Supplementary Information

Characterization of Near-Highway Submicron Aerosols in New York City with a High-Resolution Aerosol Mass Spectrometer

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1 Investigation of silicone contamination

During the first roadside measurements on 27 July, highly elevated *m/z*'s, e.g., 73, 147, 201, 221, and 281 in the mass spectra of ambient OA were frequently observed (Fig. S5). Previous studies have confirmed that such *m/z* series are mainly from the fragmentation of polymer of dimethylsiloxane ($SiO(CH_3)_2$), e.g., the conductive silicone tubing (Schneider et al., 2006; Timko et al., 2009; Yu et al., 2009). Consistently, we observed similar *m/z* series in the mass spectra of vehicle exhaust from the gasoline generator (Fig. S5). The contamination from silicone tubing not only absorbs gaseous CO_2, but also artificially increases organic mass (Timko et al., 2009). This contamination is particularly substantial for the engine exhaust measurements since the emissions are enhanced when the tubing is heated. In our study, a 12-feet silicone coated fiberglass hose (Hi-Tech Duravent) was directly connected to the gasoline generator and a second 25-feet thermoplastic rubber reinforced with a wire helix (Flex-Flyte L-9) was then connected to the first hose. The generator exhausts were emitted to the atmosphere after passing the first and second hoses. A detailed check of the *m/z* series of silicon fragmentation during the four measurements (e.g., Fig. S5) suggests the most significant impact of silicone contamination on 27 July, and then much reduced influences on 28 July, and almost no impacts on the following two deployments. This indicates that the contamination is mainly from the fist hose coated with silicones inside, which are emitted significantly when heated (temperature > 70ºC though we never measured it). To further evaluate the contribution of contaminations, we performed PMF analysis to the unit mass resolution (UMR) spectra with *m/z* up to 300. Four components were identified including HOA, SV-OOA, LV-OOA and a component representing the silicone-contaminated OA. The small component of NOA was not able to be resolved from UMR-PMF analysis; instead another component showing the *m/z* series of silicon contamination was identified. The mass spectra and time series for each component are shown in Fig. S6. As HOA, SV-OOA, and LV-OOA show very similar spectral patterns to those from HR-PMF analysis, the exhaust component however resembles that of vehicle exhaust with silicone contamination over *m/z* >= 73.The differences at small *m/z*'s below 73 suggest that the generator exhausts might have mixed with ambient aerosols. It's interesting to note that the silicone contamination is very consistent with the operations of gasoline generator used as the power supply (Fig. S7). When the generator was on, clear silicone contamination from the exhaust tubing was observed, however, when Li batteries were used as a surrogate and no emissions of vehicle exhaust at the same time, the silicone contamination correspondingly went away. As shown in Fig. S7, the silicone contamination was most significant and sporadic on 27 July with the contribution up to 45% to the OA. Such substantial silicone contamination might affect the composition and properties of aerosol particles. The data on 27 July is therefore not discussed in the text, but presented in Fig. S8. Although silicone contamination on 28 July still existed, the contribution was much reduced, generally less than 10%. The contamination was not observed in the two following measurements on 30 July and 1 August, respectively.

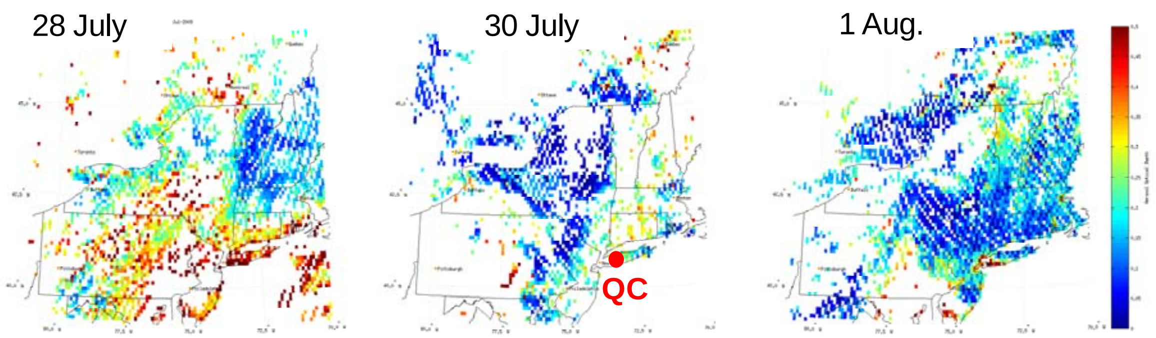

Fig. S1. Aerosol Optical Depth (AOD) retrieved from Terra MODIS at 550 nm on 28 July, 30 July and 1 August, respectively.

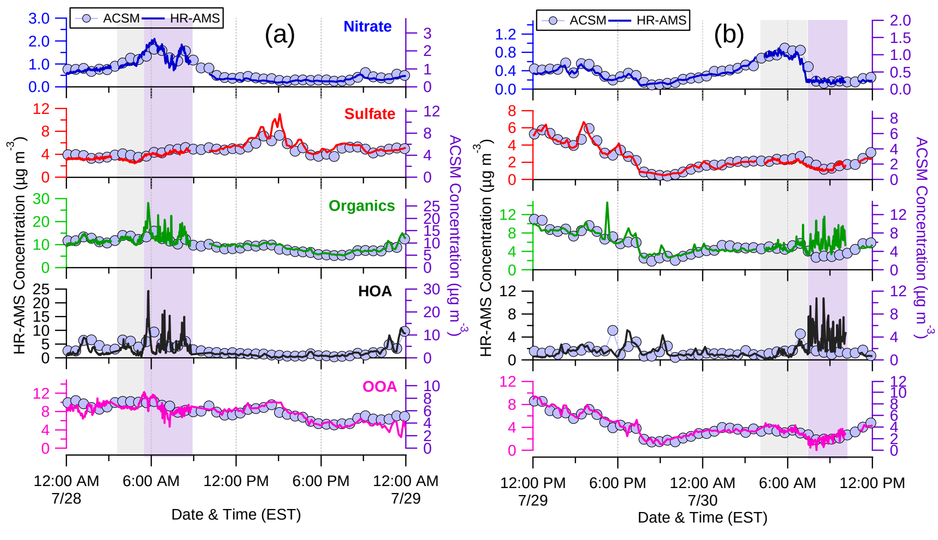

Fig.S2. Comparisons of time series organics, sulfate, nitrate, HOA and OOA measured by HR-AMS and ACSM on (a) 28 July and (b) 30 July. The shaded areas indicate the HR-AMS measurements near roadway.

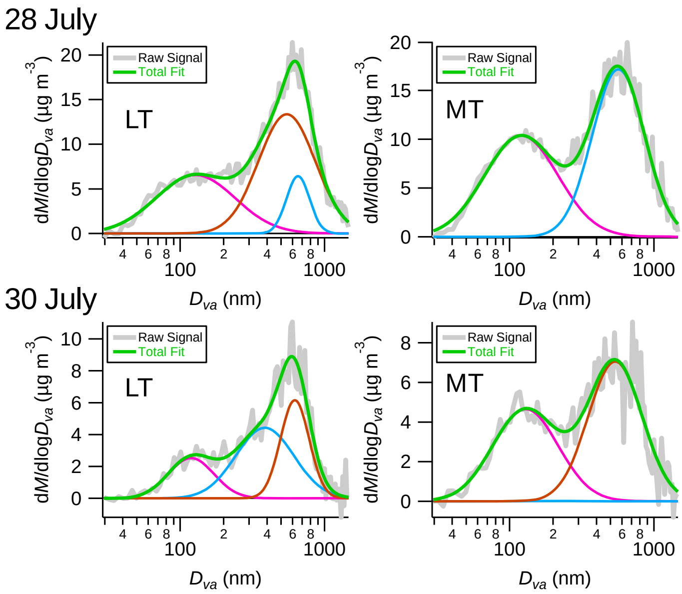

Fig. S3. Lognormal fittings of average size distributions of organics during LT and MT periods on 28 and 30 July, respectively.

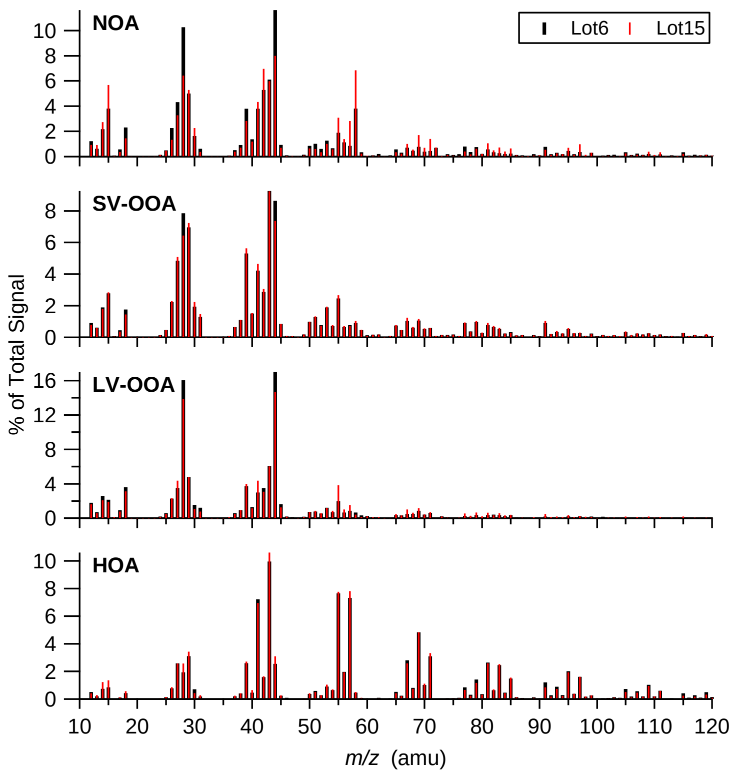

Fig. S4. The mass spectral comparisons of OA components between Lot 6 and Lot 15. Note that the mass spectra of OA components at Lot6 and Lot15 are from five and four component analyses, respectively because the OA near roadway didn't resolve the cooking-related OA.

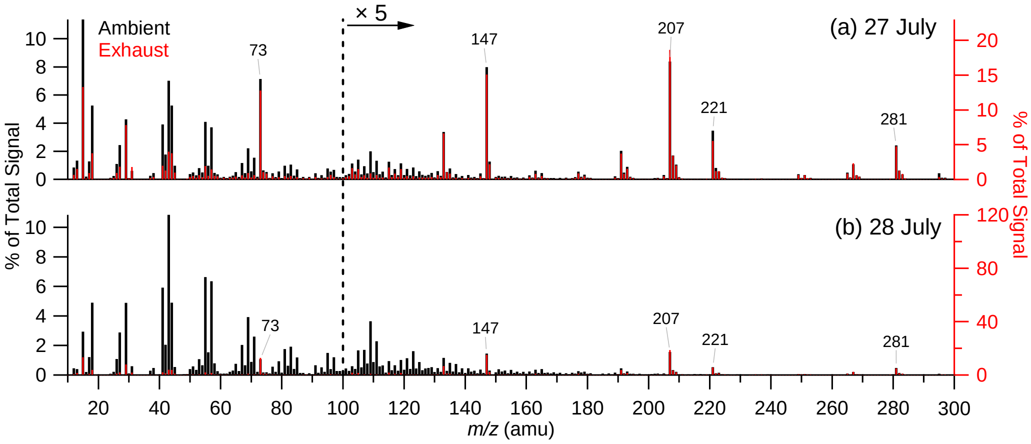

Fig. S5. Mass spectral comparisons between ASRC-ML generator exhaust and ambient OA with silicone contamination on (a) 27 July and (b) 28 July.

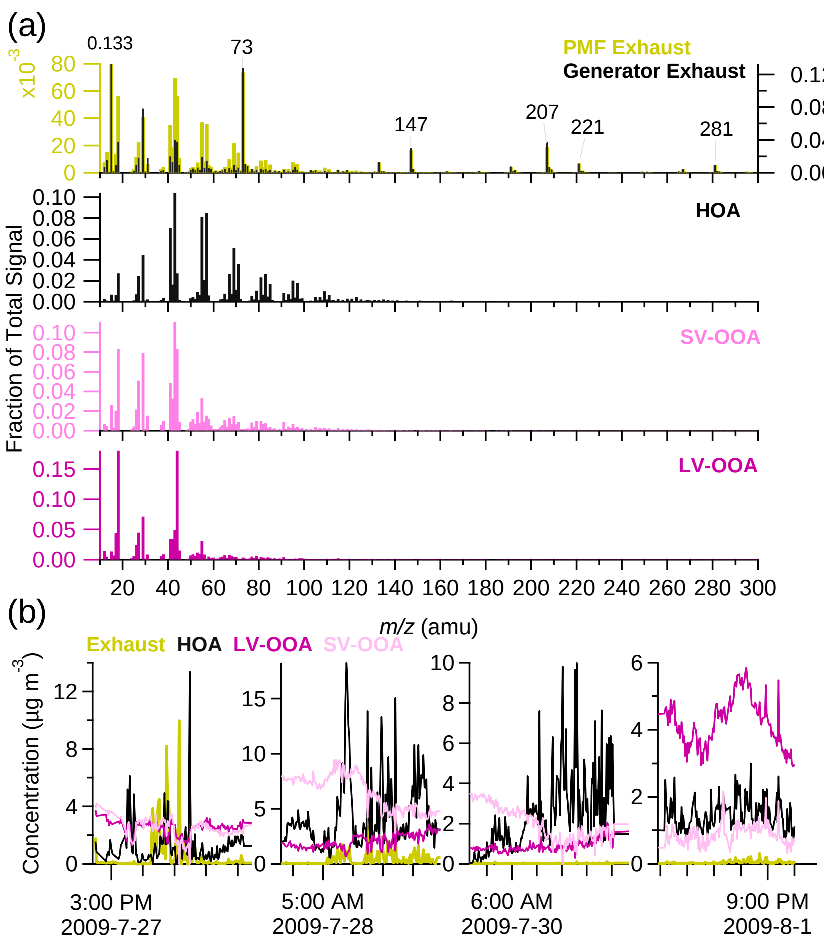

Fig. S6. (a) Mass spectra of OA components from UMR-PMF analysis. The mass spectrum of generator exhaust is also shown for a reference; (b) time series of mass concentrations of OA components for the four roadside measurements.

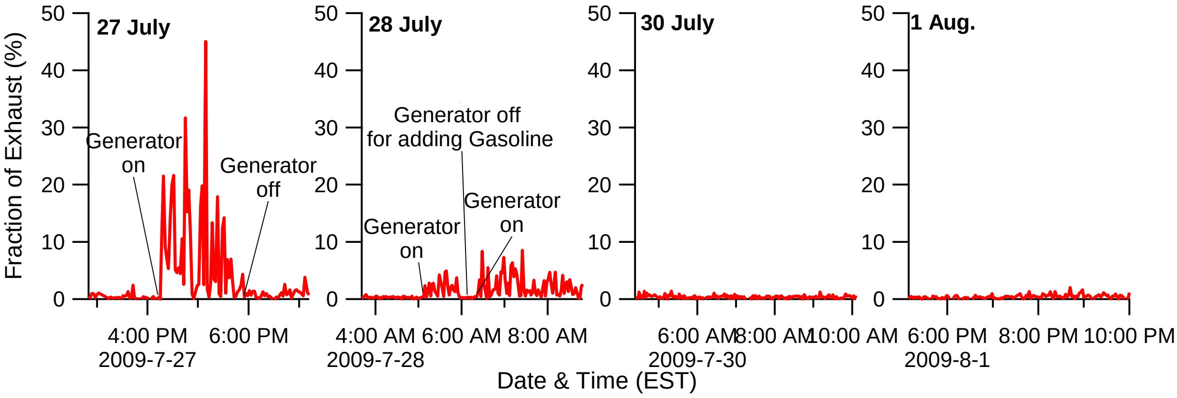

Fig. S7. Contribution of ASRC-ML generator exhaust to the total OA during four roadside measurements. The operations of generator are also marked on the plot as a reference.

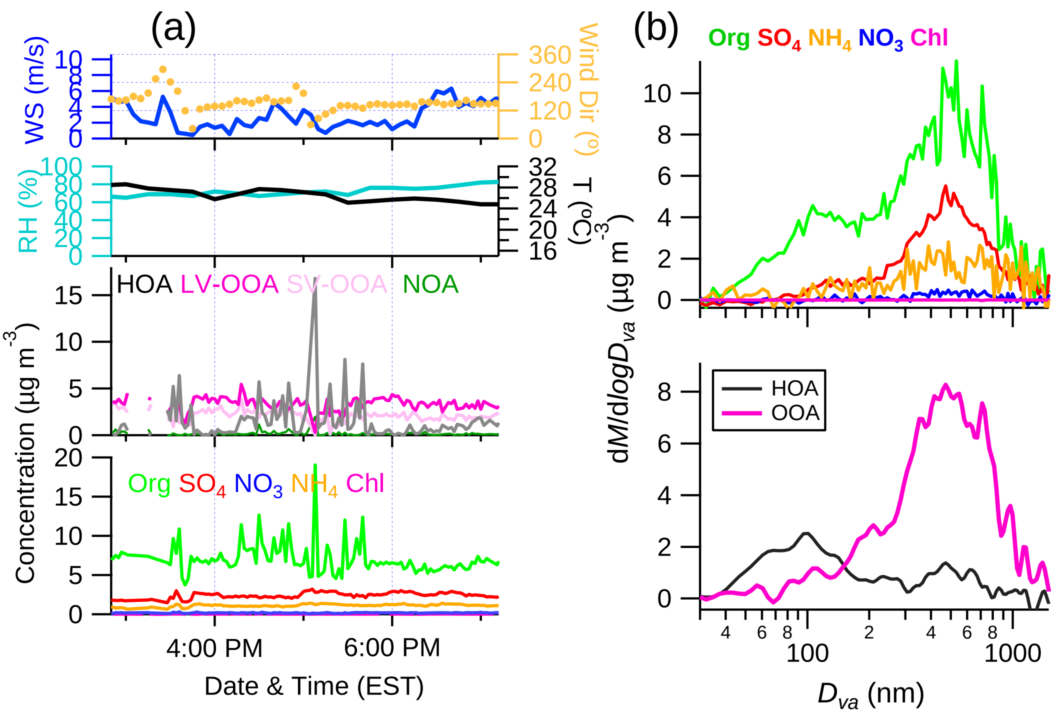

Fig. S8. (a) Time series of mass concentrations of NR-PM_1 species (organics, sulfate, nitrate, ammonium, and chloride), OA components (LV-OOA, SV-OOA, HOA, and NOA) and meteorology, and (b) Average size distributions of NR-PM_1 species, HOA and OOA during the roadside measurement of 27 July. Note that the size distributions of HOA and OOA were smoothed by 3 points using binominal algorithm.

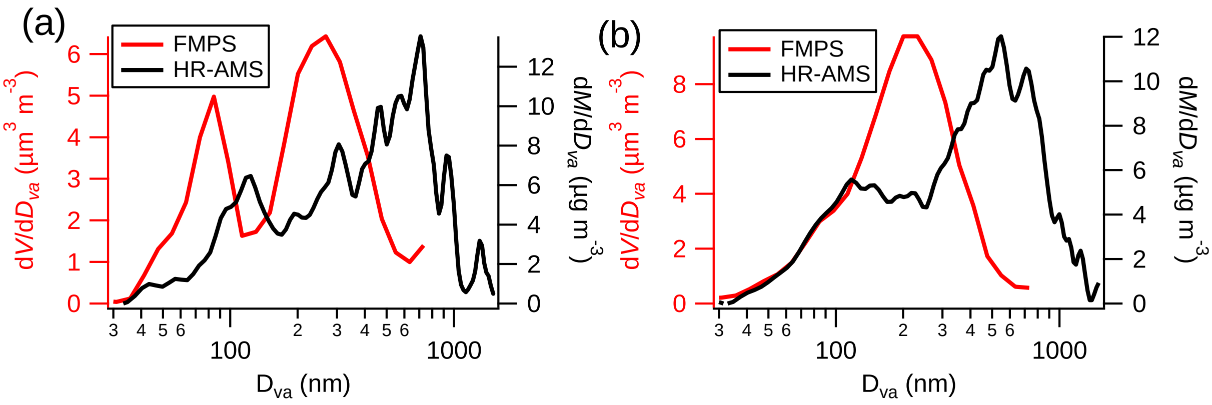

Fig. S9. The comparison of average volume-weighted size distributions from the FMPS measurements and mass-weighted size distributions from the HR-AMS measurements during (a) LT and (b) MT periods on 30 July. The D_m was converted to D_{va} by multiplying the estimated density of 1.4 μg m^{-3} assuming that the particles are sphere.

References:

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