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Supplement of

Mist cannon trucks can exacerbate the formation of water-soluble organic aerosol and $PM_{2.5}$ pollution in the road environment

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S1. Parameter Calculation and Compound Categorization

The value of double-bond equivalent (DBE) was calculated to reflect the sum of π-bonds and rings in a neutral molecule (Lechtenfeld et al., 2014; Qiao et al., 2020). The equation was shown below.

$$\mathrm{DBE} = 1 + N_\mathrm{C} - N_\mathrm{H}/2 + N_\mathrm{N}/2 \tag{1}$$

where the N_C, N_H, and N_N denote the number of carbon, hydrogen, and nitrogen atoms in a molecular formula, respectively.

The modified aromaticity index (AI_mod) can be used to reflect the aromaticity of organic molecules, which was calculated according to the following equation (Schmidt et al., 2017; Koch and Dittmar, 2006).

$$\mathrm{AI}_\mathrm{mod} = (1 + N_C - 0.5 \times N_\mathrm{O} - N_\mathrm{S} - 0.5 \times N_N - 0.5 \times N_H)/(N_\mathrm{C} - 0.5 \times N_\mathrm{O} - N_\mathrm{S} - N_\mathrm{N}) \tag{2}$$

where the N_C, N_H, N_O, N_N, and N_S denote the number of carbon, hydrogen, oxygen, nitrogen, and sulfur atoms in a molecular formula, respectively.

The carbon oxidation state (OSc) is an indicator to describe the evolving composition of aerosol organics undergoing oxidation processes (Kroll et al., 2011). For assignable molecular formulas, OS_C was calculated with following equation.

$$\mathrm{OS}_\mathrm{C} \approx 2 \times N_\mathrm{O}/N_\mathrm{C} - N_\mathrm{H}/N_\mathrm{C} \tag{3}$$

where the N_C, N_H, N_O, and N_N denote the number of carbon, hydrogen, oxygen, and nitrogen atoms in a molecular formula, respectively. Although the heteroatoms (N, S, and P) can introduce some uncertainties to the OS_C value of a given molecule in the measurement of ultrahigh resolution ESI-MS, the influence of these heteroatoms on the OS_C value of orgainc aerosols is generally small (Kroll et al., 2011).

In this sutdy, the molecular formulas of organic molecules were classified into five categories according to the ranges of AI_{mod} values and the values of H/C and O/C. Specifically, these categories include unsaturated aliphatic-like (UA) ($1.5 \leq H/C < 2.0$), highly unsaturated-like (HU) ($AI_{mod} \leq 0.5$ and $H/C < 1.5$), highly aromatic-like (HA) ($0.5 < AI_{mod} \leq 0.67$), polycyclic aromatic-like (PA) ($AI_{mod} > 0.67$), and saturated-like (Sa) ($H/C \geq 2.0$ or $O/C \geq 0.8$) molecules (Sihui et al., 2021; Seidel et al., 2014). Considering the presence of isomers for identified formula, the divided categories only represent the compounds cantaining the most likely functional structure (Butturini et al., 2020; Xie et al., 2021).

S2. Aerosol Liquid Water (ALW) Prediction

The model ISORROPIA-II was used to estimate the mass concentration of ALW with particle-phase concentrations of Na^+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}, SO_4^{2-}, NO_3^-, and Cl^-, as well as meteorological data (ambient temperature and relative humidity) as inputs (Guo et al., 2015; Nguyen et al., 2016; Tan et al., 2017). In this study, the model was run in the “reverse mode” without inputs of gas-phase parameters (Nguyen et al., 2015; Xu et al., 2020). In addition, the thermodynamically metastable state was set in the subsequent calculation (Guo et al., 2015; Nguyen et al., 2015; Nguyen et al., 2016). The “forward mode” was also run with inputs of only particle-phase ion concentration data, temperature, and relative humidity. The calculation results of water concentrations showed little difference irrespective of the mode used, which is consistent with the previous measurements (Guo et al., 2015; Hennigan et al., 2015).

Aerosol organics typically have complex compositions. It is difficult to directly quantify the mass concentration of water associated with organic fraction (Nguyen et al., 2016; Sareen et al., 2013; Cruz and Pandis, 2000). Accordingly, the mass concentration of water derived from organic compounds was predicted using a simplified model with the Zdanovskii−Stokes−Robinson (ZSR) mixing rule, as suggested by previous studies (Nguyen et al., 2016; Nguyen et al., 2015). Briefly, the hygroscopic growth of aerosol mixtures can be estimated using weighted hygroscopicity of each component according to their dry volume fractions (Bian et al., 2014; Nguyen et al., 2016; Nguyen et al., 2014). The detailed calculation was shown below (Petters and Kreidenweis, 2007; Kreidenweis et al., 2008).

$$V_{\mathrm{w,\,o}} = V_{\mathrm{o}}\,\kappa_{\mathrm{org}}\,a_{\mathrm{w}} / (1 - a_{\mathrm{w}}) \tag{4}$$

where $V_{\mathrm{w,\,o}}$ and V_{o} are the volumes of water and organics, respectively. κ_{org} is dimensionless and represents the hygroscopicity parameter of the organics. a_{w} is dimensionless and indicates water activity. The typical value of 1.4 g cm^{-3} for organic density was used to calculate the V_{o} value (Davidson et al., 2005; Turpin and Lim, 2001). A κ_{org} value of 0.08 was used in this study, which has been considered as a representative κ_{org} value for urban aerosols (Cerully et al., 2015; Dusek et al., 2010; Gunthe et al., 2009; Nguyen et al., 2016). The a_{w} value can be treated as relative humidity to simplify the calculation (Nguyen et al., 2015). This consideration was based on the following assumptions. The effect of aerosol curvature is insignificant. Furthermore, the effect of aerosol water uptake on ambient vapor pressure is also negligible (Bian et al., 2014). However, this assumption may lead to the overestimation of hygroscopicity (4–11%) (Nguyen et al., 2014).

Table S1. The arithmetic and peak-intensity-weighted averages of the elemental ratios and DBE values for different compound subgroups in different $PM_{2.5}$ samples.

$PM_{2.5}$ sample	All compounds			CHO			CHON		
	O/C ± SD O/C$_w$	H/C ± SD H/C$_w$	DBE ± SD DBE$_w$	O/C ± SD O/C$_w$	H/C ± SD H/C$_w$	DBE ± SD DBE$_w$	O/C ± SD O/C$_w$	H/C ± SD H/C$_w$	DBE ± SD DBE$_w$
Air spray	0.52 ± 0.21	1.35 ± 0.36	6.93 ± 3.33	0.46 ± 0.17	1.24 ± 0.35	7.61 ± 3.19	0.49 ± 0.19	1.21 ± 0.31	8.61 ± 3.17
(March 23)	0.56	1.56	4.51	0.49	1.27	7.04	0.46	1.24	8.22
Ground aspersion	0.53 ± 0.21	1.27 ± 0.41	7.44 ± 3.64	0.47 ± 0.17	1.09 ± 0.38	8.45 ± 3.52	0.47 ± 0.16	1.05 ± 0.31	9.79 ± 2.98
(March 23)	0.55	1.56	4.57	0.49	1.05	8.95	0.43	1.13	9.18
Air spray	0. 49± 0.2	1.33 ± 0.36	7.15 ± 3.32	0.44 ± 0.17	1.21 ± 0.36	8.06 ± 3.59	0.45 ± 0.17	1.20 ± 0.30	8.71 ± 2.73
(March 24)	0.52	1.56	4.58	0.47	1.23	7.57	0.42	1.24	8.28
Ground aspersion	0.55 ± 0.21	1.29 ± 0.40	7.30 ± 3.61	0. 48 ± 0.15	1.08 ± 0.38	9.50 ± 4.13	0.50 ± 0.16	1.07 ± 0.31	9.40 ± 2.79
(March 24)	0.57	1.57	4.45	0.50	1.06	9.21	0.47	1.14	8.88
Air spray	0.46 ± 0.19	1.23 ± 0.41	8.36 ± 4.18	0.42 ± 0.15	1.09 ± 0.40	9.57 ± 4.38	0.42 ± 0.15	1.06 ± 0.33	10.47 ± 3.60
(March 25)	0.45	1.48	5.54	0.43	1.07	9.27	0.39	1.10	9.88
Ground aspersion	0.18 ± 0.21	1.25 ± 0.39	12.00 ± 3.48	0.49 ± 0.17	1.14 ± 0.38	7.68 ± 3.18	0.49 ± 0.16	1.07 ± 0.31	9.66 ± 3.10
(March 25)	0.57	1.59	4.23	0.52	1.10	8.07	0.46	1.15	8.88
No water spray (I)	0.55 ± 0.22	1.29 ± 0.38	7.49 ± 3.69	0.47 ± 0.16	1.14 ± 0.36	8.96 ± 4.14	0.51 ± 0.17	1.09 ± 0.31	9.44 ± 2.97
(March 26)	0.54	1.54	4.79	0.50	1.14	8.54	0.45	1.14	8.97
No water spray (II)	0.52 ± 0.21	1.31 ± 0.38	7.18. ± 3.37	0.47 ± 0.17	1.18 ± 0.36	8.07 ± 3.45	0.48 ± 0.16	1.10 ± 0.31	9.27 ± 2.88
(March 26)	0.61	1.56	4.47	0.49	1.17	8.09	0.47	1.15	8.83

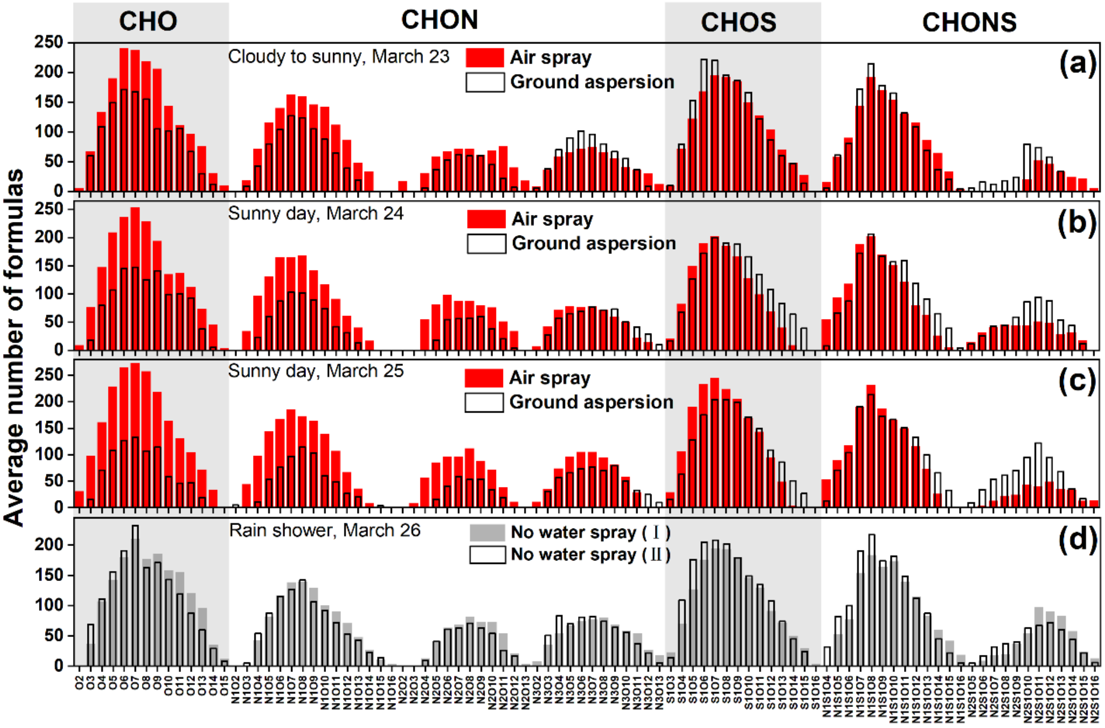

Figure S1. Classification of CHO, CHON, CHOS, and CHONS species into subgroups according to the number of O atoms in their molecules in WSOM in $PM_{2.5}$ collected from different cases: (**a, b, c**) air spray vs ground aspersion and (**d**) no water spray (I) vs no water spray (II).

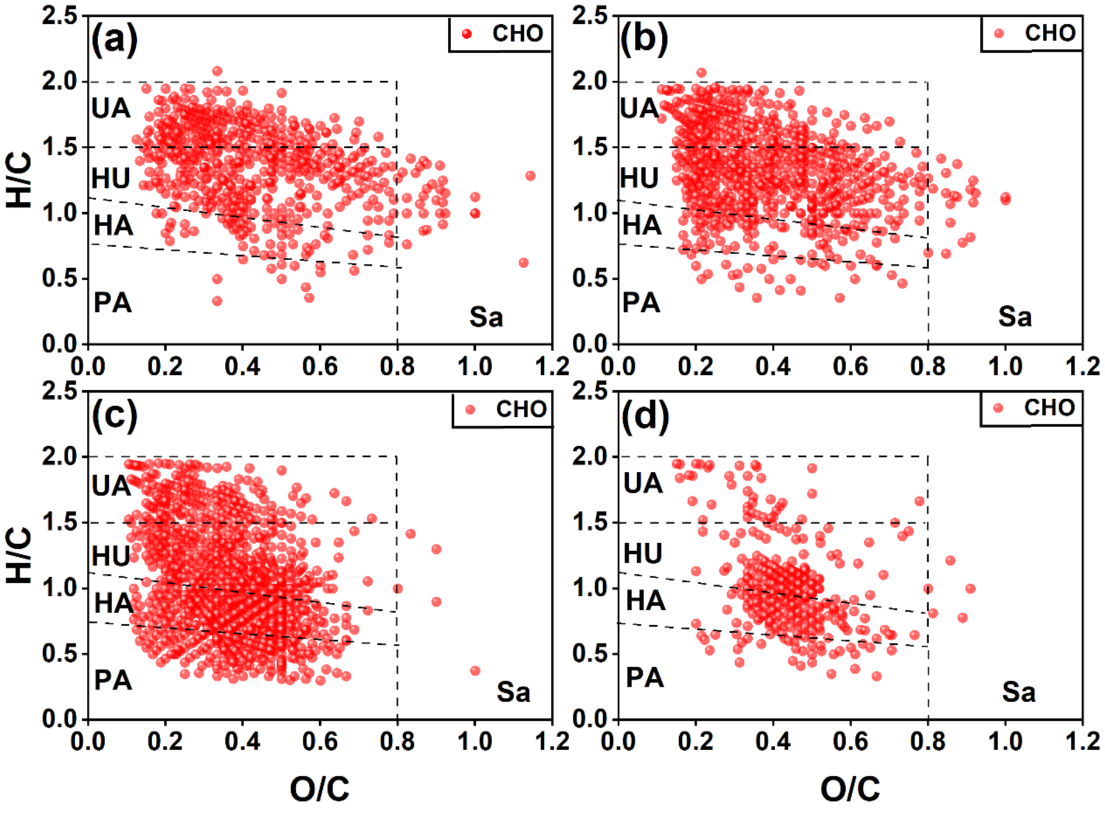

Figure S2. Van Krevelen diagrams of unique CHO compounds in WSOM in $PM_{2.5}$ collected from different cases: air spray vs ground aspersion on (**a**) March 23, (**b**) March 24, and (**c**) March 25 and two road segments without water spray (I vs II) on (**d**) March 26. For the above comparative cases, the unique CHO compounds indicate the CHO molecules identified in $PM_{2.5}$ collected from the air spray (/no water spray-I) road segments. The classifications of compounds include unsaturated aliphatic-like (UA), highly unsaturated-like (HU), highly aromatic-like (HA), polycyclic aromatic-like (PA), and saturated-like (Sa) molecules.

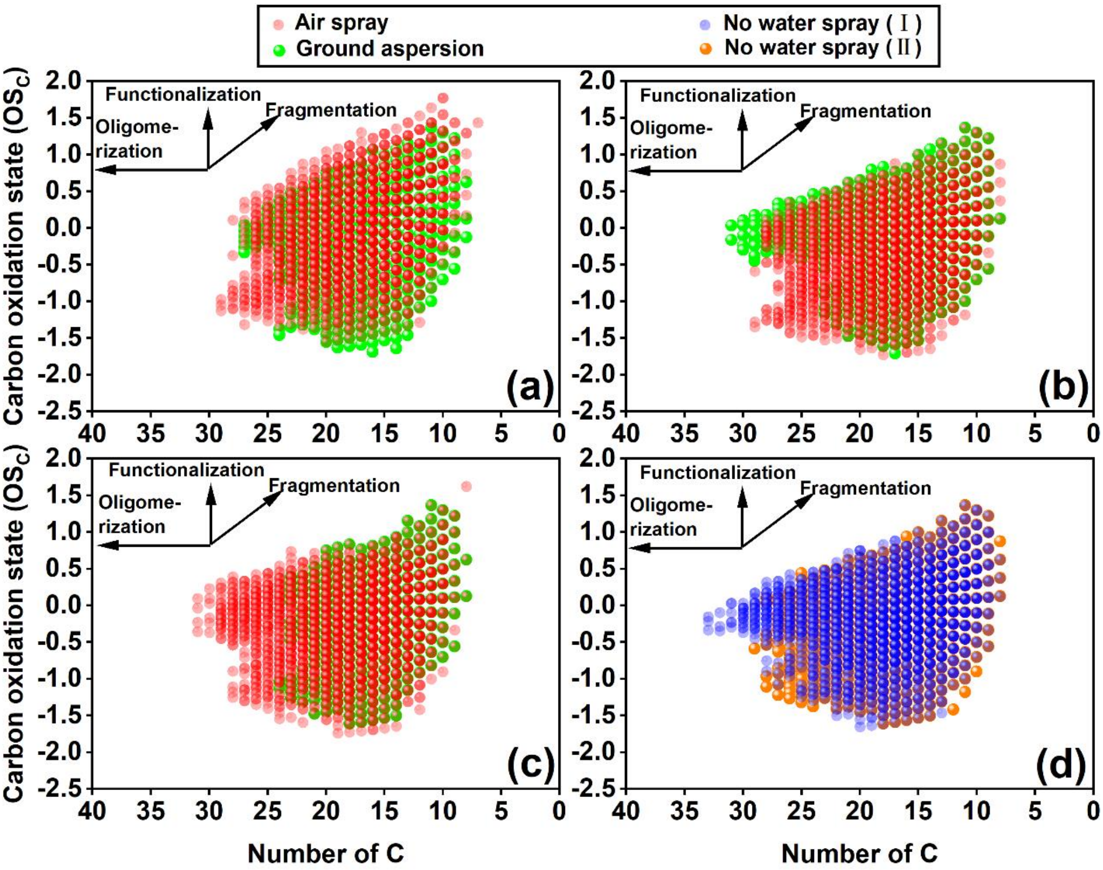

Figure S3. OS$_c$ of each CHO molecule in WSOM in PM$_{2.5}$ collected from different cases: air spray vs ground aspersion on (**a**) March 23, (**b**) March 24, and (**c**) March 25 and no water spray (I) vs no water spray (II) on (**d**) March 26.

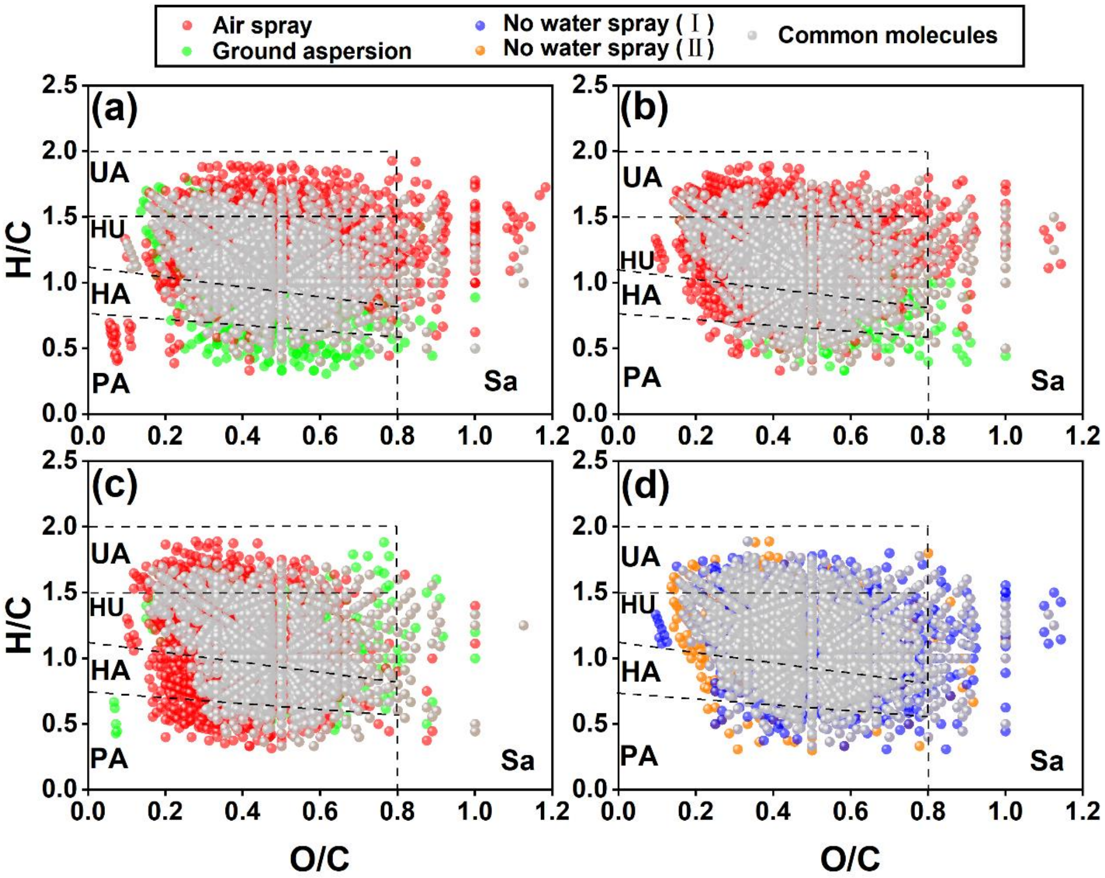

Figure S4. Van Krevelen diagrams of CHON compounds in WSOM in $PM_{2.5}$ collected from different cases: air spray vs ground aspersion on (**a**) March 23, (**b**) March 24, and (**c**) March 25 and two road segments without water spray (I vs II) on (**d**) March 26. The classifications of compounds include unsaturated aliphatic-like (UA), highly unsaturated-like (HU), highly aromatic-like (HA), polycyclic aromatic-like (PA), and saturated-like (Sa) molecules.

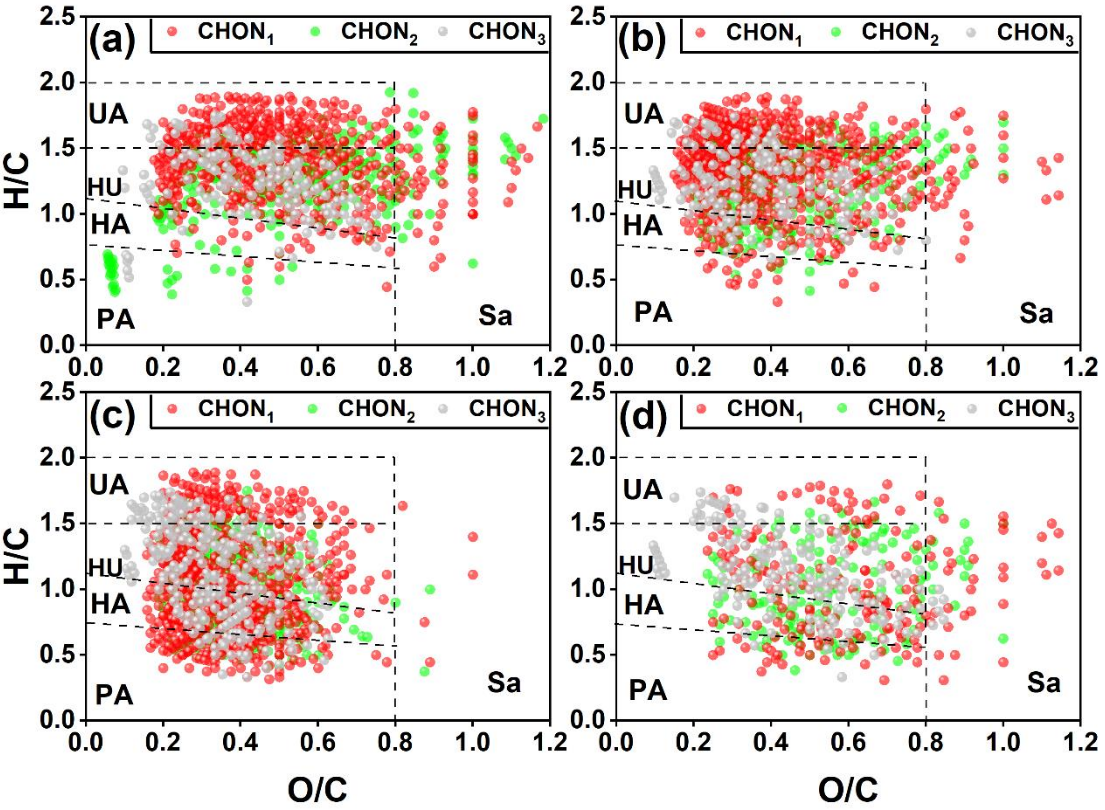

Figure S5. Van Krevelen diagrams of unique CHON compounds in WSOM in $PM_{2.5}$ collected from different cases: air spray vs ground aspersion on (**a**) March 23, (**b**) March 24, and (**c**) March 25 and two road segments without water spray (I vs II) on (**d**) March 26. For the above comparative cases, the unique CHON compounds indicate the CHON molecules identified in $PM_{2.5}$ collected from the air spray (/no water spray-I) road segments. The classifications of compounds include unsaturated aliphatic-like (UA), highly unsaturated-like (HU), highly aromatic-like (HA), polycyclic aromatic-like (PA), and saturated-like (Sa) molecules.

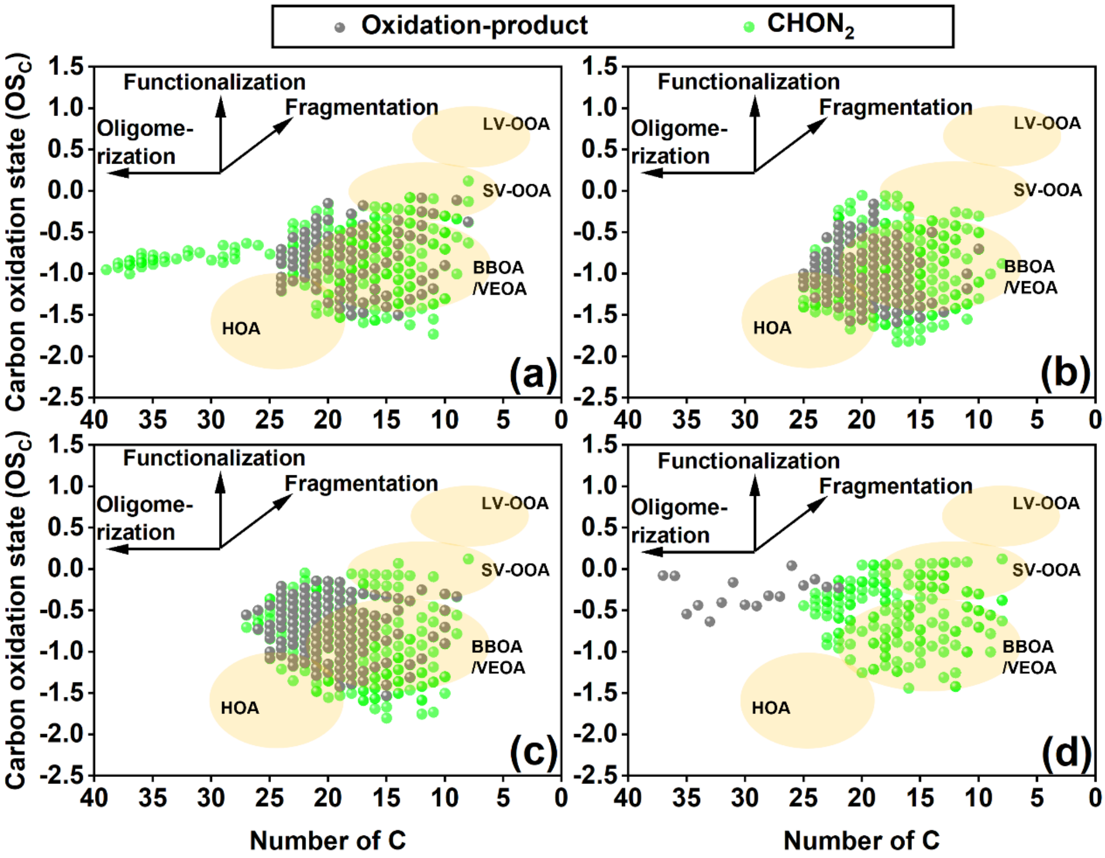

Figure S6. OS_c of unique $CHON_2$ molecules in WSOM in $PM_{2.5}$ collected from different cases: air spray vs ground aspersion on (**a**) March 23, (**b**) March 24, and (**c**) March 25 and two road segments without water spray (I vs II) on (**d**) March 26. For the above comparative cases, the unique $CHON_2$ compounds indicate the $CHON_2$ molecules identified in $PM_{2.5}$ collected from the air spray (/no water spray-I) road segments. The light orange background indicates areas of HOA (hydrocarbon-like organic aerosol), BBOA and VEOA (biomass burning and vehicle emission organic aerosols) (Kroll et al., 2011; Tong et al., 2016), SV-OOA (semivolatile oxidized organic aerosol), and LV-OOA (low-volatility oxidized organic aerosol) (Kroll et al., 2011). The grey circles refer to the identified oxidation-product pairs.

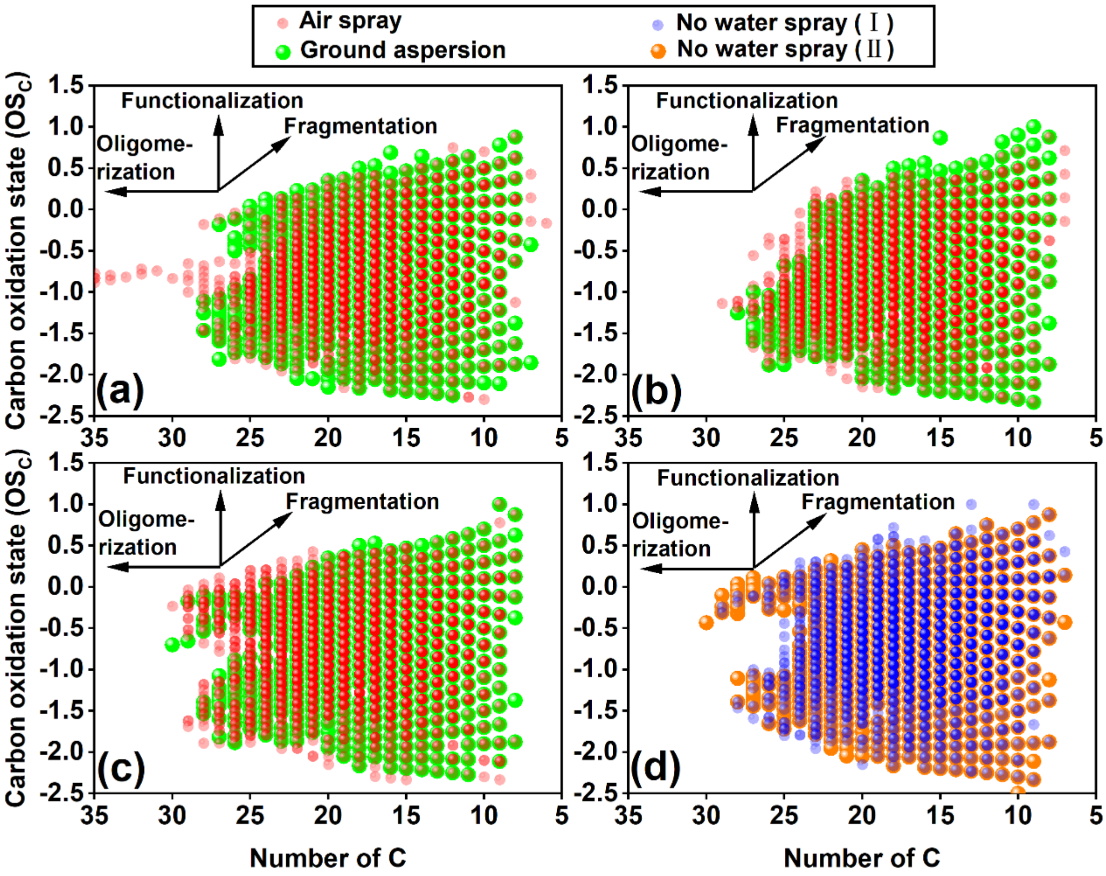

Figure S7. OS_c of each CHON molecule in WSOM in $PM_{2.5}$ collected from different cases: air spray vs ground aspersion on (**a**) March 23, (**b**) March 24, and (**c**) March 25 and no water spray (I) vs no water spray (II) on (d) March 26.

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