

## Supplementary Information to

# Secondary Aerosol Formation in Incense Burning Particles by Ozonolysis and Photochemical Oxidation via Single Particle Mixing State Analysis

Zhancong Liang<sup>1,2</sup>, Liyuan Zhou<sup>1,2</sup>, Xinyue Li<sup>1</sup>, Rosemarie Ann Infante Cuevas<sup>1,2</sup>, Rongzhi Tang<sup>1,2</sup>, Mei Li<sup>3,4</sup>, Chunlei Cheng<sup>3,4</sup>, Yangxi Chu<sup>5</sup>, Patrick. K.H. Lee<sup>1</sup>, Alvin. C.K. Lai<sup>1</sup>, Chak K. Chan<sup>1,2,6\*</sup>

<sup>1</sup> School of Energy and Environment, City University of Hong Kong, Hong Kong, China

<sup>2</sup> City University of Hong Kong Shenzhen Research Institute, Shenzhen, China

<sup>3</sup> Institute of Mass Spectrometry and Atmospheric Environment, Guangdong Provincial Engineering Research Center for On-line Source Apportionment System of Air Pollution, Jinan University, Guangzhou 510632, China

<sup>4</sup> Guangdong-Hongkong-Macau Joint Laboratory of Collaborative Innovation for Environmental Quality, Guangzhou 510632, China

<sup>5</sup> State Key Laboratory of Environmental Criteria and Risk Assessment, Chinese Research Academy of Environmental Sciences, Beijing, 100012, China

<sup>6</sup> Low-Carbon and Climate Impact Research Centre, City University of Hong Kong, Hong Kong, China

\*To whom the correspondence should be addressed: [chak.k.chan@cityu.edu.hk](mailto:chak.k.chan@cityu.edu.hk)

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**Text S1.** Estimation of the OH exposure.

SO<sub>2</sub> was used to calculate the OH exposure in the Go:PAM. The UVC lamps were turned on to warm up for ~30 min and turned off. Then, O<sub>3</sub> (300, 800, 1500 ppb) and SO<sub>2</sub> (~200 ppb) were introduced to the Go:PAM with the UVC lamps turned off until its initial concentration remained constant at steady-state conditions, which typically took around 5 min. The [SO<sub>2</sub>] was recorded as [SO<sub>2</sub>]<sub>Initial</sub>. After that, the UVC lamps were turned on until the final [SO<sub>2</sub>] stabilized and was recorded as [SO<sub>2</sub>]<sub>Final</sub>. The time scale for the stabilization of [SO<sub>2</sub>] was around 4 min. The OH exposure at each condition is calculated using Eq. (A1):

$$OH\ exposure = \frac{1}{k_{OH,SO_2}} \times -\ln\left(\frac{[SO_2]_{Final}}{[SO_2]_{Initial}}\right) \quad (A1)$$

where  $k_{OH,SO_2} = 9 \times 10^{-13} \text{ cm}^3 \text{ molec}^{-1}$  is the bimolecular rate constant between OH and SO<sub>2</sub> (Davis et al., 1979). The equation above is the result of integrating the differential rate equation for SO<sub>2</sub> and assuming pseudo-first order kinetics. The estimation of external OH reactivity (i.e., the OH reactivity with VOCs) requires VOC analysis and is not available in our study. Therefore, the OH exposure shown in this study may have been underestimated.

**Text S2.** Estimation of the mass hygroscopic growth factor (GF) by AIOMFAC model.

Based on the AIOMFAC model (Zuend et al., 2008), we obtained the weight fraction  $w$  of water and solutes (i.e., dry particles) in KNO<sub>3</sub> and KCl particles, respectively. Then, the GF is estimated as equation A2:

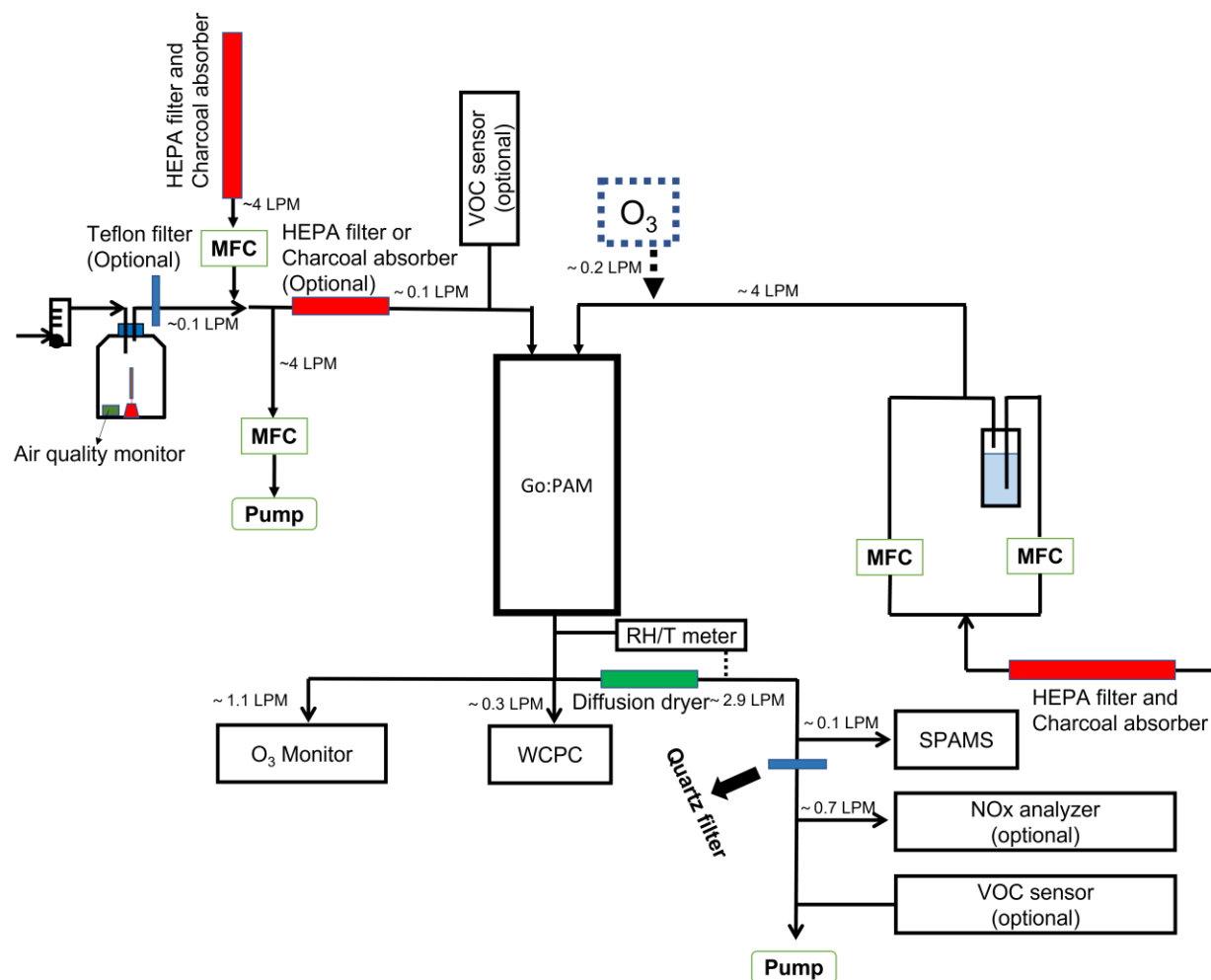
$$GF = \frac{m_{wet}}{m_{dry}} = \frac{m_{dry} + m_{water}}{m_{dry}} = 1 + \frac{m_{water}}{m_{dry}} = 1 + \frac{w_{water}}{w_{dry}} \quad (A2)$$

where  $m_{wet}$ ,  $m_{dry}$  and  $m_{water}$  are the mass of wet particle, dry particle, and particulate water at different relative humidity (RH), respectively. The GF of  $\text{KNO}_3$  and  $\text{KCl}$  particles as a function of time is shown in Figure S18. We did not consider efflorescence in the figure since the efflorescence RH for the  $\text{KNO}_3$  and  $\text{KCl}$  are around 50%, much lower than 80% used in this study.

**Text S2.** The evolution of organic fragments.

We only showed NF ratios larger than 1 to focus on SOA formation. The positive spectra of both  $\text{O}_3$ -Dark-aged and  $\text{O}_3$ -UV-aged particles show NF increases for +30[NO] or  $[\text{CH}_2\text{NH}_2]$  (possibly due to nitrates, oxidized NOC, or amine), +44[ $\text{CO}_2$ ] or  $[\text{N}_2\text{O}]$  (oxidized organics), +53[ $\text{C}_4\text{H}_5$ ] and +69[ $\text{C}_5\text{H}_9$ ] (aromatic hydrocarbons) (Wang et al., 2009; Silva et al., 2000; Dall'osto et al., 2013) (Figure S19). The negative spectra show increases for -137[ $\text{C}_8\text{H}_9\text{O}_2$ ] (possibly methyl guaiacol) (Pagels et al., 2013; Gaie-Levrel et al., 2012) and -57[ $\text{C}_2\text{HO}_2$ ] (a glyoxylate fragment) (Sullivan et al., 2007; Cheng et al., 2017), as well as -16[O].  $\text{O}_3$ -UV-aged particles showed 200-folds and 10-folds NF increases for -137[ $\text{C}_8\text{H}_9\text{O}_2$ ] and -57[ $\text{C}_2\text{HO}_2$ ], respectively, significantly greater than that for UV-aged particles (~2-folds). Compared to OH chemistry, the UV photoactivity of compounds in particulates contributes minorly to organic chemistry (Figure S19).

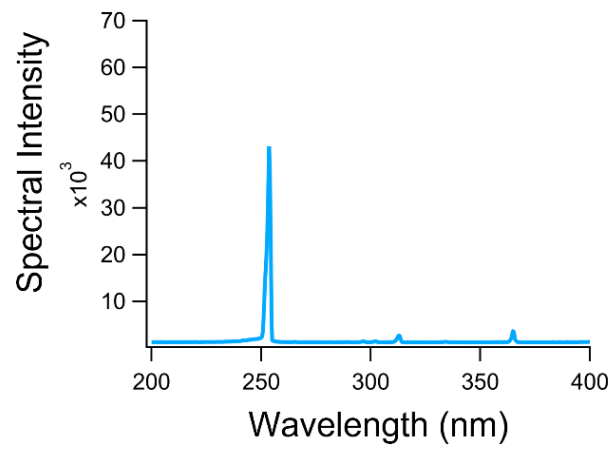
Figure S20 show the NF of the new peaks in aged particles, which cannot (Dall'osto et al., 2009) be shown in the NF ratio plot due to their absence in fresh particles (zero denominator).  $\text{O}_3$ -Dark-aged particles show NF decreases of  $m/z$  +186 to +189 (probably PAHs) (Dall'osto et al., 2009) with increasing  $[\text{O}_3]$ . In contrast,  $\text{O}_3$ -UV-aged particles show NF increases of -31[ $\text{CH}_3\text{O}$ ] or [HON], +123[ $\text{C}_7\text{H}_7\text{O}_2$ ] and +124[ $\text{C}_7\text{H}_8\text{O}_2$ ] (probably guaiacol) (Diab et al., 2015), and  $m/z$  +140 (probably HULIS) (Qin et al., 2006) with increasing OH exposure. These apparent changes in NF of organics fragments indicate the oxidative evolution of organics and likely formation of SOA, although the molecular characterization was hindered by severe fragmentation.



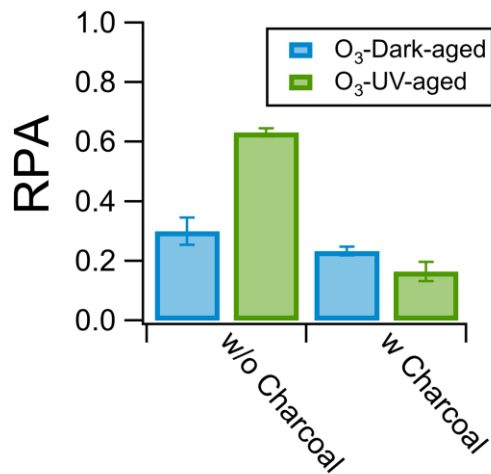
**Figure S1.** The schematic of the experimental set-up. The NO<sub>x</sub> analyzer and the VOC sensor were used only in the experiment for determining the gas removal efficiency and NO<sub>x</sub> decay under OH exposure.



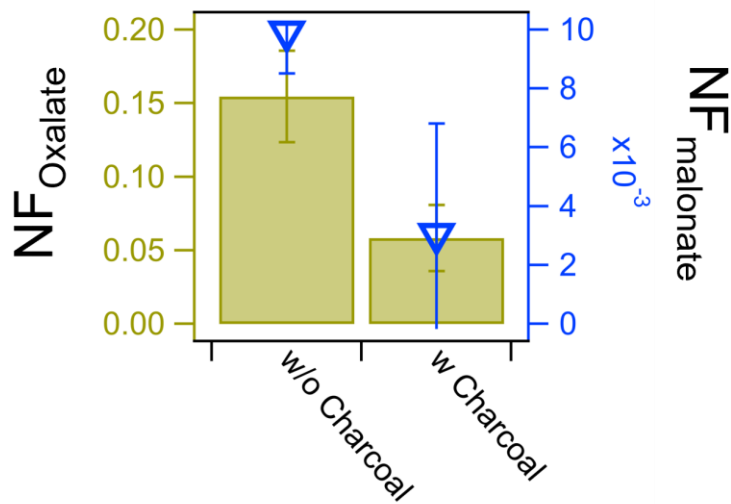
**Figure S2.** The appearance of the incense sticks.



**Figure S3.** The emission spectrum of the UVC lamps.

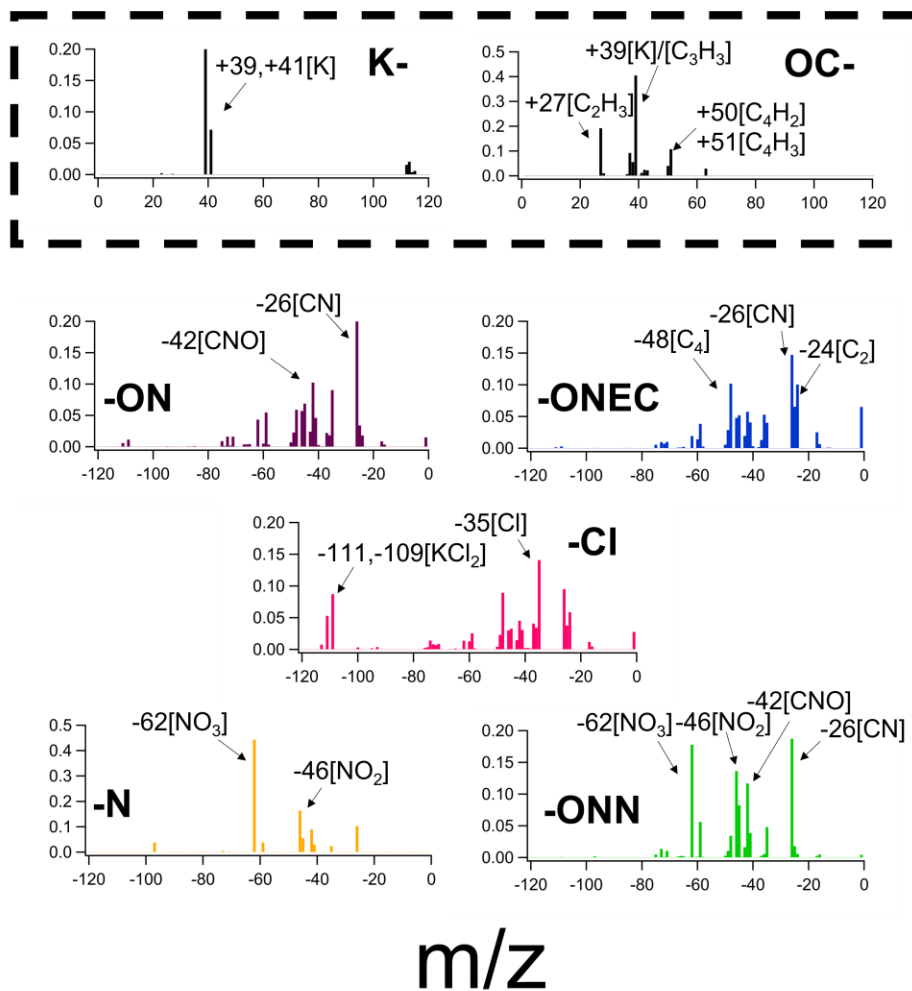


**Figure S4.** RPA of summed nitrate peaks of aged incense burning particles in the presence and absence of charcoal absorber.



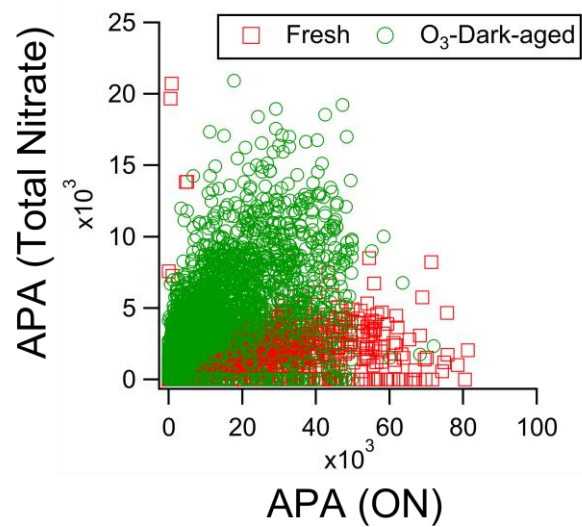
**Figure S5.** NF of oxalate and malonate of O<sub>3</sub>-UV-aged incense burning particles (1500ppb O<sub>3</sub> + UV) in the presence and absence of a charcoal absorber.

RPA

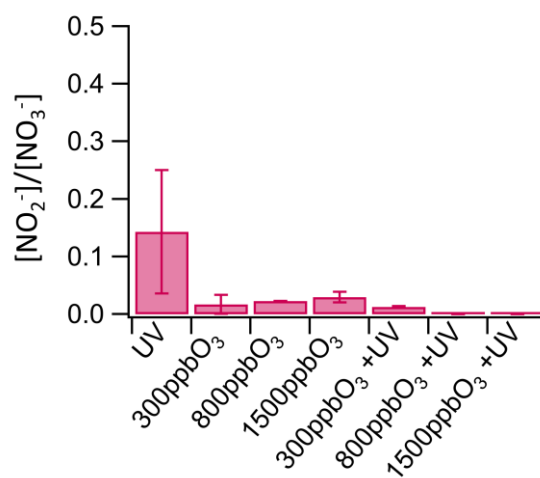


**Figure S6.** Positive (K- and OC-) and negative spectra (-ON, -ONEC, -Cl, -N, and -ONN) of different categorizations of particles. The spectral characteristics of these categories are similar under different conditions.

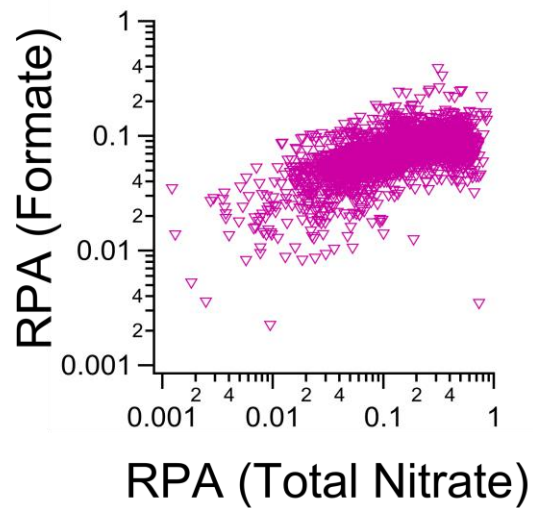




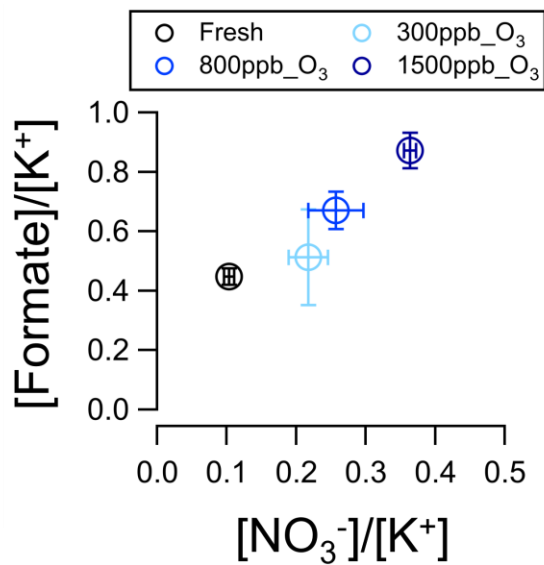
**Figure S7.** APA of ON as a function of APA of total nitrate in fresh and O<sub>3</sub>-Dark-aged particles.



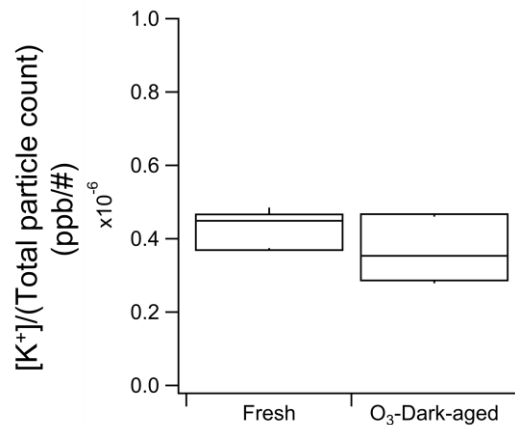
**Figure S8.**  $[\text{NO}_2^-]/[\text{NO}_3^-]$  in water-extract of aged particles. Noted that the  $\text{NO}_2^-$  in 800ppb O<sub>3</sub>+UV and 1500ppb O<sub>3</sub>+UV experiments are undetectable.



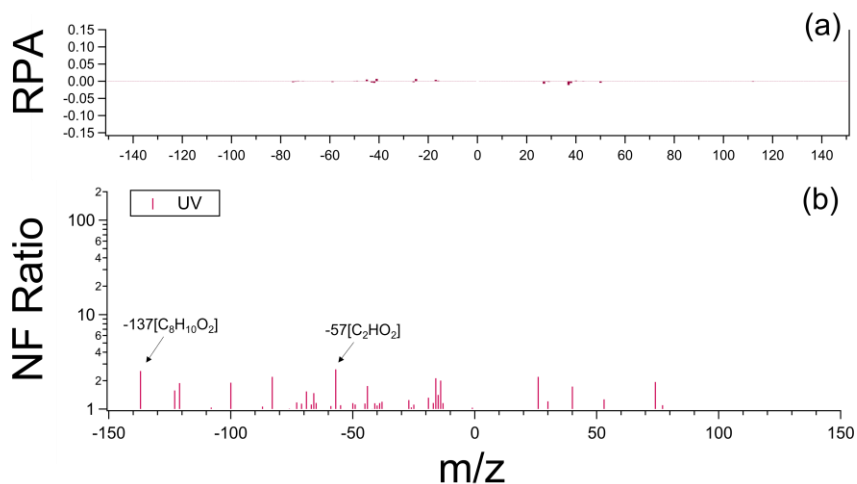
**Figure S9.** RPA of total nitrate as a function of RPA of formate.



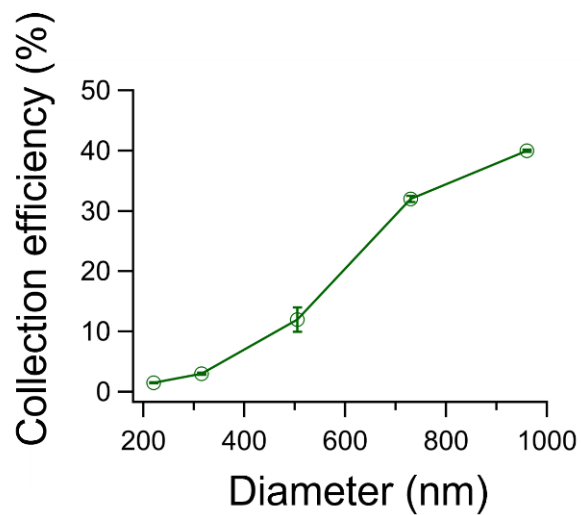
**Figure S10.**  $[\text{Formate}]/[\text{K}^+]$  as a function of  $[\text{NO}_3^-]/[\text{K}^+]$  in the water-extract of fresh and  $\text{O}_3$ -Dark-aged incense burning particles.



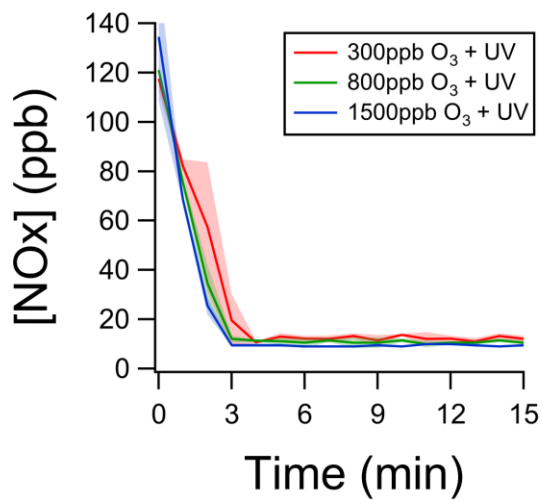
**Figure S11.** The whisker-box plot of the [K<sup>+</sup>] in the particle water-extract measured by IC normalized by the total counts of collected particles for IC measurement. The error bar shows one standard deviation. We assume there was no new particle formation under ozone exposure since the WCPC showed comparable particle number concentration in the presence and absence of ozone.



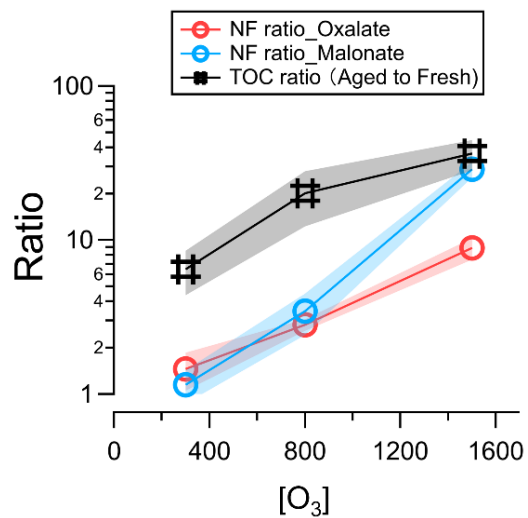
**Figure S12.** (a) The difference (aged minus fresh) of the average organic spectra of UV-aged particles; (b) The NF ratio (aged to fresh) of UV-aged particles to fresh particles, as a function of m/z [-150, 150].



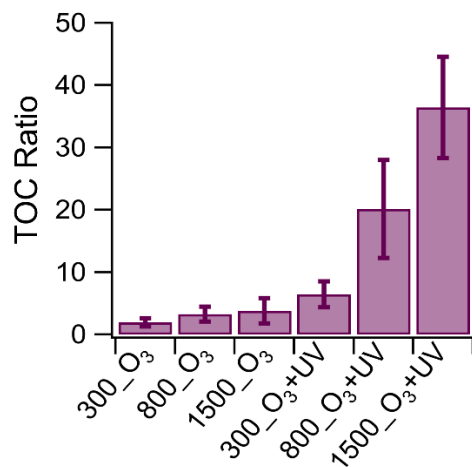
**Figure S13.** Relation between the particle collection efficiency of Polystyrene Latex particles and diameter in SPAMS.



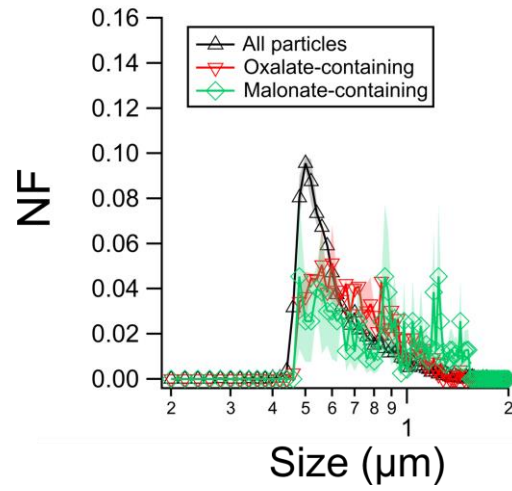
**Figure S14.** Evolution of [NOx] under different OH exposure as a function of time. The shadings show one standard deviation. The [NOx] was equilibrated without O<sub>3</sub> and UV at 0 min.



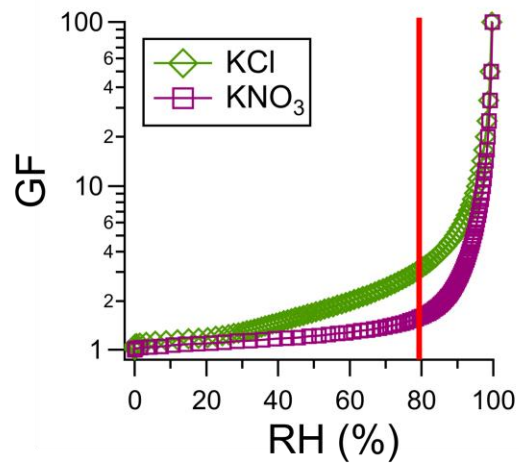
**Figure S15.** The NF ratios (aged to fresh) of oxalate and malonate, and TOC ratios as a function of [O<sub>3</sub>] in O<sub>3</sub>+UV aging experiments.



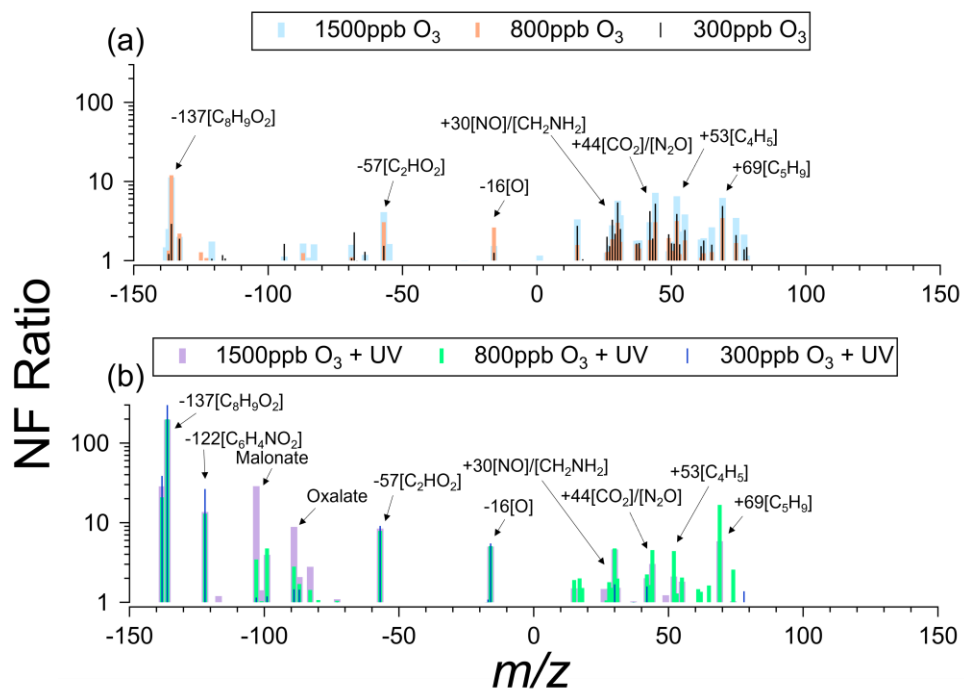
**Figure S16.** The total organic content (TOC) ratios of aged to fresh particles extract.



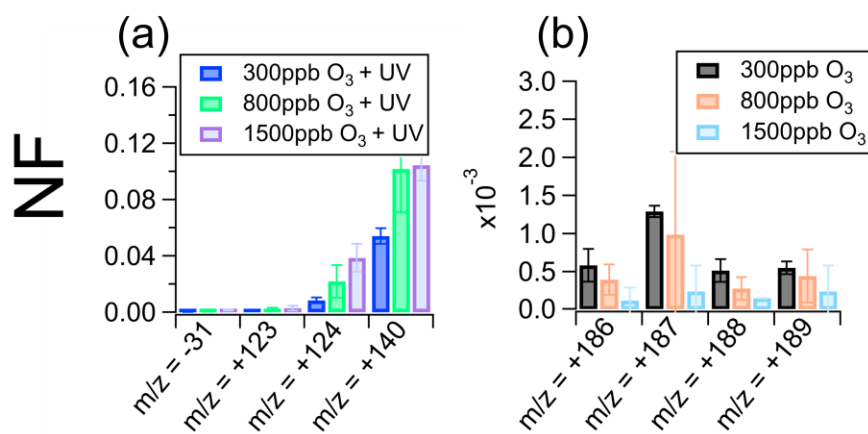
**Figure S17.** Size distribution (0.2-2  $\mu\text{m}$ ) of  $\text{O}_3$ -UV-aged particles (at 1500 ppb  $\text{O}_3$  and UV) and their dicarboxylate-containing fractions. The shadings show one standard deviation.



**Figure S18.** The GF of KCl and  $\text{KNO}_3$  particles as a function of RH. The red line denotes 80% RH.



**Figure S19.** The NF ratio (aged to fresh) of (a) O<sub>3</sub>-Dark-aged, (b) O<sub>3</sub>-UV-aged particles to fresh particles, as a function of  $m/z$  [-150, 150].



**Figure S20.** The NF of new organic peaks in aged particles.

**Table S1.** The classification of particles and RPA of total nitrate peaks of aged particles at different laser fluences.

Experimental Conditions	Laser fluence (mJ)	OC-ON (%)	OC-N (%)	K-ON (%)	K-ONN (%)	K-N (%)	K-ONEC (%)	K-Cl (%)	RPA <sub>Total nitrate</sub>
O <sub>3</sub> -dark-aged (800 ppb)	0.6	(7.6±1.0)	(21.9±1.7)	(29.0±0.7)	(11.2±0.9)	(2.5±0.5)	(22.8±1.8)	(3.9±0.4)	(0.40±0.15)
	1.2	(7.2±1.4)	(22.6±4.0)	(28.5±4.3)	(10.6±0.2)	(3.3±1.3)	(22.8±2.5)	(4.0±0.1)	(0.37±0.09)
O <sub>3</sub> -UV-aged (800 ppb)	0.6	(0.0±0.0)	(36.7±7.2)	(0.0±0.0)	(26.7±2.1)	(35.5±4.2)	(0.0±0.0)	(0.0±0.0)	(0.66±0.03)
	1.2	(0.0±0.0)	(35.6±2.3)	(0.0±1.0)	(28.6±1.9)	(33.6±4.7)	(0.2±0.2)	(0.0±0.0)	(0.67±0.07)

**Table S2.** Potential peaks from the inorganics and elemental carbons.

	m/z	Formula	Ref
Inorganic salts	-163	KNO <sub>3</sub>	(Pratt et al., 2011)
	-155	Na <sub>2</sub> Cl <sub>3</sub>	(Harrison et al., 2012)
	-153		
	-151		
	-147	Na(NO <sub>3</sub> ) <sub>2</sub>	(Ault et al., 2014)
	-131	NaNO <sub>3</sub> NO <sub>2</sub>	(Ault et al., 2014)
	-125	H(NO <sub>3</sub> ) <sub>2</sub>	
	-113	KCl <sub>2</sub>	(Bi et al., 2011)
	-111		
	-109		
	-101	KNO <sub>3</sub> /NaNONO <sub>2</sub>	(Ault et al., 2014)
	-97	HSO <sub>4</sub> /NaCl <sub>2</sub>	(Liang et al., 2022)
	-96		
	-95	NaCl <sub>2</sub>	(Ault et al., 2014)
	-93		
	-62	NO <sub>3</sub>	(Cheng et al., 2017)
	-58	NaCl	(Zhu et al., 2020)
	-46	NO <sub>2</sub>	(Cheng et al., 2017)
	-37	Cl	(Ault et al., 2014)
	-35		
	+23	Na	(Dall'osto et al., 2004)
	+39	K	(Bi et al., 2011)
	+41		
	+63	Na <sub>2</sub> OH	(Yang et al., 2009)
	+81	Na <sub>2</sub> Cl	(Dall'osto et al., 2004)
	+83		
+97	NaKCl	(Gross et al., 2000)	
+113	K <sub>2</sub> Cl	(Silva et al., 1999)	
+115			



	+139	Na <sub>3</sub> Cl <sub>2</sub>	(Dall'osto et al., 2004)
	+141		
Elemental carbons	±12n	C <sub>n</sub>	(Zhou et al., 2022)

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