Characterization of Aerosol Hygroscopic Growth Properties During the DC3 Campaign

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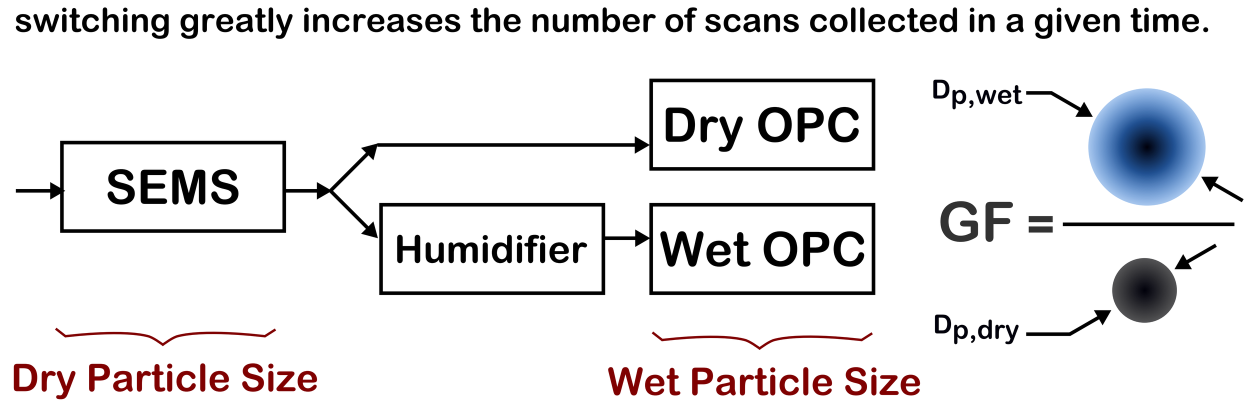
The DASH-SP on the NASA DC-8

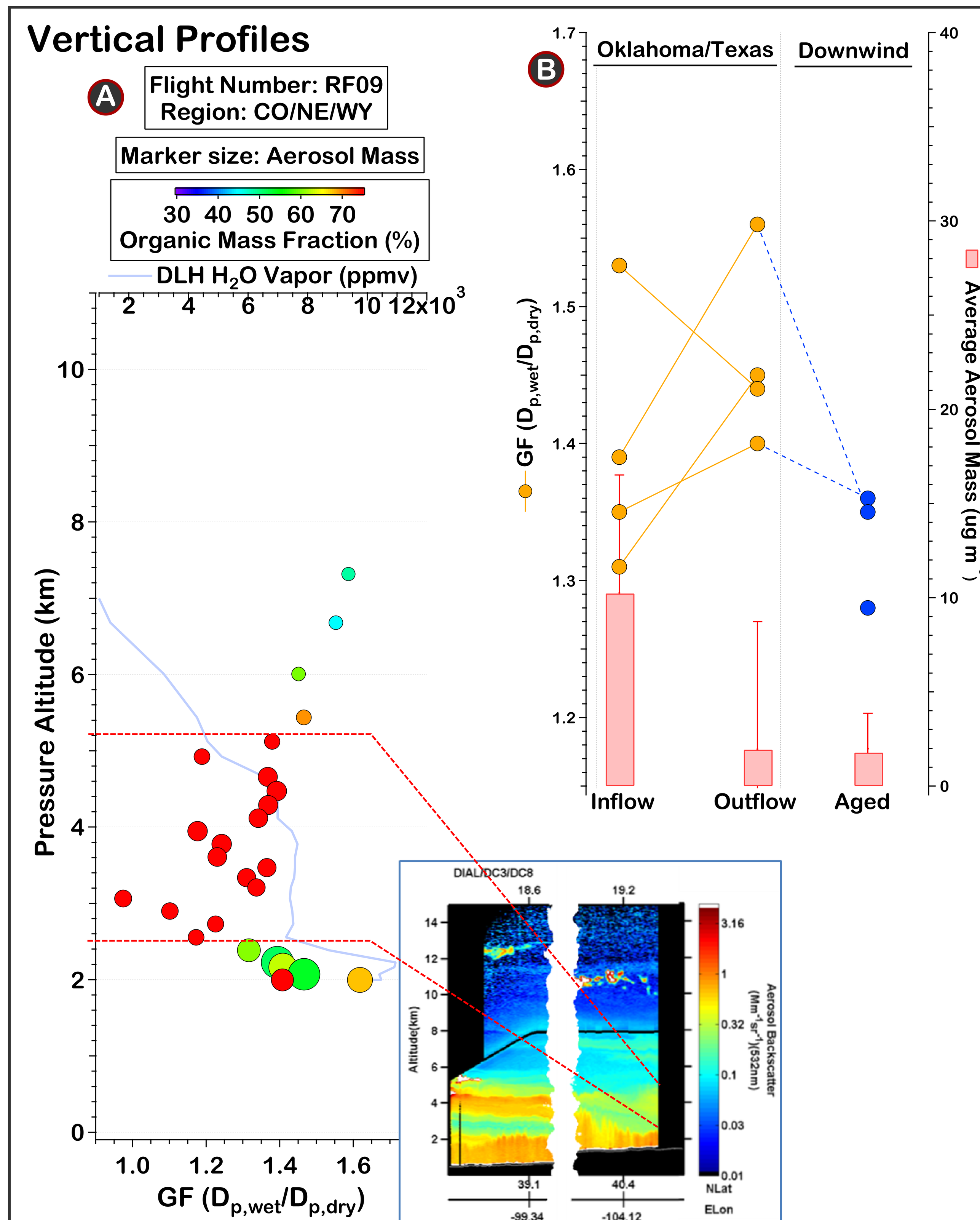
Aerosol particles can scatter and absorb solar radiation and serve as cloud condensation nuclei (CCN), thereby influencing cloud properties and precipitation. The radiative and CCN-relevant properties of particles depend on their hygroscopicity. As particles are exposed to changing relative humidity (RH), they undergo a change in their size owing to hygroscopic growth (via water-uptake) with the magnitude of this growth dependent on their chemical composition.

The Differential Aerosol Sizing and Hygroscopicity - Spectrometer Probe (DASH-SP; Brechtel Mfg. Inc.; Sorooshian et al., 2008) is capable of rapidly measuring size-resolved hygroscopic growth factors (GFs) of ambient aerosol particles. GF values are defined as the ratio of a particle's diameter when exposed to elevated RH values (typically ~70-95 %) vs the particle's diameter when dried to <15 % RH. The polydisperse aerosol sampling stream is first size selected using a Scanning Electrical Mobility and Sizer (SEMS) to create a monodisperse aerosol flow at a single diameter between 175 - 350 nm. A flow splitter separates the monodisperse aerosol sample into two paths with equivalent total residence times through the "dry" and "wet" optical particle counters (OPCs). The humidifier has the ability to quickly equilibrate the sample flow to the desired RH before reaching the OPC without diluting the sample with dry/filtered air.

The DASH-SP instrument has much faster time resolution than widely used alternative hygroscopicity measurement techniques, including the HTDMA. Single scans can be acquired in as little as 3 s. The average uncertainty associated with DASH-SP GF measurements is ± 4.3 %. A range of scans covering $D_{p,dry}$ sizes 175 - 350 nm at a single RH can be completed in under 5 minutes with the majority of the time being required by the SEMS between size changes. Performing replicate scans at each dry size before switching greatly increases the number of scans collected in a given time.



Vertical Profiles



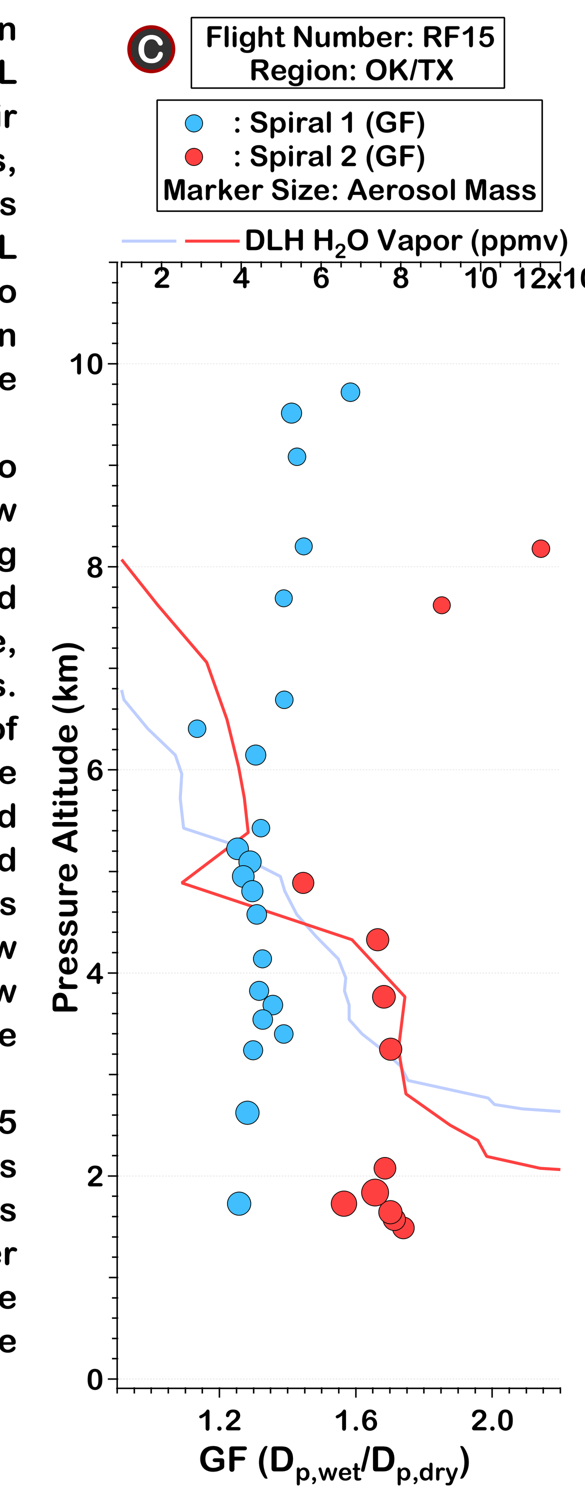


Figure 1.

A) The GF response during a spiral with multiple aerosol layers is shown. A separate air mass between ~ 2.5 - 5 km was also confirmed by the DIAL instrument. There appears to be three distinct air masses present: a surface layer with higher total mass, a central aerosol layer with suppressed GFs and less aerosol mass, and the upper layer being above the PBL - with notably higher GFs. An area of interest is to investigate the prevalence of external mixtures in different air masses and to quantify GFs for multiple modes in single scans.

B) One of main objectives of the DASH-SP is to measure differences in GF values between the inflow and outflow of active convection during thunderstorms. Regions of each flight were classified as "inflow" or "outflow" based on the DC-8's altitude, proximity to thunderstorm, and operational flight notes. Average GF values were plotted for each regime of each flight and the dashed blue lines connect the same air masses to the following day's downwind aged sampling. Only GFs for the OK/TX region are displayed here, though the other regions had similar patterns with varied magnitudes (in total, 7 flights had outflow GF values greater than the corresponding inflow values; 4 of the 5 flights with lower outflow GFs were flights with smoke present).

C) Two spirals were conducted during RF15 (RH ~85 %, $D_{p,dry}$ = 225 nm) yielding a significant shift towards higher GF values in the second spiral. These changes will be analyzed in an attempt to determine whether this is a result of air parcel processing during active convection, or a direct result of spatial variances in the sampling regions.

Restructuring of Biomass Burning Particles

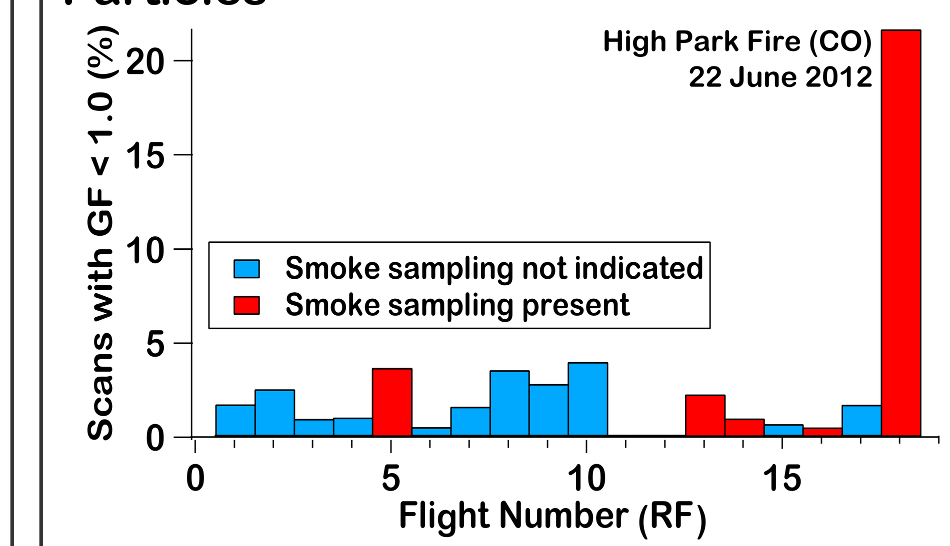
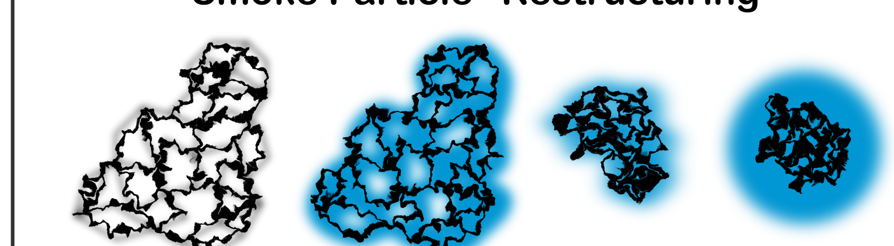


Figure 3. Above: Percentage of scans during each flight with GF values less than 1.0. This is a possible indication that particles are undergoing restructuring in smoke-influenced aerosol particles.

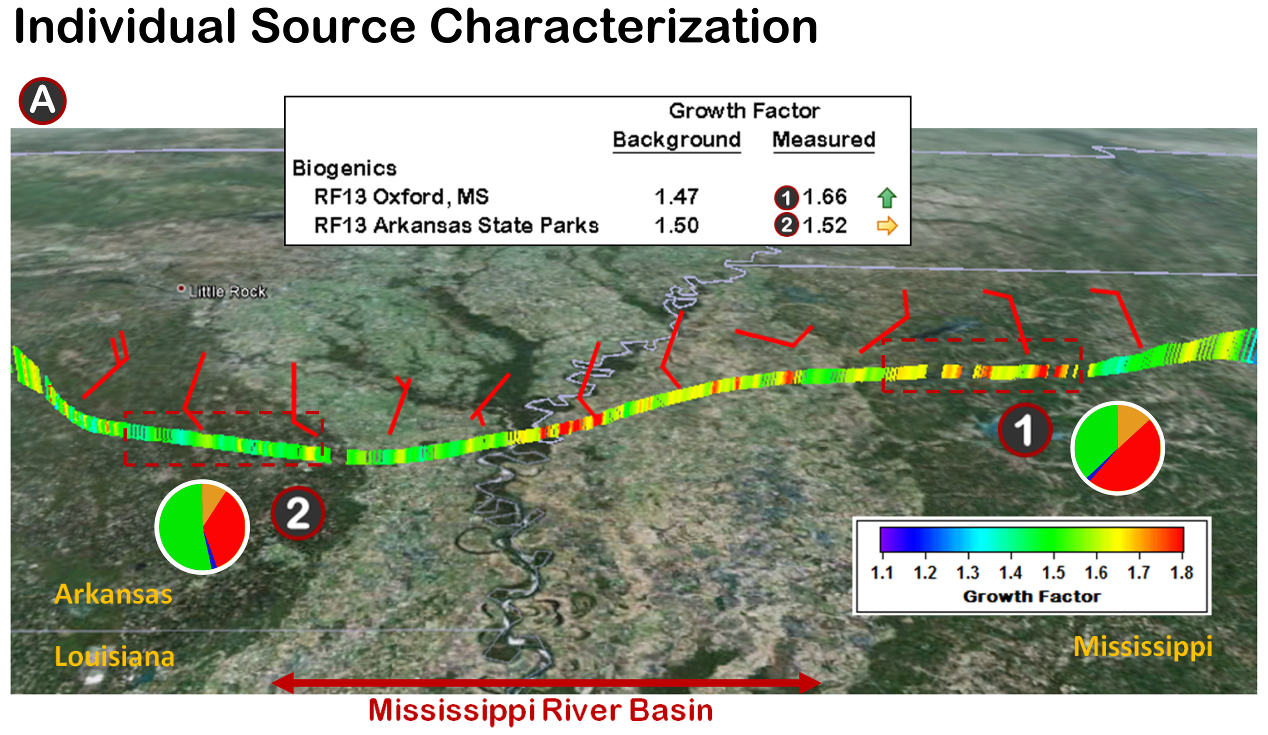
Below: Soot particle restructuring has been seen previously (Martin, 2013) and occurs when the soot's fractal structure collapses during hydration, yielding a smaller wet size than the initial dry particle size. It is believed that this process may be taking place inside the DASH-SP instrument and the apparent suppression in GFs will be investigated to determine if restructuring accounts for these results.

Smoke Particle "Restructuring"



Fractal structure collapse upon hydration

Individual Source Characterization



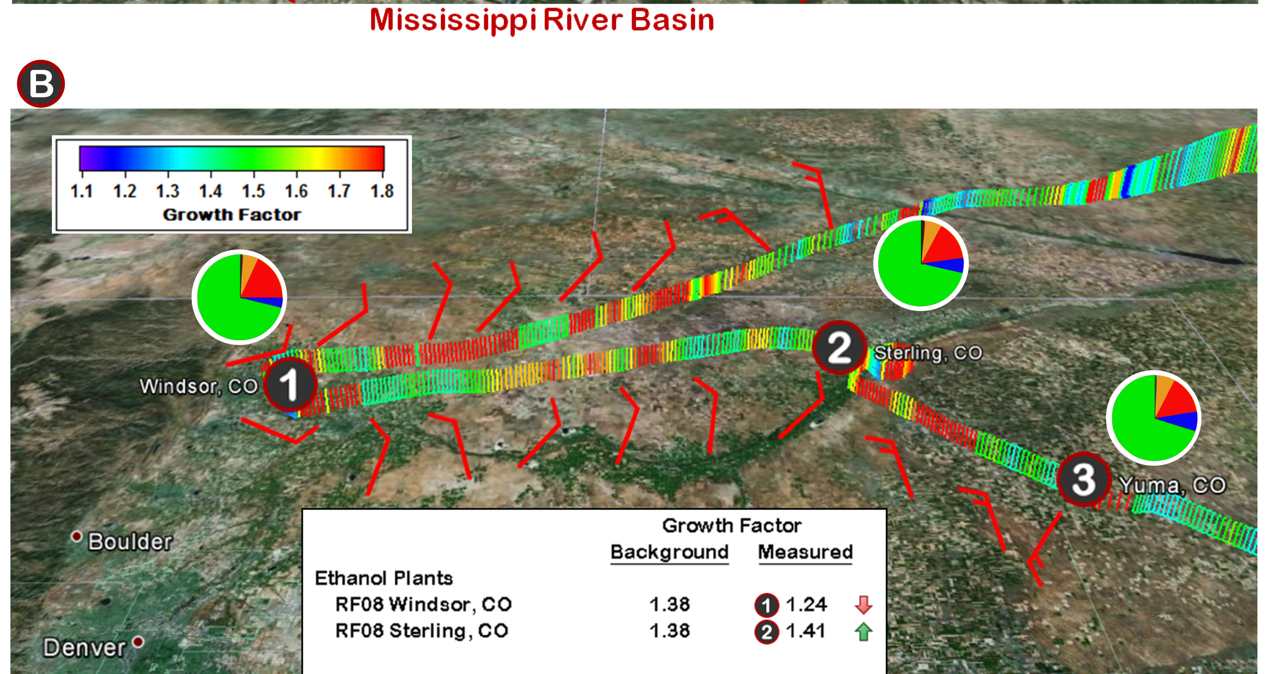


Figure 2. A) Biogenic source emissions during RF13 sampled from both sides of the Mississippi river in Mississippi and Arkansas. The two different sources were identified by very high isoprene concentrations (>2 ppbv). Region 1 is located between Tombigbee and Holly Springs National Forests and the flight path transected the town of Oxford, MS. Both regions were sampled at a DASH-SP RH of ~85%. The increase in GF over the background in region 1 is likely due to the enhanced sulfate mass fraction (48%) which is due to anthropogenic emission contributions. However, in all regions with isoprene concentrations above 1 ppbv, the GFs are above normal (1.48 vs 1.40, respectively at ~85 % RH). Further analysis will be directed at determining various biogenic constituents' role in modeling GF values.

B) Specific source locations were targeted in RF08 in NE CO, including large ethanol production facilities (in all three towns displayed) and feed lots (primarily in Yuma, CO). All three sampling sites are easily identified by total aerosol mass concentration spikes (>60 % above background) and Yuma by an additional minor spike in nitrate mass fraction (>10 %). Despite very similar bulk species concentrations, GF values at a fixed DASH-SP RH (~88 %) respond differently in each region. These results will be investigated further with size-resolved AMS species data to determine what source contributions have more of an influence on the actual GF values in the size range of the instrument.

C) Characteristic source tracers were used to classify certain regions as under the influence of a specific source. Emissions from biogenic regions were categorized by increased concentrations of isoprene (>1 ppbv) and smoke sources were detected by increased levels of acetonitrile (> background level) and corroborated by in-flight operations notes. All source region tags are identified by GPS location near sampling. Differences in measured source GF values over background values listed.

D) Smoke influences on tropospheric air masses were investigated during RF18 in NE CO. The High Park Fire started burning on 9 June 2012 west of Ft. Collins, CO and had burned ~70,000 acres by 22 June 2012 when the DC-8 sampled the region. The DC-8 sampled near the source of the fire and GF values of 1.28 were measured at a DASH-SP RH of 83 % and a $D_{p,dry}$ of 250 nm (compared to a regional background average of 1.38). Winds were blowing from the SW and downwind transects through the major plume were conducted ~275 km from the source. These two transects had GFs of 1.28 and 1.16 when sampled at ~83 % RH and a $D_{p,dry}$ of ~250 nm. A possible explanation for this GF suppression is discussed in Figure 3.

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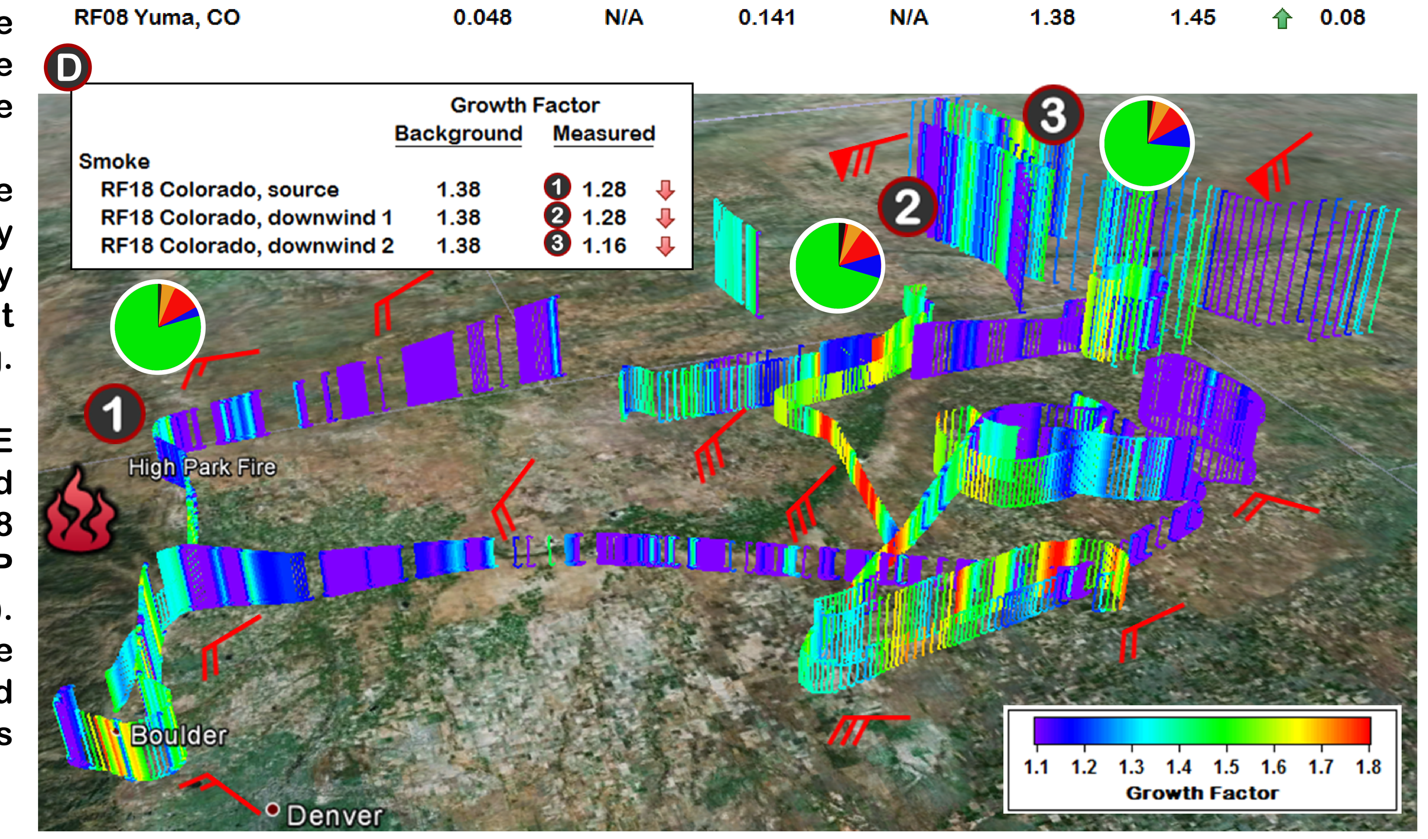
	Isoprene (ppbv)		Acetonitrile (ppbv)		Growth Factor (RH ~85 %)		
	Background	Measured	Background	Measured	Background	Measured	ΔGF
Biogenics							
RF05 Lake of the Ozarks, 1	0.687	5.067	0.117	0.149	1.50	1.52	⇨ 0.01
RF05 Lake of the Ozarks, 2	0.687	5.056	0.117	0.144	1.50	1.33	⇩ (0.18)
RF06 Central OK, 1	0.130	0.509	0.167	N/A	1.44	1.32	⇩ (0.11)
RF06 Central OK, 2	0.130	0.478	0.167	N/A	1.44	1.41	⇩ (0.03)
RF13 Savage Gulf Area	0.369	0.899	0.131	0.117	1.47	1.53	⇧ 0.05
RF13 Oxford, MS	0.369	1.621	0.131	0.124	1.47	1.66	⇧ 0.19
RF13 Arkansas State Parks	0.776	3.774	0.157	0.127	1.50	1.52	⇨ 0.02
Smoke							
RF05 Arkansas	0.687	0.007	0.117	0.219	1.50	1.09	⇩ (0.42)
RF13 Alabama	0.369	0.000	0.131	0.196	1.47	1.45	⇩ (0.02)
RF14 Colorado	0.048	0.239	0.141	0.163	1.38	1.17	⇩ (0.21)
RF18 Colorado, 1 (High Park)	0.048	0.411	0.141	0.348	1.38	1.28	⇩ (0.10)
RF18 Colorado, 2 (Downwind 1)	0.048	0.229	0.141	0.290	1.38	1.28	⇩ (0.10)
RF18 Colorado, 3 (Downwind 2)	0.048	0.200	0.141	0.259	1.38	1.16	⇩ (0.22)
Ethanol Plants							
RF08 Windsor, CO	0.048	N/A	0.141	N/A	1.38	1.24	⇩ (0.14)
RF08 Sterling, CO	0.048	0.024	0.141	0.125	1.38	1.41	⇧ 0.03
Feed Lots							
RF08 Yuma, CO	0.048	N/A	0.141	N/A	1.38	1.45	⇧ 0.08



Key Research Interests

1) How the prevalence of hygroscopic aerosol properties vary as a function of region. Quantify sensitivity of GF to individual emission point sources (large scale fuel production, agricultural operations, biogenic sources, biomass burning, urban, etc…).

2) The role of cloud processing and convective pumping in altering aerosol hygroscopic properties. How GFs vary between non-convective conditions and inflow/outflow regimes of thunderstorms.

3) The nature and degree of aerosol hygroscopicity variability as a function of altitude up to UTLS.

4) Examining the prevalence and nature of multiple hygroscopic modes in air parcels during single DASH-SP scans.

5) Critical evaluation of existing parametrization for aerosol water-uptake in the sub- and super-saturated regimes. Compare GF to CCN activation ratios and composition.

6) Soot particle restructuring in the DASH-SP and the high occurrence of sub 1.0 GFs during smoke events.

References

Martin, M. et al., J. Aerosol Sci., 56 (2013) 15-29.
Sorooshian, A. et al., Aerosol Sci. Tech., 42 (2008) 445-464.

Acknowledgements

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