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# Continuous measurements at the urban roadside in an Asian Megacity by Aerosol Chemical Speciation Monitor (ACSM): particulate matter characteristics during fall and winter seasons in Hong Kong

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## Abstract

Non-refractory submicron aerosol is characterized using an Aerosol Chemical Speciation Monitor (ACSM) in the fall and winter seasons of 2013 at the roadside in an Asian megacity environment in Hong Kong. Organic aerosol (OA), characterized by application of Positive Matrix Factorization (PMF), and sulfate are found dominant. Traffic-related organic aerosol shows good correlation with other vehicle-related species, and cooking aerosol displays clear meal-time concentration maxima and association with surface winds from restaurant areas. Contributions of individual species and OA factors to high NR-PM<sub>1</sub> are analyzed for hourly data and daily data; while cooking emissions in OA contribute to high hourly concentrations, particularly during meal times, secondary organic aerosol components are responsible for episodic events and high day-to-day PM concentrations. Clean periods are either associated with precipitation, which reduces secondary OA with a lesser impact on primary organics, or clean oceanic air masses with reduced long-range transport and better dilution of local pollution. Haze events are connected with increases in contribution of secondary organic aerosol, from 30 to 50% among total non-refractory organics, and influence of continental air masses.

## 1 Introduction

The Special Administrative Region of Hong Kong (HKSAR) is a global logistics and finance center located at the south-eastern edge of the Pearl River Delta Region (PRD), China's largest manufacturing area and one of the world's most densely populated regions. Hong Kong has been plagued by deteriorating air quality, attributed to local emissions from traffic, residential and commercial activity, regional pollution from the PRD and long-range transport (Nie et al., 2013; Wong et al., 2013; Yuan et al., 2013).

High-time-resolution online instruments can characterize ambient aerosols quickly and mitigate the influence of changing environmental conditions. Few real-time studies

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have been conducted in Hong Kong aside from recent measurement campaigns conducted by high resolution aerosol mass spectrometer (HR-AMS) (Lee et al., 2013; Li et al., 2013, 2015; Huang et al., 2015). Long-term AMS studies tend to be costly and time-consuming due to the complexity of the instrument. The ACSM, whose design is based on the AMS but has been substantially simplified, has seen a growing trend of use due to its comparative ease of operation, robustness, and sufficient time resolution (~ 20–60 min) for studies spanning months or longer. (Ng et al., 2011; Sun et al., 2012, 2013a, b; Budisulistiorini et al., 2013; Canonaco et al., 2013; Takahama et al., 2013; Bougiatioti et al., 2014; Petit et al., 2015; Ripoll et al., 2015; Tiitta et al., 2014; Minguillón et al., 2015).

This study presents the first ACSM characterization of particulate matter at a busy urban roadside station in Hong Kong with heavy traffic and commercial and residential activities. It aims to provide long-term characterization of particulate matter sources in a typical inner-city environment, enabling the identification of the relative importance of different sources and typical recurring patterns.

## 2 Experimental

The roadside measurement data were collected from 3 September to 31 December 2013 in Mong Kok (MK), an urban area with dense buildings and population in the Kowloon peninsula as part of a Hong Kong Environmental Protection Department (HKEPD) project (ref.: 13-00986). The sampling site was next to the road-side air quality monitoring station (AQMS) of HKEPD at the junction of the heavily trafficked Nathan Road and Lai Chi Kok Road (22°19'2" N, 114°10'06" E). The distribution of businesses in the vicinity varies, with restaurants mainly to the east, commercial buildings to the south and east, small shops for interior decoration, furniture and electrical goods to the west and residential buildings to the north of the sampling location (Lee et al., 2015). The sampling setup is described in detail in the Supplement, Sect. 1.

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Non-refractory PM<sub>1</sub> (NR-PM<sub>1</sub>) species (sulfate, nitrate, ammonium, chloride, and organics) were measured in-situ by an Aerodyne Aerosol Chemical Speciation Monitor (ACSM, SN: 140–154). Other data including meteorological data (wind, temperature, relative humidity, solar irradiation), volatile organic compounds (VOCs) measured by an online gas-chromatography system (GC955-611 and GC955-811, Synspec BV), and standard criteria pollutants (NO<sub>x</sub>, SO<sub>2</sub> and PM<sub>2.5</sub>) were provided by the HKEPD, with equipment details available from the HKEPD air quality reports (Environmental Protection Department, 2013).

The acquired 20 min-average data were treated according to the general ACSM data analysis protocols established in previous studies (Ng et al., 2011; Sun et al., 2012), using the standard WaveMetrics Igor Pro-based Data Analysis Software (Version 6.3.5.5) and incorporating calibrations for relative ionization efficiency (RIE), collection efficiency (CE) and detection limit (DL). Further details on data treatment can be found in the Supplement, Sect. 2.

Factors contributing to organic aerosol were explored using PMF (Paatero and Tapper, 1994; Zhang et al., 2011) with the Igor-Pro-based PMF evaluation toolkit (PET) (Ulbrich et al., 2009). In general, PMF can be used to resolve factors as organic aerosol (OA) into hydrocarbon-like OA (HOA), cooking OA (COA), semi-volatile oxygenated OA (SV-OOA), low-volatility oxygenated OA (LV-OOA), and others. The optimal factor number was determined by inter-comparing factors' mass spectra and time series, correlations between factors and related tracers, and correlations with standard mass spectra; solutions with 3, 4, and 5 factors at  $f_{\text{peak}} = 0$  were explored, after which the optimal  $f_{\text{peak}}$  value was determined by repeating the above analysis with varying  $f_{\text{peak}}$  values.

The 4-factor solution (HOA, COA, SV-OOA, LV-OOA) is optimal, with  $Q/Q_{\text{exp}} = 0.8$  and better differentiation between factor time-series ( $R_{\text{pr}} < 0.6$ ; Fig. S4). The factors also correlate well with associated inorganics and external tracers (NO<sub>3</sub>, SO<sub>4</sub>, NH<sub>4</sub>, NO<sub>x</sub>; Zhang et al., 2005, 2011; Ulbrich et al., 2009), e.g. HOA with NO<sub>x</sub>, SV-OOA with NO<sub>3</sub>, LV-OOA with SO<sub>4</sub> and NH<sub>4</sub> (Table S4). Furthermore, the resolved mass spectra of four factors exhibit good similarity (all un-centered  $R$  ( $R_{\text{uc}}$ ) > 0.80) with reference

source mass spectra from the AMS MS database (Ulbrich, I. M., Lechner, M., and Jimenez, J. L., AMS Spectral Database, url: <http://cires.colorado.edu/jimenez-group/AMSsd>; Ulbrich et al., 2009). PMF diagnostic details are shown in the Supplement (Sect. 3) and Fig. S5. We note that  $m/z$  60 and 73 were resolved not only in COA but also in SV-OOA. When PMF was run using only nighttime data (between 00:00 and 06:00 LT), when there is little COA (Fig. S6), these two ions still persist, with similar fractional intensities in SV-OOA as at other times. The time series of  $m/z$  60 and 73 also track well with SV-OOA, with  $R_{pr}$  of 0.92 and 0.93 respectively; hence, we believe that their presence in SV-OOA is not the result of artifacts in PMF.

### 3 Results and discussion

#### 3.1 Mass concentration and chemical composition

Figure 1a and b displays meteorological data (relative humidity, temperature, and precipitation) and mass concentrations of non-refractory  $PM_1$  (NR- $PM_1$ ) species and organic aerosol (OA) components, respectively, between September and December 2013. Total NR- $PM_1$  concentrations vary from 2.1 to 76.4  $\mu\text{g m}^{-3}$  with an average of  $25.9 \pm 13.0 \mu\text{g m}^{-3}$ . ACSM NR- $PM_1$  concentrations co-vary with that of  $PM_{2.5}$  measured by TEOM ( $R^2 = 0.64$ , slope = 0.59; Fig. S1); the low slope value may be caused by the different size cuts of ACSM and TEOM and the presence of refractory materials such as elemental carbon (and to a lesser extent mineral dust and sea salt) which the ACSM cannot detect. Overall, daily  $PM_{2.5}$  concentrations range from 3.7 to 106.0  $\mu\text{g m}^{-3}$  and are largely (90.0 %) within the 24 h air quality standard of 75  $\mu\text{g m}^{-3}$  set by the Hong Kong Air Quality Objectives (HKAQO). Days with better air quality ( $PM_{2.5} < 35 \mu\text{g m}^{-3}$ ) are mainly observed in the month of September and in rainy periods of other months. The prevailing winds from the ocean in September not only bring in less polluted air mass but also dilute the local air pollutants compared with other seasons (Yuan et al.,

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2006; Li et al., 2015). Precipitation has an obvious impact on total NR-PM<sub>1</sub> concentrations, but as we will discuss, has a lesser effect on primary organics.

Overall, NR-PM<sub>1</sub> is dominated by organics and sulfate with relative contributions of 58.2 and 23.3% and average concentrations of  $15.1 \pm 8.1 \mu\text{g m}^{-3}$  and  $6.0 \pm 3.5 \mu\text{g m}^{-3}$ , respectively (Fig. 2a). Other inorganic species (ammonium, nitrate and chloride) amount to approximately 20% of NR-PM<sub>1</sub>. The dominance of organics and sulfate is consistent with previous on-line studies in urban areas (e.g., Salcedo et al., 2006; Aiken et al., 2009; Sun et al., 2012, 2013b) as well as previous filter-based studies in MK (e.g., Louie et al., 2005; Cheng et al., 2010; Huang et al., 2014). The measured composition is consistent with earlier HR-AMS measurements carried out at the same site in spring and summer 2013 (Lee et al., 2015) with very similar overall species distribution, but slightly lower measured concentrations as compared to the ACSM, likely due to the fact that sampling took place in different time periods (spring-summer 2013 for the AMS campaign, fall-winter 2013 for the ACSM campaign). In the AMS study, 6 PMF aerosol factors were identified (one additional OOA factor and one additional COA factor). A marked difference is observed in the distribution of primary OA (POA) and secondary OA (SOA); whereas in spring and summer (AMS), POA makes up 65% of total organics, the reverse is observed for fall and winter (ACSM) where POA only amounts to 42% overall. A possible reason for this discrepancy is the fact that impacts of regional pollution and long-range transport are usually higher during fall and winter (Yuan et al., 2013; Li et al., 2015), thus contributing more SOA.

Elemental carbon (EC) concentrations are significant at the Mong Kok site but not measureable by ACSM due to its high refractory temperature. EC has been discussed extensively in the previously mentioned filter-based studies and a brief comparison of online ECOC measurements to the results of HR-AMS measurements has been presented in an HR-AMS study (Lee et al., 2015). We therefore do not discuss EC in detail in this work.

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19:00–21:00 LT) and non-meal times (00:00–06:00 LT); the non-meal period is defined by the periods of low concentration ( $< 2 \mu\text{g m}^{-3}$ ) in the COA diurnal pattern. During dinner time, the average concentration of organics increases by about  $11 \mu\text{g m}^{-3}$  and its contributions in total NR-PM<sub>1</sub> increase to 70 %, while the concentrations of other species do not change much (Fig. 6b). As shown in Fig. 6c, the increase in organic concentrations results from the increase in COA from  $1.7$  to  $7.8 \mu\text{g m}^{-3}$  ( $\sim 360$  % increase), and to a lesser extent increases in SV-OOA (from  $1.5$  to  $4.5 \mu\text{g m}^{-3}$ , a  $\sim 200$  % increase) and in HOA (from  $1.4$  to  $3.2 \mu\text{g m}^{-3}$ , a  $\sim 130$  % increase). As shown in Table 1, the average concentration of organics during dinner time is  $5 \mu\text{g m}^{-3}$  higher than that during lunch, and this increase is attributed to the increase of COA and SV-OOA mass but not of HOA. This is consistent with the expectation that the cooking activities at MK are higher during dinner than during lunch, while traffic during dinner is comparable to or smaller than that during lunch (Fig. 4f and h). The increase of SV-OOA during dinner time may be the result of enhanced cooking emissions and possibly less evaporation due to lower ambient temperature; contributions from traffic emissions are not likely to be important since there is little increase of HOA during meal time.

### 3.2.3 Oxygenated OA (OOA)

The LV-OOA spectrum correlates well with the standard LV-OOA spectrum (Fig. 3), with a  $R_{\text{uc}}$  of 0.97. The LV-OOA time series is associated with that of  $\text{SO}_4^{2-}$  with a  $R_{\text{pr}}$  of 0.86 (Fig. 1), consistent with reports in the literature (DeCarlo et al., 2010; He et al., 2011; Zhang et al., 2014; Tiitta et al., 2014). The LV-OOA diurnal pattern varies little, suggesting that it is part of the background aerosol, possibly resulting from long range transport (Li et al., 2013, 2015).

The mass spectrum of SV-OOA closely resembles that of “standard” SV-OOA (Fig. 3). Some marker fragments of COA and HOA, for example,  $m/z$  41, 43, 55, and 57, are present in the SV-OOA mass spectrum. SV-OOA concentrations are also weakly associated with those of HOA and their co-emitted precursors (benzene and toluene), with  $R_{\text{pr}}$  of 0.58, 0.65 and 0.51 respectively. In fact, the correlation between





those observed in other periods. Thus, HOA overall has a stronger relationship to SV-OOA than COA has. Cooking emissions reflected by COA are not as important to SV-OOA in the BT periods but they can be important during MT periods. Some SV-OOA might also have converted to LV-OOA under  $\text{HiO}_x$ , although overall most LV-OOA is considered to be from transport.

### 3.3 Diurnal patterns

The diurnal profiles of NR-PM<sub>1</sub> species and OA components are depicted in Fig. 4. Total organics display a diurnal pattern with two pronounced peaks during 12:00–14:00 and 19:00–21:00 LT, corresponding to the peaks of COA at lunch and dinner time respectively. In addition, organics increase at about 10 a.m., which may be related to the increase of local emissions of HOA and COA by 2.3 and 1.1  $\mu\text{g m}^{-3}$  respectively from 06:00 to 10:00 LT.

The mass concentration of sulfate (Fig. 4b) does not show any diel variation. It is likely that sulfate, as a regional pollutant, is mainly formed during long-range transport, leading to the lack of a specific diurnal pattern at MK; a similar flat diurnal pattern for sulfate has also been found at the HKUST supersite in Hong Kong (Lee et al., 2013; Li et al., 2015). These results differ significantly from observations in Beijing and Lanzhou in China and Welgegund in South Africa (Sun et al., 2012, 2013b; Xu et al., 2014; Tiitta et al., 2014) where sulfate displays an obvious increase at noon-time in summer and wet seasons due to either photochemical reaction or aqueous oxidation of  $\text{SO}_2$ . The difference may result from the much lower level of sulfur dioxide ( $\text{SO}_2$ ) with an average of 4.6 ppb in MK compared to for example, ~ 32 ppb in Beijing, where coal combustion leads to a much higher  $\text{SO}_2$  concentration (Lin et al., 2011); sulfate and relative humidity (RH) have almost no correlation ( $R^2 = 0.06$ ) in MK, suggesting that local aqueous processing may not be significant for sulfate observed at Mong Kok.

Nitrate shows a slight dip around noontime, corresponding to the increase of the ambient temperature (Fig. 4j); evaporative loss of particulate nitrate might outweigh the secondary production of nitrate during this time. The diurnal pattern of ammonium

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(Fig. 4d) is very similar to that of sulfate, as expected based on their commonly observed association in atmospheric particles. Chloride (Fig. 4e) has rather low concentrations and shows a similar diurnal variation to that of nitrate, likely due to its volatility.

### 3.4 Day-of-week patterns

Figure 8a shows the average concentration trends on individual days of the week for NR-PM<sub>1</sub> species and Fig. 8b describes the diurnal patterns of the OA components for weekdays, Saturdays and Sundays, respectively. Because of the small datasets on Saturdays and Sundays, data beyond one standard deviation from the mean ( $25.9 \pm 13.0 \mu\text{g m}^{-3}$ ) were removed from the whole dataset to remove the influence of episodic events in this analysis. Overall, total NR-PM<sub>1</sub> concentrations have no obvious variation (average variation less than 5%) from Monday to Saturday, but drop by 16% on Sundays compared to Saturdays. This weekend difference is opposite to the result found in Beijing where higher concentrations occur on Sundays than Saturdays (Sun et al., 2013b). On the other hand, some others such as Lough et al. (2006) and Rattigan et al. (2010) reported that both Saturdays and Sundays had obvious traffic emissions reduction due to less human activities on weekends in Los Angeles and New York, respectively.

Organics and secondary inorganics (SO<sub>4</sub>, NH<sub>4</sub> and NO<sub>3</sub>) contributed 54 and 46% respectively to the concentration difference between Sundays and Saturdays in MK. The difference in organics is mainly attributed to the variation of HOA, which shows very similar diurnal variations on Saturdays and weekdays, but has an average decrease of 23% after 07:00 LT on Sundays. A 37% reduction of traffic-related carbonaceous aerosol on Sundays compared with weekdays in MK has been reported (Huang et al., 2014). In Hong Kong many people work on Saturday, which leads to a traffic pattern similar to normal weekdays. COA shows nearly the same diurnal patterns on all days, and LV-OOA and SV-OOA do not show obvious variations. Overall, local emissions from traffic contribute most to the day-of-week variations in organics.

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high hourly PM levels, LV-OOA/SV-OOA are responsible for episodic events and high day-to-day PM levels.

To analyze the difference in particle composition and meteorological conditions among episodic periods and clean periods, three heavy polluted episodes (19–22, 23–26 October and 10–13 December) and two clean periods (17–18 September and 14–18 December), highlighted in shaded regions in Fig. 1, were analyzed. The average concentrations of these chosen periods are larger than one standard deviation from the average concentration of the campaign ( $25.9 \pm 13.0 \mu\text{g m}^{-3}$ ). The composition, meteorological features ( $T$  and RH) and oxidation index ( $\text{O}_x$  and  $f_{44}$ ) of these five events are shown in Table 4. Clean period 1 (C1) is characterized by low NR-PM<sub>1</sub> concentrations (below  $13 \mu\text{g m}^{-3}$ ), prevailing coastal wind (easterly wind), lack of rain, high ambient temperature ( $\sim 28^\circ\text{C}$ ) and high relative humidity ( $\sim 70\%$ ). Another clean period (C2) features continuous precipitation with the coldest and most humid weather condition in the period studied. Haze period 1 (H1) has similar temperature and humidity as C1 but is marked by mixed continental/oceanic winds. From H1 to the following haze period (H2), the observed wind direction shifts to reflect continental transport, with a significant decrease in RH to 36%. Haze period 3 (H3), just before C2, is also dominated by continental winds but with lower temperatures ( $\sim 19^\circ\text{C}$ ) than during other haze events.

The total NR-PM<sub>1</sub> of C1 ( $12.2 \mu\text{g m}^{-3}$ ) is only 25–30% of that during haze periods and this is mainly attributed to easterly wind bringing less air pollutants and diluting local air pollutants. HOA, COA and SV-OOA in C1 are lower than in C2 despite the lack of rain during C1; their low concentrations during C1 may be influenced by both particle evaporation during high temperatures, especially for SV-OOA, and dilution of local emissions

Compared to the adjacent period H3, precipitation in C2 dramatically reduces the concentration of secondary species such as  $\text{SO}_4$ ,  $\text{NH}_4$ ,  $\text{NO}_3$ , SV-OOA and LV-OOA, but not primary HOA and COA. The total organic mass reduces by 68% to an average of  $8.1 \mu\text{g m}^{-3}$  (Table 4). Precipitation effectively removes secondary particles but is less efficient for primary particles that are continuously generated locally.

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The contributions of individual species and OA factors to high NR-PM<sub>1</sub> were analyzed based on hourly data and daily data. It suggests that while cooking is responsible for the hourly high concentrations during meal times, LV-OOA/SV-OOA are responsible for episodic events and high daily PM concentration. Three heavily polluted episodes and two clean periods were recorded during sampling and attributed to different meteorological and circulatory conditions. The analysis of clean periods shows that precipitation has an obvious deposition impact on total NR-PM<sub>1</sub> concentrations, but has a lesser effect on primary organics. Clean ocean wind not only brings in less polluted air mass, but also dilutes the local air pollutants. During this campaign, high-PM events are generally related to continental air mass influence or land-see breeze circulatory conditions, which has less influence on primary emissions but significant effects on secondary particles, with a pronounced increase in the secondary OA contribution during haze events (from 30 to 50 %).

**The Supplement related to this article is available online at doi:10.5194/acpd-15-19405-2015-supplement.**

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**Table 1.** Average concentrations of NR-PM<sub>1</sub> and OA components during lunch time, dinner time and non-meal times.

Species $\mu\text{g m}^{-3}$	Lunch	Dinner	Non-meal
Org	18.8	23.7	10.3
SO <sub>4</sub>	5.8	6.1	6.3
NH <sub>4</sub>	2.6	2.9	3.0
NO <sub>3</sub>	1.4	1.8	1.6
Chl	0.1	0.2	0.2
Organic aerosol components			
HOA	3.2	3.2	1.4
COA	6.2	9.6	1.7
LV-OOA	5.8	5.4	5.6
SV-OOA	3.6	5.5	1.5

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**Table 2.** Regression of SV-OOA on HOA, COA and LV-OOA and concentrations of OA factors and  $O_x$  under high and low temperature (LT and HT) of the three chosen periods (MT, BT and OT).

Period Temperature	Meal time (MT)		Background time (BT)		Other time (OT)	
	LTemp	HTemp	LTemp	HTemp	LTemp	HTemp
	Coefficients <sup>a</sup>					
HOA	0.80	0.56	0.70	0.43	0.48	0.23
COA	0.29	0.15	0.22	0.00	0.31	0.11
LV-OOA	0.25	0.23	0.23	0.24	0.25	0.28
Adjusted $R^2$	0.90	0.81	0.83	0.57	0.85	0.73
	Average Concentration ( $\mu\text{g m}^{-3}$ , ppb)					
HOA	3.71	2.85	1.60	1.18	3.51	2.88
COA	7.34	7.40	1.61	1.54	3.50	3.74
LV-OOA	5.46	5.57	5.91	5.07	5.85	5.99
SV-OOA	6.30	3.89	2.68	1.44	4.1	2.39
$O_x$ (ppb)	83.12	85.23	58.71	53.45	75.06	76.77

<sup>a</sup> The coefficient of HOA, COA and LV-OOA in the regression equation reconstructing SV-OOA under LTemp ( $T < 22.5^\circ\text{C}$ ) and HTemp ( $T > 22.5^\circ\text{C}$ ) during meal time (12:00–14:00, 19:00–21:00 LT), background time (00:00–06:00 LT) and other time. The average temperature of the whole campaign is  $22.5^\circ\text{C}$ . All entries of coefficients are significant at the 1% level (two-tailed), except that of HOA/OT, which is significant at the 5% level.

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**Table 3.** Regression of SV-OOA on HOA, COA and LV-OOA and concentrations of OA factors and Temperature under high and low  $O_x$  ( $HiO_x$  and  $LO_x$ ) of three chosen periods (MT, BT and OT).

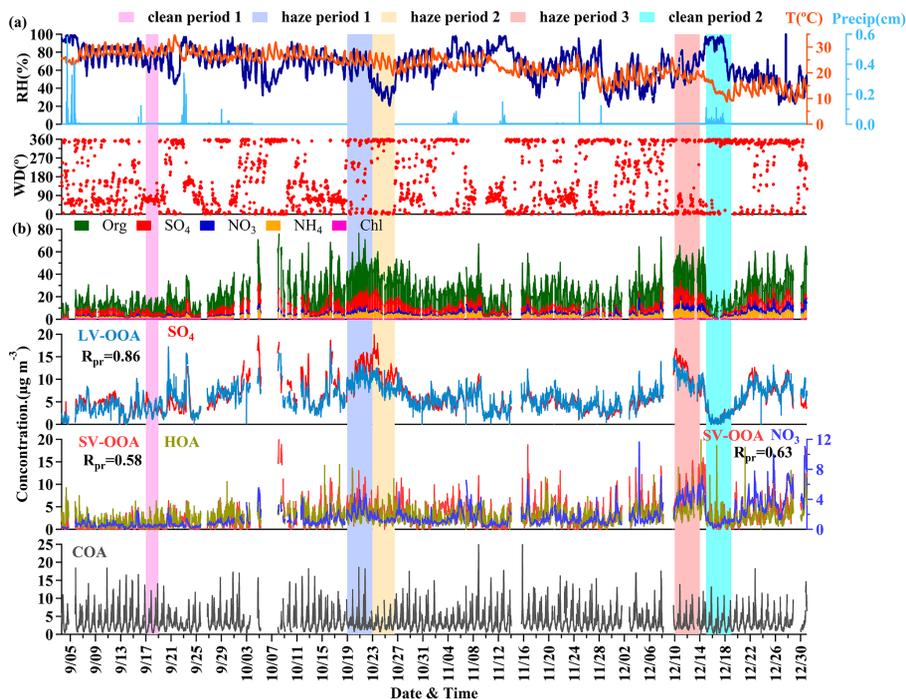
	Meal time (MT)		Background time (BT)		Other time (OT)	
	$LO_x$	$HiO_x$	$LO_x$	$HiO_x$	$LO_x$	$HiO_x$
	Coefficients <sup>a</sup>					
HOA	0.50	1.13	0.62	0.64 <sup>b</sup>	0.08 <sup>b</sup>	0.52
COA	0.13	0.14	0.00	0.15	0.14	0.14
LV-OOA	0.33	0.10 <sup>b</sup>	0.26	0.18	0.34	0.21
Adjusted $R^2$	0.73	0.86	0.73	0.80	0.67	0.78
	Average Concentration ( $\mu\text{g m}^{-3}$ , ppb)					
HOA	2.24	3.41	1.20	2.03	2.11	3.55
COA	7.31	7.57	1.59	1.73	2.77	3.71
LV-OOA	3.50	5.92	5.07	7.22	4.06	6.77
SV-OOA	3.22	5.29	1.85	2.79	1.8	3.56
Temp ( $^{\circ}\text{C}$ )	23.30	23.80	21.48	20.39	22.01	22.74

<sup>a</sup> The coefficient of HOA, COA and LV-OOA in the regression equation reconstructing SV-OOA under  $LO_x$  ( $O_x < 70$  ppb) and  $HiO_x$  ( $O_x > 70$  ppb) during meal time (12:00–14:00, 19:00–21:00 LT), background time (00:00–06:00 LT) and other time. 70 ppb is the average  $O_x$  of the whole study. All entries of coefficients are significant at 1 % level (two-level), except those indicated with <sup>b</sup>, which indicates significance at the 5 % level.



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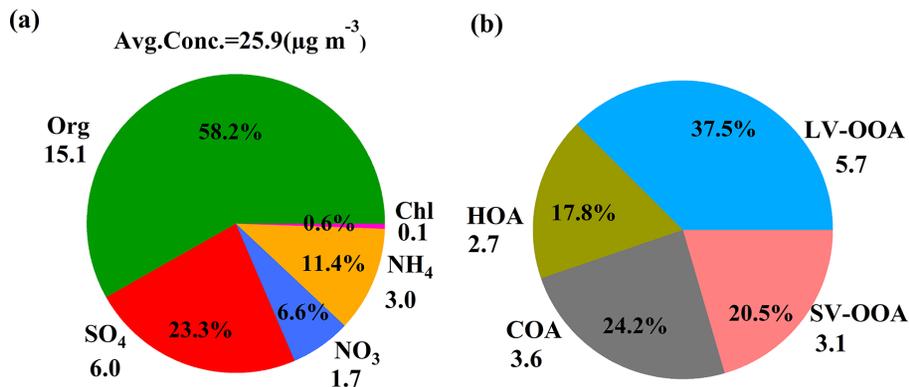
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**Figure 1.** Overview of temporal variation of (a) meteorological factors (Relative Humidity, Temperature and Precipitation) and (b) non-refractory PM<sub>1</sub> species (Org, SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub> and Chl) and organic aerosol components (LV-OOA, SV-OOA, HOA and COA). Five periods: clean period 1 (C1), haze period 1 (H1), haze period 2 (H2), haze period 3 (H3) and clean period 2 (C2) are highlighted.

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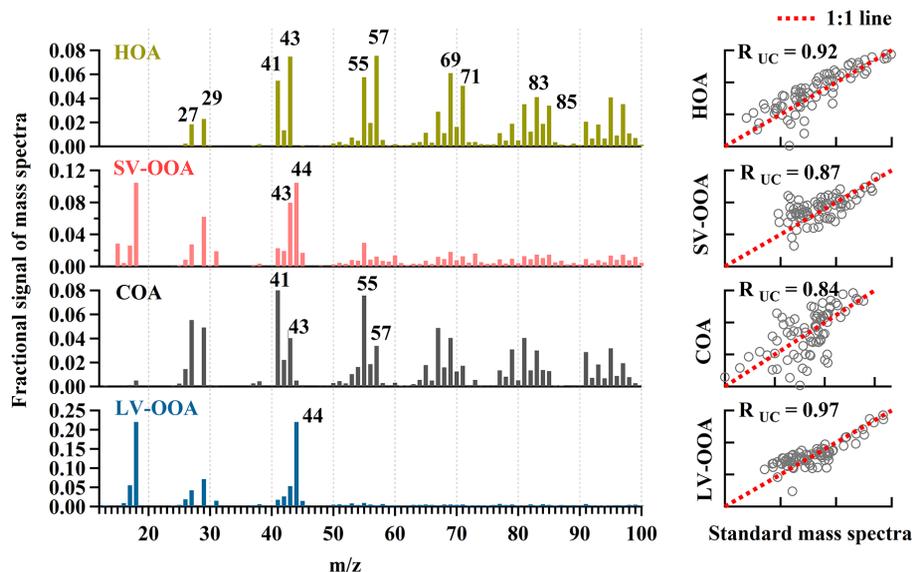


**Figure 2.** Average concentration of each chemical composition of (a) NR-PM<sub>1</sub> (Org, SO<sub>4</sub>, NO<sub>3</sub>, NH<sub>4</sub> and Chl) and (b) organic aerosol (LV-OOA, SV-OOA, HOA and COA).

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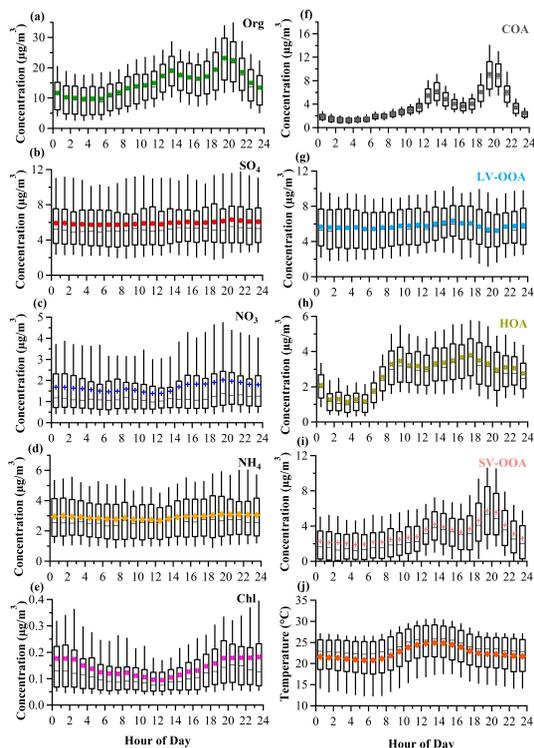
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**Figure 3.** Mass spectra of resolved OA components (HOA, SV-OOA, LV-OOA, COA) and the correlation with standard mass spectral profiles available on the AMS MS database (Ulbrich, I. M., Lechner, M., and Jimenez, J. L., AMS Spectral Database). The x and y axes in the right-hand graphs are mass spectra of resolved factor and the standard, respectively.

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**Figure 4.** Diurnal profiles of NR-PM<sub>1</sub> species, OA components and Temperature for the entire study with 25 and 75 percentile boxes, 10 and 90 percentile whiskers, mean as colored marker and median as black line in the whisker box.

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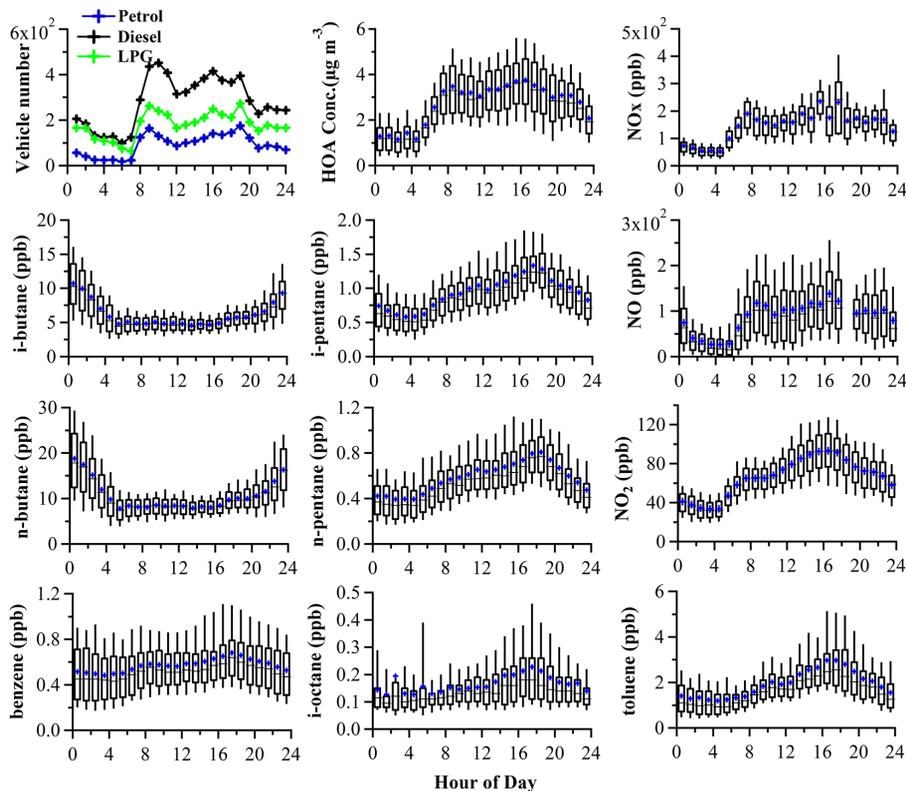
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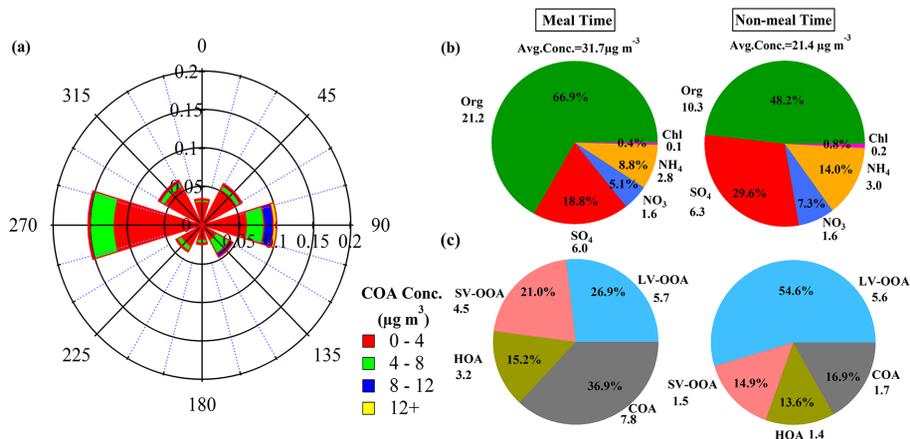


**Figure 5.** Diurnal patterns of vehicle numbers at the Mong Kok site in 28–31 May 2013 and concentrations of HOA,  $\text{NO}_x$ ,  $\text{NO}_2$ , NO, i-pentane, n-pentane, i-octane, i-butane, n-butane, benzene and toluene during the whole study.

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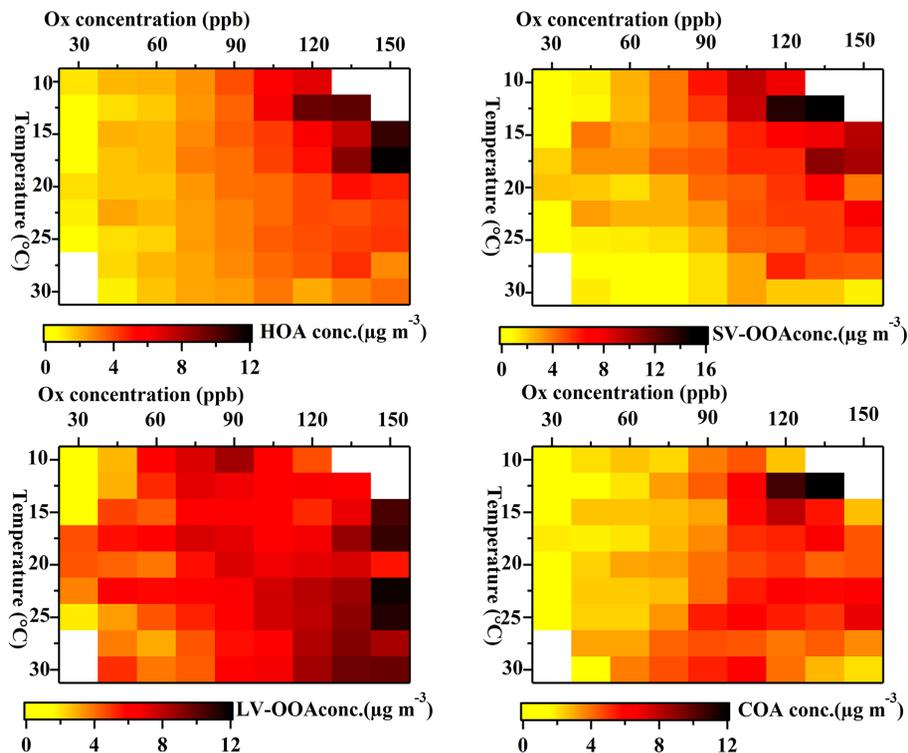
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**Figure 6.** (a) Wind rose plot of COA concentration. The angle and radius present the wind direction and its probability, respectively, while color indicates COA concentration. (b) The fractional composition of NR-PM<sub>1</sub> species during meal time (12:00–02:00, 19:00–21:00 LT) and non-meal time (00:00–06:00 LT). (c) The fractional composition of OA during meal time and non-meal time, respectively.

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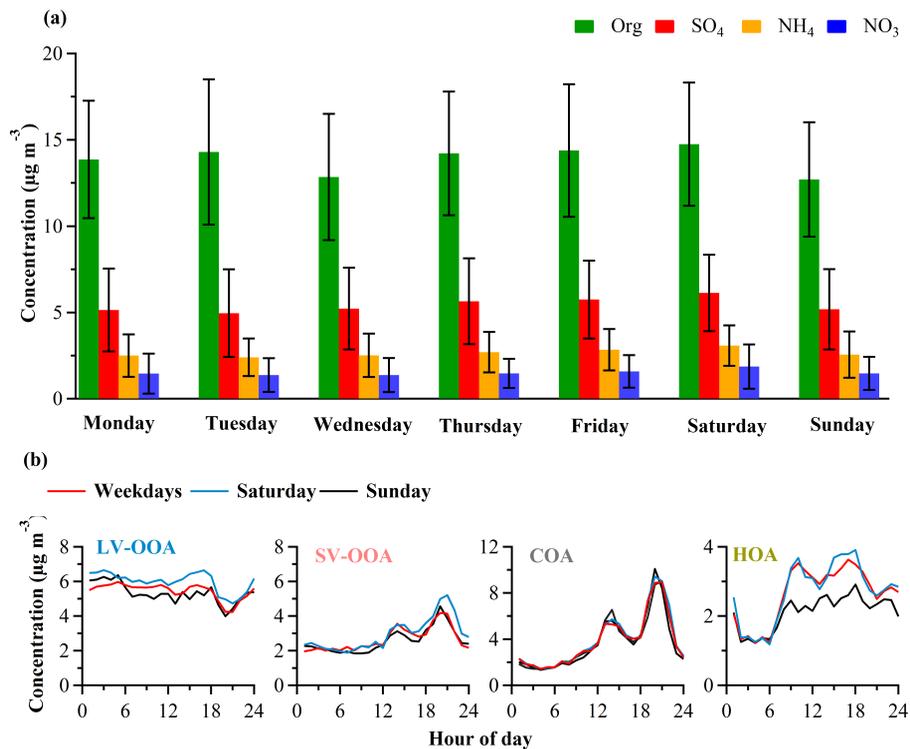


**Figure 7.** Variation of the average concentration of OA components (HOA, SV-OOA, LV-OOA and COA) coded by color as a function of binned  $O_x$  concentration (ppb) and binned temperature ( $^{\circ}C$ ).

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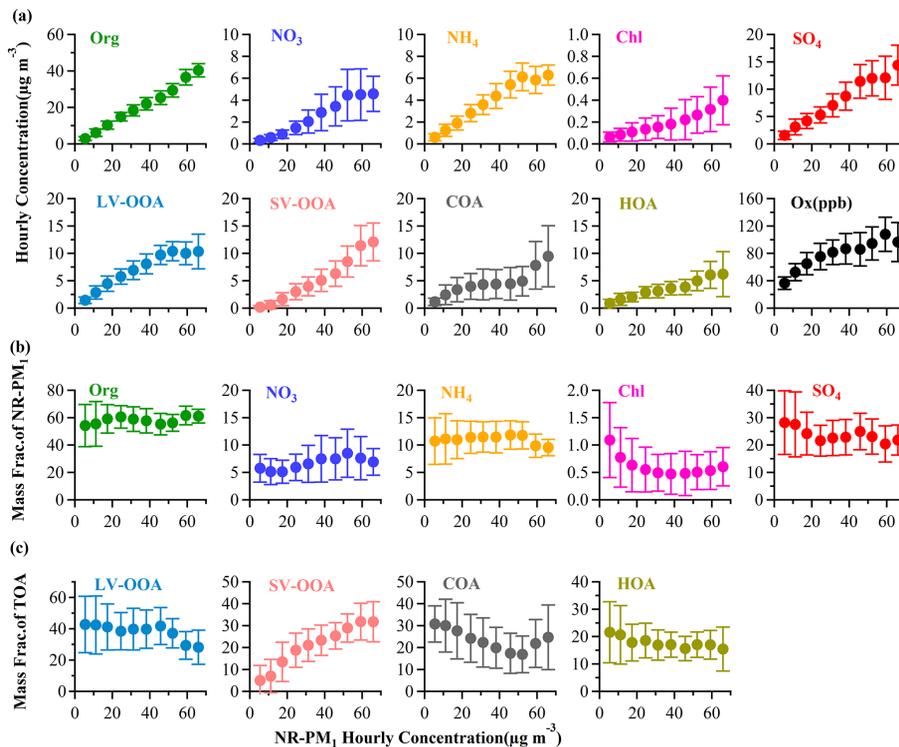
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**Figure 8.** (a) Day-of-week variations of NR-PM<sub>1</sub> species (standard deviation as vertical line) and (b) average diurnal patterns of OA components for weekdays, Saturdays and Sundays.

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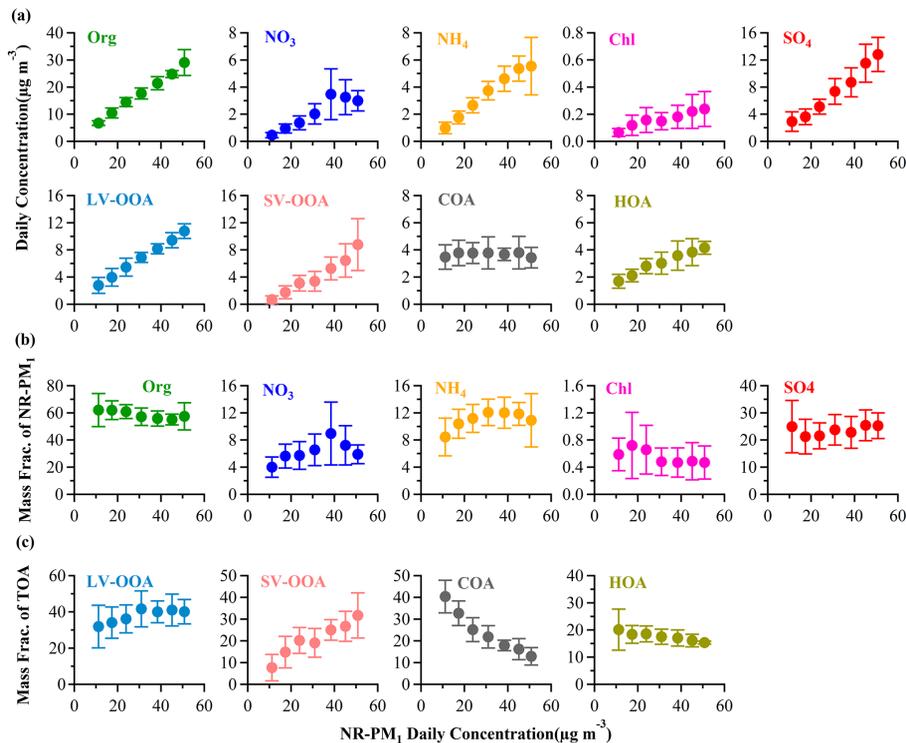


**Figure 9.** (a) Variation in mass concentration of NR-PM<sub>1</sub> species and OA components as a function of total NR-PM<sub>1</sub> mass loading, and (b) mass fraction of total NR-PM<sub>1</sub> for NR-PM<sub>1</sub> species, and (c) mass fraction of total organics for OA components, as a function of total NR-PM<sub>1</sub> mass loading. The data were binned by the hourly average NR-PM<sub>1</sub> mass with a range of 7 μg m<sup>-3</sup>, and the vertical lines are the standard deviation.

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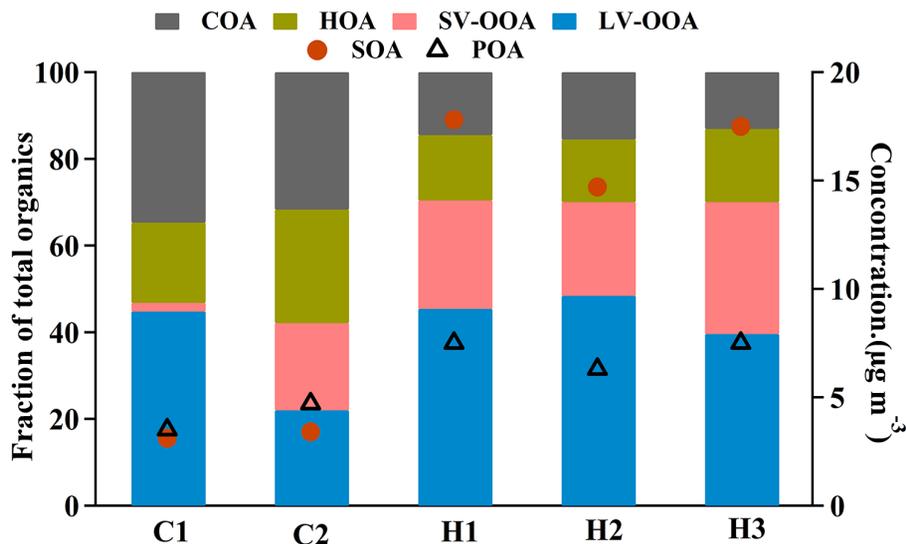
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**Figure 10.** (a) Variation in mass concentration of NR-PM<sub>1</sub> species and OA components as a function of total NR-PM<sub>1</sub> mass loading, and (b) mass fraction of total NR-PM<sub>1</sub> for NR-PM<sub>1</sub> species, and (c) mass fraction of total organics for OA components, as a function of total NR-PM<sub>1</sub> mass loading. The data were binned by the *daily average* NR-PM<sub>1</sub> mass with a range of 7 μg m<sup>-3</sup>, and the vertical lines are the standard deviation.

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**Figure 11.** Mass fraction of hydrocarbon-like organic aerosol (HOA), cooking organic aerosol (COA), semi-volatile oxygenated organic aerosol (SV-OOA) and low-volatility oxygenated organic aerosol (LV-OOA) in color, and the mass concentration of POA and SOA marked by triangles and circles, respectively, during five periods: clean periods (C1 and C2), and haze periods (H1, H2 and H3).

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