particle linear depolarization ratio of a two-component aerosol mixture δ_p with biomass burning aerosol or marine aerosol is calculated as follows,

$$[\%_{\delta}] = \frac{D_B}{D_B + D_A}, \quad (1)$$

with the coefficients D_A and D_B ,

$$D_A := \frac{\delta_{p,DU} - \delta_p}{S_{DU}(1 + \delta_{p,DU})}; \quad (2)$$

$$D_B := \frac{\delta_p - \delta_{p,BB|MA}}{S_{BB|MA}(1 + \delta_{p,BB|MA})}. \quad (3)$$

Here, S_{DU} and $S_{BB|MA}$ are the known lidar ratios of Saharan mineral dust (50 ± 4 ; Groß et al., 2011, 2013) as well as of aged tropospheric biomass burning aerosols (72.5 ± 7.5 ; Haarig et al., 2018) and marine aerosol (18 ± 5 ; Groß et al., 2013). $\delta_{p,DU}$ and $\delta_{p,BB|MA}$ are the corresponding known particle linear depolarization ratios (Saharan mineral dust: 27 ± 2 %; aged biomass burning aerosol: < 3 %; marine aerosol: 3 ± 1 %; Tesche et al., 2009; Groß et al., 2011, 2013; Haarig et al., 2017, 2018). For S the percentage is calculated using,

$$[\%_S] = \frac{S_{BB|MA}(S_{DU} - S)}{S(S_{BB|MA} - S_{DU})}. \quad (4)$$

3 Results

140 3.1 Saharan dust research flights during EUREC⁴A

Overview of measured profiles during the HALO-0130 (a), HALO-0131 (b) and HALO-0202 (c) research flights as measured by lidar and dropsondes. The left panels show median profiles of 532 nm backscatter ratio ($R_{532||}$, black), lidar ratio (S , teal), temperature (T , black), equivalent potential temperature (Θ_e , teal, dashed) and potential temperature (Θ , teal, solid) during the respective flights. Shaded regions indicate the inter-quartile region. The right panels show the measured profiles of 532 nm particle linear depolarization ratio ($\delta_{p(532)}$) and water vapor mass mixing ratio (r_m). Since measurements of the WALES DIAL are masked at altitudes < 200 m (contamination from surface echoes), no water vapor measurements near ground level are available. Transported Saharan mineral dust was present in the research area on three measurement days. Times of take-off and landing of the flights conducted on these days, as well as the number of launched dropsondes are summarized in Table 1. In the following, a detailed overview of the measurements during these three flights is given.

150 a. HALO-0130 on 30 January 2020:

An objective of this relatively short research flight was an under-flight of the GPM (Global Precipitation Measurement

Mission; Hou et al., 2014) satellite for concurrent airborne and spaceborne radar and radiometer measurements of sub-tropical clouds. For this purpose the flight track was designed to lead to a region in the Southeast of Barbados, where long-range-transported Saharan dust aerosol prevailed. The time-height cross section of $\delta_{p(532)} \delta_p$ highlights a highly depolarizing aerosol regime which ranged from approximately 0.7 km to 3.5 km altitude during the whole flight (Figure 3 (a)). Pure and non-mixed long-range-transported Saharan mineral dust aerosol usually takes typical values around 30 % (Freudenthaler et al., 2009; Groß et al., 2013). This value does not change with transatlantic transportation (Groß et al., 2015). However, $\delta_{p(532)} \delta_p$ between 0.7 km to 3.5 km altitude took slightly smaller values that ranged from 15 % to 30 %. Depolarization ratios in this range are typical for aerosol regimes where mineral dust particles are mixed with less depolarizing particles, i.e. marine aerosol or biomass burning aerosol (Teschke et al., 2009, 2011). Mixing of different aerosol types is also evident in derived lidar ratios at 532 nm. From ground level to approximately 0.7 km altitude they took values around 30 sr. In intermediate altitudes (0.7 km to 2.0 km) where $\delta_{p(532)} \delta_p$ took values around 15 % lidar ratios were around 40 sr. At the top levels of the depolarizing layer (2.0 km to 3.5 km) where $\delta_{p(532)} \delta_p$ was highest, the derived lidar ratios were also highest and took values around 60 sr. This is a clear indication that at the top levels of the aerosol layer highly absorbing biomass burning aerosols (S of 6372.5 ± 77.5 ; Haarig et al., 2018) are mixed with mineral dust particles (typical S of 50 ± 4). On the other hand, at the lower levels of the aerosol layer, values of $\delta_{p(532)} \delta_p$ and S point towards a mixture of marine aerosols (S of 18 ± 5) with dust particles (Ben-Ami et al., 2009; Groß et al., 2011).

Ensembles of 7-day backward trajectories that are calculated utilizing HYSPLIT show that the sampled dust-containing aerosol layer traveled for one week at a nearly constant altitude from Africa towards the Caribbean (Figure 3 (b)). Thermal anomalies derived from VIIRS (Visible Infrared Imaging Radiometer Suite) indicate strong activity of fires in West Africa. The low-latitude transportation route of the aerosol layer (5°N to 15°N) together with these fires favored the entrainment of biomass-burning aerosols into the boundary layer. A detailed discussion and analysis of the observed aerosol mixtures is given in subsection 3.2.

During HALO-0130 air masses at low atmospheric levels (trajectories starting at 0.4 km altitude) took a more northerly route over the North Atlantic Ocean (10°N to 15°N) than the dust-laden air masses at 1.2 km and 2.0 km. This could

Table 1. Overview of the conducted research flights over Saharan dust aerosol during EUREC⁴A in 2020 including dates, times of take-off and landing, total duration as well as number of launched dropsondes (times given in UTC - note: Atlantic Standard Time = UTC-4).

ID	Date	Take-off	Landing	Duration	Sondes
0130 HALO-0130	30 Jan	11:19	15:09	04:50 h	4
0131 HALO-0131	31 Jan	14:07	00:10 (+1 d)	10:03 h	74
0202 HALO-0202	02 Jan	11:28	20:15	08:47 h	89

explain why the MBL at lowermost altitudes (<0.7 km) is characterized by relatively small $\delta_{p(532)}\delta_p$ and S compared to the overlying atmosphere. The altitude of ~ 0.7 km altitude marked a transition inside the MBL. The MBL changed from a moist and neutrally stratified convective boundary layer near the surface ($r_m \sim 18 \text{ g kg}^{-1}$; $\Theta = \text{const.}$) to a dust-laden, drier and more stable layer with lower wind speeds ($r_m = 4\text{-}10 \text{ g kg}^{-1}$; $d\Theta/dz \geq 0$). This layer is then capped at approximately 2.5 km altitude by the trade wind inversion (TWI). The TWI was characterized by a hydrolapse, gradients in wind speed as well as temperature and the transition from an aerosol-laden to an aerosol-free atmosphere.

b. *HALO-0131 on 31 January 2020:*

HALO's second dust-flight aimed at measurements of large-scale divergence from dropsondes. This is why a total of seven circles were flown upstream of Barbados and dropsondes were launched at a very high rate. After 3.5 circles a short excursion towards the Northwest Tropical Atlantic Station - a meteorological buoy - was performed. The sampled atmosphere below the circles was characterized by an almost dust-free region in the Northwest and a dusty regime in the Southeast (see Figure 2 (b)). As a result the lidar depolarization data shows recurring features (Figure 4 (a)). Highly depolarizing and dust-laden regions were observed between 0.7 km and 3 km altitude ($15\% < \delta_{p(532)}\delta_p < 30\%$), with greatest depolarization ratios in 2 km altitude. ~~In-At~~ these altitudes the lidar ratio S took values around 60 sr, indicating the presence of both mineral dust and biomass burning aerosol. At lower levels (0.0 km to 0.7 km) S was smaller and around 20 sr. This points towards an additional contribution of marine aerosol ~~in-at~~ these altitudes.

Similar to HALO-0130, backward trajectories with starting points at low altitudes originate from more northern latitudes than the trajectories starting from altitudes affected by long-range-transported Saharan mineral dust, i.e. at 1.2 km and 2.0 km altitude (Figure 4 (b)). The low latitudes together with low transportation altitudes can again explain that not only mineral dust, but also biomass burning aerosol from West African fires, marine aerosol and Saharan mineral dust, were observed. This is substantiated by the evident forest fires detected from VIIRS thermal anomalies. The MBL was characterized by a moist and well-mixed convective layer from surface to approximately 0.7 km altitude ($r_m \approx 18 \text{ g kg}^{-1}$; $\Theta = \text{const.}$) and a dryer, calmer and statically more stable layer from 0.7 km to 2.2 km which was capped by the TWI. A relatively moist and dust-containing aerosol layer was observed atop of the MBL. It was vertically extending for roughly one kilometer and showed water vapor mass mixing ratios of approximately 4 g kg^{-1} .

c. *HALO-0202 on 2 February 2020:*

Circular flight patterns for large-scale divergence measurements have also been the focus of this research flight. For an additional characterization of the cloud and aerosol scene in the interior of the circles a clover-pattern was flown. As a result WALES lidar measurements during this research flight show recurring features (Figure 5 (a)).

The MBL was again characterized by a well-mixed convective layer from surface to approximately 0.7 km altitude ($\Theta = \text{const.}$). However, water vapor mixing ratios were slightly smaller compared to the other two flights ($\sim 16 \text{ g kg}^{-1}$). The MBL was capped at approximately 2.5 km altitude by a pronounced TWI. Depolarizing aerosol was found along the whole vertical extent of the MBL. However, $\delta_{p(532)}\delta_p$ -values were highest at the very top of the MBL ($\sim 30\%$ at 2.3 km altitude). Lidar ratios again indicate that the aerosol layer was characterized by a particle mixture of mineral

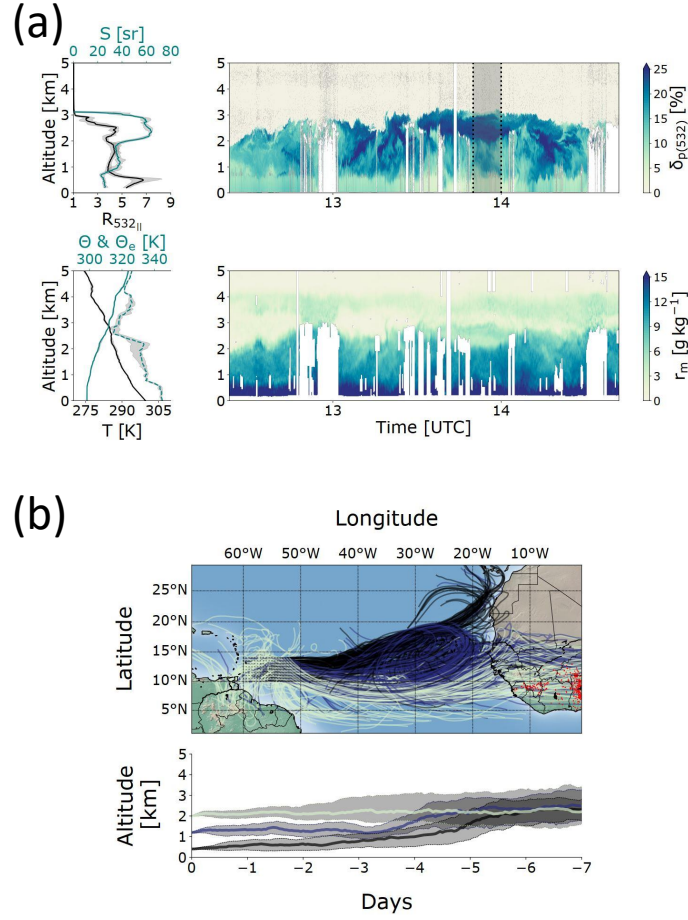


Figure 3. (a) Overview of measured profiles during the HALO-0130 research flight as measured by lidar and dropsondes. The left panels show median profiles of 532 nm backscatter ratio ($R_{||}$, black), lidar ratio (S, teal), temperature (T, black), equivalent potential temperature (Θ_e , teal, dashed) and potential temperature (Θ , teal, solid) during the flight. The profiles of $R_{||}$ and S refer to the grey shaded regions in the respective right panel. Shaded regions indicate the inter-quartile region. The right panels show the measured profiles of 532 nm particle linear depolarization ratio (δ_p) and water vapor mass mixing ratio (r_m). (b) The 7-d backward trajectories of the air masses that were sampled during the HALO-0130 (a), HALO-0131 (b) and HALO-0202 (c) research flights. Trajectories are indicated for starting points in 0.4 km (black), 1.2 km (blue) and 2.0 km (light-green) altitude that are regularly distributed over the research areas (0.5° spacing) and are calculated using the Hybrid Single Particle Lagrangian Integrated Trajectory model HYSPLIT. Red markers indicate detected thermal anomalies related to active fires detected by VIIRS (Visible Infrared Imaging Radiometer Suite) 7 days before the respective research flights. The lower panels show the median altitude of the backward trajectories. Shaded regions indicate the inter-quartile region.

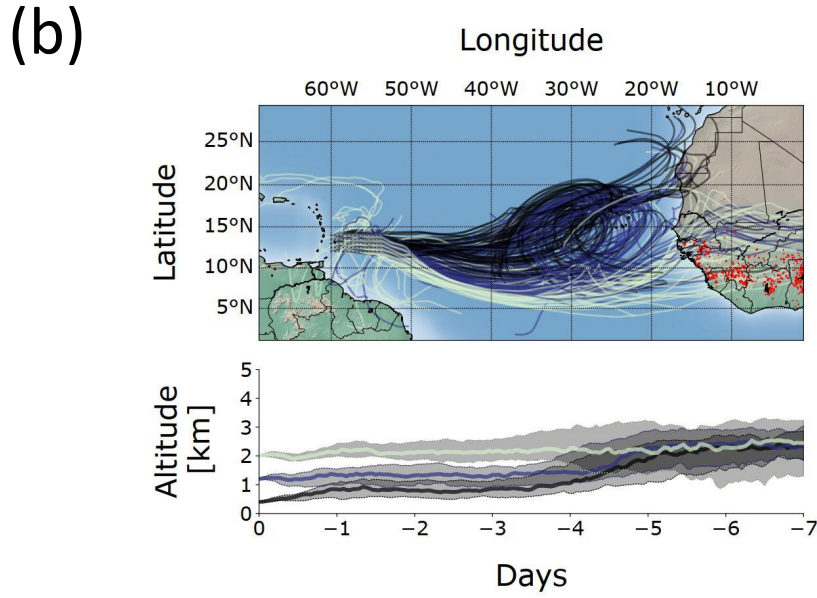
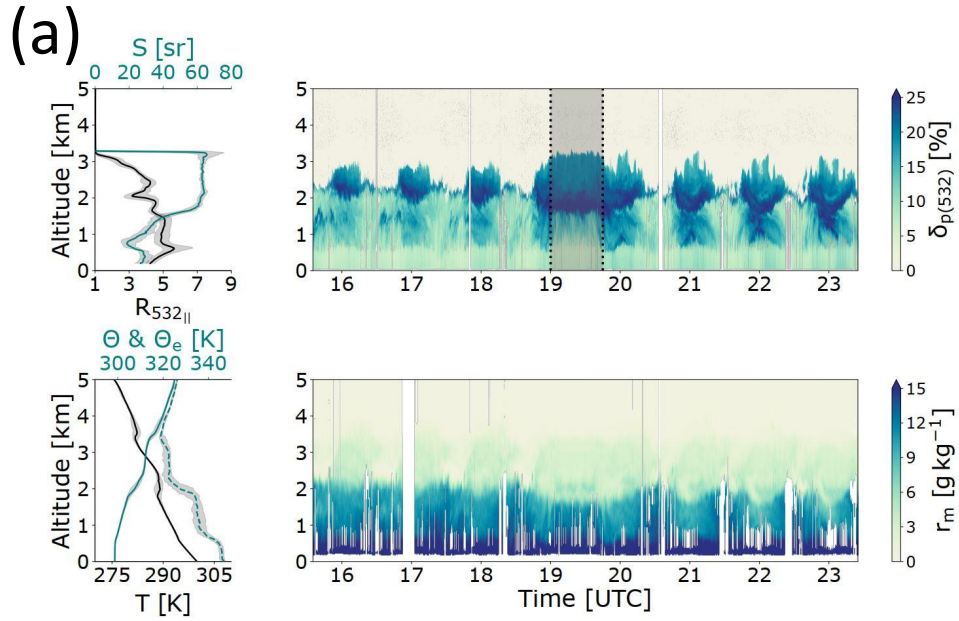


Figure 4. Same as Figure 3 for HALO-0131.

dust aerosol, [aged](#) biomass-burning aerosol and marine aerosol. S took highest values inside the aerosol layer atop of the MBL (~ 60 sr) and was lowest near ground level (~ 30 sr). This indicates that the contribution of biomass burning aerosol to the aerosol mixture was greatest at the top levels and marine aerosol mostly contributed to the aerosol mixture at lower levels. Regions inside the MBL that were impacted by this aerosol mixture came along with reduced atmospheric moisture content and were characterized by small water vapor mixing ratios that ranged from 5 g kg^{-1} to 10 g kg^{-1} .

7-d backward trajectories for this research flight again indicate that the dust-laden air masses in 1.2 km and 2.0 km altitude traveled at nearly constant altitude and that they took a southerly route that favored mixing processes with biomass burning aerosols and marine aerosols (Figure 5 (b)).

3.2 Aerosol and water vapor composition of the observed dust layers

Measurements of lidar ratios and particle linear depolarization ratios during the three research flights over transported Saharan mineral dust indicated that dust particles have been mixed together with [aged](#) biomass burning and marine aerosols. An aerosol regime containing only mineral dust particles was never observed during EUREC⁴A. To investigate these mixed regimes in more detail, the contribution of Saharan mineral dust aerosol to the aerosol mixture is calculated. ~~The well-established aerosol-separation technique for two-component aerosol mixtures shown by Tesche et al. (2009) and Groß et al. (2011) allows for the calculation of the contribution (in percent) of mineral dust to both the measured particle linear depolarization ratio $\delta_{p(532)}$ and the measured lidar ratio S .~~

~~The percentage that Saharan mineral dust is contributing to a measured particle linear depolarization ratio of a two-component aerosol mixture $\delta_{p(532)}$ with biomass burning aerosol or marine aerosol is calculated as follows,~~

$$[\%_{\delta}] = \frac{D_B}{D_B + D_A},$$

~~with the coefficients D_A and D_B ,~~

$$D_A := \frac{\delta_{p(532),DU} - \delta_{p(532)}}{S_{DU}(1 + \delta_{p(532),DU})},$$

$$D_B := \frac{\delta_{p(532)} - \delta_{p(532),BB|MA}}{S_{BB|MA}(1 + \delta_{p(532),BB|MA})}.$$

~~Here, S_{DU} and $S_{BB|MA}$ are the known lidar ratios of Saharan mineral dust (50 ± 4) as well as of biomass burning aerosols (63 ± 7) and marine aerosol (18 ± 5) (mean values of observations by Groß et al., 2013). $\delta_{p(532),DU}$ and $\delta_{p(532),BB|MA}$ are the corresponding known particle linear depolarization ratios (Saharan mineral dust: $27 \pm 2\%$; biomass burning aerosol: $14 \pm 2\%$; marine aerosol: $3 \pm 1\%$). For S the percentage is calculated using,~~

$$[\%_S] = \frac{S_{BB|MA}(S_{DU} - S)}{S(S_{BB|MA} - S_{DU})}.$$

~~Using these equations~~ [Using the equations presented in](#) subsection 2.3, one can derive mixing lines between the pure Saharan mineral dust aerosol regime and the marine and biomass burning aerosol regime in a two-dimensional space of S and $\delta_{p(532)}$

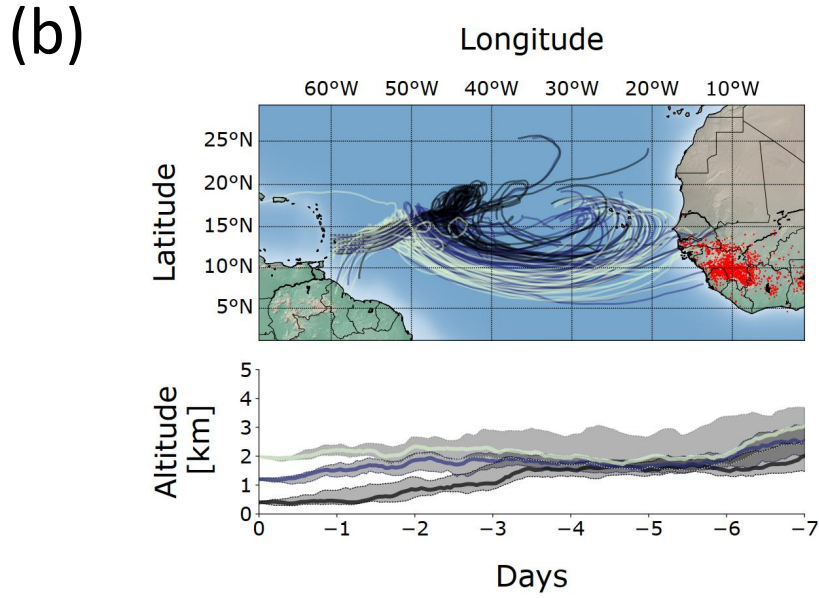
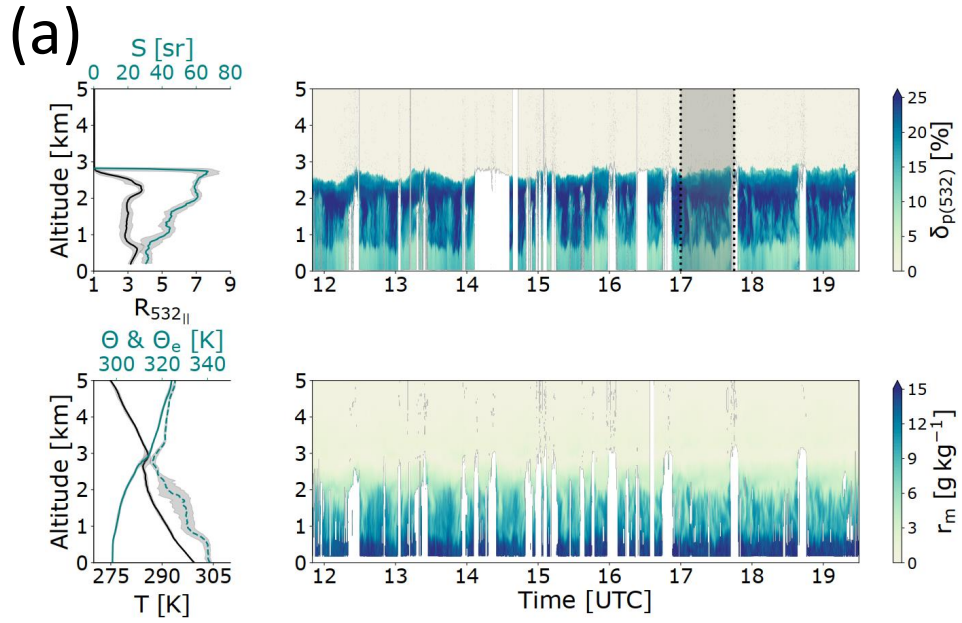


Figure 5. [Same as](#) Figure 3 [for HALO-0202](#).

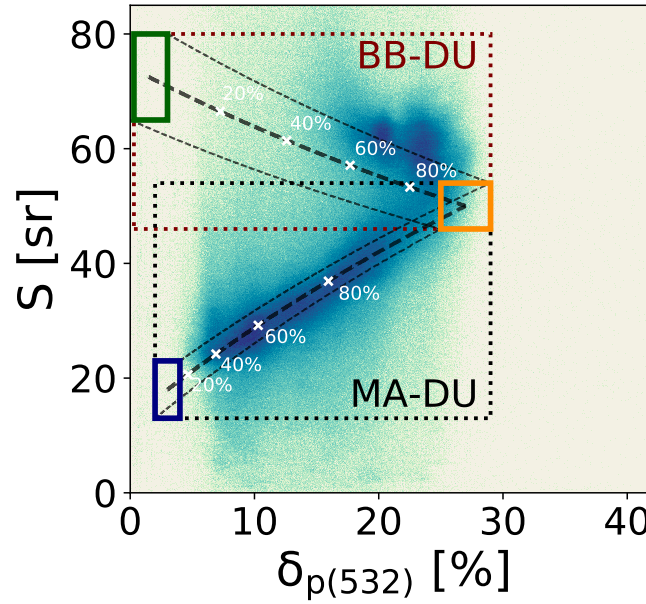


Figure 6. Two-dimensional histogram showing the joint distribution of measured particle linear depolarization ($\delta_{p(532)}\delta_p$) and lidar ratios (S) during the three research flights over long-range-transported Saharan mineral dust. Solid rectangles mark the typical positions of the pure aerosol regimes: marine aerosol (MA, blue), transported Saharan mineral dust (DU, orange) and biomass burning aerosol (BB, green). The dashed rectangles indicate the mixing regimes of biomass burning aerosols with transported Saharan dust aerosols (BB-DU, maroon) as well as of marine aerosols with transported Saharan dust aerosols (MA-DU, blue). Grey lines indicate the calculated mixing lines (dashed: mean, solid: \pm standard deviation) of the two aerosol mixtures MA-DU and BB-DU. White crosses and percentages indicate the contribution of dust to $\delta_{p(532)}\delta_p$ of the respective two-component mixtures. The histogram is normalized to the maximum bin count.

δ_p (Figure 6). The joint histogram of the lidar measurements during the three flights over long-range-transported Saharan mineral dust aerosol fits well to the calculated mixing lines. Maximum frequencies of measurements correlate very well with the mixing lines between mineral dust and biomass burning aerosol as well as mineral dust and marine aerosol derived from the calculations. The histogram also shows that biomass burning aerosols are not mixed to low atmospheric levels as this would
 245 cause a shift of the mixing line between marine aerosol and dust aerosol towards higher lidar ratios. Mineral dust contributions between 40 % and 80 % were observed most frequently in mixtures of marine aerosol with dust aerosol (MA-DU). At higher atmospheric levels mineral dust ~~contributed between 20~~ mostly contributed between 60 % and 80 % to the mixture of biomass burning aerosol and dust aerosol (BB-DU).

Gutleben et al. (2019a, 2020) observed enhanced water vapor concentrations of approximately 4 g kg^{-1} in elevated and
 250 long-range-transported SALs compared to the surrounding atmosphere during the boreal summer months. One objective of the lidar measurements during EUREC⁴A is to answer the question whether this is also the case for transatlantic dust transport during the boreal winter season.

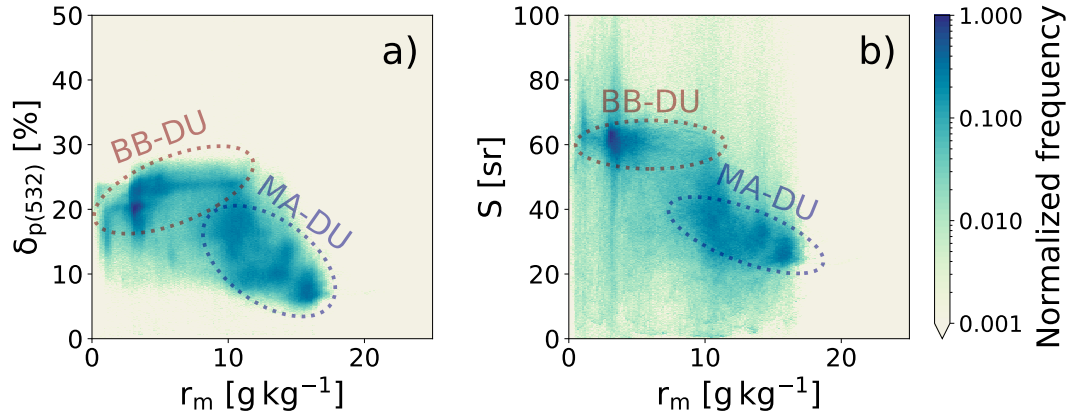


Figure 7. Two-dimensional histograms showing the joint frequency distribution of measured particle linear depolarization $\delta_{p(532)}$ (a) and lidar ratios S (b) with simultaneously measured water vapor mass mixing ratios r_m during the three research flights over long-range-transported Saharan mineral dust. The dashed ellipses indicate an outline of the mixing regimes of aged biomass burning aerosols with transported Saharan dust aerosols (BB-DU, maroon) as well as of marine aerosols with transported Saharan dust aerosols (MA-DU, blue). The histograms are normalized to their respective maximum bin count.

When looking at the individual lidar curtains of the three research flights ~~in~~ one can already identify the slightly enhanced water vapor mass concentrations compared to the dry free troposphere in regions of highest $\delta_{p(532)}$. Measurements during HALO-0131 and HALO-0202 highlight that the regions of maximum $\delta_{p(532)}$ - which are associated with a great mineral dust loading - on top of the MBLs are not dry, but carry water vapor around 4 g kg^{-1} .

This can also be seen in the joint distributions of r_m with $\delta_{p(532)}$ and S (see Figure 7). It indicates that regions affected by mineral dust are not completely dry, but always come along with enhanced water vapor concentrations. Only in mixed regions, where biomass-burning aerosol ~~dominates the dust aerosol~~ ($\delta_{p(532)} \approx$ ~~affects the aerosol mixture~~ ($\delta_p \approx 20 \%$; $S \approx 60 \text{ sr}$), water vapor mixing ratios drop to values smaller 4 g kg^{-1} . In the lower MBL, where a mixture of mineral dust with marine aerosol predominates, water vapor concentrations are high in general and take values between 8 g kg^{-1} and 18 g kg^{-1} .

4 Discussion and Conclusions

While during the summer months Saharan dust particles are predominantly transported westwards in SALs at altitudes as great as 6 km (e.g. Prospero and Carlson, 1972; Prospero et al., 2010), Saharan dust transport in the winter months happens at lower atmospheric levels (~~Chiapello et al., 1995~~) (e.g. Ansmann et al., 2009; Groß et al., 2011; Tesche et al., 2011). This agrees well with the findings in this study, as mineral dust particles were never observed in altitudes higher than $\sim 3.5 \text{ km}$. Low level transport also favors mixing processes of mineral dust particles with other aerosol species like biomass burning aerosol or marine aerosol. As a consequence, pure dust aerosol regimes were never observed during EUREC⁴A and mineral dust

particles could only be observed in mixed aerosol regimes. At lowermost altitudes inside the MBL the dust particles were
270 predominantly mixed with marine aerosol. Above the MBL in altitudes from 2 km to 3.5 km the dust particles were mixed with
biomass burning aerosol from fires in West Africa.

During their travel over the Atlantic Ocean these dusty aerosol regimes can impact the Earth's radiation budget by directly
interacting with radiation via absorption and scattering, by changing cloud microphysical properties or by modifying the
atmospheric stratification. However, up to now the radiative impact of long-range Saharan mineral dust transport over the
275 Atlantic Ocean during boreal winter has never been investigated in detail. Needed highly resolved observations of the vertical
and horizontal aerosol distribution were missing. For summertime transport Gutleben et al. (2019b, 2020) highlighted that not
the aerosol, but the water vapor embedded in long-range-transported SALs is the dominant driver for radiative heating and the
subsequent modification of the atmospheric stability. They used a radiative transfer model together with airborne lidar data
collected during NARVAL-II (Next-generation aircraft remote sensing for validation studies II) for their calculation. During
280 summertime the dust transport usually occurs in elevated layers. Due to this spatial separation of the different aerosol layers,
the impact of enhanced concentrations of water vapor in SALs on atmospheric heating could easily be quantified. However,
wintertime low-level transport - as observed during EUREC⁴A - hampers such a separation as the water vapor concentration
inside the MBL is high on principle. In addition, the indirect radiative effect of Saharan mineral dust and biomass burning
aerosols (i.e. the modification of marine cloud microphysics by aerosol particles) has to be considered in future simulations
285 of radiative transfer in the winter season. Haarig et al. (2019) demonstrated that smoke particles play a crucial role for cloud
condensation especially during boreal winter as they dominate the concentration of available cloud condensation nuclei in
the mixture. Moreover, the potential of mixed dust aerosol regimes during winter-time transport for ice nucleation should be
investigated in future studies.

This study highlighted the characteristics of the observed dusty aerosol layers during EUREC⁴A. Although it is based on
290 data collected during only three measurement flights, enhanced water vapor concentrations could be linked to the dust-laden
aerosol layers for the first time. In near future the composition of the dust layers and their radiative impact on the subtropical
environment should be investigated in more detail. Finally, it is highly recommended that future and ongoing analyses with
focus on radiative transfer, cloud physics or cloud occurrence based on observations during EUREC⁴A consider the impacts
of the pronounced aerosol layers described in this paper.

295 *Data availability.* The data used in this publication were collected in the framework of the field study EUREC⁴A and are publicly available
online in the AERIS database (<https://observations.ipsl.fr/aeris/eurec4a/>)

Author contributions. MG, SG and MW contributed in carrying out the airborne measurements during the EUREC⁴A field campaign. Initial
lidar data processing was performed by MW. MG and CH performed all analytic computations and analyzed the measured data set. MG took

the lead in writing the manuscript under permanent consultation with SG. All authors discussed and reviewed the findings and contributed to
300 the final manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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