Supplementary Information for:

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

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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 LS particle counts as a function of d_{va-LS} , which are normalized to the ratio of LS counts in LS mode to the adjacent MS open mode to account for the extra time needed to acquire and save chemical ion signals and the consequence of less particles recorded in LSSP mode. This

20 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:

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

where $N_{d_{va-LS}}$ in LSSP mode (#. m^{-3}) is number of all particles (prompt+delayed+null) with size above light scattering threshold as a function of size d_{va-LS} ;

25 *N* in *LSSP* mode $(\#. s^{-1})$ is total particle number recorded in each LSSP mode divided by the sampling time period (30 sec); *N* in *MS* mode $(\#. s^{-1})$ is interpolated value of total particle number recorded in adjacent MS mode divided by corresponding chopper open recording time and multiply by chopper duty cycle (2%).

Flight	Water ^a	Water ^a	O16 ^b	Air ^b	CO2 ^b
Date	16/18	17/18	16/14	29/28	44/28
5/31/2013	0.035	0.247	0.340	0.0058	0.00028
6/3/2013	0.018	0.260	0.323	0.0050	0.00047
6/10/2013	0.035	0.250	0.325	0.0035	0.00047
6/11/2013	0.040	0.250	0.324	0.0037	0.00031
6/12/2013	0.020	0.279	0.344	0.0054	0.00038
6/16/2013	0.023	0.263	0.384	0.0043	0.00051
6/18/2013	0.024	0.255	0.344	0.0056	0.00007
6/19/2013	0.022	0.270	0.343	0.0057	0.00025
6/22/2013	0.019	0.260	0.333	0.0054	0.00022
6/23/2013	0.020	0.253	0.339	0.0051	0.00029
6/25/2013	0.019	0.260	0.342	0.0049	0.00027
6/26/2013	0.030	0.256	0.343	0.0065	0.00041
6/29/2013	0.021	0.262	0.336	0.0067	-0.00004
7/2/2013	0.020	0.264	0.338	0.0072	0.00039
7/3/2013	0.025	0.258	0.343	0.0070	0.00041
7/5/2013	0.032	0.258	0.344	0.0068	0.00044
7/6/2013	0.025	0.259	0.344	0.0072	0.00036
7/8/2013	0.022	0.261	0.344	0.0069	0.00039
7/10/2013	0.033	0.254	0.339	0.0071	0.00026

Table S1. Coefficients Changed in the Standard Fragmentation Table (Allen et al., 2004).

^a Since this AMS instrument is pumped down starting 3 hours before each flight, the background water signals decrease over the course of the day. The fragmentation pattern of water for each flight was determined by plotting the closed m/z 16 and m/z 17 signals versus closed m/z 18.

5 ^b About 5 minutes of filtered air measurements were taken just prior to take off, at high altitude shortly after take off, and just after landing for each flight. The contributions of various peaks to the particle-free air signals were determined by averaging the ratio of the signals from the difference spectra at the indicated m/z.



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. 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 total optically detected particles for research flights from June 26 to July 8 (except the flight shown in Figure 8).