Radiative Effects of Increased Water Vapor Associated with Enhanced Dustiness in the Saharan Air Layer

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Abstract

The Saharan Air Layer (SAL) has been shown to be an elevated, well-mixed, warm, dry, frequently dusty layer. The structure of the SAL plays an important role in regional climate and in long-range dust transport. A new analysis of aircraft observations shows that although increased dustiness in the SAL is associated with drier conditions in the lower-SAL as expected, dustiness is also associated with increased moisture in the upper-SAL. We assess the radiative effects of the observed dust and increased water vapor (WV) using a radiative transfer model. The observed WV in the upper-SAL affects the top-of-atmosphere (TOA) direct radiative effect (DRE), while lower-SAL WV affects the surface DRE and column atmospheric heating. TOA DRE is negative for dust-only, while including both the observed dust and WV reduces the magnitude of the negative TOA DRE by 11%. The observed WV structure increases the negative surface DRE from dust by 8% and increases atmospheric heating by 17%. These effects are driven by longwave (LW) radiation, whereby WV changes increase the positive TOA LW DRE by 30%, decrease the surface LW DRE by 52% and change the sign of LW atmospheric heating from negative to positive. The observed WV profile leads to enhanced cooling in the moist upper-SAL and heating in the dry lower-SAL under dustier conditions. Increased WV in the SAL is consistent with other studies demonstrating a trend of increased WV over the Sahara. This work demonstrates the importance of the upper-SAL WV profile in determining the radiative effect dust.

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7 **Key Points:**

- 8 • Observations show enhanced moisture in the upper Saharan Air Layer (SAL) associated 9 with dust, counter to the conventional dry layer model
- Enhanced moisture reduces the magnitude of the negative direct radiative effect from 10 11 dust at the top of atmosphere by 11%
- 12 • Observed water vapor structure leads to enhanced cooling in the moist upper-SAL and 13 heating in the dry lower-SAL under dustier conditions
- 14

15 Abstract

16 The Saharan Air Layer (SAL) has been shown to be an elevated, well-mixed, warm, dry,

17 frequently dusty layer. The structure of the SAL plays an important role in regional climate and

18 in long-range dust transport. A new analysis of aircraft observations shows that although

19 increased dustiness in the SAL is associated with drier conditions in the lower-SAL as expected,

20 dustiness is also associated with increased moisture in the upper-SAL. We assess the radiative

21 effects of the observed dust and increased water vapor (WV) using a radiative transfer model.

22 The observed WV in the upper-SAL affects the top-of-atmosphere (TOA) direct radiative effect

23 (DRE), while lower-SAL WV affects the surface DRE and column atmospheric heating. TOA

24 DRE is negative for dust-only, while including both the observed dust and WV reduces the

25 magnitude of the negative TOA DRE by 11%. The observed WV structure increases the negative

26 surface DRE from dust by 8% and increases atmospheric heating by 17%. These effects are

27 driven by longwave (LW) radiation, whereby WV changes increase the positive TOA LW DRE

28 by 30%, decrease the surface LW DRE by 52% and change the sign of LW atmospheric heating 29

from negative to positive. The observed WV profile leads to enhanced cooling in the moist

30 upper-SAL and heating in the dry lower-SAL under dustier conditions. Increased WV in the

31 SAL is consistent with other studies demonstrating a trend of increased WV over the Sahara. This work demonstrates the importance of the upper-SAL WV profile in determining the

32

33 radiative effect dust.

34 **Plain Language Summary**

35 During summer, warm, dry, dusty air is transported from the Sahara across the Atlantic Ocean in

36 an elevated plume. This plume has many impacts on climate, such as suppressing convection

- 37 which may be important to hurricane development, and transporting dust particles which can
- 38 supply nutrients to the oceans and degrade respiratory health. Previously the plume has been
- 39 found to be very dry, but by using new observations from a research aircraft, we show that high

40 levels of moisture are found at its top. This is important because it alters the balance of heating

41 and cooling at different altitudes within the atmosphere, which has not been accounted for in this

- 42 way before, and may impact how Saharan air interacts with weather in the Atlantic region and
- 43 how easily dust is transported over long distances. We find that the addition of moisture at the
- 44 top of the plume adds a warming of 11% compared to the effects from dusty air, and decreases in
- moisture at lower altitudes can change the nighttime effect of the plume from cooling to heating.
 Therefore future work should take into account the vertical distribution moisture in Saharan dust
- 47 plumes as well as the dust itself.

48 **1 Introduction**

49 The Saharan Air Laver (SAL) is conventionally considered to be a deep warm, dry, elevated 50 layer of air which exists over the tropical north Atlantic Ocean from late spring to early fall, frequently containing mineral dust transported from North Africa (Braun, 2010). The SAL 51 52 originates from strong surface heating over the Sahara desert, where a deep, well-mixed, warm, 53 dry boundary layer develops, typically extending up to 5 to 6 km, known as the Saharan 54 Boundary Layer (Engelstaedter et al., 2015; Marsham et al., 2013). The SAL, and any embedded 55 mineral dust, is transported westwards by prevailing winds and steered by African Easterly 56 Waves. As it moves over the Atlantic Ocean, it is undercut by a moist marine boundary layer 57 (MBL), forming an elevated mixed layer known as the SAL, characterized by near-constant 58 potential temperatures and water vapor (WV) mixing ratios, and retaining its structure as it is 59 transported westwards (Carlson, 2016; Dunion & Velden, 2004).

60

61 In the original conceptual model of the SAL (e.g. (Carlson & Prospero, 1972; Dunion, 2011; 62 Karyampudi et al., 1999; Prospero & Carlson, 1972) the SAL is shown to be warm, dry and to 63 varying degrees, dust laden. For example, reported values of WV mixing ratios within the 64 eastern SAL are around 2 to 5 g/kg (Carlson & Benjamin, 1980; Carlson & Prospero, 1972; Ismail et al., 2010; Karyampudi et al., 1999), and 1 to 6 g/kg in the western SAL (Dunion, 2011; 65 66 Dunion & Velden, 2004). Kanitz et al. (2014) present measurements across the width of the 67 tropical Atlantic, finding WV mixing ratios between 3 to 7 g/kg within the SAL. Warm, dry 68 anomalies in the SAL between 800 to 900 hPa have been shown to be related to increased dust 69 optical depth. Together the dusty, dry anomalies are responsible for heating the lower SAL and 70 maintaining the temperature inversion its base, as well has playing a role in suppressing deep 71 convection and increasing low cloud fraction (Doherty & Evan, 2014; Wong & Dessler, 2005; 72 Wong et al., 2009).

73

74 Around 50% of dust events in the summertime Sahara are driven by cold pool outflows 75 (haboobs), with the dust being associated with elevated moisture levels deriving from moist 76 downdrafts (Marsham et al., 2013; Marsham et al., 2008). Several studies have described the role 77 of cold pool outflows and density currents in transporting dust and moisture into the arid Sahara 78 (Allen et al., 2013; Engelstaedter et al., 2015; Flamant et al., 2007). Marsham et al. (2016) 79 examined the relative radiative effects of dust and WV in the Saharan heat low (SHL), finding 80 that WV variations exert a dominant role on the top of atmosphere (TOA) radiation budget, 81 while dust variability was dominant in driving atmospheric heating and surface radiation. 82 Gutleben et al. (2019) found that SAL WV mixing ratios over the western Atlantic for one dust 83 event were elevated compared to those in the surrounding free atmosphere, and found that the 84 altered WV profile within the dust led to substantial changes in radiative heating rates.

Here we examine dust and WV aircraft observations in the tropical north Atlantic Ocean region. During research flights, it was noticed that specific humidity often increased when dust layers were penetrated, and was particularly noticeable during a few extremely large dust events (Marenco et al., 2018). Since this increased moisture within the dusty SAL is counter to the conventional model of a dry, dusty SAL, here we examine the evidence from airborne observations and evaluate the radiative effect of the observed changes in dust and enhanced water vapor.

93

94 **2** Methods

95 **2.1 Aircraft Observations**

We present meteorological and aerosol in-situ aircraft observations from the AERosol Properties
 - Dust (AER-D) airborne field campaign (Marenco et al., 2018) which took place during August

- 98 2015 in the region of the Cape Verde Islands (Figure 1). The Facility for Atmospheric Airborne
- 99 Measurements BAe146 research aircraft sampled a series of dust events in the SAL between 7
- and 25 August 2015, described in more detail by Ryder et al. (2018), Liu et al. (2018), Marenco
- 101 et al. (2018), where microphysical, chemical, optical and radiative properties of the dust samples
- 102 are presented.

103



104

Figure 1: Locations of AER-D aircraft profiles analysed in this study. Lines show locations sampled during profile ascents or descents, solid dots indicate mean profile location. Colors

107 indicate AOD as shown in the legend.

108

Here we use in-situ observations from 24 aircraft profile descents and ascents as shown in Figure 110 1, sampled during 6 flights (also shown in Ryder et al. (2018), their table 2). Profiles cover

111 altitudes from close to the ocean surface to above the SAL, generally ~5.5 km. All the dust

112 events sampled during AER-D were driven by outflows from mesoscale convective systems 113 ('haboobs,' identified in Ryder et al. (2018), which in most cases were mixed vertically 114 throughout the Saharan boundary layer by convective mixing before being advected over the 115 ocean in an elevated SAL. However, two flights (b923, b924) sampled a large dust plume with 116 aerosol optical depths (AODs) at 550 nm greater than 1, further to the north close to the Canary 117 Islands. Two particular profiles sampled an extremely unusual dust structure with very high dust 118 concentrations peaking at low altitudes of around 1 km, constituting a clear 'dust front' which 119 kept its structure (low altitude, intense dust concentrations) while being advected out over the 120 Atlantic Ocean (brown lines in Figure 1). This event is investigated in more detail by Marenco et 121 al. (2018), and is referred to here as the 'dust front' case since it is different to the other, more 122 typical elevated dust profiles sampled.

123

Airborne measurements of temperature are taken from a Rosemount/Goodrich type 102 Total Air Temperature probe, using the non-deiced models for all flights except b934 when data is not available, and when the deiced model was used instead. Dew point temperature measurements, used to calculate water vapor mixing ratios are taken from the Sky Phys Tech Inc. Nevzorov total water content probe (Korolev et al., 1998), except for flights b923, b934 and b932 Profile 1 where they are taken from the General Eastern 1011B Chilled Mirror Hygrometer. Pressure was measured by a reduced vertical separation minimum data system.

131

132 Ryder et al. (2018) have shown that the aerosol load sampled was predominantly mineral dust, 133 including the aerosol in the marine boundary layer. Here we take a straightforward indication of 134 dust loading from the dust extinction at 550 nm, calculated by summing dust scattering and 135 absorption. Scattering is measured by a TSI 3563 integrating nephelometer (550 nm wavelength 136 used here) and absorption measurements were made by a Radiance Research particle soot 137 absorption photometer (PSAP) at 567 nm. Further details and processing information can be 138 found in Ryder et al. (2018). Extinction is integrated vertically from aircraft profiles to provide 139 AODs at 550 nm.

140

141 **2.2 Radiative Transfer Calculations**

We employ the Suite Of Community RAdiative Transfer codes based on Edwards and Slingo (SOCRATES, Edwards and Slingo (1996), Manners et al. (2017)) radiative transfer model (RTM) in order to calculate radiative fluxes. SOCRATES is the RTM implemented by the family of UK Met Office numerical weather prediction and climate models. Here we use the spectral setup analogous to that from HadGEM3 model Global Atmosphere 6 configuration (Walters et al., 2011). A two stream practical improved flux method is used (Zdunkowski et al., 1980).

148

149 Shortwave fluxes are calculated over 6 spectral bands from 0.2 to 10 μ m and longwave fluxes 150 are calculated in 9 spectral bands from 3.3 to 100 μ m. Gaseous absorption is represented 151 according to Cusack et al. (1999) using a correlated-k method. Water vapor terms are based on 152 the HITRAN 2001 database (Rothman et al., 2003) for gaseous absorption coefficients, with 153 updates up to 2003. The water vapor continuum is represented using version 2.4 of the CKD 154 model. Top of atmosphere incoming solar radiation is set to 1365 Wm⁻².

155

156 SOCRATES requires aerosol vertical profiles in terms of mass mixing ratios. These are 157 calculated to represent dust from the measured in-situ aerosol extinction profiles, converted to a mass loading by using a field campaign specific mass extinction coefficient of $0.36 \text{ m}^2\text{g}^{-1}$ (Ryder

et al., 2018). This value represents the full size distribution up to 100 μ m diameter.

160

161 We use 122 vertical levels in SOCRATES, covering 1007 to 0.0005 hPa, with resolution ~20 162 hPa in the lower atmosphere, including the SAL. Observations of dust mass mixing ratio, water

163 vapor mixing ratio and temperature are regridded to the required vertical grid. Only dust aerosols

164 are included. At altitudes above where aircraft data were measured, dust concentrations are set to

165 zero and temperature and water vapor values are set to revert to values from a tropical standard

166 profile (Anderson et al., 1986). Ozone, methane, carbon monoxide, carbon dioxide, nitrous oxide

and oxygen profiles are taken from Randles et al. (2012). Shortwave surface albedo is set to 0.05
 representative of an ocean surface and LW surface emissivity to 0.982 in all spectral intervals.

169

170 Dust optical properties are calculated spectrally in the longwave (LW) and shortwave (SW) 171 spectra using lognormal size distribution parameters representing the AER-D SAL average, as given in Ryder et al. (2018). In the SW, complex refractive index data are taken from Colarco et 172 173 al. (2014) and LW values from Volz (1973) since these datasets fall centrally in the range from 174 literature (Ryder et al., 2019) and cover the full spectral range required. Mie scattering code (and therefore a spherical particle assumption) is used to calculate spectral optical properties of mass 175 176 extinction coefficient, single scattering albedo (SSA) and asymmetry parameter, which are 177 applied in SOCRATES and are shown in Table 1. The SSA at visible wavelengths is 0.86, considerably lower than the AER-D campaign mean value of 0.95 at 550 nm (Ryder et al., 2018). 178 179 The difference occurs due to the spectral averaging applied here which incorporates absorption 180 increasing significantly towards smaller solar wavelengths, as well as the use of the Colarco et 181 al. (2014) imaginary refractive index of 0.0024 at 550 nm compared to a value of 0.0010 derived

182 in Ryder et al. (2018). SOCRATES includes both absorption and scattering in the LW spectrum.

183

Spectral Range	Lower wavelength,	Upper wavelength,	MEC/m ² g ⁻¹	SSA	Asymmetry Parameter
C	μm	μm			
SW	0.20	0.32	0.36	0.70	0.83
	0.32	0.69	0.36	0.86	0.76
	0.69	1.19	0.36	0.95	0.71
	1.19	2.38	0.35	0.93	0.69
	2.38	10.00	0.26	0.82	0.71
LW	25.0	10000.0	0.05	0.28	0.29
	18.2	25.0	0.15	0.36	0.32
	12.5	18.2	0.13	0.42	0.52
	13.3	16.9	0.10	0.37	0.54
	8.3	12.5	0.17	0.42	0.56
	8.9	10.1	0.28	0.45	0.46
	7.5	8.3	0.09	0.41	0.74
	6.7	7.5	0.16	0.62	0.72
	3.3	6.7	0.24	0.87	0.68

184 **Table 1:** Optical properties applied in SOCRATES for each spectral band estimated using

refractive index data from Colarco et al. (2014) for the SW, Volz et al. (1973) for the LW and the AER-D mean size distribution (Ryder et al., 2018).

187 Sensitivity tests have also been carried out using more and less absorbing dust at solar 188 wavelengths. These were calculated using the more absorbing OPAC dataset refractive indices 189 with the OPAC transported mineral dust size distribution (Hess et al., 1998), and the less 190 absorbing refractive index dataset of Balkanski et al. (2007) combined with a smaller dust size 191 distribution (Dubovik et al., 2002). At infrared wavelengths, sensitivity to dust is tested by 192 varying the dust size distribution to the smaller one of Dubovik et al. (2002) and the refractive 193 index to that from OPAC. These calculations yield SSA values of 0.74 and 0.94 for the more and 194 less absorbing cases respectively for the spectral band spanning wavelengths of 0.32 to 0.69 195 microns.

196

197 Surface temperatures for the LW calculations are set based on values observed from aircraft in-198 situ observations or dropsondes. SW diurnal averages are calculated by running SOCRATES at 199 three solar zenith angles based on the location and time of year of observations, which are 200 multiplied by a gaussian weight and summed.

201

202 For each of 24 profiles, 4 radiative transfer simulations are conducted, as outlined in Table 2, in order to isolate the effects of the altered WV profile in the SAL, the presence of dust, and their 203 204 combined effect. The temperature profile is always taken from aircraft measurements. For the 205 CONTROL, no radiative effects of dust are included. In order to create a WV profile 206 representative of background conditions for the region for the control, a median of the aircraft 207 WV profiles where AOD is less than 0.3 is taken (blue lines in fig 1e and 2a), which represents a 208 non-SAL background WV profile. We do not use the tropical standard WV profile (Anderson et 209 al., 1986), because the non-SAL atmosphere in the region is significantly moister than this (see 210 Figure 1e). The three perturbation experiments shown in Table 2 allow the quantification of the 211 radiative impacts relative to a typical background, non-dusty state in the region. For the dust only 212 experiment (DU), dust observations from aircraft measurements are included, but the background WV profile maintained, such that only the radiative impact of dust is assessed. For 213 214 the water vapor only experiment (WV), no dust is included, but the water vapor profile used is 215 taken from the aircraft observations. Finally, for DU+WV, both the aircraft-observed dust and 216 water vapor profiles are included to assess their combined effect. For each experiment, vertical 217 profiles of SW and LW radiative fluxes and heating rates are calculated for all aircraft profiles. 218

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- 219 220

Experiment Name	Abbreviation	Temperature Profile	Dust Profile	Water Vapor
				Profile
Control	CONTROL	Aircraft observations	None	Background WV profile: median of WV profiles where AOD<0.3
Dust only	DU	Aircraft observations	Aircraft observations	Background WV profile: median of WV profiles where AOD<0.3
Water vapor only	WV	Aircraft observations	None	Aircraft observations
Dust and water vapor	DU+WV	Aircraft observations	Aircraft observations	Aircraft observations

221 Table 2: SOCRATES experiments. Each experiment is performed for each of 24 aircraft 222 profiles.

The direct radiative effect (DRE) due to each experiment relative to the control is calculated as defined in equation 1.

$$DRE_{lev}^{spect} = NET_EXPT_{lev}^{spect} - NET_CONTROL_{lev}^{spect}$$

Equation 1

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226 NET refers to the net (downwards minus upwards) radiative flux at a level (lev) either at the top of atmosphere (TOA) or surface (SFC), for spect which can be either the SW spectrum, LW 227 spectrum, or total (SW+LW). EXPT refers to the experiment in question, either DU, WV or 228 229 DU+WV. DREs calculated are instantaneous, diurnally averaged calculations. Positive values at 230 the TOA indicate a warming of the earth atmosphere system. The atmospheric (ATM) radiative effect, also sometimes referred to as atmospheric radiative convergence and indicating 231 atmospheric column heating when positive, is calculated from $DRE_{ATM}^{spect} = DRE_{TOA}^{spect} -$ 232 DRE_{SFC}^{spect} . 233

234 **3 Results**

235 **3.1 Aircraft observations of dust and water vapor**

Figure 2 shows the profiles of aerosol extinction and water vapor observed by the aircraft. Each is subset by the measured AOD, selected so that each range contains a reasonable number of profiles. The dust load is clearly be seen to increase as AOD increases, and the expected vertical profile of the SAL develops, with (mostly) enhanced dust centered around 700 hPa.

240

Water vapor profiles are also shown. It is evident that even in cases without much dust (AOD<0.3, blue), the atmosphere is moister than the tropical standard. This is perhaps not surprising given the large amount of variability across tropical regions for which the tropical standard atmosphere represents (e.g. Dunion (2011)).



Figure 2. Aircraft profile observations of aerosol extinction (top row) and water vapor mixing ratio (bottom row). Each variable is subset by measured AOD: blue (AOD<0.3, 5 profiles), green (0.3<AOD<0.6, 9 profiles), orange (0.6<AOD<1.0, 6 profiles) and red (AOD>1.0, 4 profiles). Brown lines indicate dust front event. Note that panel (d) extinction x-axis is nearly 5 times as large as (a)-(c). Black diamonds indicate the tropical standard profile. WV profiles shown in panel e are used to create the background WV profile as in Table 2 for the CONTROL experiment.

As AOD increases, the encroachment of the SAL is evident with a deepening of the elevated, well-mixed layer. This can also be seen in Figure 3 where medians for each AOD category, and anomalies relative to the background state are shown. As AOD increases, a well-mixed layer of near-constant potential temperature and water vapor develop, as expected. The well-mixed nature of the SAL means that at lower altitudes, the SAL is warmer and drier than the background state, whereas towards the top of the SAL the atmosphere is cooler and moister than the background state.

262

263 Figure 3b clearly shows that under increased AODs (or dust loading), enhanced WV in the 264 upper-SAL and decreased WV in the lower-SAL is observed. This becomes more pronounced as 265 the AOD increases. For AODs over 0.6, enhanced WV is observed for heights above 750 hPa, 266 with anomalies exceeding 2 g/kg for the 1<AOD<2 category (red line). An interesting doublepeaked water vapor anomaly structure is seen, whereby anomalies peak at a lower level of 267 around 700 hPa, coincident with the center of the SAL and the anomalies it brings, but anomalies 268 269 also peak at around 500 hPa, due to the dustier SAL cases extending deeper vertically with more 270 moisture at higher altitudes.

271

Dust extinction (Figure 3e) and potential temperature (Figure 3c) as a function AOD are also shown, and show the well-mixed and relatively warmer structure of the SAL becoming more clearly defined as it becomes dustier, as expected. It is also notable that the highest AOD category (red) shows a cold anomaly in the upper SAL, at heights above 520 hPa.

276

277 The case of the dust front, shown in brown in Figures 2 and 3, is particularly interesting. The 278 extremely high dust loadings are reflected by extinction values of up to nearly 2500 Mm⁻¹, 279 peaking at around 900 hPa (~1km), unusually low altitudes for a dust event over the tropical 280 north Atlantic during summer. The figures show that the peak in dust loadings at ~900 hPa are 281 coincident with an increase in WV content (Figure 2h) up to around 13 g/kg relative to the more 282 well-mixed layer above this with WV mixing ratios of around 5 g/kg. Ryder et al. (2018) and 283 Marenco et al. (2018) have identified this dust event as being an intense haboob which 284 propagated out over the Atlantic at low altitudes, rather than being mixed vertically through the 285 Saharan boundary layer first, as usually occurs. Thus the intense concentration of high moisture 286 and dust loadings at low altitudes can be traced back to cold-pool outflow driving the dust uplift 287 in the form of a recent haboob.

288

Figure 3f shows the relationship between AOD and precipitable water vapor (PWV) for the whole column (black), for the lower SAL (PWV_lower, p>820 hPa, red), and for the upper SAL (PWV_upper, pressure from 780 to 480 hPa, orange). Overall, as AOD increases, PWV decreases, though there is a large amount of variability and the linear fit has a low correlation (0.18). More insight is gained into the impact of increased dust through viewing the SAL lower and upper WV changes separately. Figure 3f shows that as AOD increases, PWV_upper increases by 3.8 mm per unit AOD, while PWV_lower decreases by 6.7 mm per unit AOD, with the linear fits showing correlations of 0.58 and 0.59 respectively. For the upper-SAL, PWV_upper increases from 18 mm to 23 mm between the AOD categories of AOD<0.3 and 1<AOD<2 respectively for the profiles shown in Figure 2a (blue vs red). Therefore there is a clear relationship between increased dust load (AOD), decreased lower-SAL moisture, and increased upper-SAL moisture.

301

Note that the two highest AOD data points (relating to the dust front case) can be viewed to some extent as outliers and are not included in the linear fits. As Figure 3f shows, the two in-situ aircraft profiles sampling this dust front feature demonstrate low PWV_upper and high PWV_lower values, different to the trend demonstrated by the more conventional elevated SAL structure of the other profiles.





Figure 3. Aircraft observations of (a) water vapor mixing ratio, (c) potential temperature and (e) aerosol extinction; anomalies of (b) water vapor mixing ratio and (d) potential temperature relative to the background state (profiles where AOD<0.3). Profiles are regridded vertically as applied in the RTM. Profiles are grouped by AOD range as indicated with bold lines indicating the median and dashed lines the 10^{th} and 90^{th} percentile ranges for AOD categories where AOD<1. 1<AOD<2 and dust front categories (red and brown) are represented by means as

categories contain only 2 profiles. Black line with symbols indicates tropical standard profile. (f)
Relationship for all individual profiles between AOD and PWV, PWV_lower (pressure >820
hPa) and PWV_upper (480 hPa < pressure < 780 hPa), and linear fits where AOD< 2, with
Pearson's correlation coefficients of 0.18, 0.58 and 0.59, and dPWV/dAOD values of -2.4, -6.7
and +3.8 mm/AOD respectively.

320

Although the structure of the SAL has been extensively examined and documented, some of the changes found here are not consistent with the conventional view of the SAL's structure. Figures 2 and 3 show much moister values than those documented in most cases, up to 11 g/kg depending on altitude, and with a clear upper SAL moistening with increasing dustiness relative to less dusty cases. Previously, the radiative roles of dust and *dry* SAL air have been assessed (e.g. Wong et al., 2009). Next, we evaluate the radiative effects of the measured dust, and *moist* upper SAL air shown in the observations.

328

329 **3.2 Impact of altered WV structure on Radiative Effect**

Having demonstrated that increased dust loadings were associated with increased upper-SAL
WV (PWV_upper) but decreased lower-SAL WV (PWV_lower) during AER-D, the DREs from
dust only, WV only and both combined are now described in order to investigate the radiative
impact of the WV changes relative to those from the dust.

334

Figure 4 shows the shortwave DRE as a function of AOD for each experiment at the TOA, surface and for the atmospheric column. DREs are relative to the CONTROL experiment, where WV is derived from conditions where AOD<0.3. The results show that in the shortwave spectrum, the main driver of the DRE is always dust, since the WV DREs are comparatively small. Increased dust AOD leads to a more negative TOA DRE, a more negative surface DRE, and larger atmospheric heating.

341



343 344 Figure 4. Shortwave DRE relative to control experiment for the inclusion of dust only (DU, 345 orange), WV only (blue) and dust and WV (DU+WV, green), as a function of 550 nm AOD. 346 Panels show (a) DRE at the TOA, (b) surface and (c) atmospheric column. Vertical dashed lines 347 indicate AOD categories.

348 Figure 5 shows DREs as a function of AOD, but for the longwave spectrum. Here, the WV DRE exerts a large enough effect to alter the DRE significantly. At the TOA dust generates a positive 349 350 LW DRE (orange points), which increases with AOD. (The exception are the two 'dust front' 351 cases with the highest AOD, where the dust was lower in the atmosphere resulting in weaker LW 352 DREs). The WV TOA DRE is also mostly positive and also increases with AOD (blue data 353 points). This is consistent with increased WV, as observed in the upper-SAL, and will be 354 investigated further in Section 3.3. As a result of the positive DRE due to WV, the DU+WV LW 355 TOA DRE (green data points) is significantly larger than due to dust only, with values increasing 356 by an average of 30% and a wide range of -27% to 292%.

- 357
- 358 At the surface and through the atmospheric column, dust causes a surface warming and an 359 atmospheric cooling. The WV DRE is the opposite, resulting in a negative surface DRE and 360 atmospheric heating. This is consistent with a net reduction in WV, and Section 3.3 will demonstrate that these changes are driven by a reduction in lower-SAL WV. At the surface, this 361 362 results in a reduced LW DRE from DU+WV compared to DU alone, with values reducing by 363 52% on average. In the atmosphere, WV changes the sign of atmospheric heating, from a mean of -1.5 Wm⁻² for DU to 5.5 Wm⁻² for DU+WV. This could influence SAL dynamics at nighttime 364 365 when shortwave influences are inactive.





Figure 5. Longwave DRE relative to control experiment for the inclusion of dust only (DU, 368 369 orange), WV only (blue) and dust and WV (DU+WV, green), as a function of 550 nm AOD. 370 Panels show (a) DRE at the TOA, (b) surface and (c) atmospheric column. Vertical dashed lines 371 indicate AOD categories.

372 Figure 6 shows the total (SW+LW) DREs as a function of AOD, and demonstrates how the dust

373 and WV shortwave and longwave impacts from Figures 4 and 5 combine. The total TOA DRE

374 for dust is negative, reflecting the larger magnitude originating from the SW dust DRE compared

- to the LW. WV TOA DREs are positive, driven by the LW DRE from WV. The net effect of 375
- 376 including the observed WV profiles is to reduce the total negative DRE for DU+WV compared
- to DU alone. Overall including WV effects (excluding the dust front case) increases the TOA 377 DRE from dust by -1.5 to 7.3 Wm⁻² (mean of 1.3 Wm⁻²) or increases the negative DRE by -17 to
- 378
- 379 64% (mean of 11%).



Figure 6. Total (shortwave plus longwave) DRE relative to control experiment for the inclusion of dust only (DU, orange), WV only (blue) and dust and WV (DU+WV, green), as a function of 550 nm AOD. Panels show (a) DRE at the TOA, (b) surface and (c) atmospheric column. Vertical dashed lines indicate AOD categories.

Figure 6 shows that at the surface, the main driver of the total DRE is dust, where the large negative SW DREs are slightly reduced in magnitude by positive values from the LW spectrum. WV DREs at the surface are small and negative and reduce the SFC DRE by a mean of 8% for DU+WV compared to DU. In the atmospheric column, dust is also the main driver of atmospheric heating, originating from large positive SW values shown in Figure 3. However, here the WV change can act to enhance this heating from -1 to 16 Wm⁻² (mean of 5 Wm⁻² or 17%), as a result of the LW WV effect.

392

393 **3.3 Impact of WV in the lower and upper-SAL**

Figure 7 and Figure 8 show the LW DREs as a function of PWV in the lower-SAL and upperSAL respectively, to isolate the impacts of the changed WV structure with increasing dust,
whereby WV in the upper-SAL increases, but decreases in the lower-SAL.

397

In Figure 7, clear relationships between PWV_lower and both surface and atmospheric LW DRE can be seen for water vapor (blue data points). In the atmosphere (panel c), a drier lower-SAL results in relatively more atmospheric heating (or equivalently less atmospheric cooling), while 401 at the surface, drier lower-SAL conditions result in a more negative LW DRE because less 402 radiation is emitted downwards by the drier atmosphere.

403

404 Under dustier conditions when the lower-SAL is drier, the DU experiment mostly results in 405 slightly negative ATM DRE values (atmospheric cooling, orange points in Figure 7c). The 406 addition of the WV DRE changes are large enough to change the sign of ATM DRE and result in 407 atmospheric heating for DU+WV (green points). Thus the change in sign from cooling due to 408 heating (also seen in Figure 5c) for DU+WV compared to DU is explained by lower moisture 409 content in the lower-SAL, and the LW ATM DRE for WV increases as moisture in the lower-410 SAL decreases. In Figure 6b, a drier, dustier lower-SAL results in a positive surface DRE for 411 DU. Since the WV surface DRE is negative, values for DU+WV are sometimes large enough to 412 change the sign compared to DU. In contrast the TOA DRE shows no dependence on WV in the 413 lower SAL in Figure 6a.

414



415

418 (DU+WV, green). Panels show (a) DRE at the TOA, (b) surface and (c) atmospheric column.

Figure 8 shows the DREs as a function of PWV in the upper-SAL. Although PWV_upper has no observable effect over the LW DRE at the surface and on the total atmospheric column, it has a strong effect on the LW DRE at the TOA. Larger PWV_upper (associated with increased

⁴¹⁶ Figure 7. Longwave DRE relative to control experiment, as a function of the lower SAL PWV
417 (PWV_lower), for the inclusion of dust only (DU, orange), WV only (blue) and dust and WV

dustiness) results in larger, more positive LW TOA DREs due to reduced outgoing longwave
radiation. This is seen for both the WV and DU experiments. When WV in the upper-SAL is
higher, its proximity to the TOA (compared to the lower SAL) means that enhanced upper-SAL
WV results in less LW radiation being emitted and therefore a warming effect results. At the
same time, since dust AOD and PWV_upper are related, the LW dust TOA DRE also increases
for high PWV_upper values. As a result, the two effects act in the same direction and increase
the DU+WV TOA DRE.

429

This also explains the trend seen in Figure 5a, and confirms that the TOA WV effect is controlled by changes to PWV_upper, rather than PWV_lower. Thus increases in dust and increased PWV_upper both act to cause and amplify a positive LW TOA DRE.

433





Figure 8. Longwave DRE relative to control experiment, as a function of the upper SAL PWV
(PWV_upper), for the inclusion of dust only (DU, orange), WV only (blue) and dust and WV
(DU+WV, green). Panels show (a) DRE at the TOA, (b) surface and (c) atmospheric column.

Figure 9 shows the absolute heating rates for the control and changes in heating rates for each experiment for the SW, LW and total relative to the control. It can be seen that dust (Figure 9 df) causes a SW heating, a LW cooling, and an total heating. In the SW, WV (Figure 9g) causes a slight heating at higher altitudes which is stronger and higher for larger AOD where PWV_upper is larger. In the LW (panel h), the impact of increased PWV upper and decreased PWV lower 443 can be seen. When AODs are higher, more upper-SAL cooling from enhanced moisture and less
444 mid-SAL cooling (i.e. warming) from enhanced dryness is observed. This results in changes to
445 total heating rates of up to -2.5 K/d in the upper SAL and +1.5 K/d through the lower-SAL due
446 to the dust-related WV changes (panel i).

447

448 Comparing Fig f to Fig l shows the changes in total heating rates due to DU+WV compared to 449 dust alone. We see that the total WV+DU heating rates (fig l) show extra cooling at heights 450 above 550 hPa and increased heating through the mid-SAL (~950 to 600 hPa), higher by about 2 451 K/d relative to Fig f. Although increased dust was associated with decreased PWV lower at 452 heights beneath 820 hPa, the radiative heating effects of this WV change are felt throughout the 453 wider column up to around 680 hPa. As a result, the PWV_lower changes control the ATM 454 DRE, determining atmospheric heating or cooling, as shown in Figure 6c. It is also notable that 455 only by including the enhanced WV in the upper SAL, is the cold anomaly in potential temperatures seen in Figure 3d consistent with the additional cooling rates above around 500 456 457 hPa.



460 Figure 9. Top row: absolute heating rates for the CONTROL for all profiles for (a) SW, (b) LW,
461 (c) total (SW+LW). Other rows: changes in heating rates (HR, K/day) between each experiment
462 and the control, for (d-f) DU, (g-i) WV (middle row) and (j-l) WV+DU (bottom row), for the SW

463 spectrum (left), LW spectrum (center) and total (SW plus LW, right). Profiles are colored as a 464 function of AOD.

465 **4 Discussion**

This work finds elevated moisture levels in the upper SAL associated with dust, and demonstrates that this increased moisture is important in determining the magnitude, and potentially the sign of the radiative effect exerted by the dusty SAL. The elevated moisture found in the upper SAL differs to the frequently reported SAL characteristics of it being a straightforward dry layer, suggesting that more complexity and vertical detail is required to accurately reflect the SAL and its radiative impacts.

472

473 Much previous work characterizing the SAL focuses on the its lower atmospheric structure, 474 particularly heating rates at around 850 hPa and their relation to sustaining the temperature 475 inversion between the marine boundary layer and the SAL base, due to its importance in 476 suppression of convection and potentially impeding tropical cyclone development (e.g. Dunion 477 and Velden (2004); Wong et al. (2009)). However, a few studies have focused more on the 478 structure of the entire depth of the SAL. For example, Braun (2010) find a 400-600hPa 479 moistening across Africa to Saudi Arabia relative to the very dry Saharan air below, due to deep, 480 dry convective mixing over the Sahara. This structure has sometimes been found at the west 481 coast of Africa and also downstream. Ismail et al. (2010) show a case where WV mixing ratios were around 2 to 7 g/kg throughout the SAL. They attribute this increased moisture to 482 483 midaltitude convection as a result of intensification of an AEW, pointing out that increased WV 484 leaves the SAL more amenable to convection. Thus the elevated WV in the upper SAL may have 485 a wider dynamical impact than has been possible to examine here.

486

487 Despite the fact that the observations presented here show enhanced moisture in the upper-SAL 488 compared to background conditions, we do not suggest that the SAL is not a well-mixed, 489 elevated layer. This is illustrated through the schematic shown in Figure 10, which is based on 490 the observations shown in Figure 3. The background (non-SAL) profiles of potential temperature 491 and water vapor mixing ratio are represented by gradually increasing and decreasing values, 492 respectively. Therefore, when a well-mixed layer (the SAL), with near-constant potential 493 temperature and water vapor mixing ratio, is imposed, this results in either a positive or negative 494 anomaly in both its upper and lower portions. Therefore, the results presented in this work do not 495 oppose the previous documentation of the SAL evidencing its well-mixed nature, they simply 496 expose the extension of the well-mixed SAL upwards which results in the cooler, moister 497 anomalies shown in Figure 10. The absolute magnitude of water vapor mixing ratios in the SAL 498 will determine the magnitude of the anomalies relative to the background condition, such that a 499 moister, well-mixed SAL will shift the line of constant q shown in Figure 10 to the right, 500 increasing the moist anomaly.



Figure 10: Schematic depicting anomalies observed in the dusty SAL. SAL conditions are characterized by well-mixed, elevated potential temperature (θ) and water vapor mixing ratios (q). Compared to the background, non-SAL environment, this results in a warmer, drier lower-SAL and a cooler, moister upper-SAL.

507 Gutleben et al. (2019) present airborne observations from a case study in the western Atlantic 508 SAL. Although they find that WV mixing ratios increased in the SAL relative to the free 509 troposphere, their figures show that the SAL was actually drier than the tropical standard 510 atmosphere. Nevertheless, their results show that the increase from dry to moist air descending 511 into the top of the SAL was important for driving strong LW cooling rates at the top of the SAL, 512 similar to the heating rate profiles shown here.

513

514 Adebiyi et al. (2015) present similar findings to this work, but for biomass burning aerosol 515 (BBA) layers over the southeast Atlantic containing enhanced moisture. They find that this 516 anomalous moisture leads to anomalous LW cooling in the layer counteracting the SW heating 517 from the BBA. However, due to the negligible LW radiative effect of BBA, the LW and SW 518 radiative effects of WV combine differently in the results of Adebiyi et al. (2015) to those 519 presented here. Clearly dust layers are not the only type of aerosol layer which can be collocated 520 with enhanced WV, and there may be more instances of transported aerosol layers globally 521 where WV changes exert an effect.

522

As well as influencing the radiative effect, enhanced upper SAL moisture and associated cooling at these altitudes may also influence mid-level altocumulus cloud development. For example, it could be important to the development of the frequently observed altocumulus cloud capping the SABL and SAL, as shown in Kealy et al. (2017) and Mantsis et al. (2020), which play an important radiative role.

528

529 The additional cooling at the top of the SAL due to WV may help to maintain its well-mixed 530 vertical structure, aiding subsidence at the SAL top, combined with ascent at the SAL base due 531 to the well-known lower SAL heating from both dust and water vapor. This is consistent with a 532 lowering of the SAL top towards the west, and a rising of the SAL base (e.g. Liu et al. (2008),

533 Tsamalis et al. (2013)).

534

An interesting question regarding elevated moisture in the upper SAL is whether this is a feature that has simply been under-investigated in the past, or whether the moistening is a temporal 537 trend. Evan et al. (2015) show that WV content in the Saharan heat low region over Africa has 538 increased over the last 30 years as a result of a WV-temperature feedback cycle. Thus it seems 539 plausible that the SAL, outflowing from the Sahara, may be moistening as a consequence. Cold pool outflows (haboobs) play a key role in the transport of both water vapor and dust into the 540 541 Saharan heat low region (Allen et al., 2013; Engelstaedter et al., 2015; Flamant et al., 2007; 542 Marsham et al., 2013; Yu et al., submitted) where it is mixed throughout the deep Saharan 543 boundary layer with dust, before being transported westwards as the SAL. Therefore another 544 question which arises is whether the intensity and/or frequency of cold pool outflows over west 545 Africa has changed over time.

546

547 The ability of enhanced moisture in the upper-SAL to reduce the magnitude of the negative TOA 548 radiative effect of the SAL, shifting it towards positive values, is particularly important when 549 considered with other recent developments in the literature. Adebiyi and Kok (2020) have shown 550 that climate models miss most coarse dust in the atmosphere, and including this missed coarse dust adds a TOA radiative warming effect of 0.15 Wm⁻² globally. Di Biagio et al. (2020) show 551 552 that updated dust optical properties and inclusion of dust sizes larger than 20 µm further shifts 553 the global direct radiative effect of dust away from a cooling effect, to -0.03 Wm⁻². When 554 combined with potential increases in upper SAL moisture, this will further shift the regional 555 radiative effect of dust towards a net warming effect over the tropical Atlantic. The importance 556 of these combined effects will be further increased in regions where dust emissions are 557 increasing over time, exerting a radiative forcing which may tend towards positive values.

558

559 Another pertinent question concerns potential mechanisms which may enable coarse dust 560 particles to be transported further than expected (Mallios et al., 2020; O'Sullivan et al., 2020; Ryder et al., 2019; van der Does et al., 2018; Weinzierl et al., 2017). This work shows that as 561 562 well as a significant day-time shortwave atmospheric column heating from dust, the reduced 563 lower-SAL moisture which occurs under dustier conditions can lead to increased atmospheric 564 longwave heating throughout most of the SAL depth, which would dominate at night. Both these 565 mechanisms may contribute to upward dynamic effects which may increase the lifetime and 566 long-range transport of dust particles, including the coarser sizes. 567

568 Finally there are several uncertainties and limitations inherent in this work. Firstly, the 569 observations, although constituting 24 profiles over the course of 19 days and 5 dust events, are 570 only representative of August 2015 and the eastern Tropical Atlantic, and a wider spatial and 571 temporal analysis would be beneficial. For example, questions such as whether the upper-SAL 572 WV enhancement is present throughout the summer dust season, and how this may vary year to 573 year and westwards across the Atlantic are important.

574

575 The main uncertainty in this work stems from the optical properties of the dust applied in the 576 RTM, which were setup specifically to match the in-situ size and composition results measured 577 during the AER-D campaign (Ryder et al., 2018). As a sensitivity study, optical properties were 578 also selected to represent more extreme values of absorbing dust and less absorbing dust. The 579 impact of the WV perturbation to the DRE is quite sensitive to the optical properties assumed. 580 For example, at the TOA, the total DRE due to dust approaches zero when dust is very absorbing 581 in the SW. Therefore, when the WV changes are also included, which are positive, the final TOA 582 DRE from DU+WV can sometimes change sign, becoming positive. Conversely, for less

absorbing dust with a high SSA, the dust DRE at the TOA is more negative. Therefore,
proportionally the addition of a positive WV DRE results in a smaller fractional decrease in the
negative DRE. Nevertheless, whatever the chosen optical properties of dust are, the radiative
changes due to the WV profile remain important.

587

588 Finally, this work does not account for any changes in humidity upon the microphysical 589 properties of the dust, such as possible hydrophilic growth and changes in scattering properties. 590 Although it is possible that hydrophobic dust particles become more hydrophilic during transport 591 through chemical coatings such as sulfate, there is much uncertainty in understanding of this 592 process and some studies show that this is not an important factor during trans-Atlantic dust 593 transport (Denjean et al., 2015). Additionally, measurements of scattering and absorption at 594 different humidities were not taken during AER-D.

595

596 **5 Conclusions**

597 This work presents results from 24 in-situ aircraft profiles from 5 dust events over the eastern 598 tropical Atlantic during August 2015. It has been shown that as dust loadings increase and the 599 thermal characteristics of the SAL develop, WV mixing ratios decrease in the lower-SAL (p>820 600 hPa) and increase in the upper SAL (480 hPa to 780 hPa) at a mean rate of 3.8 mm per unit 601 AOD.

602

603 Conventionally the SAL is considered to be a warm, dry, elevated, well-mixed layer which is 604 frequently dusty (e.g. Carlson and Prospero (1972)). Therefore the observations of enhanced 605 moisture in the upper SAL under dusty conditions are somewhat contradictory to this 606 description. This can be reconciled by observations presented here showing that the SAL has 607 well-mixed characteristics, but by extending them in altitude where the SAL then becomes 608 cooler and moister than the background conditions.

609

The radiative impact of the observed WV and dusty profiles has been quantified using a radiative transfer model. The enhanced upper-SAL moisture was found to have most effect at the TOA, causing a positive DRE, counteracting the negative TOA DRE exerted by the dust. When both WV and dust were included, the negative DRE at the TOA from dust was reduced in magnitude by up to 64% with a mean of 11%. Thus, accounting for the complex WV profile is crucial in determining the radiative effect of the dusty SAL, and is significantly different to simply assuming a dry profile.

617

The total DRE at the surface and in the atmosphere were dominated by dust effects, but were enhanced by WV reductions in the lower-SAL. Decreases in lower-SAL WV acted to increase the negative surface DRE from dust by 8% and increase atmospheric heating by 17% on average.

621

622 Changes in in the total DRE were driven by WV changes from the longwave spectrum. Less WV

623 in the lower-SAL under dusty conditions resulted in a more negative surface LW DRE due to

624 less emission of LW radiation from WV towards the surface from the lower-SAL. At the same

time, less LW cooling occurred in the drier lower-SAL due to smaller LW absorption. In the LW

626 spectrum, the inclusion of the dry lower-SAL results in change in sign from net cooling to net

heating compared to dust only, and could be important for nighttime dynamics and dust transportwithin the SAL.

629

Heating rate profile changes due to WV and dust in the SAL were calculated. Dust was found to cause a net heating, driven by SW heating, whereas the WV structure generated cooling in the upper-SAL and heating throughout the mid to lower-SAL compared to background conditions, amplifying the heating occurring here due to dust. The upper-SAL cooling under dustier conditions due to WV is consistent with colder upper SAL temperatures in dusty conditions.

635

636 Although this work only represents dust events in the SAL from August 2015, increased 637 moisture in the SAL is consistent with an observed moistening of the Saharan heat low region 638 over west Africa over the last 30 years as part of a feedback cycle driven by increased 639 temperatures (Evan et al., 2015). Cold pool outflows play a crucial role in transporting WV into 640 the Saharan heat low region (Allen et al., 2013; Engelstaedter et al., 2015; Flamant et al., 2007; 641 Marsham et al., 2013). Therefore future work should evaluate the prevalence of moisture on 642 wider temporal and spatial scales than has been possible here, and also examine whether there 643 are temporal trends in the transport of WV and dust northwards into the Sahara by cold pool 644 outflows, potentially as a result of climate change, as well as examining the ability of models 645 with explicit dust schemes to reproduce the dust-WV relationship shown here.

646

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653 **Data**

654 Aircraft data is available at the Centre for Environmental Data Archive at: 655 http://catalogue.ceda.ac.uk/uuid/d7e02c75191a4515a28a208c8a069e70

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