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Background

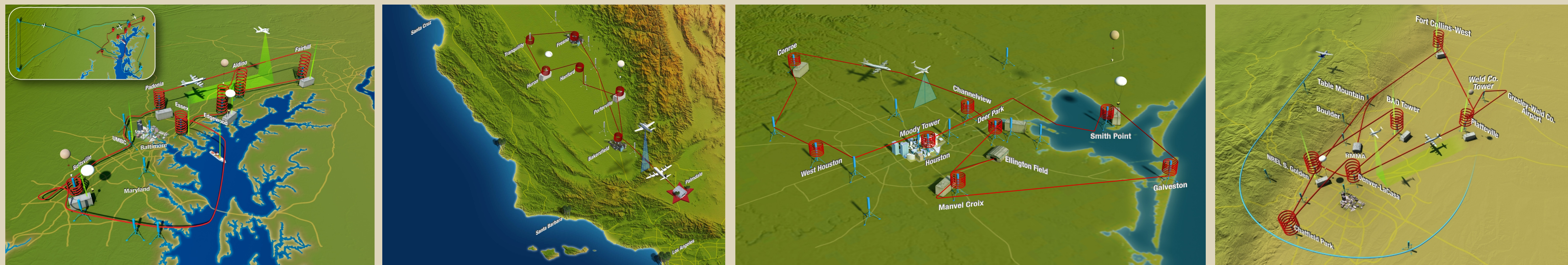
Light absorbing carbonaceous aerosols are known to be an important climatic driver with a global radiative forcing of about half (IPCC, 2013) to two-thirds (Bond et al., 2013) that of the dominant greenhouse gas, carbon dioxide. While the mass absorption coefficient of pure black carbon (BC) is fairly well known, observational evidence suggests that BC rapidly mixes with other aerosol chemical components within hours after emission (Moffet and Prather, 2009; Moteki et al., 2007). These other components may include predominantly scattering organic, sulfate, and nitrate species, as well as light-absorbing, so-called "brown carbon" (BrC). It has been suggested that the presence of these BC-mixed components may induce mixing-state-dependent lensing effects that could potentially double the BC direct radiative forcing (Jacobson, 2001). The key to better understanding how BC-rich aerosols are distributed in the atmosphere is to examine an unbiased set of measurements covering broad spatial and temporal scales; however, many past airborne field campaigns have specifically targeted source plumes or other scientifically-relevant emissions sources. The recent NASA DISCOVER-AQ campaign is unique in that approximately the same flight pattern was performed over a month-long period in each of four different U.S. metropolitan areas, ensuring an unbiased, or at least less biased, data set with both wide horizontal and vertical (up to 5 km altitude) coverage. More details as well as data from the four deployments can be obtained from the project website (<http://discover-aq.larc.nasa.gov>).

Baltimore-Washington (July 2011)

San Joaquin Valley (Jan-Feb 2013)

Houston, Texas (September 2013)

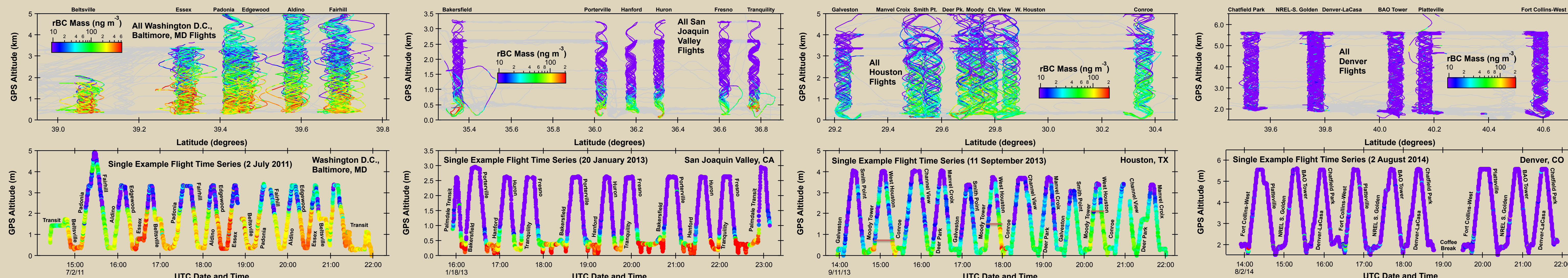
Denver, Colorado (July-August 2014)



Shown above are the NASA P-3B flight paths for each of the four, DISCOVER-AQ deployments (red lines). Labels denote the location of the vertical spiral profiles and missed approaches, which were collocated with ground-based measurement stations and balloon sondes.

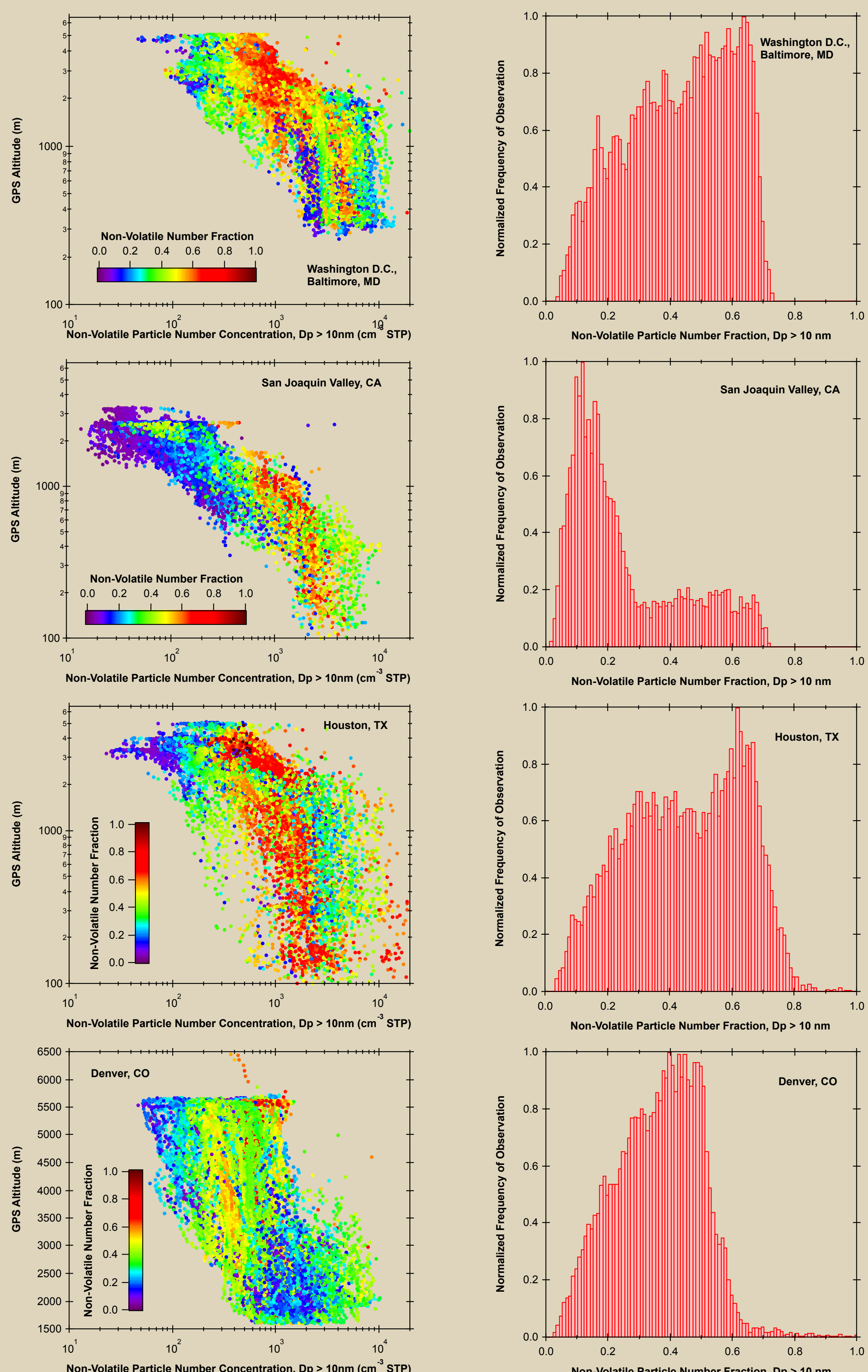
Horizontal Variability of SP2 Refractory Black Carbon

Figures below show the vertical and horizontal variability of refractory black carbon (rBC) measured by the DMT Single Particle Soot Photometer (SP2). The top row of figures reflects all flights conducted during each deployment, while the bottom figures show example time series for a single flight. Typically, each flight consists of three circuits (morning, mid-day, and afternoon) in order to capture the relationship between compositional variability and boundary layer dynamics, which are apparent, e.g., for the Moody Tower profile in the Houston, TX, timeseries below (red lines).



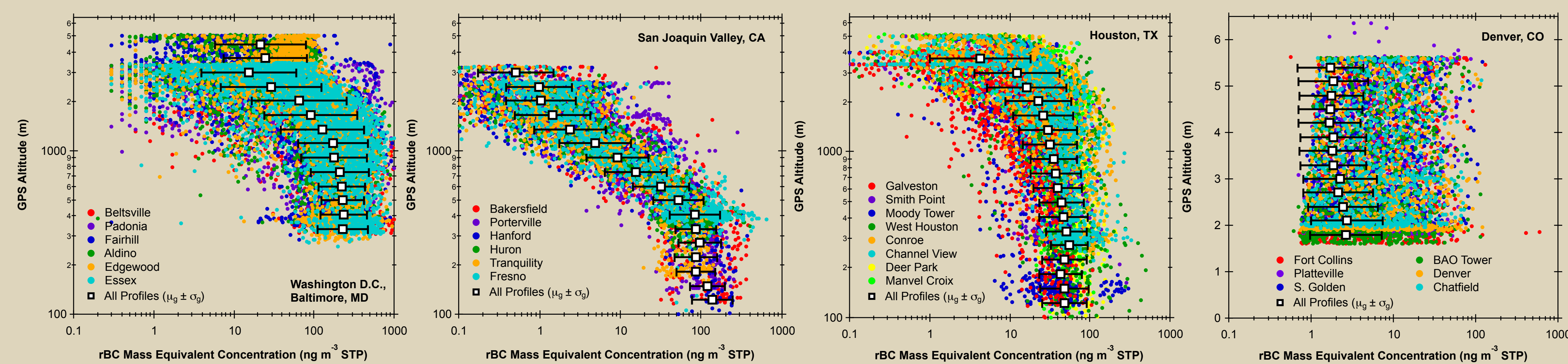
Vertical Profiles of Non-Volatile Aerosol Number Concentration (Denuded CPC)

Non-volatile particle number concentration (diameter > 10 nm) was measured using a TSI CPC 3772 after thermally denuding the particles at 350 degrees Celsius; total particle number was measured with an un-denuded CPC 3772. The vertical distribution of non-volatile number (left column below) mirrors that for mass (far right), with between 20-80% of particles containing a non-volatile core observed in the major metropolitan sites (right column below). This suggests a high degree of internal mixing for these locations. The wintertime San Joaquin Valley is different and exhibits low non-volatile number fractions (<20%), suggesting a lesser degree of internal mixing on a number basis. Future work with SP2 rBC coating thickness inferences will inform these findings as they relate to the aerosol accumulation mode.



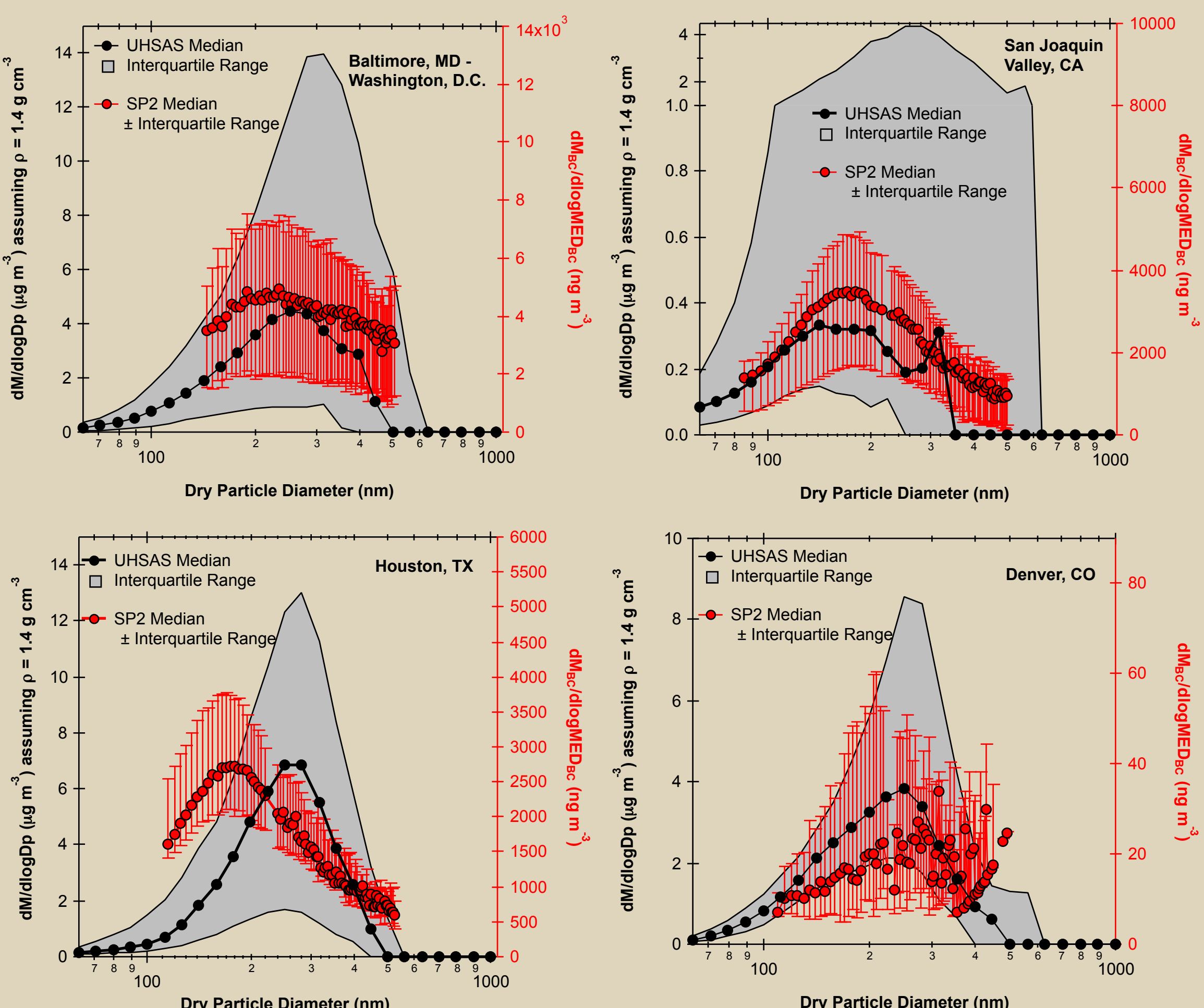
SP2 Refractory Black Carbon Vertical Profiles

The figures below show the vertical distribution of refractory black carbon (rBC) measured by the SP2. The altitude range shows the observational sampling strategy for each deployment (for example, low level legs were not conducted in the congested airspace near Washington D.C. and the surface is much higher in Denver, CO). All spiral data for a given deployment are shown color coded by the spiral site, including morning, mid-day, and afternoon profiles. A well-mixed layer of varying height is apparent in all sets of profiles, but is especially noticeable in the wintertime San Joaquin Valley with its characteristic strong inversion and shallow boundary layer. While there are some notable differences between spiral location for a given deployment (e.g., Galveston vs. downtown Houston sites), most spirals within a deployment show a similar structure, which reflects the strong influence of regional meteorology and dynamics in driving the aerosol spatial distributions.



Campaign-Avg. SP2 rBC Mass Size Size Distributions Superimposed on UHSAS Distributions

Campaign-median aerosol size distributions from the DMT Ultra-High Sensitivity Aerosol Spectrometer (UHSAS) are shown in the figures below in black and show a mass mode diameter of around 200-300 nm for all regions. The shallow boundary layers in the San Joaquin Valley skew the median concentrations toward much lower values, but the boundary layer aerosol mode is reflected in the 75th-percentile distribution. rBC mass size distributions from the SP2 are superimposed in red and show smaller median diameters than the overall aerosol mass size distribution. This lends support to the idea that the aerosol are predominantly internally mixed (moderate non-volatile number fractions and smaller core size mode), which makes rBC important for number-dependent processes such as the activation of cloud condensation nuclei (CCN) to form cloud droplets. Yet, it is also apparent that rBC constitutes a tiny fraction of the aerosol mass. Since relatively few models track aerosols with single particle resolution, and many large-scale models treat aerosol loadings only in terms of mass with prescribed size distributions, this work motivates the need for incorporating rBC number and mixing state in order to fully capture the role of rBC in contributing to aerosol processes.



Summary and Future Work

We present measurements of non-volatile and total aerosol number concentration, mass size distribution, and refractory black carbon mass measured aboard the NASA P-3B aircraft during DISCOVER-AQ. It is shown that black carbon is a variable but significant aerosol constituent on a number basis, despite its much lower mass loading. For the regions studied, the rBC vertical distribution is generally very similar across horizontally separated spiral sites, which reflects the strong influence of regional boundary layer dynamics in driving the spatial variability of black carbon. Future work will explore single particle mixing state using advanced SP2 analysis techniques including leading edge optimization and normalized derivative fitting methods.

