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Atmospheric Chemistry and Physics Discussions

Supplement of

Molecular characterization of free tropospheric aerosol collected at the Pico Mountain Observatory: a case study with long range transported biomass burning plumes

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Supplementary Information Section

Figure S1

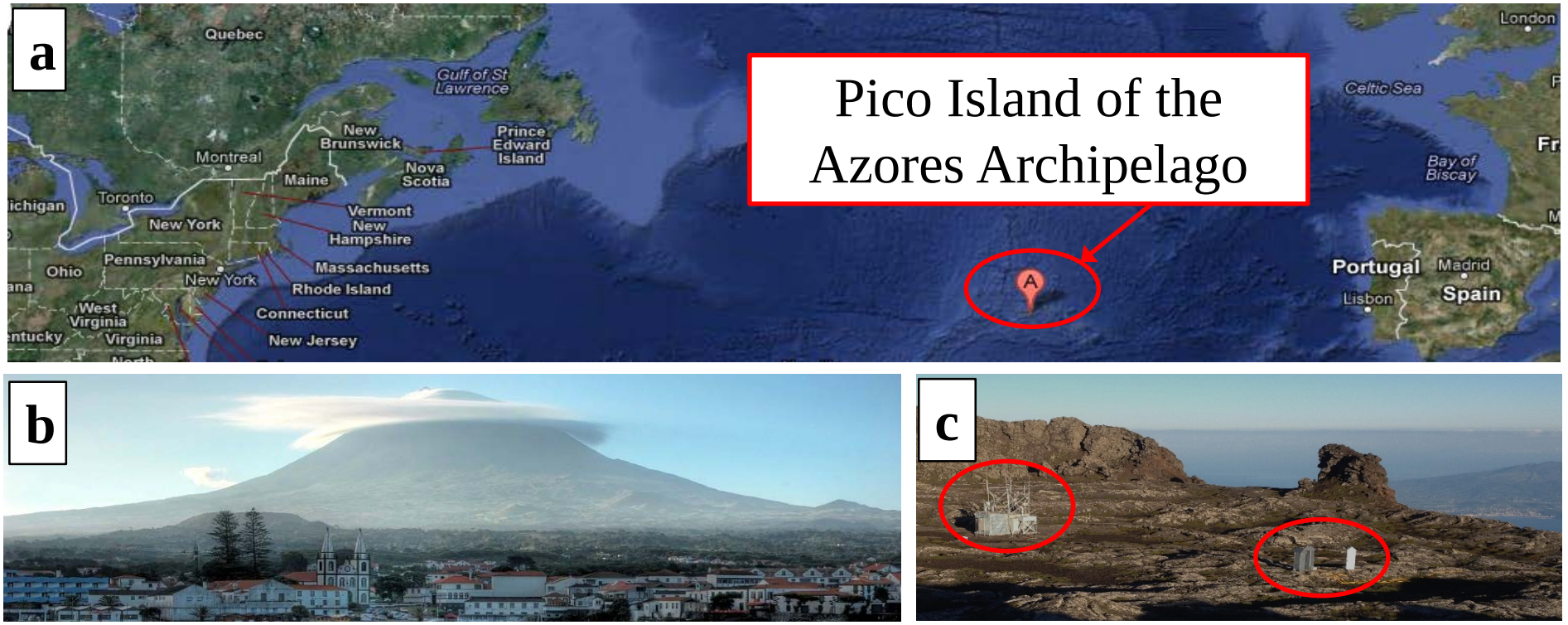

Figure S1. (a): Location of the Pico Island of the Azores Archipelago in the North Atlantic Ocean (photo credit: Google maps). Volcano on the Pico Island **(b)** (photo credit: Christer Johansson, Wikipedia) on the top of which is a crater where the Pico Mountain Observatory is located **(c)** (photo credit: Claudio Mazzoleni).

Figure S2

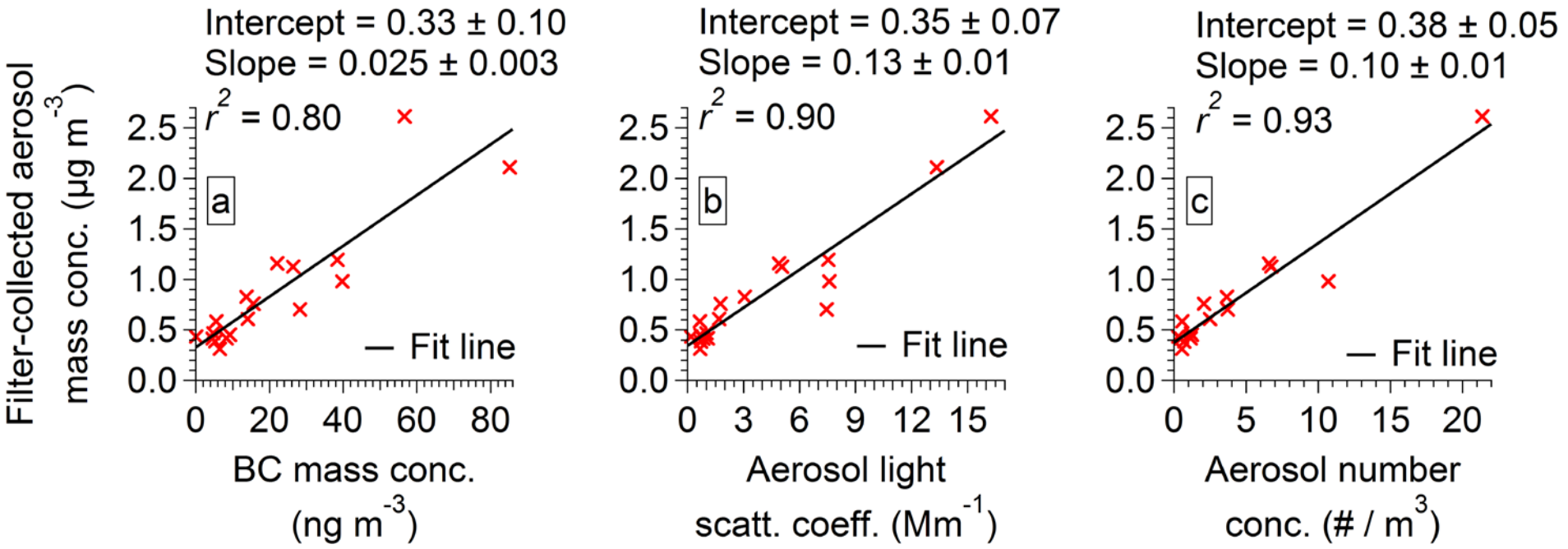

Figure S2. Scatter plots of total filter-collected aerosol mass (Org + EC + SO_4^{2-} + NO_3^- + Cl^-) shown in Figure 1a *vs.* on-line aerosol measurements of BC mass concentration **(a)**, aerosol light scattering **(b)** and number concentration **(c)** averaged over filter-collection periods. Linear regression best fit intercepts, slopes and correlation coefficients as well as their standard errors are reported.

Figure S3

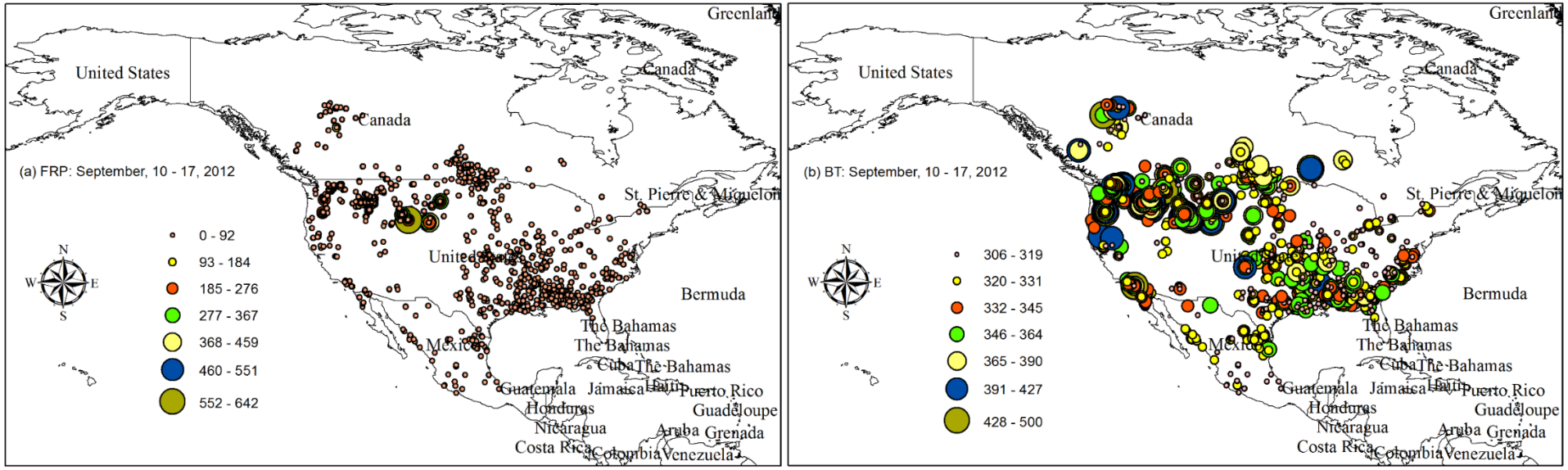

Figure S3. MODIS satellite fire counts for the period of September 10-17, 2012. MODIS fire counts are shown with the size of symbols denoting fire radiative properties in MW **(a)** and brightness temperature in K **(b)**.

Figure S4

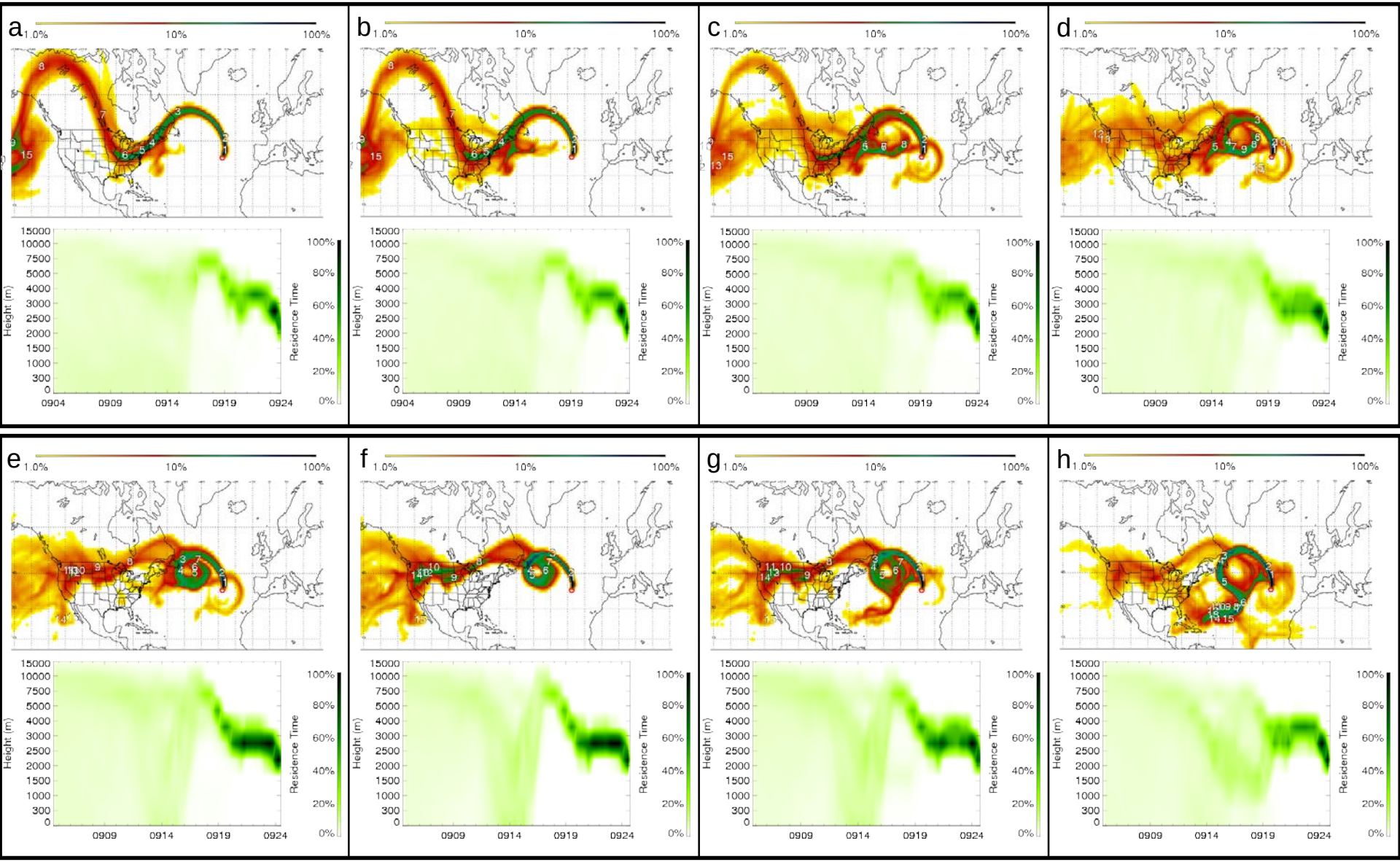

Figure S4. Simulated FLEXPART retroplumes for 9/24 sample with 3-hrs time resolution; first simulation is for Sep 24 at 15:00 UTC **(a)** and the last one for Sep 25 at noon **(h)**. Residence times of the retroplumes are normalized by each vertical intervals in the lower plots for better visualization of the distribution along an irregular height scale. Locations at each upwind time are labeled using white numbers representing days. Filter-collection period for this sample is Sep 24, 2012 at 15:00 - Sep 25, 2012 at 15:00 (Table 1).

Figure S5

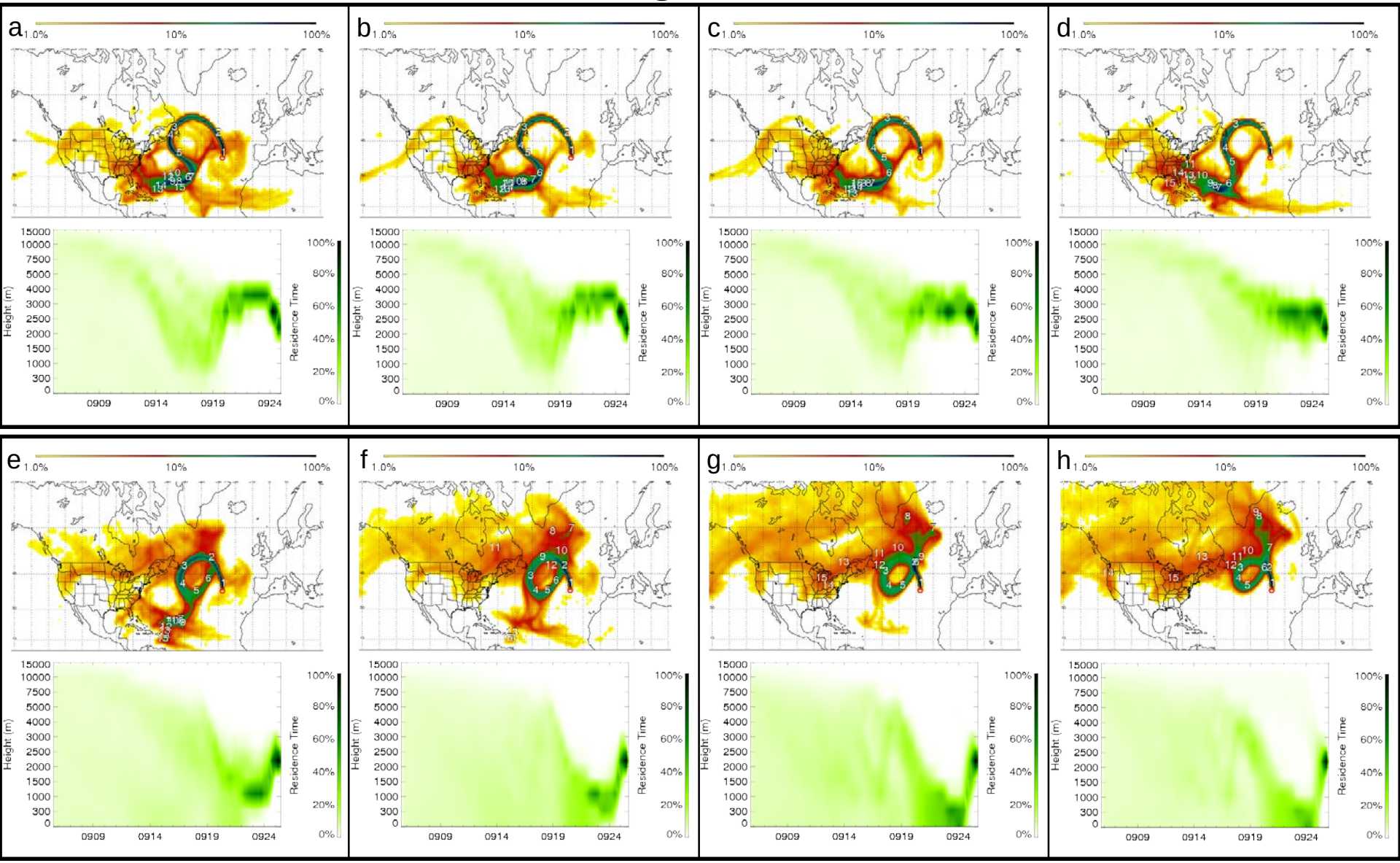

Figure S5. Simulated FLEXPART retroplumes for 9/25 sample with 3-hrs time resolution; first simulation is for Sep 25 at 15:00 UTC **(a)** and the last one for Sep 26 at noon **(h)**. Residence times of the retroplumes are normalized by each vertical intervals in the lower plots for better visualization of the distribution along an irregular height scale. Locations at each upwind time are labeled using white numbers representing days. Filter-collection period for this sample is Sep 25, 2012 at 15:00 - Sep 26, 2012 at 15:00 (Table 1).

Figure S6

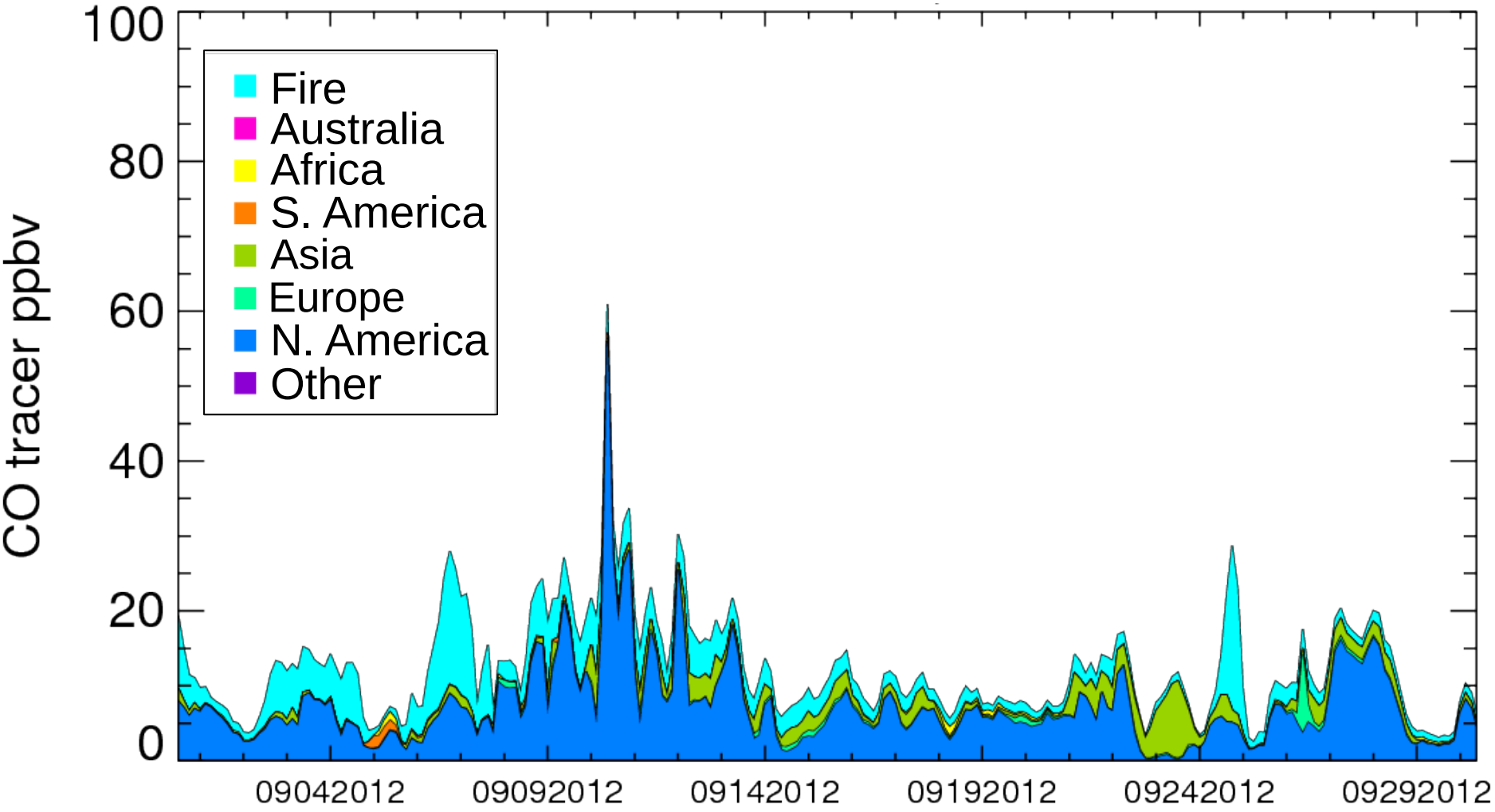

Figure S6. FLEXPART CO spectra for September 2012, as determined by multiplying the FLEXPART retroplumes with emission inventories. Fire CO includes contributions from the whole north hemisphere while the anthropogenic CO is further divided into the continent regions. Please refer to the text in Section 2.4 for the details of this calculation.

Figure S7

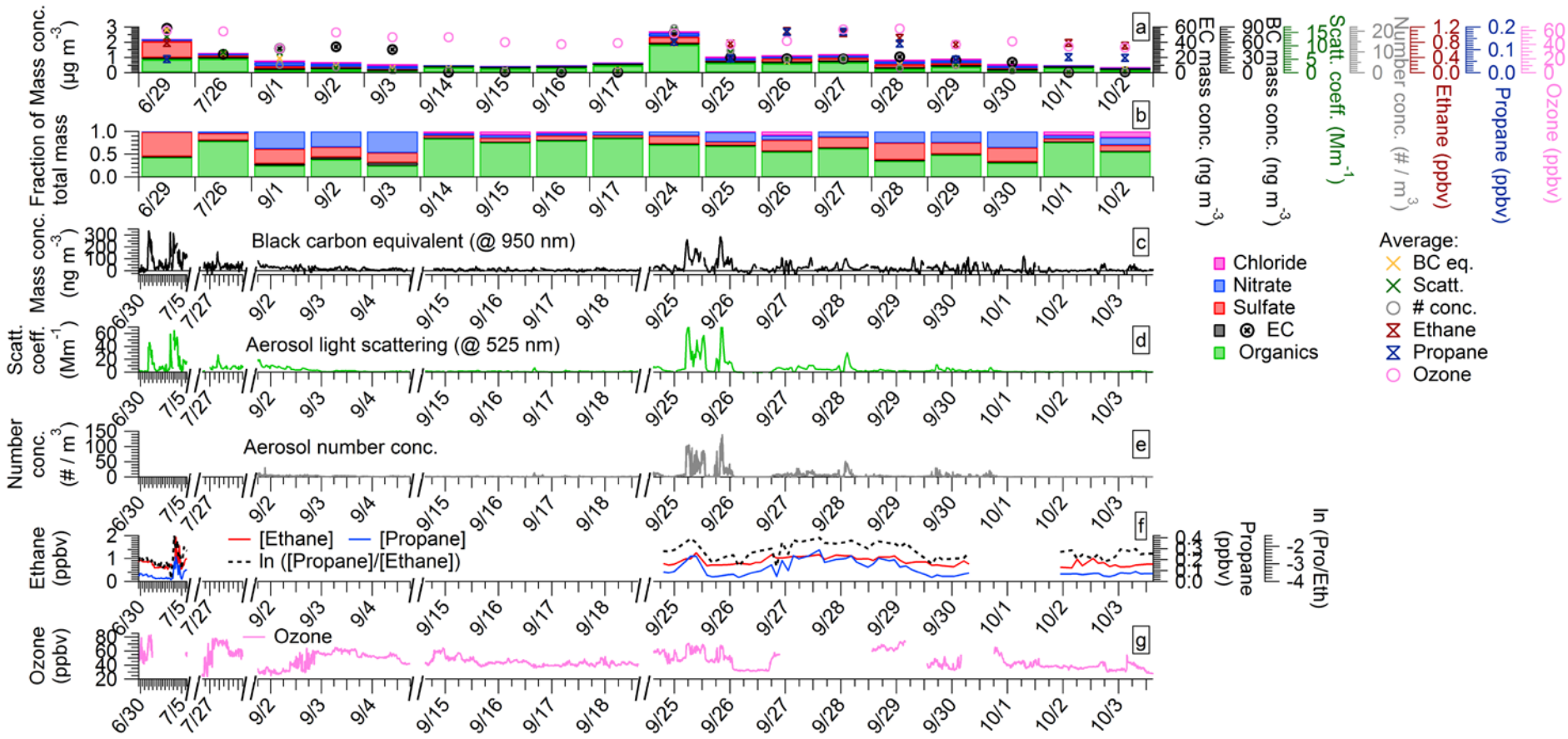

Figure S7. Time series of non-methane hydrocarbons ethane and propane and their ln(ethane/propane) ratio **(f)**, as well as ozone **(g)**. Also shown are measured mass concentrations of the filter-collected species as a percentage of the total **(b)** and all aerosol measurements shown in Fig. 1 for comparison. On-line measurements of gas-phase species are averaged for the sampling periods overlapping with filter measurements **(a)**.

Figure S8

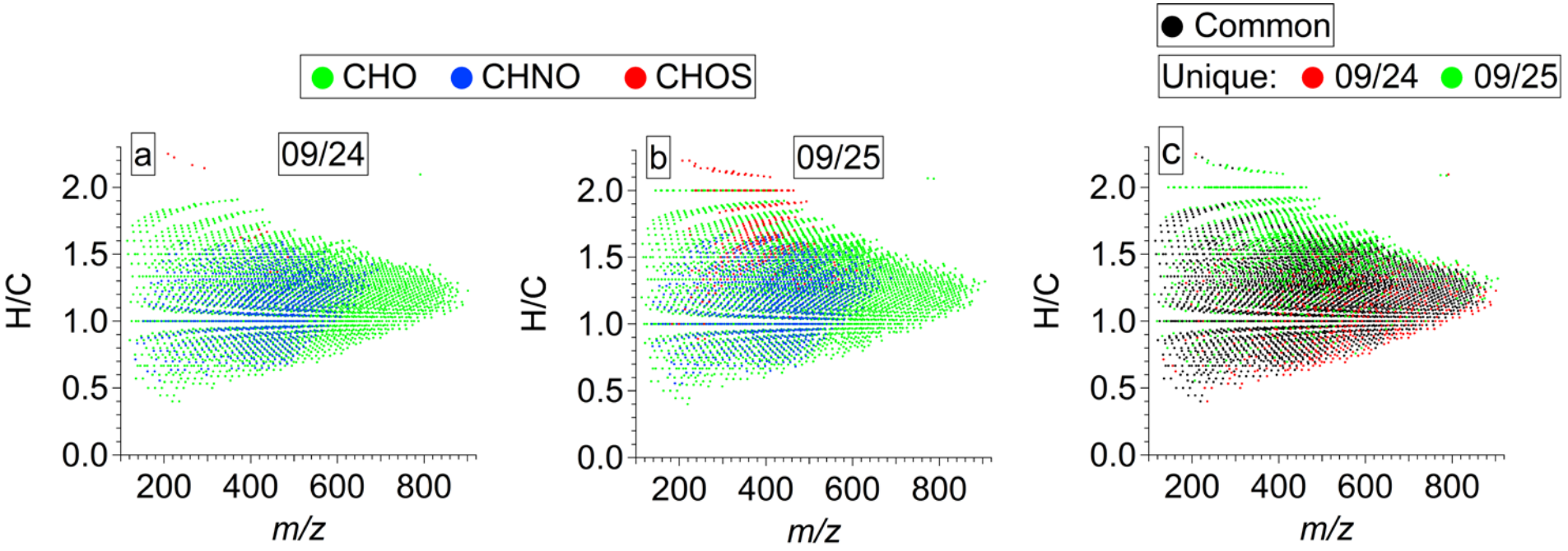

Figure S8. Mass resolved H/C elemental ratio diagram for all of the monoisotopic molecular assignments in the Pico WSOC 9/24 **(a)** and 9/25 **(b)** samples. Mass resolved H/C elemental ratio diagram is also shown for common and unique formula assignments between the two Pico samples **(c)**.

Figure S9

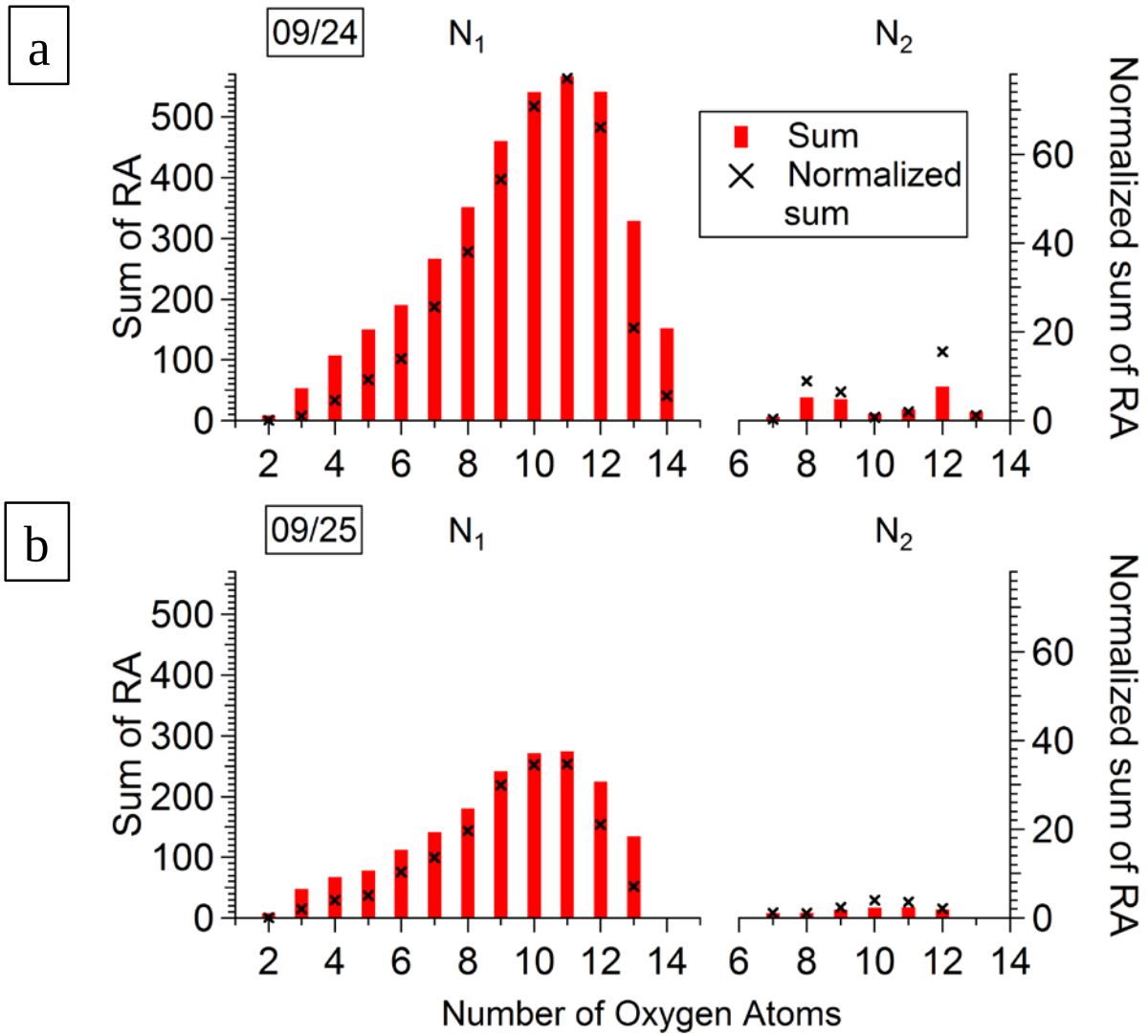

Figure S9. Sum of the relative abundance *vs.* number of oxygen atoms for CHNO compounds in the 9/24 **(a)** and 9/25 **(b)** WSOC samples. Sum of relative abundances is also normalized to the number of compounds with a certain oxygen number (x axis).

Figure S10

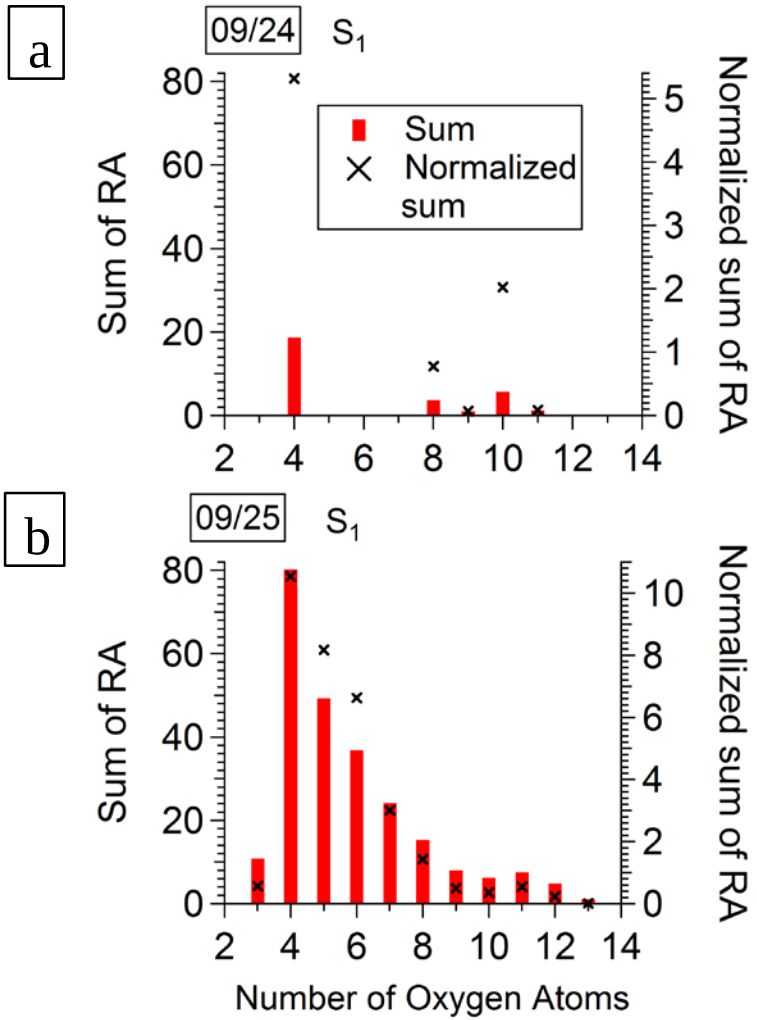

Figure S10. Sum of the relative abundance *vs.* number of oxygen atoms for CHOS compounds in the 9/24 **(a)** and 9/25 **(b)** WSOC samples. Sum of relative abundances is also normalized to the number of compounds with a certain oxygen number (x axis).