Figure S1: SP2 absolute laser power calculated from Eq. 1 vs. measured voltage from the Nd:YAG leakage detector. Each color corresponds to a separate period when the SP2 optical train was cleaned and aligned. As shown, there is good agreement between broadband high gain and low gain channels. Between separate optical cleaning and alignments, however, the SP2 absolute laser powers have a wide range of values for each each leakage detector voltage. Thus, although the leakage detector is a reliable gauge of relative power for each optical cleaning and alignment, it is not a reliable gauge of absolute laser power. This supports the recommendation that the SP2 absolute laser power should be calculated for every optical cleaning and alignment when using the SP2 as a prefilter to additional aerosol instruments.
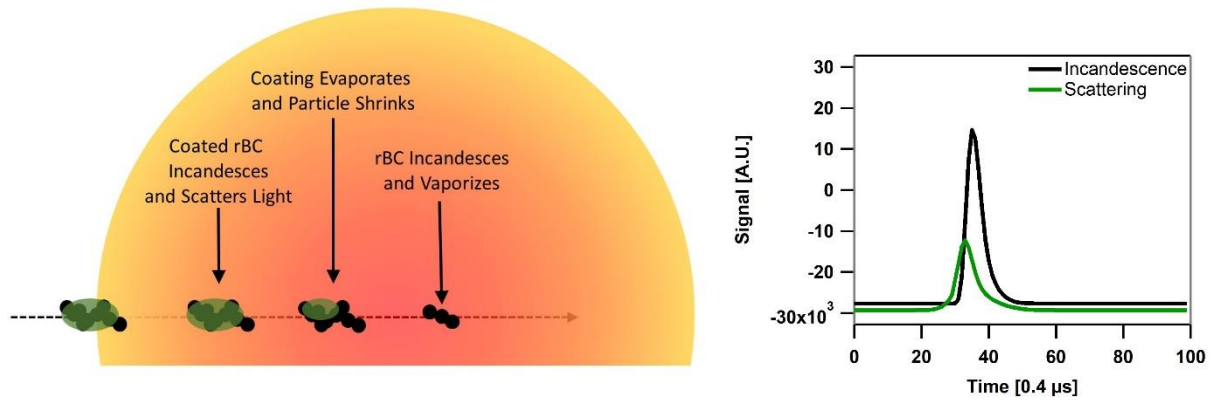
Figure S2: A cartoon of an rBC particle coated with non-refractory material as it passes through the SP2 laser and the time-dependent response from the incandescent and scattering detectors. As shown, the half-decay position of the scattering peak always appears before the half-decay position of the incandescence peak for coated particles.
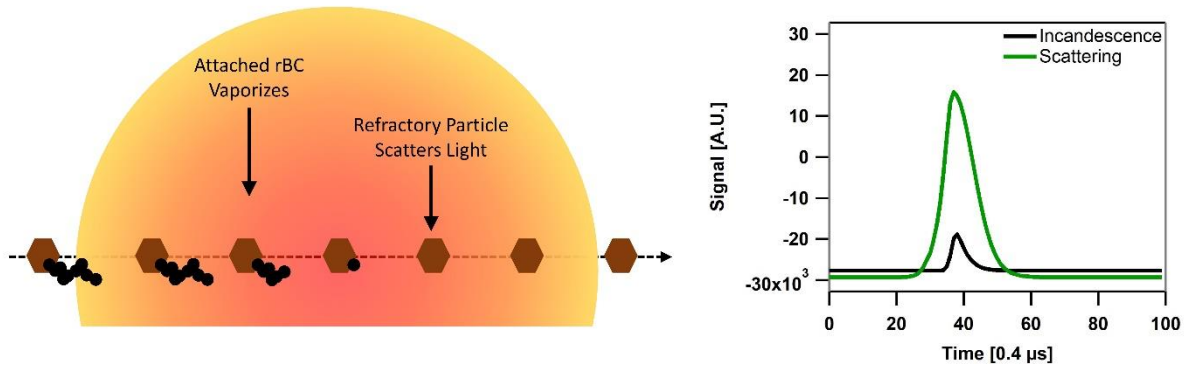
Figure S3: A cartoon of an rBC particle attached to a dust particle as it passes through the SP2 laser and the time-dependent response from the incandescent and scattering detectors. As shown, the half-decay position of the scattering peak always appears after the half-decay position of the incandescence peak for attached-type particles.
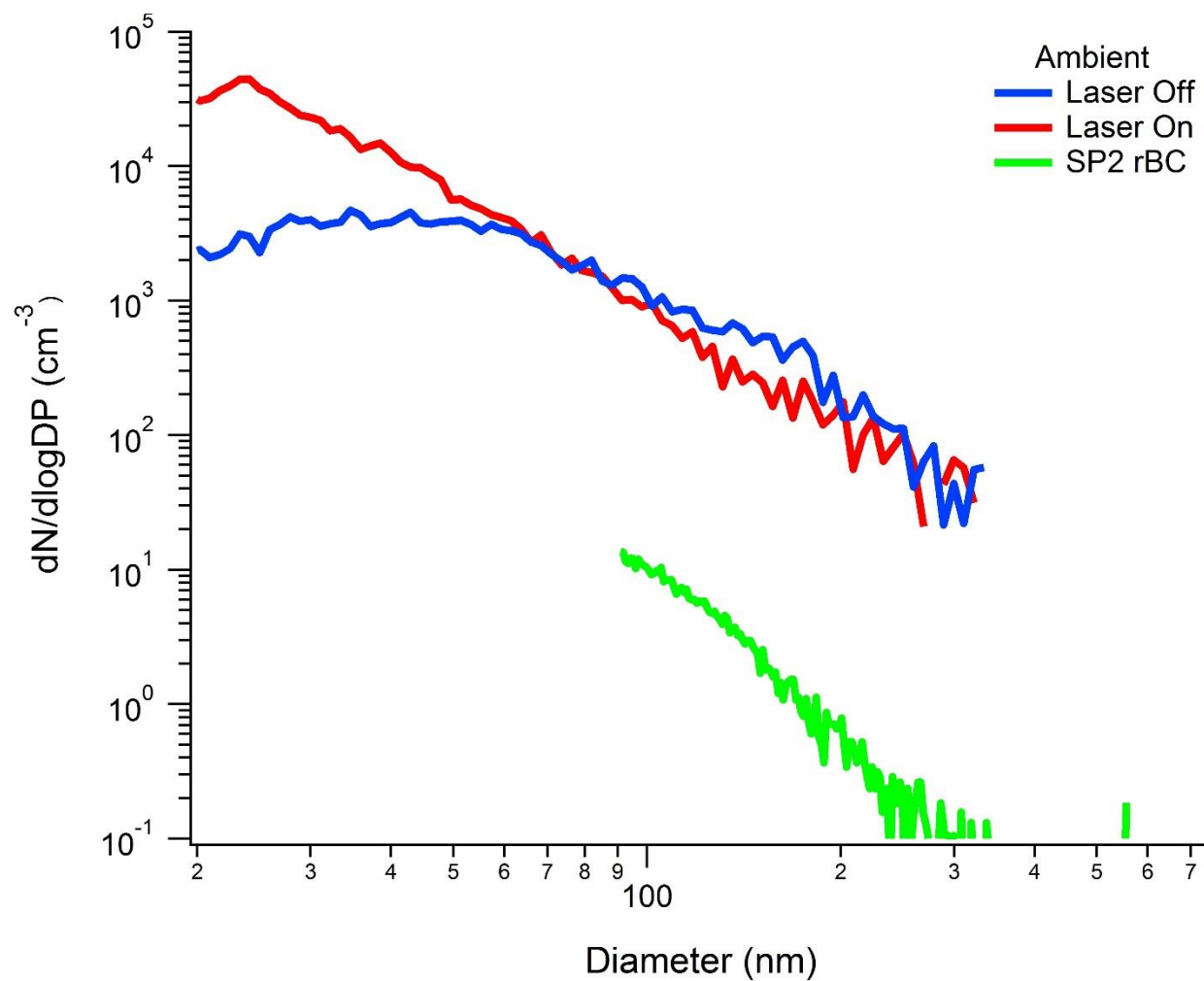
Figure S4. Size distributions of ambient air outside Colorado State University on 20150330. Size distributions were taken with an SMPS sampling the exhaust of the SP2 with the laser on and off. As shown, the "laser off" distribution diverges from the "laser on" distribution at ~60 nm. This suggests that the shoulder of the new particle peak from ambient rBC-SP2 exhaust is below the sizes expected of insoluble INP. The size distribution of rBC from the SP2 is also shown. SMPS data were corrected for dilution.