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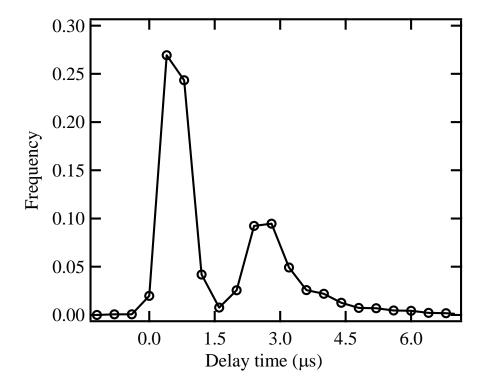
Measuring morphology and density of internally mixed black carbon with SP2 and VTDMA: new insight to absorption enhancement of black carbon in the atmosphere

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In our study, two types of ambient BC particles is divided based on the delay time 1 distribution, shown in Figure S1. The observed two peaks of delay time distribution is 2 due to the difference in behavior of Ex-BC and In-BC particles during SP2 3 measurement. The first peak with lower delay time represents the Ex-BC particles. 4 Corresponding, the second peak is considered as the proxy of In-BC particles. 5 Therefore, the particles with delay time more than 1.6 µs is chose to investigate their 6 core morphology and density. 7 8 The LEO fit used for In-BC particles is indirectly checked by that used for ambient non-BC particles (Figure S2). It is widely known that the initial scattering properties of 9 In-BC particles are changed with evaporation of In-BC coatings in SP2 measurement. 10 However, no changes in scattering properties of non-BC particles occurs during SP2 11 detection due to rarely absorbing components in non-BC particles. Therefore, the 12 scattering properties of non-BC particles derived from LEO fit should be similar with 13 that from scattering signal of SP2 measurement. In our study, the validity of LEO fit 14 used to determine the initial scattering properties of ambient In-BC particles is checked 15 by that used for ambient non-BC particles. 16 The scattering amplitude obtained from a LEO fit is compared with a full Gaussian 17 fit to the scattering signal detected by the SP2 for ambient non-BC particles, shown in 18 Figure S2. The regression line has a slope of 1.0673, indicating that the scattering 19 properties of non-BC particles from LEO fit was similar with that from SP2 20 measurement. The good agreement demonstrated that the LEO fit method is valid for 21 22 ambient particles with an uncertainty of ~6%. The RI_{nonBC} is determined by combining SP2 and VTDMA measurements in our 23 24 study. In previous SP2 studies, the RI_{nonBC} is usually given an assumption value of around 1.50. The RI_{nonBC} shown in Figure S3 is calculated using the scattering 25 properties from SP2 measurement and particle sizes from DMA1, respectively. The 26 RI_{nonBC} distribution ranges from 1.2 to 1.8 for single charged non-BC particles at 200-27 28 350 nm. We selects the peak value of 1.42 as the RI_{nonBC} for Mie theory calculations.



 ${\small 2\qquad \qquad \ \ Fig.\ S1.\ The\ distribution\ of\ delay\ time\ in\ our\ SP2\ measurement.}$

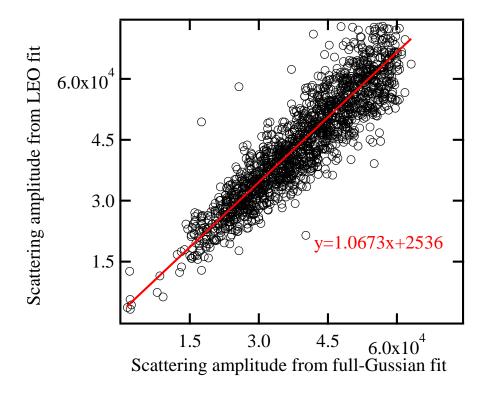


Fig. S2. Comparison of scattering amplitude obtained from full Gaussian fit and LEO fit methods for ambient non-BC particles.

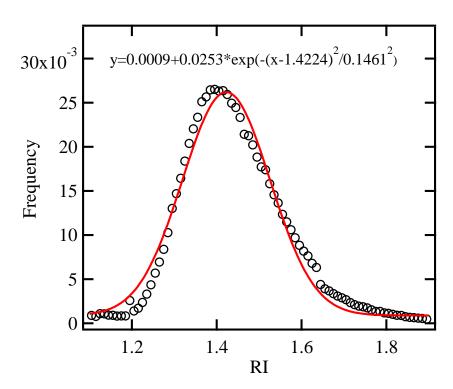


Fig. S3. The RI distribution for non-BC particles.