



Characterization of
submicron aerosol
chemical
composition

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Seasonal characterization of submicron aerosol chemical composition and organic aerosol sources in the southeastern United States: Atlanta, Georgia and Look Rock, Tennessee

S. H. Budisulistiorini¹, K. Baumann², E. S. Edgerton², S. T. Bairai³, S. Mueller⁴, S. L. Shaw⁵, E. M. Knipping⁶, A. Gold¹, and J. D. Surratt¹

¹Department of Environmental Sciences and Engineering, Gillings School of Global Public Health, The University of North Carolina at Chapel Hill, Chapel Hill, NC, USA

²Atmospheric Research and Analysis, Inc., Cary, NC, USA

³Battelle, Pueblo, CO, USA

⁴Ensafe, Nashville, TN, USA

⁵Electric Power Research Institute, Palo Alto, CA, USA

⁶Electric Power Research Institute, Washington, DC, USA

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Correspondence to: J. D. Surratt (surratt@unc.edu)

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A yearlong near-real-time characterization of non-refractory submicron aerosol (NR-PM₁) was conducted at an urban (Atlanta, Georgia) and rural (Look Rock, Tennessee) site in the southeastern US using the Aerodyne aerosol chemical speciation monitor (ACSM) collocated with established air-monitoring network measurements. Seasonal variations in organic aerosol (OA) and inorganic aerosol species are attributed to meteorological conditions as well as anthropogenic and biogenic emissions in this region. The highest concentrations of NR-PM₁ were observed during winter and fall seasons at the urban site and during spring and summer at the rural site. Across all seasons and at both sites, NR-PM₁ was composed largely of OA (50–76 %) and inorganic sulfate (12–31 %). Six distinct OA sources were resolved by positive matrix factorization applied to the ACSM organic mass spectral data collected from the two sites over the one year of near-continuous measurements at each site: hydrocarbon-like OA (HOA), biomass burning OA (BBOA), semi-volatile oxygenated OA (SV-OOA), low-volatility oxygenated OA (OOA), isoprene-derived epoxydiol (IEPOX) OA (IEPOX-OA), and 91Fac OA (a factor dominated by a distinct ion at m/z 91 fragment ion previously observed in biogenic influenced areas). LV-OOA was observed throughout the year at both sites and contributed 30–66 % of total OA mass. HOA was also observed during the entire year only at the urban site (15–24 % of OA mass). BBOA (15–33 % of OA mass) was observed during winter and fall, likely dominated by local residential wood burning emission. Although SV-OOA contributes quite significantly (~ 27 %), it was observed only at the urban site during colder seasons. IEPOX-OA was a major component (27–41 %) of OA at both sites, particularly in spring and summer. An ion fragment at m/z 75 is proposed as an additional marker for IEPOX-OA, as it is shown to correlate well with the m/z 82 ion shown to be associated with the aerosol mass spectrum of IEPOX-derived secondary organic aerosol (SOA). The contribution of 91Fac to the total OA mass was quite low at the urban site (~ 9 %), but was significant (21–23 %) at the rural site during warmer months. Comparison of 91Fac OA time series with SOA tracers measured from

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filter samples collected at Look Rock suggests that isoprene oxidation through a pathway other than IEPOX SOA chemistry may contribute to its formation. Other biogenic sources could also contribute to 91Fac, but there remains a need to resolve the exact source of this factor based on its significant contribution to rural OA mass.

1 Introduction

Characterization of the chemical composition of atmospheric fine aerosol is important, because of its adverse human health effects (Pope III and Dockery, 2006) and possible impacts on the Earth's climate system (Forster et al., 2007). Aerosol with aerodynamic diameters $\leq 1 \mu\text{m}$ (PM_{10}) play a significant role in scattering and/or absorbing solar radiation as well as cloud formation (IPCC, 2013). Long-term regional characterizations of ambient PM_{10} is required to understand their sources, formation, and aging mechanisms, as well as their atmospheric lifetimes. This information will lead to more accurately constrained air quality models for making regulatory decisions to mitigate the potential adverse impacts of PM_{10} .

Over the past decade, online aerosol mass spectrometry (AMS) has been used to extensively characterize ambient non-refractory (NR)- PM_{10} (Zhang et al., 2007; Jimenez et al., 2009; Ng et al., 2010; Crippa et al., 2014); however, prior studies were limited by short measurement periods (weeks to a several months) because the need for intensive instrument maintenance required the continuous on-site presence of skilled personnel in order to generate high quality data. The Aerodyne aerosol chemical speciation monitor (ACSM) based on the AMS technology has been modified to allow for long-term operation with less maintenance (Ng et al., 2011b). The ACSM has been recently used for long-term NR- PM_{10} measurements (Petit et al., 2015; Ripoll et al., 2015; Parworth et al., 2015; Zhang et al., 2015) and shown to be durable and comparable to data collected from existing fine aerosol monitoring networks (Budisulistiorini et al., 2014).

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role of inorganics in NR-PM₁, in accord with observation in other major urban areas (Y. Sun et al., 2011; Petit et al., 2015).

The lowest seasonal average pH were observed in summer (1.45) for JST and in fall (1.53) for LRK, as illustrated in Fig. 1. On the other hand, the highest seasonal average pH were 2.01 for JST and 1.81 for LRK which were observed during winter. Overall, seasonal aerosol pH is 1.5–2.0 at both sites indicates that NR-PM₁ in the southeastern US is acidic year round, which is consistent with a recent study by Guo et al. (2015). Figure 1 shows that no direct correlation exists between aerosol pH and OA. However, this does not necessarily rule out the potential role of aerosol acidity in enhancing SOA formation in light of laboratory studies demonstrating a significant pH effect (Gao et al., 2004; Surratt et al., 2007; Y. Lin et al., 2013). Uncertainty of aerosol acidity estimation by ISORROPIA-II by omission of organic sulfate as input (Lin et al., 2014) could lead to under-prediction of aerosol acidity and the observed lack of correlation with OA. Seasonal averages of LWC were highest during summer at both JST (33.97 molL⁻¹ of aerosol) and LRK (38.17 molL⁻¹) sites. It should be noted that the possible LWC contributions from OA are not included because a technique, such as that recently used by Guo et al. (2015) to measure this quantity is lacking at our sites. Studies have suggested that reactive uptake decreases with enhanced RH (Nguyen et al., 2014; Gaston et al., 2014); however, some isoprene-derived SOA tracers were elevated by high RH (Zhang et al., 2011). Although organic water fraction in total LWC was found to be significant by Guo et al. (2015); pH prediction using ISORROPIA-II based on inorganic ions alone was found to give a reasonable estimate. Hence, the lack of correlation in this study between OA and LWC based only on inorganic water suggests that LWC is not a limiting factor in OA production in this region.

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60 and 73 ions (Bougiatioti et al., 2014). However, the BBOA factor observed at JST and LRK displayed enhanced signal at m/z 44 ion but retained signals at m/z 60 and 73 ions, suggesting that it was not as oxidized as the aged BBOA factor.

LV-OOA is characterized by high fraction of total ion intensity at m/z 44 (f_{44}) resulting from high oxygen content (Ng et al., 2011a) and is the most abundant OA type at both JST and LRK (Table 1). Maxima around midnight at JST and in the mid-afternoon at LRK (Fig. 4b) were not significant, indicating that LV-OOA concentration is relatively constant throughout the day in this region. LV-OOA has been shown to correlate with non-volatile secondary species (Jimenez et al., 2009). Lack of temporal correlation between LV-OOA and sulfate has been observed to depend on oxidation degree of urban ambient aerosol (Y. L. Sun et al., 2011). We observed weak correlations ($r^2 < 0.2$) between LV-OOA and sulfate at both sites. Ratio of organic-to-sulfate ratio was used to track photochemical aging of biogenic SOA (Henry and Donahue, 2012). Large variability of LV-OOA : sulfate ratio (Fig. S10) at LRK and JST could indicate gradual photochemical aging process in winter. This might cause lack of correlation with sulfate, as previously seen in New York City (Y. L. Sun et al., 2011). On the other hand, the mass spectral comparison of LV-OOA from both sites were strongly correlated ($r^2 \sim 1$; Fig. S11), possibly suggesting similar sources of LV-OOA at these sites.

SV-OOA, which was observed only in urban OA, is the most abundant OA. An f_{44} smaller than that of LV-OOA indicates the factor is less oxidized and thus semi-volatile (Ng et al., 2011a). The temporal variation of SV-OOA was moderately correlated ($r^2 \sim 0.4$) with nitrate (Fig. 3; Table S1) while the mass spectrum was well correlated with previously resolved 82Fac and IEPOX-OA factors (82Fac and IEPOX-OA are equivalent and are characterized by a distinct ion at m/z 82) (Robinson et al., 2011; Budisulistiorini et al., 2013, 2015). However, low isoprene emissions during winter do not support the association of the SV-OOA factor is associated with IEPOX-derived SOA. The diurnal profile of SV-OOA showed an increase in the evening and decrease in the morning, similar to the BBOA profile and HOA factors. This variability suggests an influence of nitrate-radical chemistry on nighttime SOA formation (Xu et al., 2015).

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($r^2 \sim 0.4$) at JST, whereas at LRK, stronger correlations ($r^2 > 0.5$) with inorganics (i.e., sulfate, nitrate and ammonium) were observed.

3.2.4 Fall

At JST, PMF analysis of fall OA resulted in similar a four-factor solution similar to that of winter OA (i.e., HOA, BBOA, SV-OOA, and LV-OOA), while at LRK a three-factor solution was resolved (i.e., LV-OOA, 91Fac, and IEPOX-OA) (Fig. 2d). The concentration of HOA increased to a level comparable to that in winter (Fig. 3). The correlations of the time series of HOA with BC, CO, and NO_x ($r^2 > 0.7$) were similar to spring and summer and slightly stronger than in winter (Table S1). The diurnal profile of HOA appears similar to that in winter (Fig. 4).

At JST, the BBOA factor was resolved again from OA in fall with average concentration and fractional contribution to total OA less than from winter. BBOA was not resolved from OA at LRK. The diurnal profile of BBOA during fall at JST appeared similar to that in winter, suggesting similar emission sources as well as possible PBL effect during these two colder seasons. SV-OOA was also resolved from JST OA in fall with slightly higher average concentration and fractional contribution than in winter. The diurnal profile of fall SV-OOA was similar to that in winter, suggesting similar sources and role of PBL.

LV-OOA was resolved from OA at both JST and LRK. Average concentrations of LV-OOA remained relatively constant from summer to fall at both the urban and rural sites (Fig. 3). However, the contribution of LV-OOA to total OA at LRK increased due to decreasing concentrations of other OA factors (i.e., IEPOX-OA and 91Fac) (Table 1). JST LV-OOA did not show diurnal variation, whereas LRK LV-OOA increased in mid-morning and reached a maximum around mid-afternoon. Temporal variation of LV-OOA was weakly correlated ($r^2 \sim 0.2$) with inorganics at JST, but moderately correlated ($r^2 = 0.4\text{--}0.5$) at LRK. Strong correlation of LV-OOA mass spectra ($r^2 \sim 1$, slope = 0.8–1.1; Fig. S1) at JST and LRK indicates a similar or identical source.

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The concentration and fractional contribution of 91Fac resolved from OA at LRK were much lower in fall than summer (Fig. 3, Table 1). No significant diurnal variation or correlations with inorganics was observed. IEPOX-OA concentration decreased in fall as expected from reduced isoprene emissions. Time series of IEPOX-OA were not correlated with any secondary species. The diurnal profile of IEPOX-OA showed a small peak around noon, suggesting that SOA formation was still occurring.

3.3 Seasonal changes and contribution of OA sources

Figure 3a and b illustrates annual temporal variations of PMF factors resolved from the JST and LRK. HOA was observed throughout the year at JST and contributes significantly to total OA, on average 20–24 % (Table 1). Seasonal average mass concentration of HOA varies from 0.5–2 $\mu\text{g m}^{-3}$ (Fig. 3c). Wider standard deviations in winter and fall suggest more variability in HOA mass in Atlanta during these seasons. LV-OOA, which was also observed throughout the year, contributes on average 30–43 % of the total OA. Seasonal average concentration of LV-OOA is relatively constant between 1–3 $\mu\text{g m}^{-3}$ with less mass variability in summer and more variability in winter. PBL might have a role in the higher variability of HOA and LV-OOA at JST during winter. At LRK, LV-OOA was also observed throughout the year, accounting for a large proportion of total OA in winter, up to 66 % (Table 1). Similar to JST, seasonal average concentration at LRK was 1–3 $\mu\text{g m}^{-3}$, with slightly more mass variability in winter and fall than in spring and summer.

BBOA and SV-OOA were both observed during winter and fall at JST. BBOA seasonal average concentration is about 1–2 $\mu\text{g m}^{-3}$, accounting on average 15–19 % of total OA. Standard deviations of mass concentrations indicate large variability of BBOA in winter, which narrows in fall (Fig. 3c). BBOA was observed only during winter at LRK. The average concentration of BBOA at LRK was consistently $\sim 1 \mu\text{g m}^{-3}$ (Fig. 3b), but several episodes of high levels resulted in a large standard deviation (Fig. 3b and c). SV-OOA was resolved only at JST and only in the fall and winter (Fig. 3a), implying that it might be formed from local aging process or transported from nearby areas. Seasonal

monoterpene oxidation. This might indicate that 91Fac at LRK could be influenced by isoprene chemistry.

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Table 1. Seasonal averaged mass concentrations of non-refractory PM₁ (NR-PM₁) in $\mu\text{g m}^{-3}$, percent contributions of organic and inorganic species measured by the Aerodyne ACSM and PMF factors resolved from Atlanta, GA (JST site) and Look Rock, TN (LRK site) during 2012 and 2013.

	Winter		Spring		Summer		Fall	
	JST	LRK	JST	LRK	JST	LRK	JST	LRK
NR-PM ₁	10.50 ± 7.32	4.77 ± 3.32	6.19 ± 2.85	5.59 ± 3.47	8.78 ± 4.46	8.39 ± 4.44	12.47 ± 6.72	4.55 ± 2.55
OA	69.0 %	50.2 %	75.9 %	57.8 %	70.0 %	63.4 %	65.9 %	62.1 %
SO ₄ ²⁻	13.4 %	30.6 %	12.0 %	26.9 %	17.4 %	24.5 %	15.8 %	21.6 %
NO ₃ ⁻	9.3 %	9.2 %	5.6 %	6.1 %	4.5 %	3.8 %	9.3 %	7.2 %
NH ₄ ⁺	7.9 %	9.9 %	6.3 %	9.0 %	7.9 %	8.2 %	8.6 %	9.1 %
Cl ⁻	0.3 %	0.1 %	0.2 %	0.1 %	0.1 %	0.1 %	0.3 %	0.0 %
OA speciation								
HOA	24 %	NA	17 %	NA	15 %	NA	20 %	NA
BBOA	19 %	33 %	NA	NA	NA	NA	15 %	NA
SV-OOA	26 %	NA	NA	NA	NA	NA	28 %	NA
LVOOA	30 %	66 %	43 %	37 %	39 %	36 %	37 %	63 %
91Fac	NA	NA	9 %	21 %	9 %	23 %	NA	9 %
IEPOX-OA	NA	NA	32 %	41 %	37 %	40 %	NA	27 %

NA is not available or resolved from PMF analysis. PMF analysis yielded some residuals of unresolved OA mass that make up the remaining percentage of OA factors.

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Table 2. Correlations of PMF factors resolved from OA measurements at LRK, TN against SOA tracers from monoterpene chemistry and isoprene ozonolysis quantified during 2013 SOAS.

	IEPOX-OA	LV-OOA	91Fac	Ref.
Monoterpene SOA tracers				
C ₁₀ H ₁₈ O ₅ S	0.28	0.26	0.39	(1)
C ₁₀ H ₁₆ O ₇ S ^a	0.42	0.26	0.37	(1)
C ₁₀ H ₁₇ NO ₇ S	0.00	0.00	0.01	(1)
C ₉ H ₁₅ NO ₈ S ^a	0.12	0.22	0.22	(1)
C ₁₀ H ₁₇ NO ₁₀ S	0.11	0.15	0.26	(1)
C ₈ H ₁₂ O ₄ (Terpenylic acid) ^a	0.32	0.36	0.41	(2)
C ₉ H ₁₄ O ₄ (Pinic acid)	0.12	0.21	0.19	
C ₁₀ H ₁₆ O ₄ (Hydroxy pinonic acid)	0.15	0.21	0.25	
C ₁₀ H ₁₆ O ₃ (Pinonic acid)	0.10	0.17	0.20	
C ₇ H ₁₀ O ₄ (Terebic acid)	0.21	0.32	0.27	(3)
C ₈ H ₁₂ O ₆ (MBTCA)	0.15	0.27	0.14	(4)
C ₁₀ H ₁₆ O ₆ (DTAA)	0.35	0.42	0.42	(2)
Isoprene ozonolysis tracers ^b				
C ₄ H ₈ O ₆ S	0.46	0.40	0.51	(5,6)
C ₅ H ₁₂ O ₆ S	0.39	0.19	0.35	(5,6)
C ₅ H ₁₀ O ₅ S	0.19	0.19	0.22	(6)
C ₅ H ₁₀ O ₆ S	0.33	0.38	0.41	(6)
C ₈ H ₁₀ O ₄ S	0.00	0.07	0.03	(6)
C ₆ H ₁₂ O ₇ S	0.24	0.33	0.48	(6)
C ₉ H ₁₄ O ₆ S	0.21	0.30	0.38	(6)
C ₉ H ₁₆ O ₇ S	0.38	0.50	0.46	(6)
C ₁₀ H ₂₀ O ₉ S	0.36	0.29	0.39	(6)

^a Budisulistiorini et al. (2015).

^b Only night time samples were used in PMF factor correlation with isoprene ozonolysis tracers. References: (1) Surratt et al. (2008); (2) Claeys et al. (2009); (3) Yasmeen et al. (2010); (4) Szmigielski et al. (2007); (5) Safi Shalamzari et al. (2013); (6) Riva et al. (2015).

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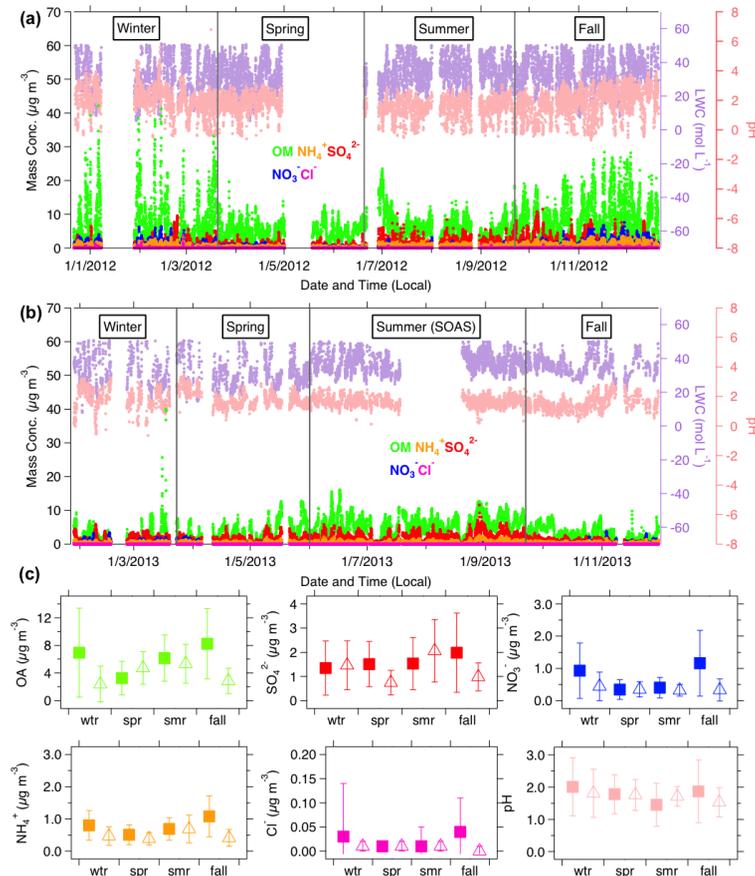


Figure 1. Annual temporal variations of OA and inorganic species measured at the **(a)** JST site in 2012; and **(b)** LRK site in 2013. Included in the plots are pH and liquid water content (LWC) estimated by ISORROPIA-II. **(c)** Seasonal average of OA, inorganic species and pH from JST (solid squares) and LRK (open triangles). Error bars show ± 1 -standard deviation.

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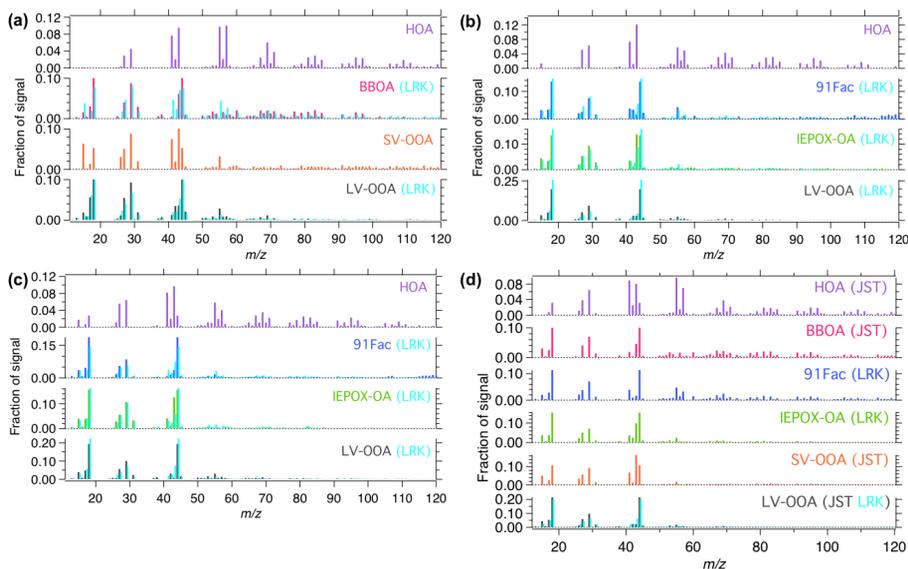


Figure 2. Mass spectra of PMF factors resolved from (a) winter, (b) spring, (c) summer, and (d) fall datasets at JST and LRK sites. The PMF factors from JST site are color-coded, while the same factor from LRK site is in light blue.

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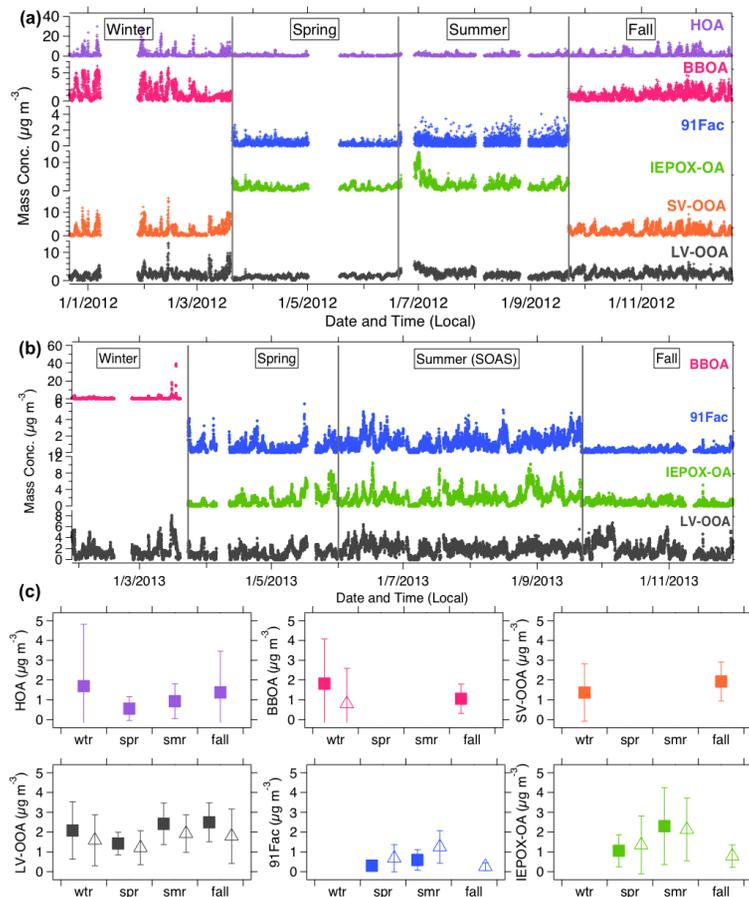


Figure 3. Annual temporal variation of PMF factors resolved from OA measurements at the **(a)** JST site, and **(b)** LRK site. **(c)** Seasonal average mass concentration of PMF factors resolved from JST (solid squares) and LRK (open triangles) with error bars shown as ± 1 -standard deviation. Seasons are classified into winter (wtr), spring (spr), summer (smr), and fall.

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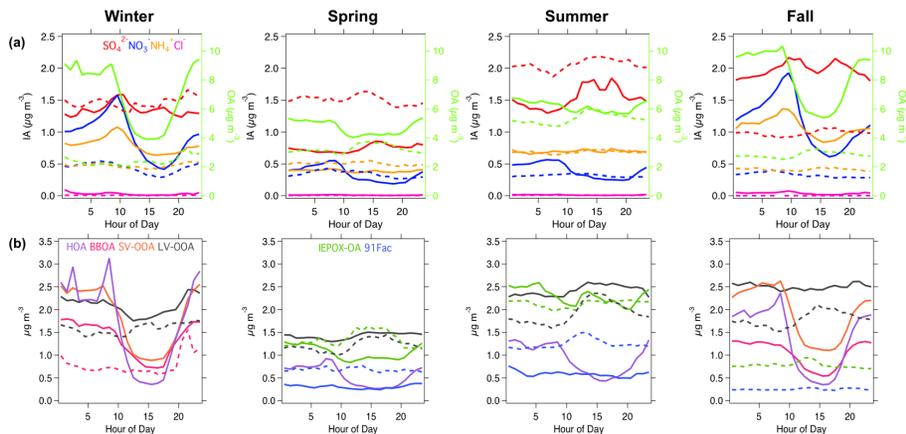


Figure 4. Diurnal variations of (a) OA and inorganic species measured by ACSM and (b) OA factors resolved by PMF analysis from winter, spring, summer, and fall measurements at the JST site (solid lines) and LRK site (dash lines).

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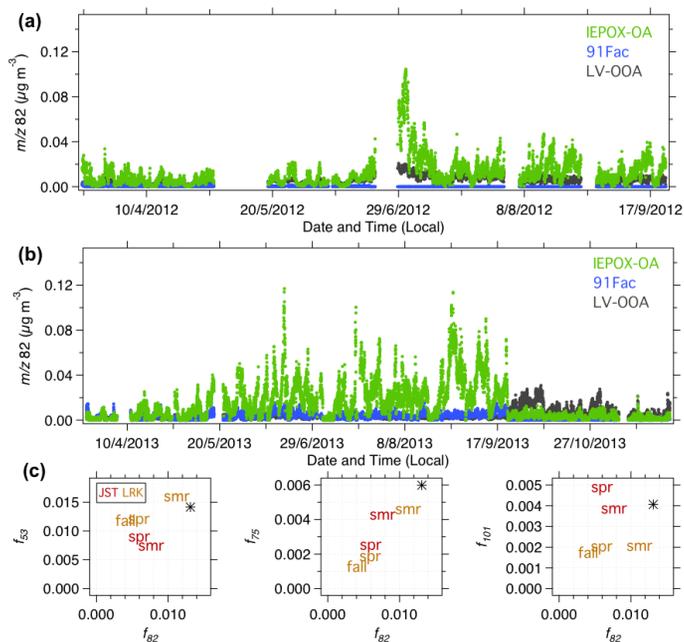


Figure 5. Temporal variations of the m/z 82 fragment ion ($\text{C}_5\text{H}_6\text{O}^+$) contribution to IEPOX-OA, 91Fac, and LV-OOA factors **(a)** at the JST site during spring and summer seasons, and **(b)** at the LRK site during spring, summer, and fall seasons. **(c)** Scatterplots of the m/z 53 (possibly C_4H_5^+), m/z 75 (possibly $\text{C}_3\text{H}_7\text{O}_2^+$), and m/z 101 (possibly $\text{C}_5\text{H}_9\text{O}_2^+$) fragment ions from the IEPOX-OA mass spectra vs. the m/z 82 fragment ion from the same mass spectra over different seasons at the JST and LRK sites. ACSM measures unit mass resolution (UMR), thus the proposed formulas are based on previous study using HR-ToF-AMS (Lin et al., 2012). The asterisk marker is the respective ion fragments of IEPOX-OA mass spectra resolved from OA measurements during summer 2011 at the JST site (Budisulistiorini et al., 2013).

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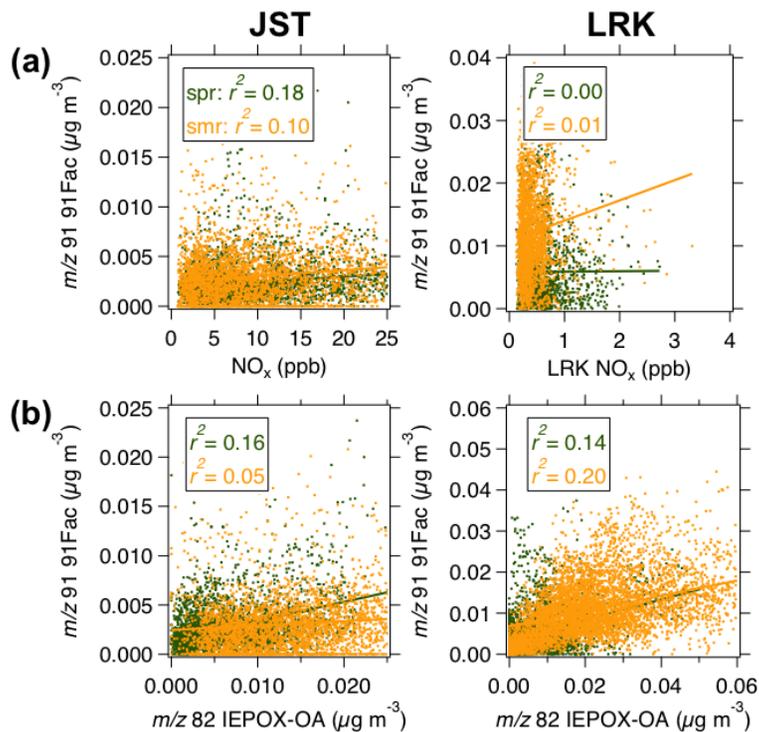


Figure 6. Scatter plots of m/z 91 of 91Fac factor vs. **(a)** NO_x and **(b)** m/z 82 of IEPOX-OA factor at JST and LRK sites during spring and summer seasons.

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