Letter of Reply to Referee 2

Thank you for carefully reading the manuscript and providing useful suggestions to improve the paper. The changes in the manuscript are marked in bold.
To strengthen the point of pure marine conditions during our measurements, we added a more sophisticated air mass analysis (Fig. 7), which takes different land covers into account. Martin Radenz who performed the calculations was added as a co-author.

Specific Comments

Figure 2 is very interesting and helpful for visualizing the sea salt particles at different stages of the transition. Initially the dust in these images confused me as I was concerned that it might be very hard to separate dry sea salt from dust if they are mixed together like this. On more careful reading, I see that the images in Figure 2 are from a completely different time of year to the case studies. This is perfectly fine, but you might consider putting in another sentence or two in key places to make sure that it is very clear (even to people who are reading a little too fast) that the images in Figure 2 are not from the same time period as the main case studies in the paper.

Thank you very much for this comment. Two phrases on p4, l19-23 and in the caption of Fig 2 have been added to make sure, that it is very clear for everyone.

Page 5, line 9-14 is confusing about whether spherical or non-spherical sea salt particles would be expected to be bigger. Possibly this confusion would be removed if you make a clearer distinction between humidified sea salt particles and droplets. Is it true that crystals are larger than solution droplets, but then the crystals continue to grow as they are re-humidified, so that humidified crystals are larger than solution droplets? Or am I misunderstanding the text in this paragraph?

To clarify this point: In general the sea salt solution droplets are larger than the sea salt crystals, because the humidified sea salt particles (=solution droplets) grow due to water uptake.

The mode radii from Sakai et al., 2010, have been removed from the manuscript. Their high standard deviation (0.08 ± 2.1 µm for the droplets and 0.19 ± 1.8 µm for the crystals) hampers a clear conclusion, which particles are larger in this laboratory study. They prepared the particles in five radius ranges between 0.15 and 2.5 µm (Sakai et al, 2010). Instead the standard deviations for the depolarization ratios have been added (p5, l12-14).

The quantities shown in the backscatter curtains Fig 3-5 are not the best ones to show. It would be more informative to show geophysical quantities, the same ones shown in Figs 8 and 9. Specifically total true backscatter coefficient rather than attenuated backscatter or the cross-polarized component of attenuated backscatter, and particulate depolarization ratio rather than volume depolarization ratio. What are the horizontal and vertical resolution of the lidar data shown in the figures?

Indeed, the total backscatter would convey more information, but the presented scenario (Fig. 3) will lose information due to a larger smoothing, that is necessary for the calculation of the backscatter. In Fig. 3 the range resolution of the lidar (7.5 m) is used with a time resolution of 10 s. We decided to show the high resolution plot of the range-corrected cross polarized signal at 1064 nm as it is very sensitive to aerosol particles.

The volume linear depolarization ratio at 1064 nm is shown in Fig. 4 and 5. The molecular contribution at this large wavelength is almost negligible, so that the shown volume depolarization ratio is almost equal to the particle depolarization ratio. This is not the case at 355 or 532 nm. In Haarig et al., ACP 2017 we discussed the advantages of measuring the depolarization ratio at 1064 nm.

The vertical and temporal resolution was added in the captions of Fig 3-5.

Figure 10 shows a larger lidar ratio for the dry particles (lower RH). Can you explain in general terms in the paper why this is, in terms of the relative size and refractive indices of the particles?

The difference of the lidar ratio at 532 nm is not significant. The higher lidar ratio at 355 nm for dry particles may be explained by the fact, that the dry sea salt particles are smaller. Absorption is very weak for sea salt particles (high single scattering albedo). Therefore, the extinction is caused almost only by scattering. The lidar ratio is then the ratio of scattering to backscattering. Smaller particles have a larger amount of sideward scattering leading to a larger lidar ratio. A sentence has been added p10, l18-19.

Page 11, lines 10-13 states that particle backscatter coefficient is an indicator of particle growth if the number concentration stays the same. However, there is no particular reason to imagine the number concentration is the same for these observations, is there? However, you have two wavelengths of extinction, and Angstrom exponent of extinction is a more usual proxy for particle size (indeed you use AOT Angstrom exponent from AERONET in Fig 7). It would be useful to show extinction Angstrom exponent from the lidar measurements to see if this supports the idea of particle growth.

The extinction Angstrom exponent cannot be derived satisfactory below 1 km due to overlap issues. Still the extinction Angstrom exponent is very uncertain, as the extinction is small. Between 1.0 and 1.5 km height (the dry marine layer) the extinction Angstrom exponent (E355/E532) increases indicating a decreasing particle size. The same holds for the backscatter Angstrom exponent (B532/B1064), which can be derived at lower altitudes as well. A sentence was added p11, l12-14. The profiles of the Angstrom exponent are shown here, but not in the paper:
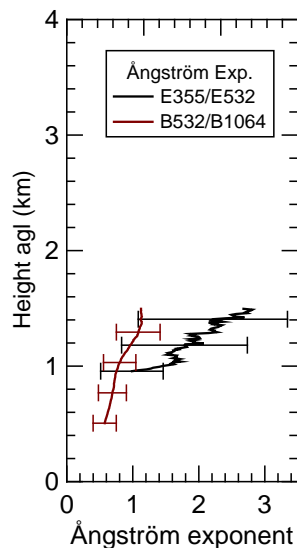

Figure 1 Vertical profiles of the Angstrom exponents measured 23 Feb 2014 23:38-01:38 UTC with uncertainty.

Same place. I also think that particle backscatter coefficient has a dependence on refractive index which may play a role in the trends seen here, although I don't know the relative strengths

of the dependencies on N, refractive index and particle size in this observation. Can you comment on this?

We cannot measure the refractive index. Simulations show, that the lidar ratio (extinction-to-backscatter ratio) is independent of refractive index in the observed size range.

Page 12 line 1-2 discusses a poor agreement when extrapolating. It's not surprising that extrapolating from an exponential fit might produce poor agreement. It would be useful to show a linearized version of Figure 13 (that is, log of enhancement factor vs log(1-RH/100)) which may make it more obvious in what regime the parameterization of Eq. 2 holds or might even actually improve the fit and the extrapolation, depending on how the parameters were fitted (that is, was the fit done on the linearized system?)

We added the suggested log-log plot in Fig. 14. It can be seen from this plot that the exponential (now linear) fit does not hold for the highest and especially for the lowest RH values. Two sentences are added (p12, l6-8).

Page 12 line 10. What does "modeled" mean? Is this the simulated data in Table 1 that is explained much later?

The simulated lidar ratios (Table 1) are used. A reference to the table was added (p12, l20).

Page 12 line 15. I don't understand this. The increase 2.05 to 3.73 to 5.37 is more than 8% from one wavelength to the next. "Zieger et al stated that the wavelength dependence for marine aerosol". Do you mean the wavelength dependence of the enhancement factor?

The 8% increase from one wavelength to the next was observed by Kotchenruther et al., 1999 for 450, 550 and 700 nm. The number was deleted. In the next sentence, the wavelength dependence is referring to the scattering enhancement factor for marine aerosol. The passage was deleted.

Same place: it seems that you could infer the expected behavior of the wavelength dependence by assuming that the angstrom exponent depends more or less linearly on size and use standard parametrization of the hygroscopic growth of particles. Would that be a reasonable assumption and would such a calculation give you similar wavelength dependence to what you see?

The small (dry) particles' influence is less pronounced for the long wavelengths (1064 nm) compared to the UV. The particle grows due to water uptake and gets more relevant in the scattering process for the long wavelengths as well. This dependence can be seen in Fig. 12 a-c. The backscatter at the dry RH is much smaller for 1064 nm. Thus, the enhancement is much stronger than at 355 nm.

Figure 14 and section 5. It would be helpful to mark a line on each panel of Figure 14 showing the value of volume equivalent radius used for the calculation of simulated results in Table 1 and to include this information also in Table 1.

For the calculations we used all values shown in Fig. 15. The different radii were weighted according to the AERONET size distribution (or an additional case assuming halved size because of drying). We make this point now clearer in Section 5. Additionally, we added the effective radius (r=1.033 µm) of the AERONET size distribution to the simulations of LR and PLDR in Fig. 15.

Page 13, lines 16-18. I got distracted trying to understand if the authors mean this last sentence of the paragraph to be related to the rest of the paragraph or not. If it is an unrelated observation, I suggest moving it into its own paragraph. If it is related, I suggest adding some kind of transition to make the reason for making this statement more obvious. Perhaps just

"However" or "Nevertheless", that is, if you are meaning to suggest that we need not worry overmuch about the disagreement between measured and modeled lidar ratios since it nevertheless represents a relatively small variation in the properties of the type compared to differences between types.

We followed your suggestion and added "Nevertheless" on p14, l2 and reshaped the paragraph.

Page 14, There are some rather vague and unconvincing conclusions here. It seems like you are struggling a little to define the relevance of the observation. I first want to say that this is a very interesting and publishable observation and analysis even in the worst case if it is not particularly relevant to other specific calculations like radiative transfer or aerosol types. If these arguments can't be strengthened, it would be better to take them out with no loss to the paper, in my opinion. More specifically, first, it's not clear to me how or whether this observation affects radiative transfer calculations. What is the mechanism by which dried sea salt aerosol would have a different radiative effect than humidified sea salt in a way that is not captured in the measurements? The differences in backscatter or extinction are directly measured so whether the backscattering comes from depolarizing particles or not is not relevant. Is it because of a poor inference of the refractive index? But you hardly discuss refractive index in this paper. Likewise, what is the mechanism (what characteristic of the dried sea salt) that would cause an error in the AERONET retrievals? Moreover, even if these layers are very common (which it is not at all obvious to me), the layer you observed is very thin compared to the aerosol below it. How much effect would this realistically have on a full column calculation of radiative transfer? The authors could do a rough, approximate calculation to show whether this is a noticeable effect or not, and then I might be more convinced. It also would be good to expand the discussion of aerosol typing to explain why it would matter to analysis such as the cited references if a small amount of aerosol is mistyped. Is this specifically about inferring lidar ratios for aerosol extinction retrievals, like CALIPSO, or is there some other implication to mistyping?

Thank you for your detailed discussion about the conclusion. We reshaped and shortened the conclusion.

The layer of dried marine aerosol is thin compared to the thicker layer of humid marine aerosol, so the radiative effect might not be that strong (p14, l22-24). But it would be interesting to see the effect in the models. The discussion is not about the radiative effect in the measurements but in the radiative transfer models, which calculate for example the radiative forcing by aerosol layers. We agree that the effect on backscatter and extinction is captured with our lidar measurements.

Inversion algorithms have to assume a shape for the particles. Due to simplicity, a spherical shape is usually assumed. It has been shown that this does not work for non-spherical dust particles. To better invert dusty scenarios, a spheroidal shape is assumed. It would not be difficult to assume a cubic shape as well to mimic non-spherical dry marine aerosol. We softened the text concerning AERONET and suggest using a cubic model (p14, l25-27). It would be interesting to see for some test cases, how a mixture of cubes and spheres would perform.

The problems in mistyping aerosol types are further discussed (p14, l28 – p15, l2). It is not only to derive extinction values for satellites. Recently estimates of mass, CCN and INP concentration profiles are provided from lidar measurements (Mamouri and Ansmann, 2016). The depolarization ratio plays a key role in these retrievals and dry marine particles with a high depolarization ratio will cause errors in the retrievals.

Furthermore, we expressed the wish that global studies with satellites (CALIPSO or EarthCARE) would estimate the global occurrence of dry marine aerosol layer (p14, l21-22). In our own lidar measurements, we found further evidence over the Southern

Atlantic, over Cyprus and Haifa. Measurements over the Ocean are rare, so it is not that easy to address the phenomena.

Technical comments

The abstract seems a little scattered. Each sentence seems to stand alone without clear relationships between them. Rearranging the sentences to be in a more logical order and adding some transitional phrases ("therefore", "however", etc.) or combining sentences would help.

The abstract was rearranged and shorted to be more convenient.

Page 6, line 5. "See AERONET web page, Barbados-SALTRACE site". Please give specific URLs.

The URL for the AERONET web page is given. The station name is Barbados_SALTRACE.

What wavelength is Fig 3-5? Please include the information in the figure labels or captions.

The wavelength information (1064 nm) is indicated in each figure and in the caption.

Figure 8. From the text, I think this is particulate depolarization ratio (rather than volume). Please confirm, and include that information in the caption.

The caption of the axis has been changed to explicitly state "Part. depolarization ratio".

Figures 8 and 9. It seems there is meant to be a dashed vertical line in all 3 panels, but it appears only in the right panels.

The crystallization RH line has been added in all 3 panels. Additionally, we added dashed lines at the crystallization RH in Fig. 11-13.

Bigger axis labels and legends are needed in the figures, big enough to be on the order of other font sizes used in the paper

Thank you for this hint. The font sizes in the figures have been increased.

Dry versus wet marine particle optical properties: RH dependence of depolarization ratio, backscatter and extinction from multiwavelength lidar measurements during SALTRACE

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Abstract. Triple-wavelength lidar profiles of the depolarization ratio and the backscatter coefficient of marine aerosol as a function of relative humidity (RH) are presented with a 5-min time resolution. The measurements were performed at Barbados (13°N, 59°W) during the Saharan Aerosol Long-range Transport and Aerosol-Cloud interaction Experiment (SALTRACE) winter campaign in February 2014. The phase transition from spherical sea salt particles to cubic-like
5 **sea salt crystals was observed under atmospheric conditions with a polarization lidar. The radiosonde and water-vapor Raman lidar observations show a drop in RH below 50% in the marine aerosol layer simultaneously with a strong increase in particle linear depolarization ratio, which reaches values up to 0.12±0.08 (at 355 nm), 0.15±0.03 (at 532 nm), and 0.10±0.01 (at 1064 nm). The lidar ratio (extinction-to-backscatter ratio) increased from 19 sr and 23 sr for spherical sea salt particles to 27 sr and 25 sr (at 355 and 532 nm, respectively) for cubic-like particle ensembles. Furthermore**
10 **the scattering enhancement due to hygroscopic growth of the marine aerosol particles under atmospheric conditions was measured. Extinction enhancement factors from 40% to 80% RH of 1.94±0.94 at 355 nm, 3.70±1.14 at 532 nm, and 5.37±1.66 at 1064 nm were found. The enhanced depolarization ratios and lidar ratios were compared to modeling studies of cubic sea salt particles.**

1 Introduction

15 Since more than 70% of the Earth are covered with water, the optical properties of marine particles must be carefully considered in radiative transfer schemes in global atmospheric models. This includes marine conditions with relative humidity (RH) < 50% so that marine particles get **increasingly** dry, change their shape and thus their optical properties as we will demonstrate in this paper. The shape of sea salt particles strongly depends on RH. At typical values of RH > 80% in the marine boundary layer, sea salt particles are liquid solution drops and thus spherical in shape. When RH decreases below 45%, they crystallize and

20 become mostly cubic-like in shape **(Wise et al., 2007), but they will not become absolutely dry**. The change in shape leads to different optical properties, especially to changes in the linear depolarization ratio. Spheres have a linear depolarization ratio of

ideally zero, non-spherical particles exhibit higher values **(e.g., Murayama et al., 1999; Sakai et al., 2010; Gasteiger et al., 2011; David et al., 2013; Kemppinen et al., 2015a, b)**. The different optical properties of dry and humid sea salt have to be considered in various applications.

Satellite passive remote sensing as well as ground-based passive remote sensing (AERONET, e.g., Smirnov et al., 2002; Sayer et al., 2012, for marine environments) may be sometimes significantly affected by dry marine particles in marine environments (coastal regions during sea breeze effects). Cubic-like particles have a different scattering phase function than spherical particles. Analogous to the mixture of Saharan dust (assumed to be spheroidal in shape) and spherical anthropogenic particles, in the case of marine particles one would need an analysis scheme which considers cubic particles (and related scattering phase function) besides the spherical ones. The same should be considered in lidar inversion methods (e.g., Veselovskii et al., 2010; Müller et al., 2013), when inverting microphysical properties over the oceans and coastal areas. Non-spherical particles can have a sensitive impact on the retrieval products, thus particle shape has to be carefully considered **(Adachi and Buseck, 2015)**.

Aerosol classification from active remote sensing (Burton et al., 2012; Groß et al., 2013) based on the depolarization ratio **will be misleading** if dried marine aerosol with a high depolarization ratio is present. The polarization lidar observation used to separate dust from non-dust (e.g. Sugimoto and Lee (2006); Tesche et al. (2009); Mamouri and Ansmann (2014, 2017)) rely on the assumption that non-dust aerosol always produces depolarization ratios around 0.05 or less and significant depolarization is only caused by dry irregularly shaped dust particles. The cubic-like sea salt particles will consequently be misinterpreted as dust leading to an overestimation of the dust concentration.

Active remote sensing from satellites (CALIOP and EarthCARE) use the polarization technique to separate aerosol types. The derived optical properties like the extinction coefficient depend on the detection of the correct aerosol type to choose the appropriate lidar ratio (Omar et al., 2009). Dry sea salt particles may not be detected as sea salt, but misinterpreted as a different aerosol type with a higher lidar ratio, which leads to an overestimation of the extinction coefficient.

This study aims to show how spherical and cubic-like sea salt particles can be separated by remote measurements with lidar. The change in shape characteristics of marine aerosol can be easily observed with polarization lidar **(Murayama et al., 1999; Sakai et al., 2000; Sugimoto et al., 2000)**. When equipped with water vapor and nitrogen Raman channels the lidar delivers profiles of specific humidity. Combined with regularly available temperature profiles from radiosondes or models, **our Raman lidar observations provide RH** together with the depolarization ratio and can thus carefully measure changing particle shape effects with RH, as a function of height. The potential of using Raman lidar to study the hygroscopic growth of aerosol particles was demonstrated by Veselovskii et al. (2009) for summer haze at the East coast of the United States. Granados-Muñoz et al. (2015) observed the decrease of depolarization ratio of marine aerosol mixtures with increasing RH over Southern Spain.

Sea salt crystallization and deliquescence was observed with a triple-wavelength polarization lidar (Haarig et al., 2017) in the marine boundary layer over the remote tropical Atlantic in the absence of any disturbing anthropogenic impact and lofted Saharan dust layer. In this way, lidar can provide valuable information on the state of the marine aerosol layer from the point of view of optical properties. **The results are presented and discussed in Section 4.1.**

By performing triple-wavelength polarization lidar observations, we provide combined information on the shape/size characteristics of marine aerosol ensembles for the modeling community dealing with the optical properties of irregularly shaped mineral dust and sea salt particles (Gasteiger et al., 2011; David et al., 2013; Kemppinen et al., 2015a). Besides the depolarization ratio of the particles, we deliver extinction-to-backscatter ratios (lidar ratios) in addition which are also sensitive to changing particle properties.**The results are presented in Section 4.2.**

RH ranged from 40% **to** more than 80% in the marine aerosol layer. Scattering enhancement factors with RH are measured under these atmospheric conditions. The enhanced backscatter coefficient depicts the hygroscopic growth of the particles in terms of changing optical properties. For pure marine aerosol the hygroscopic growth factors and thus **the backscatter and extinction enhancement is larger compared to cases with mixtures of marine and continental particles (Zieger et al., 2013). This will be presented in Section 4.3.**

At the beginning **(in Section 2),** we will give an introduction to sea salt aerosol under dry and humid conditions and show examples of sea salt particles collected above Barbados. The lidar measurements in the framework of the Saharan Aerosol Long-range Transport and Aerosol-Cloud interaction Experiment (SALTRACE) (Weinzierl et al., 2017) will be described briefly **in Section 3**. Then we will present our observations with a special focus on the phase transition from spherical solution droplets to crystalline sea salt particles and the scattering enhancement factor (Section 4). In the discussion **(Section 5),** we compare our results to model calculations.

2 Sea salt under dry and humid conditions

The oceans are the source of marine aerosol, which consists mainly of sea salt and organic compounds from the sea surface (Gantt and Meskhidze, 2013). Marine aerosol from sea spray acts as cloud condensation nuclei and ice nucleating particles over the ocean where other types of particles are rare (DeMott et al., 2016; Kristensen et al., 2016). Marine aerosols are found in a wide size range from nanometer (10^{-9} m) up to several micrometer (10^{-5} m) (Bates et al., 1998; Dadashazar et al., 2017). The Aitken mode (diameter < 70-80 nm) is dominated by organic material emitted from the ocean. New particle formation dominates the accumulation mode (80–300 nm) and the larger particles are mostly sea salt (e.g., Wex et al., 2016). The sea salt particles may contain some organics on its surface (Middlebrook et al., 1998; Facchini et al., 2008; Laskin et al., 2012; Tervahattu et al., 2002). Size-resolved studies of marine aerosol (up to 2.5 μm) have been performed on Barbados by Wex et al. (2016). They found that the sea spray mode (>300 nm) contributes 4–10% to the total number. The number size distribution is dominated by the Aitken mode. But the surface area and the volume size distribution are dominated by sea spray **(90% of the total mass and volume)** with negligible contribution of the Aitken mode, resulting in a bimodal size distribution (Wex et al. (2016), and personal communication with H. Wex). For the radiation studies as for lidar measurements the surface area and its bimodal distribution is decisive. In this study, we will focus on sea salt and its different shapes, which mainly forms the coarse or sea spray mode. The accumulation mode (later on called fine mode) consists of sea salt particles and newly formed organic particles. Marine aerosol contains sea salt and organic compounds. Sea salt is the dominant component of the coarse mode and dominates the optical properties. Therefore most of the following investigations concern the sea salt and its changes with RH.

3

Crystalline sodium chloride has a cubic shape, sulfates form frequently needle-like shapes (Kandler et al., 2007). With different compounds present in sea-salt, mixed particle geometries can occur (Wise et al., 2007). With increasing RH the hygroscopic material takes up water, deliquesces and forms a spherical solution droplet. In humid marine environments, sea salt particles are of spherical shape. Consequently sea salt exists in two shape modes, spherical and non-spherical crystalline.

The shape and thus the depolarization ratio is not only dependent on RH, but also on the chemical composition **and the rate at which the particles have dried (Wang et al., 2010),** leading to different crystalline shapes. NaCl is the major component of sea salt, but other salts such as Na_2SO_4, $MgCl_2$ and $MgSO_4$ and eventually some organics are part of atmospheric sea salt (Tang et al., 1997). These components prevent the perfectly cubic shape of dried sea salt **(Zelenyuk et al., 2006; Zieger et al., 2017)**.

In Fig. 1, the deliquescence behavior of a pure sodium chloride particle is shown. It was observed with a scanning electron microscope (SEM). The **cubic sodium chloride** particle grows with increasing RH. Once the deliquescence point (in this case at RH of approximately 75%) is reached, it turns into a droplet. The studies of Tang et al. (1997) found the deliquescence point for sea salt at 70–74%, depending on the composition of the sea salt. The particles keep the spherical shape until RH decreases to 45–48% (Tang et al., 1997). As a result of the hysteresis effect, sea salt particles may exist in both shape modes between approximately 50% and 70% RH, depending on their individual history. These conditions may occur quite often over the oceans and coastal areas, as illustrated by Kandler et al. (2011). The hysteresis behavior has been previously studied (Carrico et al., 2003; Wise et al., 2005).

Atmospheric samples of dry salt particles (Fig. 2) were collected during the SALTRACE-1 campaign (Barbados, summer 2013), **whereas the case studies shown later in this publication are for pristine marine conditions observed during February 2014.** The samples **in Fig. 2** are taken in the dust layer (2–4 km asl.)**, which was present during the summer months, but not during the winter months, when our observations of dry marine particles took place. The samples in Fig 2 were taken in the free troposphere** to ensure that the particles have been dried in airborne state to be representative for atmospheric aerosol, in contrast to wet collection followed by drying on a substrate, which might lead to substrate effects. The four dry marine particles shown in Fig. 2 were collected at RH between 28% and 39%. The X-ray fluorescence spectroscopy (XRF) pictures reveal, that particle (b) with a shape closer to a cube has a negligible contribution of sulfate, whereas particle (d) is more spherical in shape and has a larger contribution of sulfate. Also, organics **of marine origin** might be the reason for the more spherical shape **(Laskin et al., 2012)**. Particle (c) shows the outline of a **spherical marine aerosol** droplet, so this one was probable still deliquesced at the time of collection. Overall we see that sea salt particles have a non-spherical shape that could be approximated by a cube for RH below 40%. But the shape is not perfectly cubic as for pure sodium chloride (see Fig. 1). The edges of the sea salt particles dried in the atmosphere are smoother. In **this publication** we will call the shape of crystalline sea salt 'cubic-like' to separate it from the spherical sea salt droplets under humid conditions. Compared to model results of perfectly cubic particles or pure NaCl salt particles investigated in the laboratory, we should measure lower depolarization ratios for dried marine aerosol in the atmosphere, because of the smoothed cubic-like shape.

For spherical marine particles a low particle linear depolarization ratio of 0.03±0.01 at 532 nm **prevails** (Groß et al., 2013). The range of depolarization values given in the mentioned publication is 0.01 – 0.11. This indicates that not all cases classified as marine aerosol consisted of spherical sea salt particles.

Field studies on dried marine particles are very rare. First evidence of an enhanced depolarization ratio for dry marine particles was reported by Murayama et al. (1999) based on lidar measurements in Tokyo. A clear separation from a potential dust influence was not possible. They observed a peak in the particle linear depolarization ratio of 0.1 at 532 nm during sea breeze.

Sakai et al. (2000) **measured** RH and the depolarization ratio over Nagoya, Japan, from 1994–1997 with a Raman lidar. They found 532 nm particle linear depolarization ratios between 0.1 and 0.2 for 25– 45% RH at heights from 2–4 km in the free troposphere. Backward trajectories indicated pure marine conditions. These values are in good agreement with our observations. **However, it cannot be excluded that continental particles were present as well and contributed to the light depolarization**

In a laboratory study, Sakai et al. (2010) measured the particle linear depolarization ratio (PLDR) of spherical sea salt particles of 0.01±**0.001 at 532 nm**. For crystalline sea salt particles, they found a PLDR of 0.08±**0.01**. Pure crystalline NaCl has a significantly higher depolarization ratio (PLDR=0.21±**0.02**) than atmospheric sea salt (Sakai et al., 2010). In a laboratory study, Järvinen et al. (2016) observed pure NaCl particles with a depolarization ratio of 0.25.

Additionally to the change in particle shape, the size of sea salt aerosols changes with RH. Due to water uptake sea salt aerosols are much larger under humid conditions and smaller under dry conditions. The process is known as hygroscopic growth (e.g., Zieger et al., 2013; Skupin et al., 2016). The change in optical properties **(backscatter coefficient, extinction coefficient, lidar ratio)** with varying RH can be measured with a Raman lidar. **Results are shown in Section 4.3.**

Early discrete dipole approximation (DDA) modeling attempts of spherical and cubic sea salt particles have been done by Murayama et al. (1999). For cubic particles larger than 0.8 μm a PLDR of 0.08-0.22 at 532 nm was predicted. David et al. (2013) used a T-matrix approach for cubic sea salt particles to model the depolarization ratio (approx. 0.16 in the visible and UV) and the lidar ratio (19 sr in UV and 20 sr in the visible). The DDA approach for cubic particles including surface roughness (Kemppinen et al., 2015a) leads to a PLDR 0.1–0.2 and a lidar ratio of 15–20 sr for the particle radius equal to the wavelength (size parameter = 6). Our observations **will also be compared with recently performed model calculations (Section 5).**

3 Methods

3.1 The SALTRACE project

The three SALTRACE field campaigns in 2013 and 2014 are the final observational efforts of the long-term SAMUM-SALTRACE project (Heintzenberg, 2009; Ansmann et al., 2011; Weinzierl et al., 2017). During SALTRACE, we investigated the Saharan dust properties after an atmospheric travel over 5–15 days and 5000–8000 km (Weinzierl et al., 2017; Haarig et al., 2016a, 2017). In the summer seasons of 2013 and 2014 (SALTRACE-1 and SALTRACE-3 in June-July), aged dust layers were observed. To investigate aged mixtures of dust and biomass burning smoke after long-range transport, we performed an ad-

ditional campaign in February-March 2014 (SALTRACE-2, winter transport regime). In February 2014 there was a period without aerosol transport from Africa, resulting in very clean marine conditions over Barbados. The SALTRACE lidar activities were complemented by shipborne lidar observations along the main Sahara dust transport route over the tropical North Atlantic in April-May 2013 (Kanitz et al., 2013; Rittmeister et al., 2017; Ansmann et al., 2017).

5　The ground-based remote sensing station was deployed at the Caribbean Institute for Meteorology and Hydrology (CIMH) in Husbands, north of the capital Bridgetown at the west coast of Barbados (13.15°N, 59.62°W, 110 m about sea level). The BERTHA lidar system (Backscatter Extinction lidar Ratio Temperature Humidity profiling Apparatus), an AERONET sun photometer (see AERONET web page **https://aeronet.gsfc.nasa.gov/**, Barbados_SALTRACE site), and a Vaisala radiosonde station (RS92 for profiling of pressure, temperature, RH, and the vector of the horizontal wind component) were operated at
10　the field site. A second AERONET station (Ragged Point) is located at the east coast of Barbados, approximately 20 km away from the CIMH.

3.2　Triple-wavelength lidar BERTHA

The multi-wavelength polarization Raman lidar BERTHA of the Leibniz Institute for Tropospheric Research (TROPOS) is a container-based mobile lidar system. As a unique feature, it enables the measurement of the depolarization ratio at three
15　wavelengths **(355, 532 and 1064 nm)** simultaneously. A more detailed description of the lidar system and the polarization characteristics can be found in Haarig et al. (2017). Currently it operates as a 3+2+3 lidar system (3 backscatter coefficients, 2 extinction coefficients and 3 depolarization ratios) with an additional water vapor channel (407 nm) and a high-spectral-resolution channel at 532 nm. It has been used in a 3+3+2 configuration in Haarig et al. (2016b). The signals are detected with a range resolution of 7.5 m and a time resolution of 10 s.

20　The particle backscatter coefficient gives information about the **aerosol** layers. For particles with sizes comparable to the wavelength or larger, it is in first approximation proportional to the surface area of the bulk of particles. The extinction coefficient is determined from the transmission of the laser beam through the atmosphere. Both are calculated independently from the lidar signals via the Raman method (Ansmann et al., 1992). The backscatter-to-extinction ratio, also called lidar ratio, contains information about the particle size and shape, as well as about the refractive index. Therefore it is used together with
25　the particle depolarization ratio to classify aerosol types (Burton et al., 2012; Groß et al., 2013). **The 532 nm channels reach full overlap at 800–1000 m height. The 355 nm channels would reach full overlap at 2500–3000 m. Therefore, they have to be overlap-corrected according to Wandinger and Ansmann (2002).** The uncertainty especially of the extinction and the lidar ratio is therefore larger in the UV.

The particle linear depolarization ratio (PLDR) is a measure of the depolarization caused by the scattering of linear polar-
30　ized light (defined as parallel) at atmospheric particles. It is defined as the ratio of cross polarized to parallel polarized light scattered back from aerosol particles. As an intensive parameter it is characteristic for a certain aerosol type. For spherical particles (droplets, wet marine particles) the PLDR is <0.03, whereas non-spherical particles have a higher PLDR (dust approx. 0.3 (at 532 nm), ice crystals approx. 0.5) (Groß et al., 2013; Haarig et al., 2017). To ensure the good quality of the depolarization measurements, a $\Delta 90°$ calibration (Freudenthaler et al., 2009) was performed for each measurement. In the UV the

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systematic uncertainties are quite high **(0.01 in the volume linear depolarization ratio)**. The calculation of the depolarization ratio follows Freudenthaler (2016) and is described in Haarig et al. (2017), where a detailed error estimation can be found **in addition**.

For the calculation of RH, the temperature profile of the radiosonde is used. BERHTA measures the pure rotational Raman signals from nitrogen and oxygen from the 532-nm-emission wavelength to retrieve the temperature profile, but the uncertainty is too large to retrieve RH with a reasonable uncertainty (Mattis et al., 2002). During the SALTRACE campaign, a Vaisalla RS92 radiosonde was launched for each measurement. The water vapor mixing ratio of the radiosonde is used to calibrate the water vapor mixing ratio derived by the ratio of the hydrogen (407 nm) and nitrogen (387 nm) Raman signal of the lidar (Whiteman et al., 1992). Due to the weak 407 nm signal, the technique can be used at nighttime only. By using the water vapor mixing ratio profiles of the lidar and the temperature and pressure profile of the radiosonde the temporal and vertical evolution of RH can be derived. The relative error of the water vapor mixing ratio caused by calibration and signal noise was < 5% at all heights **within the aerosol layer (in these cases up to 2 km)**. The temperature of the radiosonde is used for the two hours of measurement, so an uncertainty of 1 K is reasonable. These errors lead to a maximal relative uncertainty in RH of 12%, **resulting in a** dry RH of 40±5%. A detailed error estimation for RH derived with a Raman lidar can be found in Mattis et al. (2002).

3.3 The DDA model for cubic sodium chloride

We simulate optical properties of dry sea salt particles as cubes with the refractive index m of sodium chloride, provided by Eldridge and Palik (1985), i.e. with values m=1.582 at λ=355 nm, m=1.549 at λ=532 nm, and m=1.531 at λ=1064 nm. **The size distribution is taken from the corresponding AERONET measurement. The version 2 inversions for spherical particles are used (Holben et al., 1998).**

A range of volume-equivalent particle radii up to 2 μm is covered by modeling with the Discrete Dipole Approximation code ADDA (Yurkin and Hoekstra, 2011) with logarithmically equidistant size steps of a factor of 1.1. We use the DDA formulation 'filtered coupled dipoles' (Piller and Martin, 1998) included in ADDA, which was also used for example by Gasteiger et al. (2011), and use 8 dipoles (dpl) per wavelength. To simulate random particle orientation, DDA runs for 100 orientations were carried out for each particle. The weighted distributions of particle orientations were selected according to those presented by Sloan and Womersley (2004). For each DDA run, the optical properties were averaged over 64 scattering planes rotated around the incident light direction. **The single particle properties are used in a subsequent step for the calculation of the bulk optical properties (see Sect. 5)**.

To estimate the accuracy of these model simulations, the scattering problems were modeled **in addition** with settings associated with higher accuracy, i.e. one case with increased number of dipoles per wavelength (dpl=12 instead of 8) and another case where we increased the number of orientations from 100 to 225.

The uncertainty is estimated for the lidar ratio of single randomly-oriented particles to be on the order of ±10% and for the linear depolarization ratios about ±0.02. A similar uncertainty is estimated for the final optical properties of the bulk sea salt aerosol. The estimate for the bulk properties is based on considering that on the one hand the uncertainty is reduced by averaging

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over the size distribution but on the other hand also increased due to the fact that the assumed size distribution, particle shape, and refractive index are given with an uncertainty.

4 Observations

The island of Barbados is ideal to observe pure marine conditions. It is the eastern-most island of the Caribbean and located in the trade wind zone with predominant wind direction from the east. In winter, the inner tropical convergence zone is shifted to the southern hemisphere, so the air masses originating from the African continent are transported to South America (e.g., Baars et al., 2011), leaving the Caribbean under marine influence.

The marine aerosol extends up to the strong trade wind inversion at around 2 km height (Marine Aerosol Layer - MAL). The **MAL** is defined by the predominance of marine aerosol. **It includes the convective marine boundary layer (MBL) and another residual layer of marine particles. The MAL reaches to the trade wind inversion height. The dust removal process in the MAL is very efficient as shown in Rittmeister et al. (2017). As there was no dust or other continental aerosol in the free troposphere above the MAL, it is very unlikely that dust reached Barbados during February 2014.**

During the SALTRACE-2 campaign (16 February – 8 March 2014), a layer of enhanced cross-polarized signal was observed for several days. Figure 3 shows **the period from** 23 – 25 February 2014. **A strong decrease in RH from 80% to less than 10% at the trade wind inversion height (MAL top) was continuously observed between 20 and 25 February 2014. The enhanced depolarization ratio corresponding to this RH decrease was found to be between 0.04 and 0.12 (at 532 nm).** The two night measurements of 23 and 24 February 2014 will be discussed in the following **section** to demonstrate how the decrease in RH leads to **an increase of the linear** depolarization ratio due to the change in the shape **properties** of the sea salt particles. Firstly, the observations during the two night measurements will be described. Then, the changes in shape and size with RH will be discussed **by** using the particle linear depolarization ratio and the particle backscatter coefficient, respectively.

4.1 Profile measurements of backscatter, depolarization, lidar ratio and RH

An overview of the **observations on 23 and 24 February 2014 are** given in Fig. 4 and 5. On the 23 February 2014, **the MAL was not well mixed.** RH decreased steadily from 80% at 250 m height to 35% at 1000 m height. **Above the 1000 m height level, RH increased again up to 80% at 1800 m height**. Then a fast decrease of RH (from 80% to less than 10%) occurred at the trade wind inversion **height** between 1850 and 2150 m height. **The strong decrease of RH at MAL top** was observed for most of the measurements under clean marine conditions in February 2014. The time-height display of RH is shown in Fig. 4c. The increased signal in the cross-polarized channel and the volume depolarization ratio at 1064 nm are shown in Fig. 4a+b. In parts with low RH, mostly between 1000 and 1600 m, the volume depolarization ratio is high indicating non-spherical particles.

On the 24 February 2014, only a thin layer of dried marine particles was observed. The MAL reached up to 2 km height (Fig. 5). The feature of interest is the enhanced 1064 nm cross-polarized signal in the upper 200 m of the aerosol layer. The radiosonde launched at 23:07 UTC (19:07 local time (LT)) shows a strong temperature inversion of 4 K **within 200 m around**

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2000 m height (**trade wind inversion height**). RH was about 65% to 80% **throughout the MAL and** decreased to values of about 5% 200 m **above the** MAL. RH (Fig. 5c) indicates that the **environmental conditions remained unchanged during the** two hours average (18:11-20:20 LT). Marine particles lost their spherical shape at the top of the marine aerosol layer **(efflorescence)**. This caused an enhanced depolarization ratio, as can be seen in Fig. 5b.

5   For the further discussion it is important to show, that **predominately** marine aerosol particles were present over Barbados **in the lowest 2000 m.** HYSPLIT backward trajectories (Stein et al., 2015; HYSPLIT, 2017) and AERONET (Holben et al., 1998; AERONET, 2017) observations are used to demonstrate **that pure marine conditions were given.**

The ensemble of 7-days backward trajectories for 23 February (Fig. 6a) indicate marine sources over the Atlantic for the air mass, with only a **rather small chance of aerosol uptake over** West Sahara. At the 24 February the ensemble of 7-days 10   backward trajectories **(Fig. 6b)** shows only marine aerosol sources for the MAL.

In Fig. 6, only one height level (1200 m) is considered for the MAL. A more sophisticated analysis of the ensembles of trajectories at different heights is shown in Fig. 7. To estimate the contributions of marine and continental aerosol sources to the observed air masses, ensembles of HYSPLIT backward trajectories (Stein et al., 2015) were calculated in vertical steps of 500 m. Each ensemble consists of 27 single trajectories which are initialized with a small spatial offset.
15   **An air parcel is assumed to get laden with aerosols over a given marine or continental source region when the parcel is at heights below 2 km above this region. The colors in Fig. 7 indicate the contribution of different land cover categories, taken from MODIS (Friedl et al., 2002).** Considering the ensembles of trajectories at different heights below 2000 m for both nights (Fig. 7, **only the night of 23 February shown**), none of the 10-day back trajectory **crossed a continental site at heights below 2 km. This analysis clearly indicates that our lidar observations were performed at pure marine aerosol**
20   **conditions.**

The measurements of the AERONET sun photometers at Ragged Point and at the field site (Barbados_SALTRACE) are shown in Fig. 8 (Holben et al., 1998; AERONET, 2017) . The optical thickness (AOT at 500 nm) is low **and accumulates around 0.05. Very low Ångström exponents were found and indicate the dominance of coarse mode particles. In our cases the Ångström exponents at the closest AERONET observations (after 20:00 UTC) are between 0.1 and 0.2. The**
25   **fine mode fraction (FMF) is also typical for coarse mode dominated particle ensembles.**

The transport of dust over 5000–8000 km below the trade wind inversion is extremely unlikely. The dust particles will get lost due to turbulent downward mixing or gravitational sedimentation or wet deposition. The observations of Rittmeister et al. (2017) support the conceptual model (Karyampudi et al., 1999), which implies a very efficient removal of dust from the MAL. Dust is transported over long distances in the Saharan air layer (SAL) above the trade wind
30   **inversion (above the MAL). But in late February 2014, the SAL was not present over the MAL. In conclusion, all air mass back trajectory studies and the AERONET observations indicate clean marine conditions over Barbados during 23 and 24 February 2014.**

The vertical profiles of the particle backscatter coefficient and the PLDR for the three wavelengths (355, 532 and 1064 nm) of the BERTHA lidar system are shown in Fig. 9 and 10. A good agreement is achieved between RH measured with the radiosonde
35   and the **30-minute (2-hour) mean** RH profile retrieved from the lidar observations. The profile of RH **in combination with**

the profiles of the backscatter coefficient and the PLDR **indicate the sensitive changes in the marine optical properties with changing RH.** The crystallization point for sea salt of 45-48% RH (Tang et al., 1997) is marked with a dashed line. The depolarization ratio increases strongly when RH drops below this point. The change of the backscatter coefficient with RH is less pronounced. When RH decreases below the crystallization point at 810 m height, the particle linear depolarization ratio

5   **starts to increase and takes its maximum of** 0.148±0.035 at 532 nm at around 1150 m height. RH increases again **between 1000 m and 1800 m and reaches 70–74% (deliquescence point of sea salt) at 1780 m.** The PLDR decreases below 0.02 (532 nm) at this height. This behavior will be further discussed in Section 4.2. The second decrease in RH at the trade wind inversion height leads to a less pronounced peak in the PLDR (value ± systematic uncertainty) of 0.069±0.161 at 355 nm, 0.079±0.036 at 532 nm, and 0.063±0.018 at 1064 nm around 2000 m.

10   In the 2-hour mean profiles of the 24 February 2014 (Fig. 10, **100 m vertical smoothing is applied**), a maximum PLDR of 0.055±0.109 (355 nm), 0.068±0.035 (532 nm), and 0.038±0.010 (1064 nm) is reached in the thin layer of dried marine aerosol at MAL top. In the humid marine aerosol layer below, the PLDR is below 0.02 **and thus clearly indicates the spherical shape of the sea salt particles.** The profile of RH shows the coincidence of the decrease of RH with the increase of PLDR.

The Raman lidar method allows us to derive the extinction coefficient independently of the backscatter coefficient and

15   therefore to measure the lidar ratio (extinction-to-backscatter ratio), shown in Fig. 11. In the humid MAL on 24 February the lidar ratio is **19±5 sr** at 355 nm and 23±2 sr at 532 nm, which is typical for **spherical marine particles** (Müller et al., 2007; Groß et al., 2013; Rittmeister et al., 2017). For the aerosol layer with dried marine particles on 23 February (1000-1600 m height), we obtained a lidar ratio of **27±6 sr** (at 355 nm) and 25±3 sr (at 532 nm) for the cubic-like sea salt particles. **The increase in the lidar ratio may be explained by the reduction in particle size, as the sidewards scattering (extinction**

20   **without absorption) increases for smaller particles. The results are summarized in Table 1 for dry and humid conditions. There, the results of our modeling efforts presented in Section 5 are shown as well for comparison. For cubic sea salt particles we simulated lidar ratios of 13, 20, and 36 sr at 355, 532, and 1064 nm, respectively.** David et al. (2013) simulated with the T-matrix approach a lidar ratio of 19 sr and 20 sr at 355 nm and 532 nm for cubic sea salt particles.

4.2   Observation of the phase transition in the depolarization ratio

25   **A unique opportunity is provided to study the relation between marine particle shape and RH with high temporal (5 min) and vertical (50 m) resolution under atmospheric conditions.** The correlation between PLDR and RH for the 23 February 2014 is shown in Fig. 12d-f. Only the decrease in RH is depicted here (375 – 1100 m height range). The crystallization point (45–48% RH) is more important than the deliquescence point (70–74% RH), **when the drying process is highlighted. Inorganic sea salt has multiple crystallization points due to its complex composition (Tang et al., 1997;**

30   **Zieger et al., 2017). The drying follows the upper branch of the hysteresis, as fully deliquesced sea salt particles are drying.** From 80 to 50% RH the PLDR increases slightly with decreasing RH, but remains ≤ 0.02 (532, 1064 nm) and ≤0.03 (355 nm, due to higher noise level). This observation is in line with typical depolarization values for marine aerosol as used in aerosol classification schemes (e.g., Groß et al., 2013). At around 50% RH the PLDR increases drastically indicating a significant change in particle shape from spherical to cubic-like shape. The PLDR reaches maximum values (with sys. uncertainty) of

about 0.12±0.08 (355 nm), 0.15±0.03 (532 nm), and 0.10±0.01 (at 1064 nm) at RH of around 40%. After the phase transition from spherical sea salt droplets to cubic-like sea salt crystals, the depolarization ratio remains at the high level **even for RH close to 35%**. A further drying was not observed **in the atmosphere** yet, so we can only speculate about the depolarization ratio for sea salt particles under very dry conditions (0-35% RH).

Furthermore the measurement of the 23 February 2014 contains information about the humidification process. Between 1100 and 1800 m height RH is increasing again up to 80%. **Figure 13 contrasts the PLDR dependence on RH during humidification and drying.** The sea salt keeps the cubic-like shape causing an enhanced depolarization ratio **even at RH close to 60%. The depolarization values then** slowly decrease to values below 0.02 typical for spherical sea salt particles (above around 65-70% RH). The hysteresis effect leads to the existence of both shape modes between 50% and 70% RH.

4.3 Scattering enhancement factors of pure marine aerosol

Beside the change in particle shape, the particle size is changing with decreasing RH. Sea salt particles grow by water uptake when RH increases. The particle backscatter coefficient is proportional to the surface area of the scattering particles and therefore a good indicator for particle growth. **The Ångström exponents (AE, not shown) increase monotonically between 1.0 and 1.5 km (extinction AE, lower limit due to overlap) and between 0.4 and 1.5 km (backscatter 532/1064 AE). This is a clear indication for the decrease in particles size.** In Fig. 12a-c the particle backscatter coefficient β is plotted against RH for the height interval 375–1100 m for the three wavelengths (λ= 355, 532, 1064 nm), **measured with a vertical resolution of 50 m and temporal resolution of 5 min on 23 February 2014**.

The backscatter enhancement factor $f_\beta(RH,\lambda)$ is calculated:

$$f_\beta(RH,\lambda) = \frac{\beta(RH,\lambda)}{\beta(40\%,\lambda)}. \tag{1}$$

Under atmospheric conditions it is hardly possible to **observe completely dry marine particles** (RH<10%). Therefore the backscatter coefficient at 40% RH was chosen as reference value to normalize the data, **as it is common in the literature (see discussion in Skupin et al., 2016)**. Lower values than 35% RH were not accessible under the measurement conditions over Barbados. RH of 40% is below the crystallization point, so sea salt particles should **not be affected by the hysteresis any more, i.e. significant shrinking at further decreasing RH should not be the case. Nevertheless, the sea salt particles are not completely dry at 40% RH. Laboratory studies (Tang et al., 1997; Zieger et al., 2017) show an increase of the enhancement factor of 20–30% between 0% RH (f=1.0) and 40% RH (f≈1.2–1.3).** Other lidar-based atmospheric studies used 40% or 60% as reference RH (Granados-Muñoz et al., 2015; Veselovskii et al., 2009) since it was the lowest value found in their atmospheric measurements. **In the following we will use the expression 'dry' when we refer to RH of 40%, keeping in mind, that sea salt is not completely dry, but below the crystallization point.**

To parametrize the backscatter enhancement factor, **we follow** Kasten (1969) and Hänel (1976):

$$f_\beta(RH,\lambda) = A * (1 - RH/100)^{-\gamma}. \tag{2}$$

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The parameter A gives the extrapolated value at 0% RH and the exponent γ **describes the hygroscopic behavior of the particles**. This parametrization (sometimes with $A=1$ for starting at 0% RH) has been used by various investigators (Carrico et al., 2003; Veselovskii et al., 2009; Skupin et al., 2016). **Retrieving values of $A<1$ (Table 2) for the upper branch of the hysteresis, clearly indicates that the reference RH of 40% RH for the backscatter coefficient is too high, so that the sea salt particles are not completely dry.**

The backscatter enhancement factors are shown in Fig. 14 with the parametrization according to Eq. (2). **The fit is shown on a linear scale (Fig. 14a) and on a log-log scale (Fig. 14b). The log-log plot shows that the parametrization does not hold for the lowest RH.** The fit parameters are listed in Table 2. RH has fairly reached 80% in the used height interval. $f_\beta(75\text{–}80\%)$ is the averaged value between 75% and 80% RH. Additionally, $f_\beta(80\%)$ was calculated by extrapolating the fit. Both values of the backscatter enhancement factor with respect to the reference RH of 40% can be found in Table 2. **By taking a dry value at 40% RH, we underestimate the scattering enhancement factor by approximately 25% (Titos et al., 2016).** The extrapolated values **are a lot higher than** the measured values at 75–80% RH. For the further discussion we will only use the measured enhancement factors. The error of the backscatter enhancement factor results from the standard deviation of the mean values at 40% and 75–80% RH **and the uncertainty introduced by the error in RH.**

For a better comparison to reported literature values, we convert our backscatter enhancement factors f_β to extinction enhancement factors f_α by means of the backscatter-to-extinction ratio (lidar ratio S), which was measured for wet and dry marine particles (see Table 1).

$$f_\alpha(RH, \lambda) = \frac{S_{\text{wet}}}{S_{\text{dry}}} f_\beta(RH, \lambda). \tag{3}$$

The extinction enhancement factor **is commonly used in studies of the dependence of particle optical properties on RH (Kotchenruther et al., 1999; Zieger et al., 2010; Skupin et al., 2016)**. For 1064 nm the **simulated lidar ratios (Table 1)** have to be used. The errors of the lidar ratios are included in the error of f_α **by Gaussian error propagation. Overall, the relative error of extinction enhancement factor is approximately 50% in the UV and 30% for the other wavelengths.**.

The **measured** extinction enhancement factors of pure marine aerosol range from **1.94 ± 0.94 (at 355 nm), to 3.70 ± 1.14 (at 532 nm) and 5.37 ± 1.66 (at 1064 nm)**. A clear wavelength dependence is given. Qualitatively the same wavelength dependence of f_α was observed by Kotchenruther et al. (1999) for the wavelengths 450, 550 and 700 nm.

By assuming a high single scattering albedo (ratio of scatter to extinction coefficient) for marine particle (Anderson et al., 1999; Takemura et al., 2002), the scattering and extinction enhancement factors should be almost equal. The scattering enhancement factors in the present study are in the upper range of reported literature values for marine aerosol. **This probably reflects the almost ideal marine conditions over Barbados during the winter season, when dust and pollution aerosol from Africa are at the lowest level and 5000 km upwind is only ocean.** Zieger et al. (2013) reported mean scattering enhancement factors f(85%, 550 nm) for sea salt of 2.28 at Mace Head, Ireland, 2.86 at Ny Ålesund, Svalbard, and 3.38 at Cabauw, Netherlands. **A review of mostly nephelometer derived scattering enhancement factors at around 550 nm is given in Titos et al. (2016), where the values for marine environments range between 1.5 and 3.5, depending on the amount of pollution. The hygroscopic growth of pure marine particles is strong compared to continental aerosol. The**

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7-year measurements in the Southern Great Plains, continental USA (900 km away from the ocean), yield a scattering enhancement factor f(85%, 550 nm) of 1.78 compared to a dry value of 40% RH (Jefferson et al., 2017). Skupin et al. (2016) found in a 4-year measurement period over Leipzig, central Germany (400 km away from the ocean), an extinction enhancement factor f(80%, 550 nm) of 1.75±0.4 compared to a dry value of 40% RH. For north westerly wind

5 **directions (from the North Sea) the enhancement factor was slightly higher at Leipzig (1.95±0.5).**

5 Comparison with optical modeling of sea salt cubes

The optical properties of cubic sodium chloride particles were modeled **by** using the Discrete Dipole Approximation (DDA). The model settings are described in Sect. 3.3. The lidar ratio and PLDR for each wavelength were modeled as a function of particle size (Fig. 15). If the particle diameter is **close to the laser wavelength** (λ) and smaller, the lidar ratio is high (up to

10 more than 100 sr). For particle diameters >2λ, the lidar ratio is 20±10 sr. **The lidar ratio decreases with further increasing particle size, e.g. down to 3.6 sr for the radius r=2 μm at 355 nm**. If the particle diameter is smaller than the detecting wavelength, the PLDR is very small. A significant depolarization is produced for particle diameters $\geq \lambda$. Then, values up to 0.27 are reached **however strongly varying with particle size.** In the atmosphere, we have always a distribution of particles sizes, **so that extreme optical effects are widely smoothed out**.

15 We consider the size distribution inverted by the AERONET algorithm (Holben et al., 1998) from measurements at Ragged Point on 23 February 2014 at 12:31 UTC (Fig. 16). It is version 1.5 only, but the inversion of that measurement resulted in the lowest residual error of the sky radiance on that day. We assume that the optical depth during that measurement was dominated by wet marine particles. To calculate the optical properties for dry marine particles we assume that their size is **a factor of 2 smaller** compared to the size obtained by AERONET **and assume that the lidar ratio and the linear depolarization ratio**

20 **for particles with r>2 μm is the same as for r=2 μm-particles.** The results are given in Table 1. **In the UV the simulated PLDR for the reduced radius agrees with the measurements. At 532 nm the model underestimates the PLDR, but it is still within the uncertainties of the lidar system. Whereas at 1064 nm, the model overestimate the PLDR. By using the dry radius size distribution (a factor of 2 lower radii) the agreement of the spectral slope of PLDR improves.**

The T-matrix results for cubic sea salt (David et al., 2013) are given for comparison (**Table 1**). **They agree with the max-**

25 **imum PLDR in the UV and visible (see Fig. 12d+e), whereas the 30-min mean PLDR (Table 1) are smaller. The simulations of cubic particles by Kemppinen et al. (2015a) are limited by the maximum size parameter (x=10). Taking the effective radius (1.033 μm) from the AERONET size distribution (Fig. 16) into account, the lidar ratio (18±2 sr) and the PLDR (0.15±0.05) can be determined at 1064 nm, only.**

Chamaillard et al. (2003) shows in a modeling study that a cubic shape assumption (DDA model) is necessary to

30 **reproduce the backscatter of dry marine particles. The modeled scattering coefficient could not reproduce the measurements no matter which shape is assumed.**

The modeled lidar ratios are smaller than the measured ones. **It is evident that the low modeled lidar ratios at 355 nm (Table 1) are due to the very low lidar ratios of particles r>1 μm (see Fig. 15a). Thus, the deviation between measurements**

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and the models is an indication that our applied size distributions contain too many large particles. The uncertainty in the lidar ratio for dried marine particles in the UV is high. **Nevertheless**, there is no large difference between wet and dry marine lidar ratios.

6   Conclusions

The phase transition between spherical sea salt droplets and cubic-like sea salt crystals has been observed under pure marine conditions over Barbados. The particle linear depolarization ratio, measured with a **triple-wavelength polarization lidar, significantly increased** when RH dropped below 50%. The combination of polarization and water vapor measurements with lidar offered the unique opportunity to study this behavior **with high vertical and temporal resolution.** The particle linear depolarization ratio in these dried marine layers was enhanced on 23 and 24 February 2014 **(0.05–0.12 at 355 nm; 0.07–0.15 at 532 nm and 0.04–0.10 at 1064 nm).** The systematic investigations of the depolarization ratio for dry marine particles showed maximum values (with systematic uncertainty) of 0.12 ± 0.08 (at 355 nm), 0.15 ± 0.03 (at 532 nm) and 0.10 ± 0.01 at 1064 nm.

We compared the optical properties for dry and wet marine particles at three wavelengths at 40% and 80% RH, respectively. Complete dry (0% RH) sea salt particles could not be found under atmospheric conditions. The extinction enhancement factor for the range 40 to 80% RH is **1.94 ± 0.94 (at 355 nm), 3.70 ± 1.14 (at 532 nm) and 5.37 ± 1.66 (at 1064 nm).** These results are given in Tables 1 and 2.

A layer of dried marine aerosol observed over Barbados in February 2014 probably often exists at the MAL top when dry free-tropospheric air mixes with humid air in the uppermost part of the MAL. Extended layer with dried marine particles, as observed on 23 February, may also occur frequently when the residual layer in the MAL is isolated from the convectively active MBL, and this residual layer entrains dry air from the free troposphere.

Satellite based studies, for example with CALIPSO or EarthCARE, would be helpful to assess the global occurrence of dried marine aerosol layers. **These changes in particle shape may have an impact on the Earth's radiative budget over the oceans and therefore should be studied with global atmospheric models. Although the impact of thin layers of dry marine particles may be low compared to the thicker layers of humid marine particles below.**

Assuming generally spheroidal shape for non-spherical particles causes errors in the case of marine observations. Inversion algorithms as used in AERONET (Dubovik et al., 2006) may be affected as well. For dry marine cases, we would suggest, that a cubic model could be included.

An enhanced depolarization ratio for dry sea salt particles (up to 0.15 at 532 nm) leads to an overestimation of dust in aerosol classification and separation schemes (Burton et al., 2012; Groß et al., 2013; Mamouri and Ansmann, 2014, 2017). So be careful in marine environments (decrease in RH in upper part of MAL) and at coasts (sea breeze effects and decrease in RH over land). Marine particles can be injected into the Saharan air layer (SAL) by convective cumulus convection, where RH is typically below 40% so that marine particles have a cubic-like shape. Mistyping of aerosol

layers will lead to wrong results in further retrieved products, such as the extinction coefficient, the mass concentration or the estimates of cloud condensation nuclei and ice nucleating particles (Mamouri and Ansmann, 2016).

The existence of cubic-like and spherical salt particles is known since a long time, but this study points out the atmospheric relevance. Cubic-like sea salt has been measured under atmospheric conditions. The two shape modes of sea salt (spherical and cubic-like) exist under atmospheric conditions over the ocean and should be considered in future aerosol studies.

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Table 1. Measurement and simulation of the lidar ratio and the particle linear depolarization ratio for wet and dry marine particles. **The depolarization ratio (100 m vertical smoothing) and the lidar ratio (500 m vertical smoothing) are measured around the indicated height, where the corresponding RH prevailed. The systematic uncertainties are given.** The lidar ratio at 1064 nm could not be measured in this configuration of BERTHA (see Haarig et al., 2016b). For the DDA simulation of spherical and cubic particles the AERONET size distribution (SD) from Ragged Point at 12:31 UTC on 23 February 2014 is taken. **The effective radius was 1.033 μm. In order to mimic dry marine particles, the radius of the size distribution was divided by a factor of 2.** For the wet marine particles a mixture of 7:1 parts of water to salt was used. The model uncertainties are described in section 3.3. The T-matrix results for a typical size distribution of sea salt (O'Dowd et al., 1997) are taken from David et al. (2013). The modeled uncertainties are extremely small, less than 1 for the lidar ratio.

Wavelength (nm)	**Lidar ratio** (sr) 355	532	1064	**Particle depolarization ratio** (%) 355	532	1064	Comments
Measurement							
Wet (RH=80%)	**19 ± 5**	23 ± 2	–	≤ 3.0	≤ 2.0	≤ 2.0	2014-02-24 22:11–00:21 UTC, <1800 m
Dry (RH=40%)	**27 ± 6**	25 ± 3	–	**11.5 ± 8.2**	**14.8 ± 3.5**	**9.9 ± 1.1**	2014-02-23 23:38–01:08 UTC, **1150 m**
Simulation							
Wet (Spherical)	22 ± 2	27 ± 3	35 ± 4	0	0	0	**Mie**, Aeronet SD 2014-02-23
Dry (Cubic)	10 ± 1	16 ± 2	**31 ± 3**	**8.8 ± 2.0**	**11.9 ± 2.0**	**14.2 ± 2.0**	DDA, Aeronet SD 2014-02-23
	13 ± 1	**20 ± 2**	36 ± 4	**10.8 ± 2.0**	**12.8 ± 2.0**	**12.0 ± 2.0**	DDA, Aeronet SD 2014-02-23, **radius/2**
	19	20	–	15.9 ± 0.1	16.2 ± 0.1	–	T-Matrix, sea salt SD[*], David et al. (2013)

[*] Size distribution from O'Dowd et al. (1997)

Table 2. The fit parameters for the backscatter enhancement factor according to Eq. (2). The backscatter enhancement factor $f_\beta(80\%)$ calculated with these fit parameters is compared to the measured factor $f_\beta(75\text{–}80\%)$ between 75% and 80% RH. The extinction enhancement factor $f_\alpha(75\text{–}80\% \text{ RH})$ was derived by Eq. (3) to compare with the literature.

| Wavelength	**Fit** A	γ	$f_\beta(80\%)$	**Measurement** $f_\beta(75\text{–}80\%)$	$f_\alpha(75\text{–}80\%)$
355 nm	0.532	1.077	3.01	**2.75 ± 0.94**	**1.94 ± 0.94**
532 nm	0.529	1.487	5.79	**4.03 ± 1.09**	**3.70 ± 1.14**
1064 nm	0.489	1.837	9.40	**5.52 ± 1.47**	**5.37 ± 1.66**[*]

[*] modeled lidar ratios used (S_{wet}=35 sr; S_{dry}=36 sr; Tab 1)
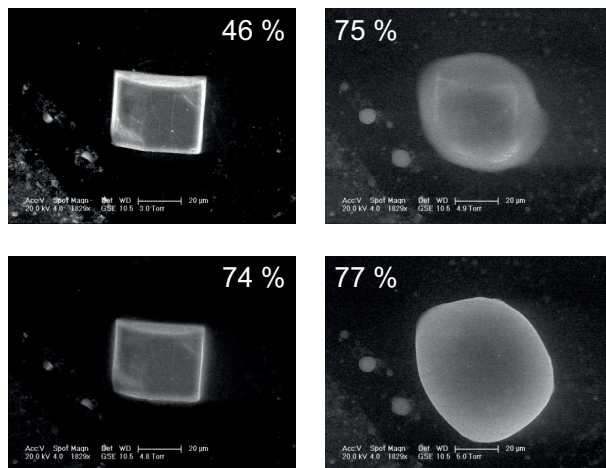
Figure 1. Sodium chloride deliquescence at 75% RH observed at laboratory conditions (at 4.9°C) **in an environmental SEM (Scanning Electron Microscope)**. The dry cubic particle with sharp edges at RH of 46% becomes surrounded by a liquid sphere when RH increases to 77%.
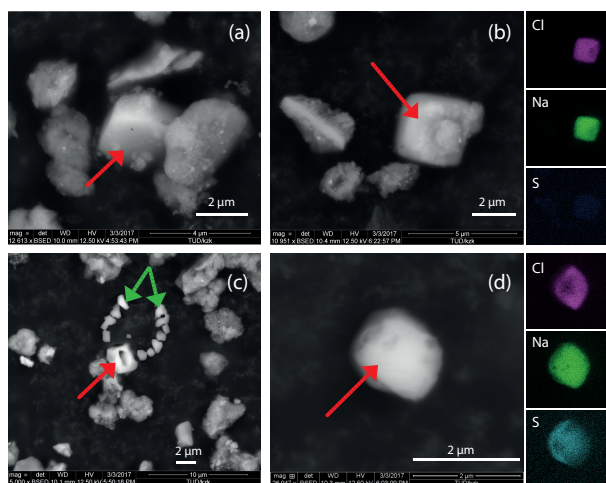
Figure 2. Images of dry atmospheric sea salt particles (red arrows) surrounded by Saharan dust particles, **collected during the summer transport regime (summer 2013), whereas the later shown case studies are from pristine marine conditions during February 2014.** The particles were sampled aboard the Falcon research aircraft (Weinzierl et al., 2017) at different heights over Barbados on (a) 21 June, (b) 10 July, (c) 26 June and (d) 30 June 2013 during SALTRACE-1. Sampling altitude and conditions are (a) 2560 m asl., 15°C, 29% RH; (b) 3550 m asl., 8°C, 28% RH; (c) 3570 m asl., 7°C, 39% RH; (d) 3230 m asl., 8°C, 34% RH. For the sea salt particles in (b) and (d) the XRF-Images are included showing that chloride (Cl) and sodium (Na) are the main components. The sulfate (S) component is negligible for particle (b) but significant for particle (d) which exhibits a more spherical shape. (c) shows an outline of a former droplet (green arrows), indicating a still (partial) deliquesced state during collection. The white bar in the bottom right corner indicates 2μm.
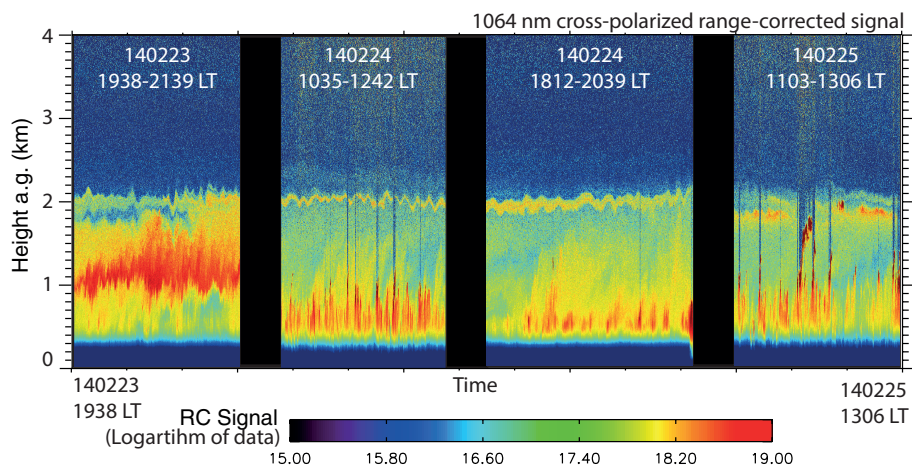
Figure 3. Three days of lidar observations (23–25 February 2014) of layers with dried marine particles in the marine aerosol layer (MAL). On 23 February (left panel), a vertically extended layer of dried sea salt particles (red area) occurred on top of the convective boundary layer. A continuously, 100–200 m thick layer with dried marine particles (yellow layer at MAL top) was present over the whole day on 24 February 2014, this layer was still present on 25 February. The convectively active MBL reaches to about 1 km on all three days and permanently pushed marine aerosol upward. The shown range-corrected 1064 nm backscatter signal (cross-polarized channel, **10 s temporal, 7.5 m vertical resolution**) is most sensitive to enhanced light depolarization produced by dry marine aerosol. LT: local time
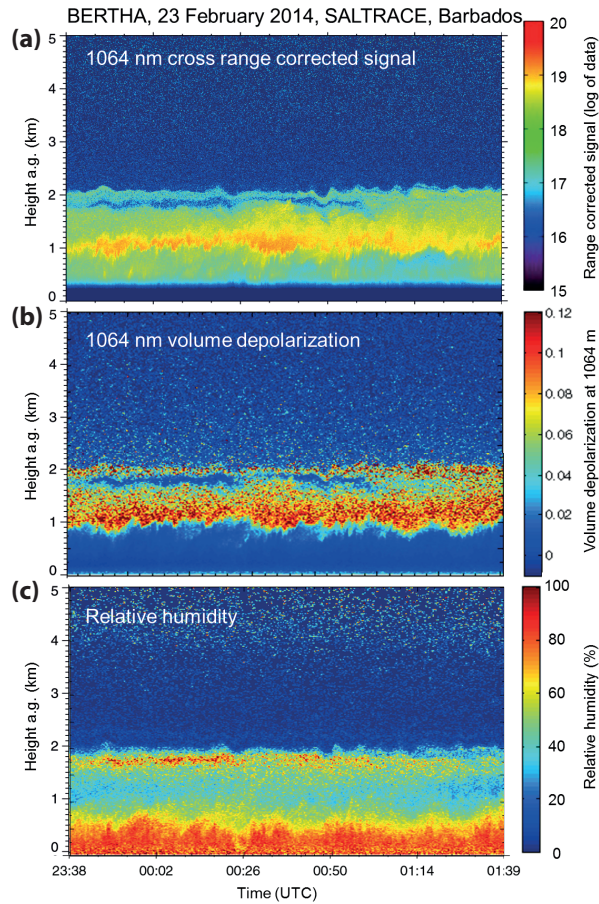
Figure 4. Marine aerosol layer (MAL) over Barbados on 23 Februray 2014, 19:38-21:39 local time with the cloudfree MBL reaching to 0.8-1 km height (indicated by a low depolarization ratio and high RH) and an extended layer between about 1 and 2.1 km height with dried marine sea salt particles causing enhanced light depolarization ((**b**), yellow-red areas) at low RH of <50% ((**c**), bluish areas). The cross-polarized 1064 nm signal ((**a**), **10 s temporal, 7.5 m vertical resolution**) highlights the layer with dried marine particles (yellow areas). **The depolarization ratio and RH are smoothed over 30 s and 22 and 52 m, respectively.**
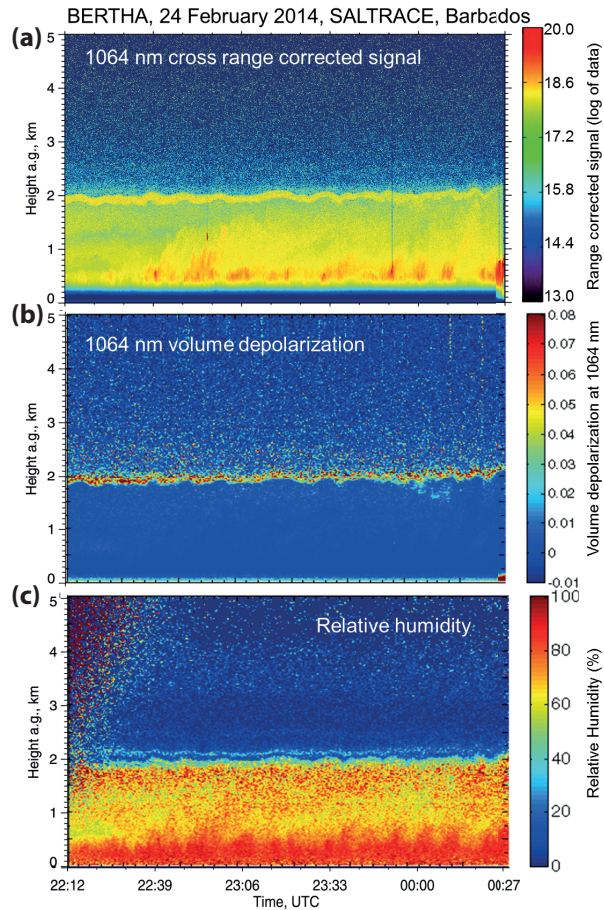
Figure 5. Same as Fig. 4, except for 24 February 2014, 18:12-20:27 local time. The MAL top is again close to 2 km height. The convective MBL reaches to about 800 m height and permanently pushes sea salt particles into the upper part of the MAL. RH is high throughout the MAL (and thus depolarization ratio caused by spherical, wet marine particles is low). Only at MAL top, dried marine particles cause a thin layer of enhanced cross-polarized signal ((**a**), **10 s temporal, 7.5 m vertical resolution**) and depolarization ratio (**b**). Daytime noise is visible in the RH (**c**) in the first half hour after sunset at 22:06 UTC. **The depolarization ratio and RH are smoothed over 30 s and 22 and 52 m, respectively.**

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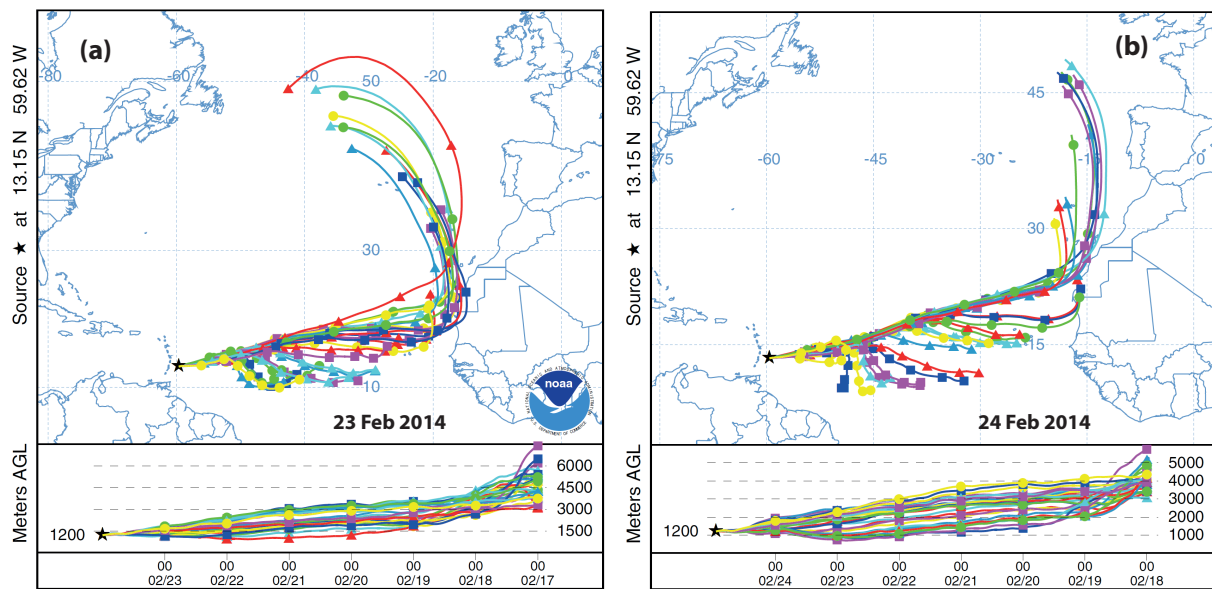
Figure 6. Ensemble of 7-day backward trajectories (HYSPLIT, 2017) for **(a)** 24 February 2014, 00:00 UTC, and **(b)** for 24 February 2014, 23:00 UTC, arriving at 1200 m over Barbados.
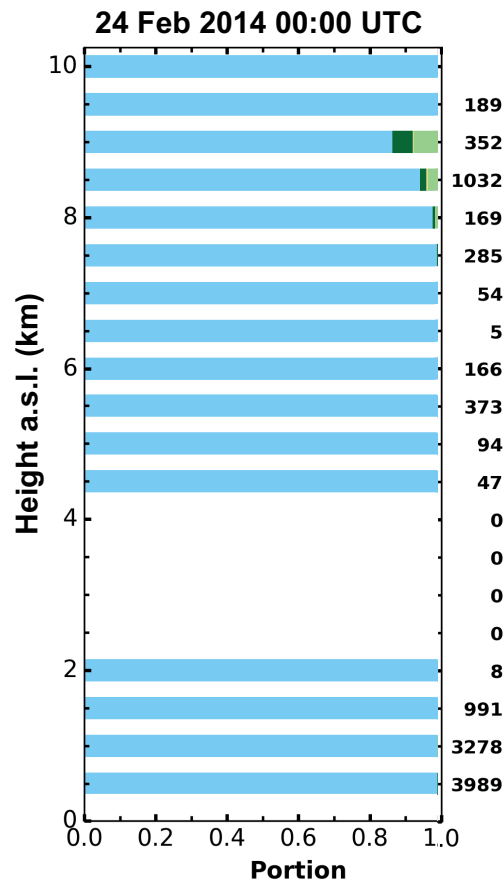
Figure 7. Relative residence time of the air masses over ground within the 10 days prior to their arrival over Barbados (24 February 2014, 00:00 UTC). For each height level (500 m) an ensemble of 27 HYSPLIT trajectories was calculated. The numbers on the right hand side indicate the total amount of hours which the ensemble of 10-day backward trajectories at each height level spend close to the ground (below 2 km height) and could be possibly laden with aerosols. The color bar indicates the portion of different surface areas, where blue stands for ocean, and green for different types of continental land cover (forest, grass, etc.). The land cover is taken from MODIS. Between 2.5 and 4.0 km height, the trajectories did not come close to the ground. The air masses in the MAL (up to 2 km height) are purely marine. The same result was found for 25 February 2014, 00:00 UTC (not shown).
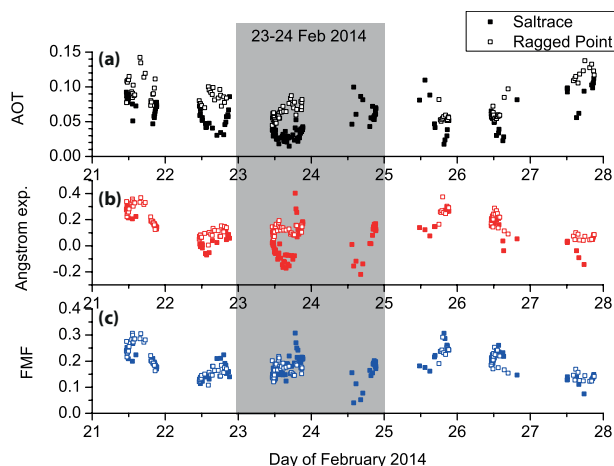
Figure 8. AERONET retrieval products ((**a**) aerosol optical thickness, **AOT,** at 500 nm, (**b**) Ångström exponent (440–870 nm) and (**c**) fine mode fraction, **FMF**) for 21–28 February 2014. AERONET level 2.0 data from Ragged Point and Barbados_Saltrace are shown. **The gray area highlights the time period of lidar observations discussed in this section.**
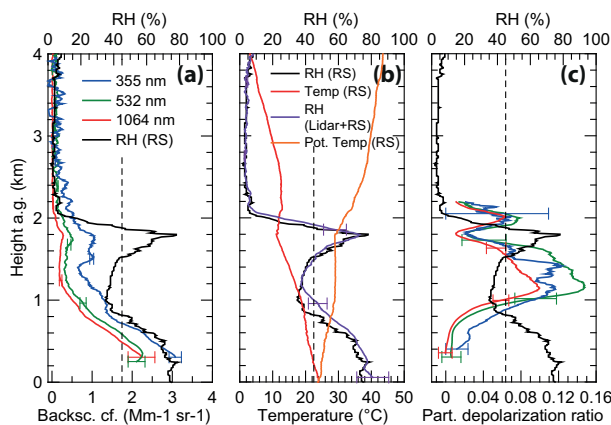

Figure 9. 30-minute mean profiles of particle backscatter coefficient (**a**) and particle linear depolarization ratio (**c**) at three wavelengths together with radiosonde RH (indicating the MAL up to 2 km height). The lidar observation was performed on 23 February 2014, 19:45-20:15 local time (23:45-00:15 UTC). **The atmospheric variability was low during the signal averaging period (Fig. 4)**. Panel (**b**) shows the potential temperature (T_{pot}, radiosonde, launch at 00:00 UTC), RH from radiosonde and from lidar (30-minute average). Note the sharp drop in RH from >70% (at 1850 m height) to <10% (at 2100 m height). The dashed line marks the sea salt efflorescence point (45% RH). Error bars indicate the lidar retrieval uncertainty. The vertical smoothing window length is 50 m for the backscatter and RH (lidar) and 100 m for the depolarization ratio.
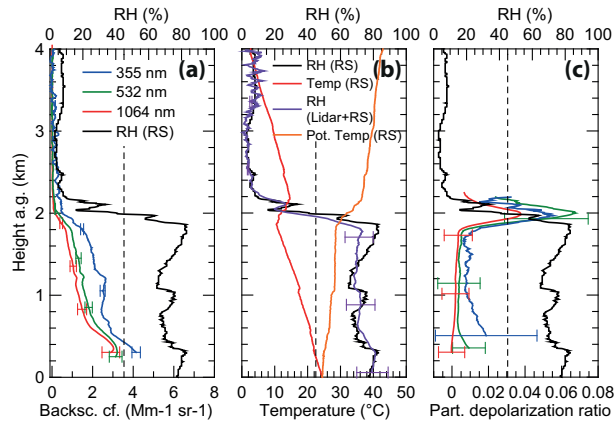
Figure 10. Same as Figure 9, except for 24 February 2014, 18:12-20:20 local time (22:12-00:20 UTC). In the central panel **(b)**, the respective 2-hour RH profile (from lidar) is shown together with the radiosonde profiles (launch at 23:07 UTC). The MAL was entirely humid on this day.
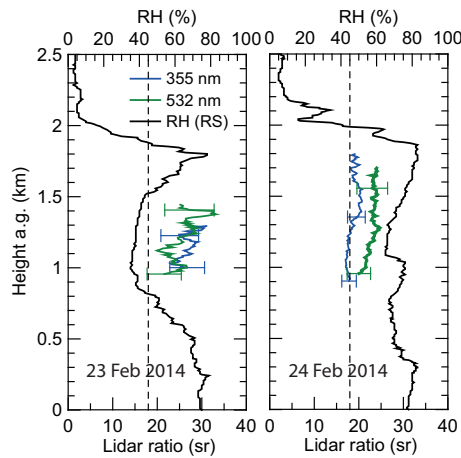

Figure 11. 2h mean profiles of the lidar ratio for the 23 February (left, 500 m vertical smoothing) and the 24 February 2014 (right, 750 m vertical smoothing). 532 (green line) without overlap correction, full overlap at 800–1000 m; 355 (blue line) with overlap correction. The RH profile of the radiosonde (black line) indicates the different layers.
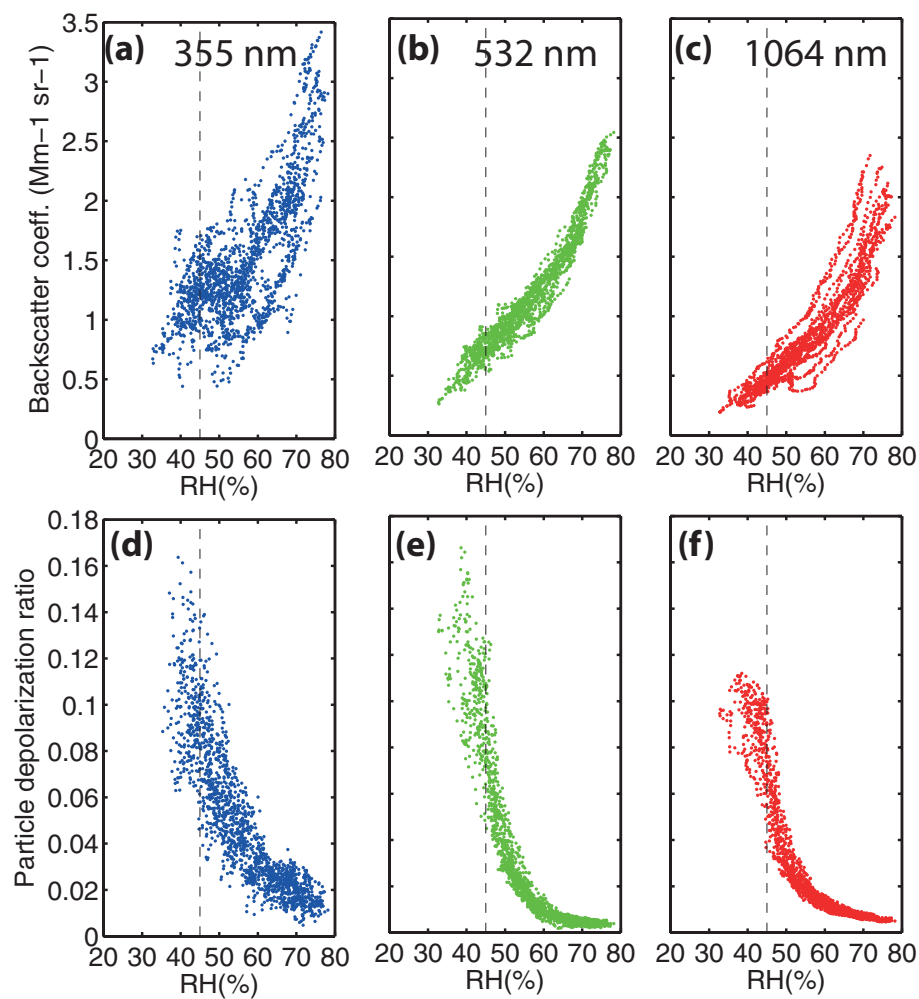
Figure 12. Correlation of the particle backscatter coefficient (top, a-c) and particle linear depolarization ratio (bottom, d-f) with RH for the three wavelengths 355, 532, and 1064 nm. The BERTHA measurement of 23 February 2014, 23:38–01:08 UTC, 375–1100 m height, are used (5 min temporal and 50 m vertical resolution). **The dashed line marks the sea salt efflorescence point (45% RH).**

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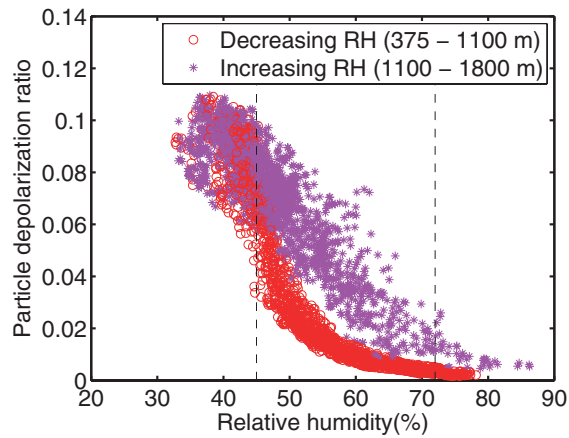
Figure 13. Correlation of the PLDR at 1064 nm and RH for the same settings as in Fig. 12, but for a different height interval (375–1800 m). Above 1100 m (purple stars) RH is increasing again, up to 80%, as can be seen in Fig. 9. The depolarization ratio decreases with increasing RH, but keeps higher values. The hysteresis effect between crystallization (45–48% RH, **dashed line**) and deliquescence (70–74% RH, **dashed line**) can be seen.
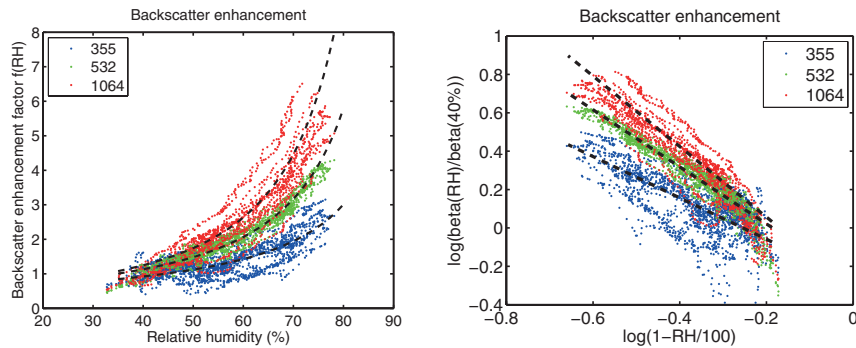

Figure 14. Backscatter enhancement factor for a dry value at 40% RH **(a) on a linear scale and (b) on a log-log scale.** The three wavelengths are fitted separately by Eq. (2).
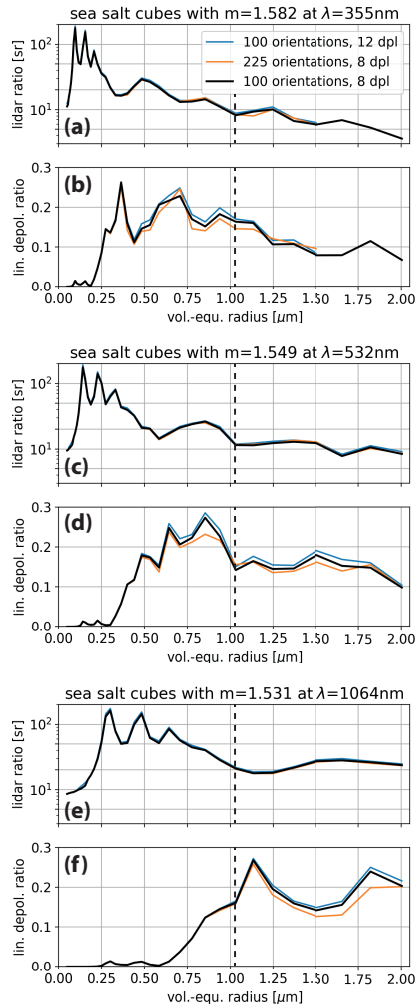
Figure 15. Simulation of the lidar ratio and the particle linear depolarization ratio for sea salt cubes depending on volume equivalent radius. The three lidar wavelengths are treated separately. The model simulations with increased number of dipoles (blue line) and orientations (orange line) are given as an estimation of the uncertainty. **A dashed line marks the effective radius (r_{eff}=1.033 μm) of the AERONET size distribution given in Fig. 16. For the simulations of the cubic sea salt particles, the radius was reduced by a factor of 2.** Details are given in the text.
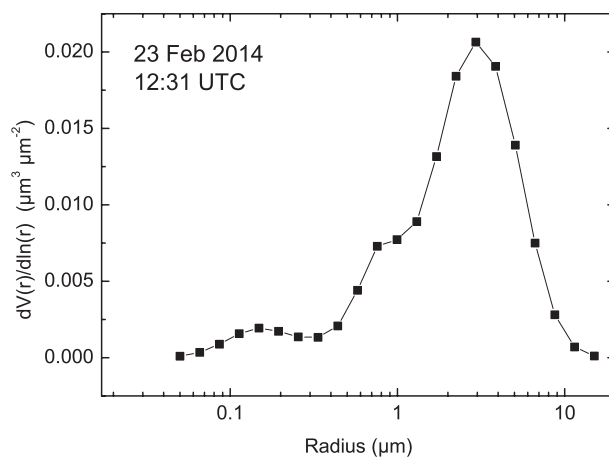
Figure 16. AERONET size distribution for 23 February 2014 12:31 UTC at Ragged Point, version 1.5 (**Holben et al., 1998; AERONET, 2017**).