Relative Humidity Dependence of Light Absorption by Mineral Dust after Long-Range Atmospheric Transport from the Sahara

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Abstract

The relative humidity (RH) dependence of light absorption for a Saharan dust-dominated air mass transported to the Gulf of Mexico was measured during the 2006 TexAQS/GoMACCS study using a photo-acoustic absorption spectrometer (PAS). Aerosol absorption was measured at low (25%) and high (73%) RH indicating a 1.5(±0.3) absorption enhancement [$f(RH_{Abs})$] under high RH conditions. $f(RH_{Abs})$ estimates, based on air-mass physical and optical properties and Mie theory modeling, were between 1.2–1.4. Reasons for differences between the measured and modeled $f(RH_{Abs})$ are discussed. The mass absorption coefficient of the long-range transported dust was calculated to be 0.04(±0.02) m^2g^{-1}.

Introduction

The optical properties of atmospheric particles can be dependent on the RH of the atmosphere in which they exist. If particles absorb sufficient water, surface area and scattered light increases. Parameterizations of this RH effect are included in climate models to better predict the radiative forcing of hygroscopic scattering aerosol [e.g. *Schmidt et al.*, 2006]. The ratios between wet and dry scattering or extinction [$f(RH_{Sca})$, $f(RH_{Ext})$] have been measured using nephelometry [*Carrico et al.*, 2003; *Li-Jones et al.*, 1998] and cavity ring down aerosol extinction spectroscopy (CRD-AES) [*Baynard et al.*, 2007] respectively. While $f(RH_{Sca})$ is

relatively well understood from theoretical, laboratory and field investigations, quantification of f(RH) on particle light absorption [$f(RH_{ABS})$] is based on modeling or limited laboratory measurements [e.g. *Lewis et al.*, 2009; *Zhang et al.*, 2008]. Light absorption can be enhanced as a particle is coated in a scattering layer that focuses the radiation onto the core [*Ackerman and Toon*, 1981; *Fuller*, 1995]. *Bond et al.* [2006] calculated absorption enhancements ($E_{Abs}=Abs_{Coated}/Abs_{Non\text{-}Coated}$) for typical atmospheric distributions of black carbon (BC) and semi-volatile coatings and found an E_{Abs} of 1.5 likely. The lack of measurements of $f(RH_{Abs})$ is due to instrument limitations; for example filter-based and photo-acoustic techniques show filter sensitivity to RH and signal loss due to evaporation of semi-volatile material respectively [*Lewis et al.*, 2009; *Murphy*, 2009; *Schmid et al.*, 2006]. Ambient measurement of $f(RH_{Abs})$ using the difference of extinction and scattering is prohibited by the uncertainties of the derived absorption.

Many studies have calculated the radiative impacts of dust and found that the atmospheric burden of 1000–5000 $Tgyr^{-1}$ [*Heintzenberg*, 2009] contributes a substantial climate influence from globally averaged cooling to altered precipitation patterns [*Rosenfeld and Rudich*, 2001; *Shell and Somerville*, 2007; *Solmon et al.*, 2008]. Saharan dust is an extensively studied dust source, can absorb 1 to 10% of the radiation it encounters (i.e. single scattering albedo (SSA) of ~0.90 at mid visible wavelengths) and has variable wavelength absorption [*Mueller et al.*, 2009; *Solmon et al.*, 2008]. The size and optical properties of lofted dust can change as transport occurs; e.g. the SSA can increase and chemical composition change as it becomes internally mixed with hygroscopic materials such as sulfate [*Andreae et al.*, 1986; *Levin et al.*, 1996], sea salt [*Levin et al.*, 2005] and ammonium nitrate [*Noble and Prather*, 1996]. Oxidation of SO_2 to sulfate on dust particles also occurs [*Usher et al.*, 2003]. Up to 80% of

transported dust can be internally mixed with other material [*Andreae et al.*, 1986; *Trochkine et al.*, 2003; *Zhang et al.*, 2003] and suspension of hygroscopic dust into the atmosphere can occur [*Rudich et al.*, 2002]. The $f(RH_{Sca})$ (and size growth due to RH changes, $g(RH_{SCA})$) of mineral dust aerosol have been measured to be between 1.1 and 1.7 [*Carrico et al.*, 2003; *Howell et al.*, 2006; *Kaaden et al.*, 2009; *Li-Jones et al.*, 1998], with lower values usually corresponding to dust measured close to the source region. *Carrico et al.* [2003] concluded that, due to the high ratio of external to internal mixing, there was no appreciable change to the radiative properties due to dust becoming internally mixed with hygroscopic material, rather the RH response in scattering was dominated by the externally mixed aerosol. *Bates et al.* [2006] reviewed the radiative impact of RH on dust aerosol and concluded that the effect was negligible and not necessary to accurately model the direct radiative effect. To date, there have not been any field measurements of $f(RH_{Abs})$ of dust or BC. Here we present $f(RH_{Abs})$ measured for dust transported from the Saharan region of Africa to the Gulf of Mexico.

Field Observations

Chemical, optical and physical properties of ≤10μm aerodynamic diameter (Dp<10μm) aerosol were measured during the 2006 TexAQS/GoMACCS study onboard the NOAA R/V *Ronald H. Brown* (RHB) and are described in *Quinn & Bates* [2005] and *Bates et al.* [2008]. Mass concentrations (±11%) of particulate sulfate, nitrate, ammonium, organic matter (POM), EC and inorganic oxidized material (IOM) (dust proxy) were measured using impactor filters over 8–12 hour periods [*Quinn and Bates*, 2005]. Aerosol extinction (±1% @ ~25% RH) and $f(RH_{Ext})$ (±5% @ 75% RH) were measured at 532nm using CRD-AES [*Baynard et al.*, 2007]. Aerosol absorption (±5% @ ~25% RH) was measured at 532nm using PAS [*Lack et al.*, 2006] and at 467nm, 530nm and 660nm using a particle soot absorption photometer[1] (PSAP, ±~30%)

[*Lack et al.*, 2008]. During a dust-dominated period, the PAS was re-configured to sample at high RH (~73%). RH was measured using a Vaisala[1] HMP50 probe with an uncertainty of ±3%. The sample was passed through a wet permeable membrane (max. RH 90%) and the RH reduced by adjusting sample flow rate. Waterproof microphones (Knowles Acoustics WP3502[1]) were used during the RH trial and the resonant frequency recalibrated at RH changes to remove instrument RH dependence. Data collection for the CRD-AES and PAS was interrupted before and after the RH trial (and during for the CRD-AES) due to the instrument plumbing changes required to elevate the RH.

Results

The $f(RH_{ABS})$ of dust-dominated particles was measured during a major dust *'event'* (27-30th August 2006). Dust was indentified as the dominant PM constituent using a combination of measurements: Figure 1a shows the wavelength dependence for absorption ($Å_{Abs}$), for the entire field campaign ranging from 0.5-6 with larger $Å_{ABS}$ usually coinciding with the enhanced dust mass. Dust shows enhanced absorption at shorter wavelengths, resulting in a $Å_{Abs}$ much greater than 1 [*Linke et al.*, 2006]. Across the event of interest dust was found to contribute 70% of Dp<10μm particle mass (41.4 μgm^{-3}) (Figure 1b) and the aerosol had a volume median aerodynamic diameter of 2.1μm (Figure 1c). Particle backscatter and vertical turbulence profiles measured using Doppler LIDAR [*Tucker et al.*, 2009] showed layers at 1-1.5 km mixing down to the surface around 1200 UTC 27th of August. Footprint emission sensitivities from the Flexpart Lagrangian dispersion model [*Stohl et al.*, 2005] indicate a North African source for the air-mass 10-15 days prior to measurement in Galveston Bay, Texas (Figure 1d). Average SSA of the event was 0.95(±0.01) and average extinction and

[1] Certain commercial equipment is identified in this article to adequately specify the experimental procedure. Such identification does not imply recognition or endorsement by NOAA nor does it imply that the equipment is necessarily the best available for the purpose.

absorption coefficients were 50 and 2.5 Mm^{-1} respectively (@ 532nm). Average $f(RH_{EXT})$ dropped from ~2.4(±0.3) prior to the event to ~1.9(±0.3) during the event (at 73% RH), higher than the expected hygroscopicity for pure dust. Hence, the following results are due to a particle ensemble of mixed composition rather than pure dust. Over a 1-hour period on the 29th August 2006 the PAS was configured to measure dry (~25%) and high RH (~73±3% RH) absorption. A difference in the absorption between RH levels was measured (Figure 2a). Dry absorption was monitored using a PSAP (Figure 2a, black line) and shown to be stable during the high RH trial (although PSAP > PAS by ~1 Mm^{-1}). Absorption data (1 sec.) 10-15min. either side of the RH change were used to produce absorption histograms. Gaussian curves were fit to each histogram and an $f(RH_{Abs})$ of ~1.50(±0.34) was calculated from the ratio of the mean values of the Gaussian distributions (Figure 2b). A 2-sample t-test was used to calculate the standard deviation in the $f(RH_{ABS})$.

Discussion

Photo-Acoustics

The photo-acoustic technique provides a lower limit on absorption due to potential PAS signal reductions introduced by evaporation of water on the particle (at high RH) and internal heat losses at large particle sizes. Utilizing the work of *Murphy* [2009] we calculate, for the size and assumed optical properties of the dust in this study, that a 7-30% reduction in $f(RH_{Abs})$ can be expected due to the above mentioned effects (see supplementary material) depending on the assumption of water accommodation coefficient.

$f(RH_{ABS})$ Simulations

Using core-shell Mie theory [*Bohren and Huffman*, 1983] and assumed particle properties we show that an $f(RH_{Abs})$ of up to 1.4 could be expected for the dust event studied. Dust particles

are rarely spherical and assuming sphericity (to simplify the calculations) tends to over estimate scattering and absorption (depending on non-spherical shape) [*Bohren and Huffman*, 1983]. As the particles are coated they will become more spherical and so our calculations will underestimate $f(RH_{Abs})$. We use optical properties representative of lofted African dust for Summer/Autumn [*Israelevich et al.*, 2003] (refractive index [RI] of 1.45 – 0.015i) and assume an aqueous sulfate or NaCl coating (RI of 1.4 + 0.0i) [*Tang and Munkjelwitz*, 1994]. We calculate that an average 25% diameter growth of 2.1μm dust particles is required to produce the measured $f(RH_{EXT})$ of 1.9. This 25% growth is used as a center point in growth for further modeling. Next we calculate the theoretical $f(RH_{Abs})$ using Mie theory, the average measured size distribution (Figure 1c), a real RI for dust of 1.45, coat RI of 1.4 + 0.0i and a range of imaginary RI for dust from literature [*Balkanski et al.*, 2007; *Israelevich et al.*, 2003; *Mueller et al.*, 2009]. The size distribution is grown by the fraction on the y-axis of Figure 3 which shows that $f(RH_{ABS})$ of 1.2-1.4 would be common across a range of particle growth and imaginary RI, even at the lower imaginary RI for dust measured in the SAMUM experiment [*Mueller et al.*, 2009]. The measured $f(RH_{Abs})$ of 1.5 (black line, Figure 3) is unlikely for the assumed properties if one considers that the 25% growth in diameter calculated above (grey line, Figure 3) would be for a full internal mixture. If we assume a diameter growth of 15% (a growth approximating ~50% internal mixture [*Trochkine et al.*, 2003]) an $f(RH_{ABS})$ of 1.25-1.30 is modeled (black dot, Figure 3). Given that studies have reported 5 to 80% internal mixing [*Andreae et al.*, 1986; *Zhang et al.*, 2003], the $f(RH_{Abs})$ expected for dust-dominated particles may range from 1 to 1.4. Modeling also showed that the $f(RH_{Abs})$ will be larger if the absorbing component of the dust is concentrated (i.e. higher imaginary RI) and mixed with non-absorbing quartz (not shown here).

Mass Absorption Coefficient (MAC)

The MAC for dust is calculated to be 0.04(±0.02) m^2g^{-1} (532nm). We estimate that the 0.1(±0.1) μgm^{-3} EC (Figure 1b) contributed 0.75 Mm^{-1} of the total 2.5(±0.2) Mm^{-1} absorption (assuming EC MAC of 7.5 m^2g^{-1}). Therefore the absorption of 37.4(±5.0) μgm^{-3} dust was 1.75(±0.75) Mm^{-1}. The calculated MAC for the long-range transported dust was therefore 0.04(±0.02) m^2g^{-1}; similar to recent studies [*Yang et al.*, 2009]. The MAC used for EC is usually applied to fresh EC emissions and can reasonably increase by 50% when coated by scattering material [*Bond et al.*, 2006]. A 50% increase in absorption by EC would contribute ~0.40 Mm^{-1} to the $f(RH_{Abs})$ shown in Figure 2b, reducing the discrepancy between measured and modeled $f(RH_{ABS})$ identified above.

Radiative Impacts

We have shown that water uptake by dust affects both scattering *and* absorption and thus SSA. SSA is an important factor that influences the global radiation balance [*Balkanski et al.*, 2007] and predicted precipitation patterns over Western Africa [*Solmon et al.*, 2008]. *Carrico et al.* [2003], *Howell et al.* [2006] and *McNaughton et al* [2009] concluded that RH-induced scattering changes within mixed dust/pollution aerosol would be negligible due to the dominance of external mixing (i.e. interaction of dust and pollution would be minimal) and a cancellation of scattering due to redistribution of hygroscopic material from the fine to coarse modes when internal mixing occurred. Those studies did not consider an $f(RH_{Abs})$ effect. To gain an insight to this the impact of RH on the SSA of the dust-dominated particles studied, we incorporated the $f(RH_{ABS})$ effect into Mie theory. Figure 3b shows that the SSA expected for humidified dust-dominated aerosol with the $f(RH_{Abs})$ effect can be reduced from 0.01 – 0.03 compared to the SSA when $f(RH_{Abs})$ is not considered. This reduction is for reasonable growth

conditions and does not consider optical changes due to redistribution of externally mixed material, which will complicate the impact of the $f(RH_{Abs})$ effect. This result is for long-range transported, well-aged dust and likely represents the end of a continuum beginning at the dust source (i.e. these results may not be as significant for less aged dust).

Summary

Photo-acoustic absorption spectroscopy was used in the field to measure an absorption enhancement from water uptake [$f(RH_{Abs})$] of 1.5(±0.3) (RH ~73%) for Saharan dust-dominated aerosol. Simulations of the $f(RH_{Abs})$ for the aerosol encountered showed $f(RH_{Abs})$ of 1.2-1.4 is possible given assumed particle properties and that SSA can be reduced by 0.01–0.03. A small amount of strongly absorbing EC, or more absorbing dust mixed with non-absorbing quartz (rather than uniformly mixed absorbing dust) may resolve the difference in modeled and observed $f(RH_{Abs})$. The observed $f(RH_{Abs})$ shows that PAS is capable of measuring an $f(RH_{Abs})$ signal and that dust aerosol can exhibit $f(RH_{Abs})$ due to water uptake by internally mixed hygroscopic material. It is recognized that this analysis is based on a small subset of data however the unique insights gained show cause for further investigation.

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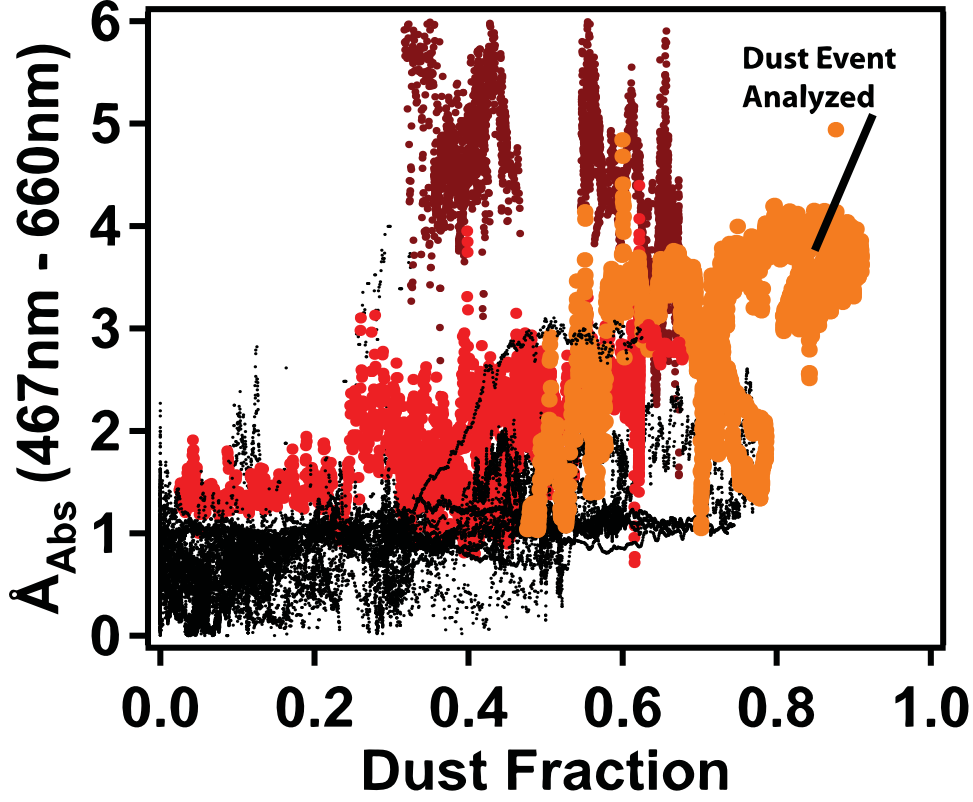
Figure Captions

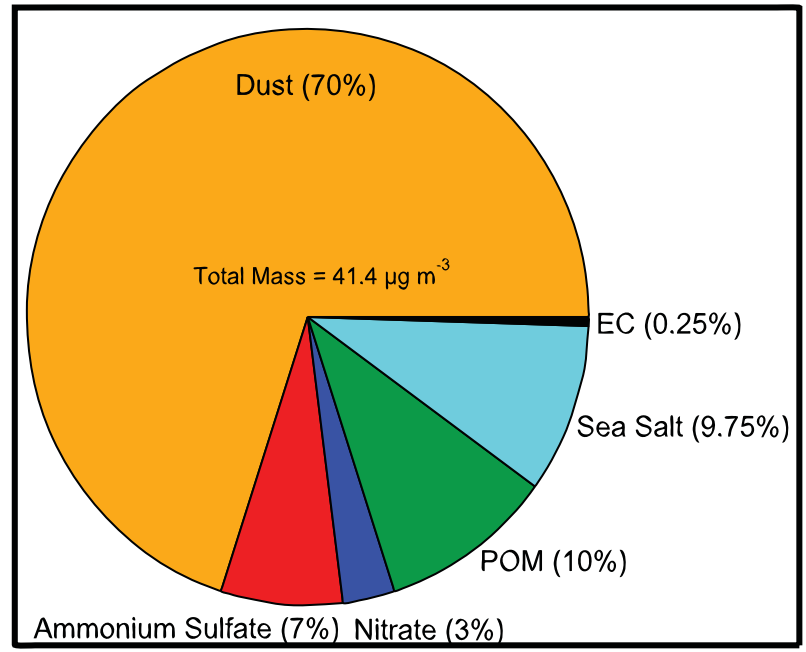
Figure 1 a) Absorption wavelength dependence ($Å_{ABS}$) vs dust fraction of total mass for all data. Major dust events shown in color, other data in black. b) Chemical composition of particles sampled during the dust event of interest. c) Average volumetric size distribution of the particles during the event of interest. d) Flexpart footprint emission sensitivity corresponding to the 29 August 2006 sampling period.

Figure 2 a) PAS absorption (grey symbols), PSAP absorption (black line) and RH (grey line) time series. b) Representation of the RH dependence of absorption of Saharan dust-dominated particles.

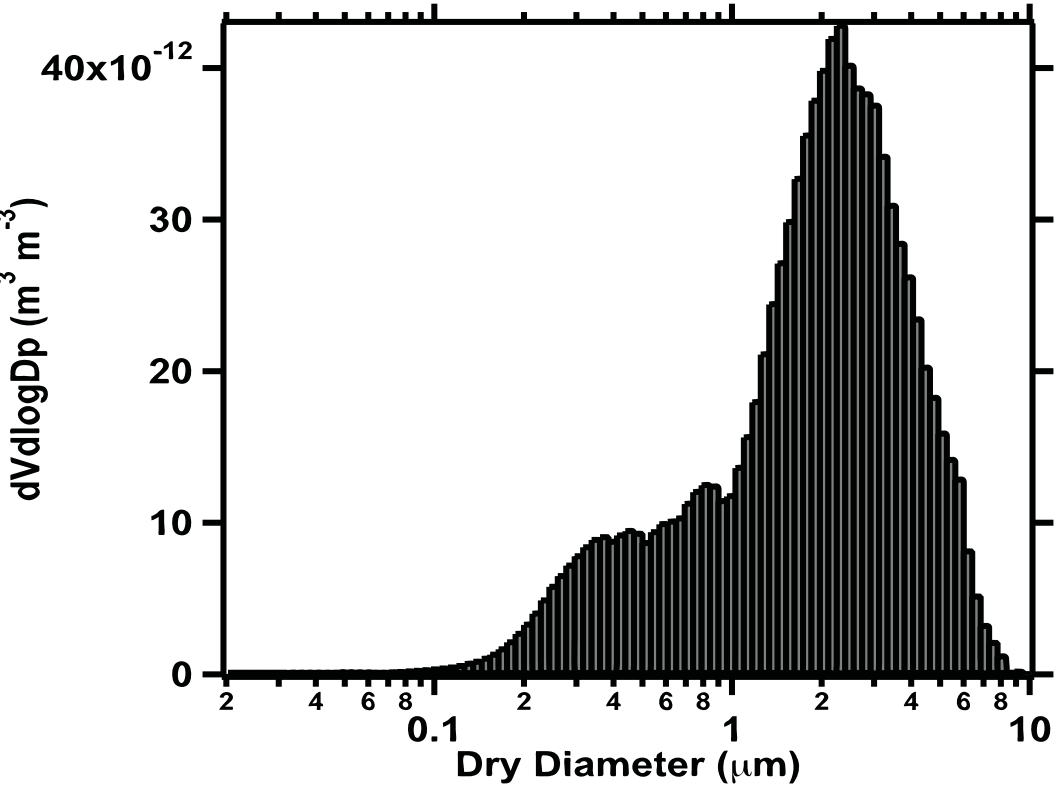
Figure 3, a) Modeled $f(RH_{Abs})$ for the dust-dominated particles encountered in this study (Figure 1c). See text for model assumptions. Dashed line: $f(RH_{ABS})$ of an internal mixture for this study. Black symbol: $f(RH_{Abs})$ for a ~50% internal mixture with dust RI from literature. Black line: $f(RH_{Abs})$ of 1.5. b) SSA reduction expected by including $f(RH_{Abs})$ effect relative to humidification of dust-dominated particles without $f(RH_{Abs})$.


Dust Event Analyzed
Å_Abs (467nm - 660nm)
Dust Fraction
0.0
0.2
0.4
0.6
0.8
1.0
0
1
2
3
4
5
6

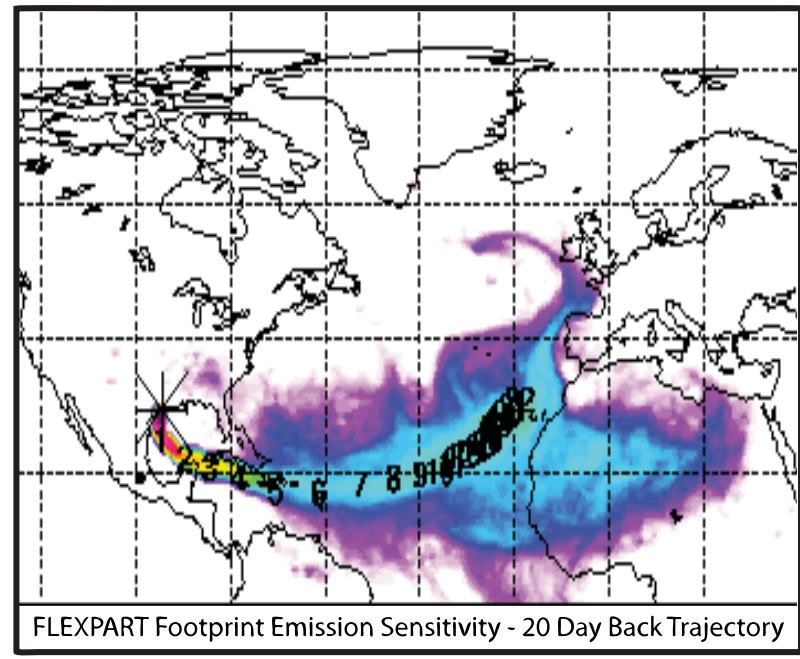
a)


Dust (70%)
Total Mass = 41.4 µg m-3
EC (0.25%)
Sea Salt (9.75%)
POM (10%)
Ammonium Sulfate (7%)
Nitrate (3%)

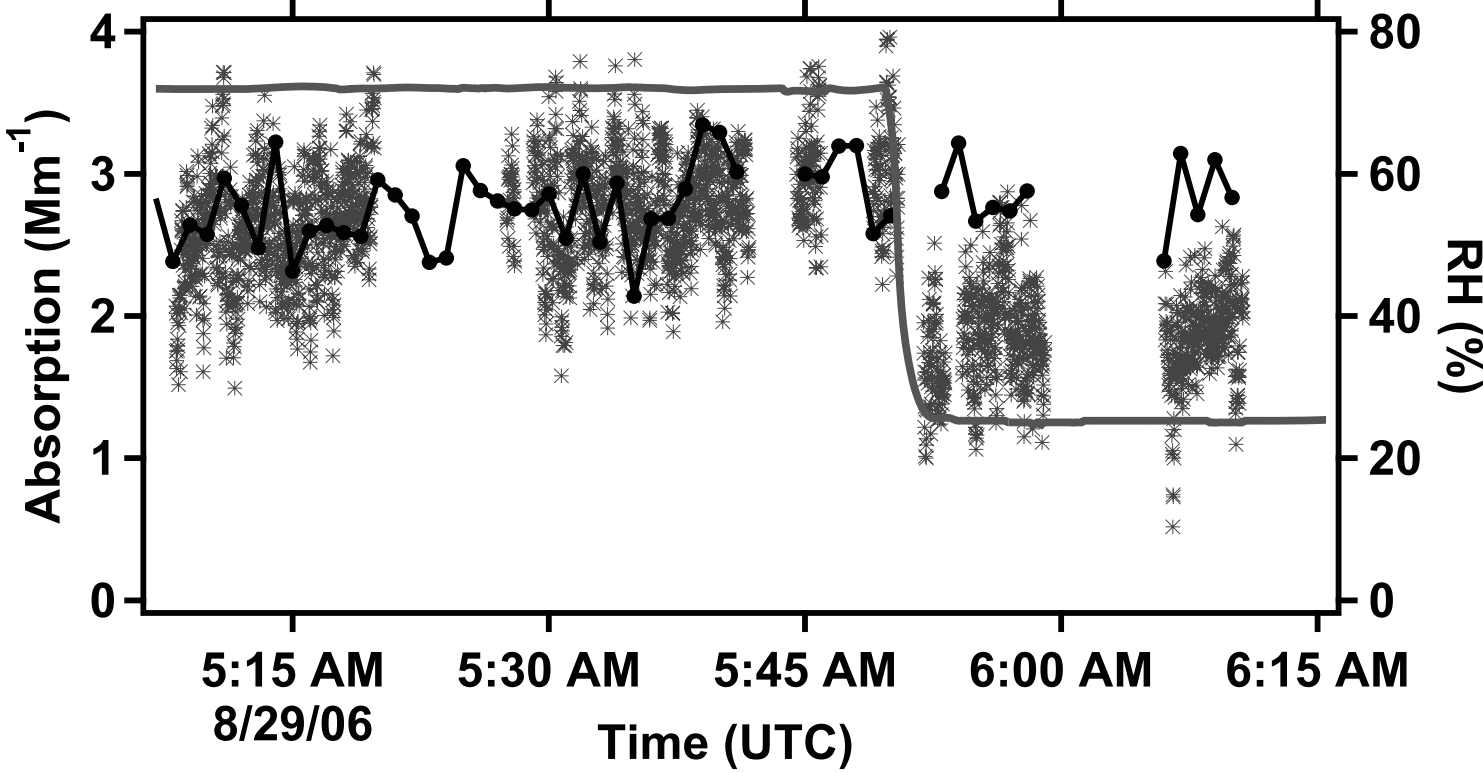
b)


40x10-12
30
20
10
0
dVdlogDp (m3 m-3)
0.1
1
10
Dry Diameter (µm)

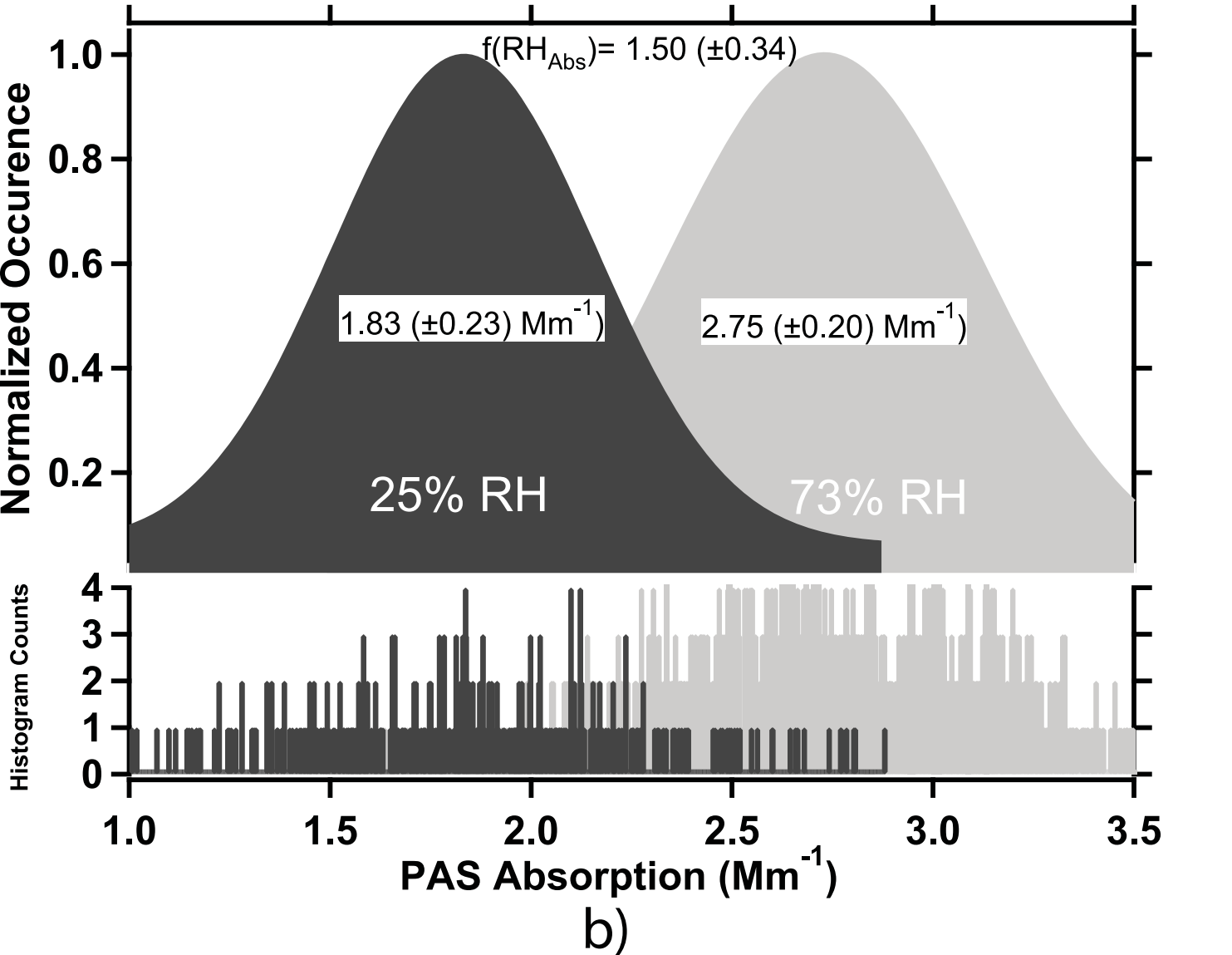
c)


FLEXPART Footprint Emission Sensitivity - 20 Day Back Trajectory

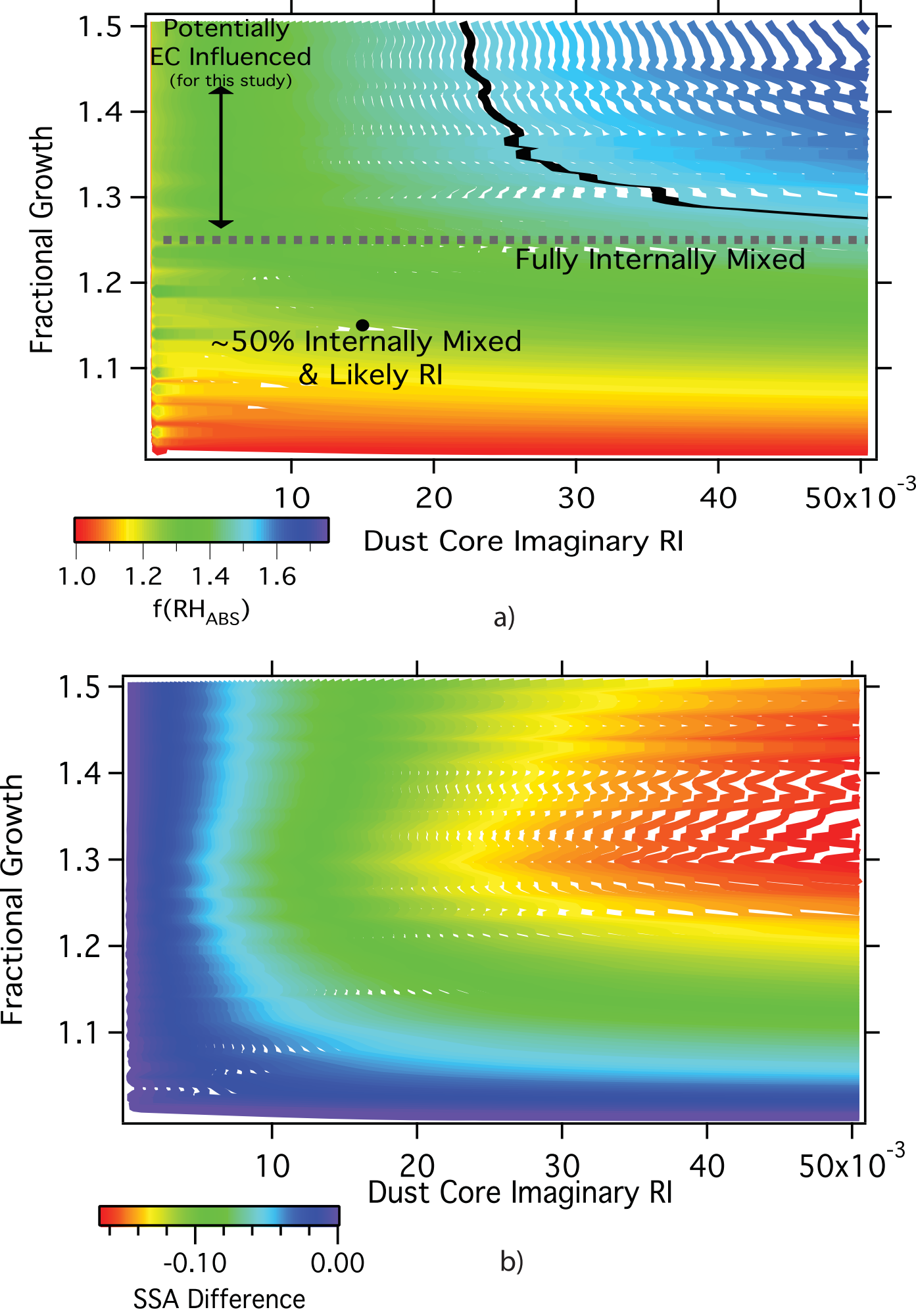
d)


Absorption (Mm-1)
RH (%)
0
1
2
3
4
0
20
40
60
80
5:15 AM
8/29/06
5:30 AM
5:45 AM
6:00 AM
6:15 AM
Time (UTC)

a)


f(RHAbs)= 1.50 (±0.34)
Normalized Occurence
1.83 (±0.23) Mm-1)
2.75 (±0.20) Mm-1)
25% RH
73% RH
Histogram Counts
PAS Absorption (Mm-1)
1.0
1.5
2.0
2.5
3.0
3.5

b)


Potentially
EC Influenced
(for this study)
Fully Internally Mixed
~50% Internally Mixed
& Likely RI
Fractional Growth
1.5
1.4
1.3
1.2
1.1
10
20
30
40
50x10-3
Dust Core Imaginary RI
1.0
1.2
1.4
1.6
f(RHABS)
a)
Fractional Growth
1.5
1.4
1.3
1.2
1.1
10
20
30
40
50x10-3
Dust Core Imaginary RI
-0.10
0.00
SSA Difference
b)