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*Supplement of*

**Single-particle measurements of bouncing particles and in situ collection efficiency from an airborne aerosol mass spectrometer (AMS) with light-scattering detection**

**Jin Liao et al.**

*Correspondence to:* Ann M. Middlebrook ([ann.m.middlebrook@noaa.gov](mailto:ann.m.middlebrook@noaa.gov))

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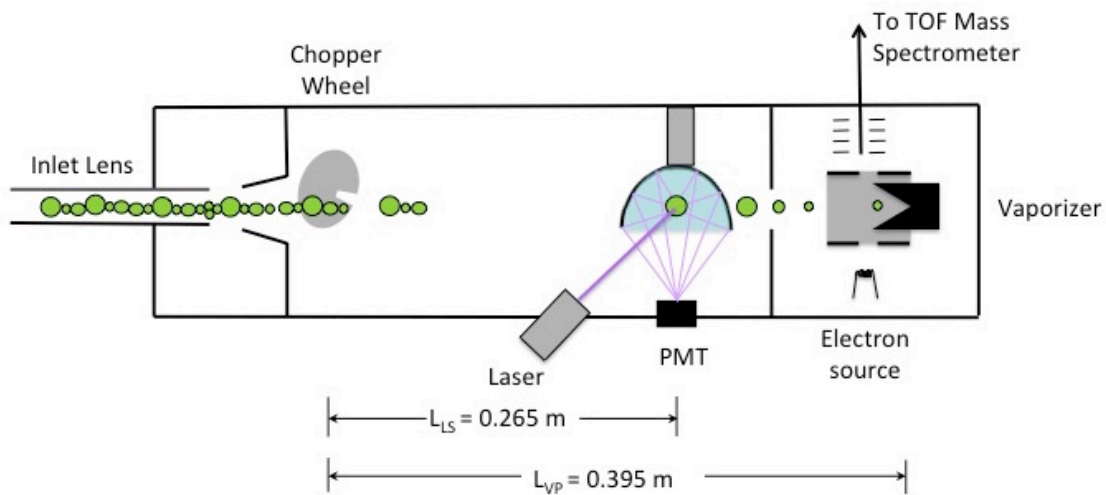
## Calculation of Number Distributions from LS Counts

Number distributions from LS counts are calculated based solely on histograms from the LSSP mode particle counts as a function of  $d_{va-LS}$ , which are normalized to the ratio of LS counts in LSSP mode to the adjacent MS open mode to account for the dead time needed to acquire and save chemical ion signals and the consequence of less particles recorded in LSSP mode.

- 5 This assumes that there are no biases in the LS events that are recorded in LSSP mode. Particle number distribution from LS counts is defined as:

$$N_{d_{va-LS}} (\#m^{-3}) = \frac{N_{d_{va-LS} \text{ in LSSP mode } (\#m^{-3})}}{\frac{N \text{ in LSSP mode } (\#s^{-1})}{N \text{ in MS mode } (\#s^{-1})}}, \quad (S1)$$

- 10 where  $N_{d_{va-LS} \text{ in LSSP mode } (\#m^{-3})}$  is the number of all particles (prompt+delayed+null) with the maximum scattered light intensity above the light-scattering threshold as a function of size  $d_{va-LS}$ ,  $N \text{ in LSSP mode } (\#s^{-1})$  is the total particle number recorded in each LSSP mode divided by the sampling time period (30 s), and  $N \text{ in MS mode } (\#s^{-1})$  is the average value of total particle counts recorded in the adjacent MS modes divided by the corresponding MS sampling time (4 s) and multiplied by the chopper duty cycle (2%).



**Figure S1. Instrument schematic diagram of the inlet, the chopper wheel, the laser beam, and the vaporizer positions for the AMS in light-scattering single-particle (LSSP) mode.**

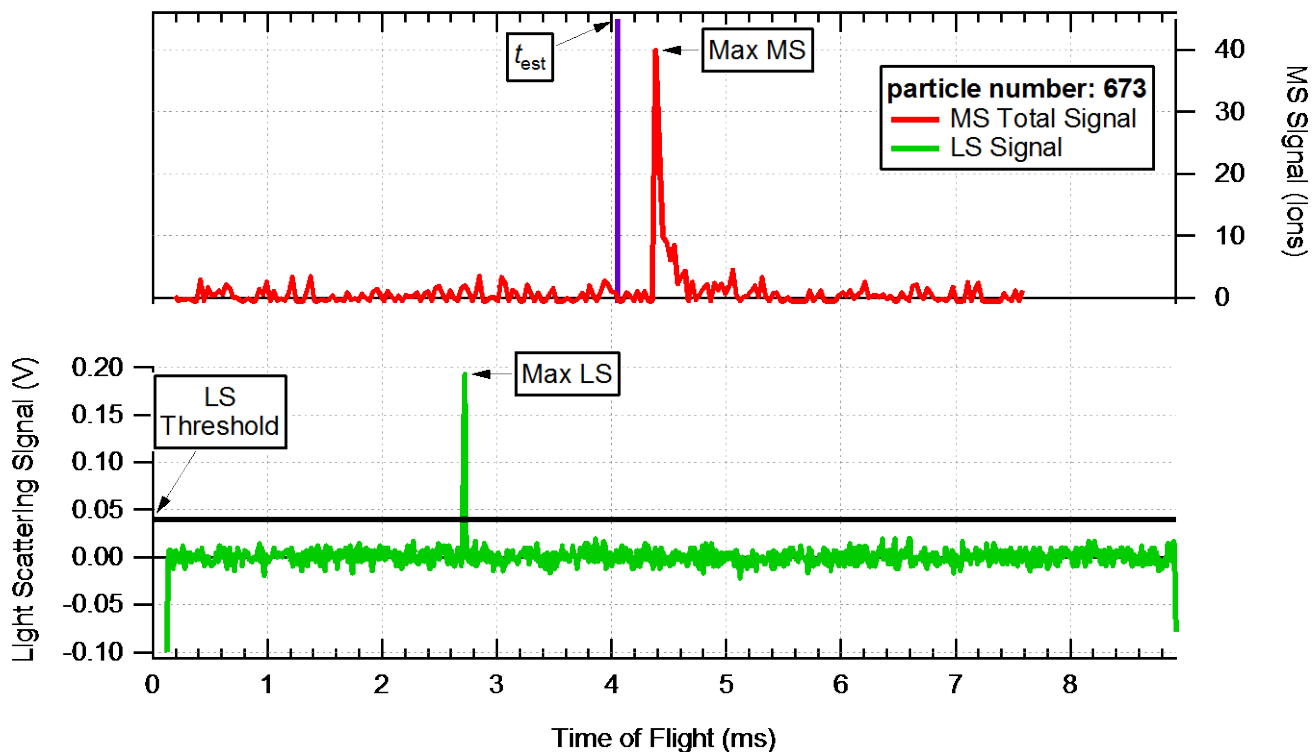
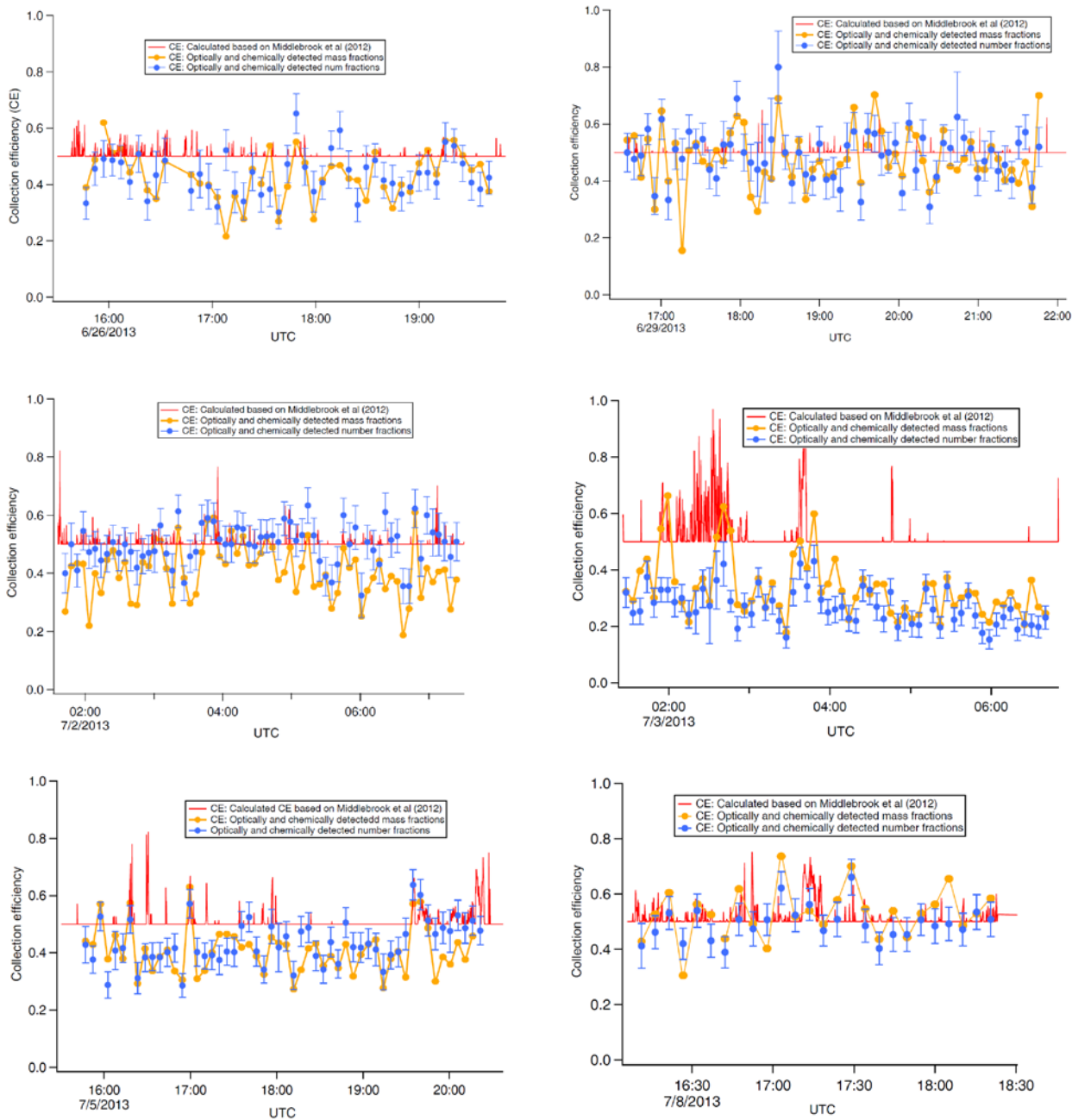


Figure S2. Sample raw data from the LSSP mode indicating the scattered light (LS) signal (bottom panel) and total mass spectral (MS) signal (top panel) for a single particle as a function of particle time-of-flight during one chopper cycle. The thick, horizontal black line indicates the threshold for optical (LS) particle detection and the thick, vertical purple line indicates the estimated arrival time at the detector ( $t_{est}$ ).

5



5 **Figure S3.** Time series plots of AMS collection efficiency calculated based on aerosol chemical composition and relative humidity method described in Middlebrook et al. (2012) (red) and measured by AMS LSSP mode based on number (blue) or mass (yellow) ratio of optically and chemically detected particles to optically detected particles for research flights from June 26 to July 8 (except the flight shown in Figure 8).