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LETTER

Large contribution of fine carbonaceous aerosols from municipal waste burning inferred from distributions of diacids and fatty acids

X Li1,2, C M Pavuluri1,2, Z Yang1,2, N He3, E Tachibana4, K Kawamura3,6 and P Q Fu1,4,7

1 Institute of Surface-Earth System Science, Tianjin University, Tianjin 300072, People’s Republic of China
2 Institute of Geochemistry, Chinese Academy of Sciences, Guiyang 550081, People’s Republic of China
3 Department of Tourism and Geography, Tongren University, Tongren 554300, People’s Republic of China
4 LAPC, Institute of Atmospheric Physics, Chinese Academy of Sciences, Beijing 100029, People’s Republic of China
5 Institute of Low Temperature Science, Hokkaido University, Sapporo 060-0819, Japan
6 Chubu Institute for Advanced Studies, Chubu University, Kasugai 487-8501, Japan
7 Authors to whom any correspondence should be addressed.

E-mail: cmpavuluri@tju.edu.cn and fupingqing@tju.edu.cn

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Abstract

To investigate the origins, other than fossil fuel combustion and biomass burning, of urban carbonaceous aerosols, we studied the fine (PM2.5) aerosols collected in Guiyang, Southwest China in winter (December 10–23) 2012 for organic carbon (OC), elemental carbon (EC) and water-soluble diacids, oxoacids, α-dicarboxyls and fatty acids as well as inorganic ions. Oxalic acid (C2) found to be the most abundant diacid species followed by succinic (C4) and terephthalic (t-Ph) acids, respectively. Even-carbon numbered fatty acids showed high abundances with a peak at C16. OC, EC and most of diacids and related compounds, but not inorganic ions, showed a similar temporal pattern with a drastic rise in December 15 day- and night-time samples during the campaign. Based on molecular distributions of diacids and fatty acids, and linear relations of OC and EC with selected marker ions and diacid species, we found that the organics and EC in PM2.5 are mainly derived from higher plant cooking and municipal waste burning in Guiyang. This study implies that municipal waste burning largely contributes to carbonaceous aerosols and warrants a need of further research on its role in aerosol loading and air quality in the urban atmosphere.

1. Introduction

Carbonaceous aerosols that are composed of EC and many organic compounds, collectively considered as OC (Contini et al 2018), account for a major fraction of PM2.5 mass (Sillanpaa et al 2005, Cao et al 2007) and received great attention in last two decades because of their serious impacts on the Earth’s climate system. They also have adverse effects on ecology, visibility and human health (Kanakidou et al 2005, Pandis et al 2013) and play an important role in atmospheric chemistry (Kolb and Worsnop 2012). Diacids and related polar compounds account for a substantial fraction of water-soluble organic aerosols (OA) (Kawamura and Ikushima 1993, Miyazaki et al 2009) and found to be ubiquitous in in the atmosphere (Kawamura and Bikkina 2016). Diacids and related compounds can be emitted directly into the atmosphere from fossil fuel combustion (Kawamura and Kaplan 1987), biomass burning (Graham et al 2002, Kundu et al 2010, Kawamura et al 2013, Sorathia et al 2018) and food cooking (Schauer et al 2002). They can also be produced in the atmosphere by oxidation of a large number of organic compounds emitted from both anthropogenic and natural sources (Kawamura and Bikkina 2016). Diacids and related compounds are further subjected to photochemical processing (aging) in the atmosphere (Kawamura et al 2005, Fu et al 2013).
Oxidation of organics in atmospheric waters are considered to be an important process that results in high abundance of oxalic (C$_2$) acid (Pavuluri et al. 2015b). Such processing of aerosols is significant under strong solar radiation and high humid environment and thus enhances the loading of short-chain diacids and the fraction of total diacids and related compounds to OA (Yang et al. 2008, Pavuluri et al. 2015b). Generally, in terrestrial or marine aerosols, C$_2$ acid was often found as the most abundant species of diacids, followed by malonic (C$_3$) or C$_4$ acids, whereas in polar aerosols, C$_4$ acid found to be higher occasionally than that of C$_2$, particularly in the samples collected under overcast conditions with brume events (Kawamura and Ikushima 1993, Kawamura et al. 2005). Moreover, some diacid species such as adipic (C$_6$), azelaic (C$_9$) and t-Ph acids are mainly derived from specific compounds originated from fossil fuel combustion, biogenic, and plastic (municipal waste) burning respectively (Kawamura and Gagosian 1987, Kawamura et al. 1996, Kawamura and Pavuluri 2010). Therefore, distributions of diacids including diurnal changes together with mass ratios of selected species would provide insights on their origins and secondary formation and transformation processes (Pavuluri et al. 2015b, Sorathia et al. 2018).

High aerosol loadings often with haze events are commonly observed in Asia, due to extensive urbanization and industrialization, particularly in China and India (Zhao et al. 2013, Zhang et al. 2014). The Asian aerosols have been studied extensively via ground-based measurements at various locale and large-scale field experiments by international research programs such as ACE-Asia, EAREX 2005, etc, as well (Fuzzi et al. 2006, Nakajima et al. 2007). However, the previous studies implied that fossil fuel combustion, biofuel and biomass burning and Asian dust are the major sources of Asian aerosols. Recently, biogenic and cooking emissions and their subsequent secondary processes have also been identified as major sources of OA (Torkmahalleh et al. 2012, Pavuluri et al. 2018, Liu et al. 2018b). Municipal waste burning is also considered as a potential source of OA (Kawamura and Pavuluri 2010). However, importance of their contribution to carbonaceous aerosols has not been fully understood yet. Hence, it is highly necessary to conduct field experiments for characterization of OA at various locations, where the biogenic, cooking and/or municipal waste burning emissions are expected to be significant, to better understand their loading, composition and sources.

We studied wintertime PM$_{2.5}$ collected in Guiyang, Southwest China, where forest vegetation is rich and most of the municipal waste is dumped into open landfills, for OC, EC and water-soluble diacids, oxoacids, α-dicarboxyls and fatty acids and inorganic ions. Based on molecular distributions of diacids and fatty acids, similarity in their temporal patterns and linear relations of OC and EC with selected markers, we discuss the possible contributions of biogenic/cooking and municipal waste burning emissions to carbonaceous aerosols. We also discuss the secondary formation/transformations of OA in the Guiyang atmosphere.

2. Experimental

2.1. Aerosol sampling

PM$_{2.5}$ sampling was performed at Guiyang (26°35’S, 106°43’E) in Yunnan–Guizhou Plateau, Southwest China (see figure S1 in supplementary material is available online at stacks.iop.org/ERC/1/071005/mmedia). A subtropical monsoon climate prevails over the region with an annual average temperature of 15.3 °C, rainfall of 1,174 mm and relative humidity (RH) of 77% in Guiyang (Xiao et al. 2012). The forest vegetation covers more than 46.5% of the Guizhou area with high mountains and two greenbelts spreading more than 100 km are existed around Guiyang. The PM$_{2.5}$ samples were collected on the rooftop (10 m AGL) of Institute of Geochemistry, Chinese Academy of Sciences building using a low-volume aerosol sampler (ME-APS100, China) and pre-baked (450 °C for 6 h) quartz fiber filters (047 mm) on day- (7:00 am ~ 7:00 pm, local time; n = 13) and night-time (∼7:00 pm ~ 7:00 am; n = 12) during December 10~23, 2012. The filter samples were wrapped in aluminum foil and sealed in zip-lock plastic bag and stored at −18 °C until analysis.

2.2. Chemical analyses

OC and EC were measured using a Sunset Lab EC/OC Analyzer following the Interagency Monitoring of Protected Visual Environments (IMPROVE) thermal evolution protocol. Typically, a 1.54 cm$^2$ punch (Ø14 mm) of the filter was placed in a quartz boat inside the thermal desorption chamber of the analyzer, and then stepwise heating was applied (Wang and Kawamura 2005). Duplicate analyses of filter samples for OC and EC showed uncertainties less than 10%.

A quarter of sample filter extracted with Milli-Q water (EMD Millipore, Merck, Germany) under ultrasonication for 30 min, and the extracts were filtered with 0.22 μm syringe filter (Pall, USA) for the measurement of Na$^+$, K$^+$, Mg$^{2+}$ and Ca$^{2+}$ using ICP-OES (MPX; Varian, Inc., USA), Cl$^-$, NO$_3^-$ and SO$_4^{2-}$ using IC (ICS-90; Dionex, USA) and NH$_4^+$ using a Skalar continuous flow analyzer (San+; Skalar, Holland).

Water-soluble diacids, oxoacids, α-dicarboxyls and fatty acids were measured in PM$_{2.5}$ following a method developed by Kawamura and Ikushima (1993). Briefly, an aliquot of filter sample was extracted with organic-free
Milli Q water (5 ml × 3) under ultrasonication for 10 min each. The extracts were adjusted to pH 8.5–9.0 using a 0.05 M KOH and then concentrated to ~0.5 ml by rotary evaporation and dried under nitrogen blow. Diacids and related compounds present in samples were then derivatized to dibutyl esters and/or acetals by reacting with 14% BF₃/ n-butanol at 100 °C for 30 min. The derivatives were extracted into n-hexane and concentrated to dryness under rotary evaporation followed by a nitrogen blow and then dissolved in 50 μl of n-hexane. Dibutyl esters and acetals were determined using GC-FID (Agilent 6890) and GC-MS system. Recovery experiments were performed using authentic free diacids and the recoveries obtained were better than 85% for C₂ acid and 90% for other species. Analytical error in duplicate analysis was less than 10% for major diacid experiments were performed using authentic free diacids and the recoveries obtained were better than 85% for C₂ acid and 90% for other species. Analytical error in duplicate analysis was less than 10% for major diacid species. Concentrations of diacids and related compounds reported here were corrected for the blanks but not for recoveries.

3. Results and discussion

3.1. Carbonaceous components and inorganic ions

Summary of concentrations of OC and EC in Guiyang PM₂.₅ samples (n = 25) are given in table 1. Those of the measured inorganic ions in day- (n = 13) and night-time (n = 12) PM₂.₅ are provided in table S1. On average, both OC and EC were higher (19.6 ± 11.2 μg m⁻³ and 6.91 ± 3.22 μg m⁻³, respectively) in daytime than that (14.4 ± 8.78 μg m⁻³ and 4.74 ± 2.75 μg m⁻³) in nighttime. Mass ratios of OC to EC were lower (2.79 ± 0.41) in daytime than those (3.05 ± 0.54) in nighttime. SO₄²⁻ was the most abundant inorganic ions followed by Na⁺ and NO₃⁻, respectively. Concentrations of Cl⁻ and Ca²⁺ were found to be almost equal and fourth most abundant ions followed by K⁺ and NH₄⁺, respectively (table S1).

3.2. Diacids, oxoacids, α-dicarbonyls and fatty acids

Concentrations of diacids, oxoacids, α-dicarbonyls and fatty acids in Guiyang PM₂.₅ (n = 25) are summarized in table 1. Their molecular distributions in day- (n = 13) and night-time (n = 12) during the campaign are depicted in figure 1. Total diacids ranged from 133–3753 ng m⁻³, with an average of 1150 ± 876 ng m⁻³ in daytime and 1076 ± 909 ng m⁻³ in nighttime. Molecular distributions of diacids, oxoacids and α-dicarbonyls in Guiyang aerosols showed that C₂ acid was the most abundant species followed by C₄, t-Ph, glyoxylic (ωC₂) and Ph acids. C₃ acid was found to be the fifth most abundant species followed by C₆ acid whereas 4-oxobutanoic (ωC₄) was the second most abundant oxoacid. Average concentrations of all these species were higher in daytime than those in nighttime (figure 1), although their diurnal trends were not clear and insignificant (not shown here). On the other hand, pyruvic acid (Pyr) showed the third most abundance in oxoacids and maleic acid (M) was the seventh most abundant diacid species followed by methylmaleic acid (mM) with their averages being higher in nighttime than in daytime (figure 1).

The relative abundances of short-chain diacids (C₇–C₁₀) in total normal saturated (C₇–C₁₁) diacids were 60.1%, 9.2% and 17.1%, respectively, whereas that of C₁₆ diacid was 9.3%. ωC₂ was the dominant species among oxoacids while methyglyoxal (mGly) was more abundant than glyoxal (Gly) (figure 2). Even carbon numbered fatty acids were found to be more abundant than odd carbon numbered homologues in Guiyang PM₂.₅ (table 1; figure 2). In fact, C₁₅ and C₁₉ fatty acids were not detected. Palmitic (C₁₆) acid was the most abundant fatty acid species followed by myristic (C₁₄) acid. Oleic (C₁₈:₁) acid was found to be almost equivalent to that of C₁₄ acid followed by stearic (C₁₈:₀) and margaric (C₁₇) acids.

3.3. Comparison with literature

OC/EC ratios (ave. 2.91 ± 0.49) in Guiyang aerosols found to be much higher than those reported for vehicular exhaust (0.71 ~ 1.1) and little higher than those reported for the particles emitted from coal combustion (2.7), but lower than those reported for the particles from biomass burning (6.6–9.0) (Cachier et al 1989, Watson et al 2001, Saarikoski et al 2008, Sandradewi et al 2008). Furthermore, the OC/EC in Guiyang were slightly higher than those reported in other Chinese cities: Beijing (1.9) (Jung et al 2009), Guangzhou (2.0) (Ho et al 2011), Shanghai (2.3) (Ye et al 2003), Chengdu (2.14) (Li et al 2015), and Seoul, Korea (1.8) (Kim et al 2007). Such large differences of OC/EC ratios in Guiyang aerosols with particularly those of the specific source materials, should have been driven by the difference in source types, rather than the different sampling and temperature protocols (e.g., NIOSH) adopted in the measurement of OC and EC in other studies.

Averages of total diacids in Guiyang were higher than those reported from Beijing (760 ng m⁻³) (Ho et al 2010), Chennai, India (673 ng m⁻³) (Pavuluri et al 2010), Ulaanbaatar, Mongolia (536 ng m⁻³) (Jung et al 2010) and Tokyo (438 ng m⁻³) (Kawamura and Yasui 2005). However, they were found to be lower than those reported from other urban localities; Chengdu (3390 ng m⁻³) (Li et al 2015), Xi’an winter haze sample (2885 ng m⁻³) (Cheng et al 2013) and Nanjing (1684 ng m⁻³) (Wang et al 2002) in China, in Hong Kong (1655 ng m⁻³) (Li and Yu 2010) and New Delhi, India (2875 ng m⁻³) (Miyazaki et al 2009), but closer to those reported in 14 Chinese
Table 1. Summary of concentrations of diacids, oxoacids, α-dicarboxyls, and fatty acids (ng m\(^{-3}\)) and OC and EC (μg m\(^{-3}\)) in PM\(_{2.5}\) in Guiyang, Southwest China*.

| Compounds                | Range       | Median  | Average ± SD |
|--------------------------|-------------|---------|--------------|
| **Saturated diacids**    |             |         |              |
| Oxalic, C\(_2\)         | 43.6–1675   | 359     | 486 ± 377    |
| Malonic, C\(_3\)        | 3.17–330    | 55.8    | 74.5 ± 73.8  |
| Succinic, C\(_4\)       | 12.5–498    | 106     | 138 ± 117    |
| Glutaric, C\(_5\)       | 2.66–86.1   | 20.1    | 22.8 ± 19.9  |
| Adipic, C\(_6\)         | 2.17–86.4   | 13.7    | 18.5 ± 18.3  |
| Pimeric, C\(_7\)        | n.d.–17.5   | 3.78    | 4.41 ± 4.12  |
| Azelaic, C\(_8\)        | 9.89–194    | 48.0    | 59.6 ± 46.0  |
| Undecanedioic, C\(_11\) | 0.97–17.0   | 4.27    | 5.24 ± 3.81  |
| Methylmalonic, iC\(_4\) | n.d.–13.7   | 2.24    | 3.55 ± 3.59  |
| Methylsuccinic, iC\(_5\) | 3.16–65.4   | 18.2    | 21.0 ± 14.1  |
| 2-Methylglutaric, iC\(_6\) | n.d.–8.12  | 0.31    | 1.67 ± 2.28  |
| **Unsaturated diacids**  |             |         |              |
| Maleic, M                | 3.98–105    | 22.0    | 27.1 ± 20.8  |
| Fumaric, F               | 0.43–23.7   | 6.64    | 7.24 ± 4.56  |
| Methylmaleic, mM         | 3.16–88.7   | 19.8    | 24.6 ± 18.0  |
| Phthalic, Ph             | 13.2–279    | 76.8    | 89.4 ± 58.2  |
| Isophthalic, iPh         | n.d.–121    | 4.47    | 9.87 ± 23.7  |
| Terephthalic, tPh        | 14.0–533    | 76.4    | 105 ± 107    |
| **Hydroxy or keto diacids** |           |         |              |
| Malic, hC\(_4\)         | n.d.–3.70   | 0.04    | 0.49 ± 0.89  |
| Ketomalonic, iC\(_5\)   | 1.18–46.4   | 11.6    | 13.6 ± 10.4  |
| 4-Ketopimelic, iC\(_7\) | n.d.–13.0   | 0.00    | 1.97 ± 3.29  |
| **Total diacids**        | 133–3753    | 914     | 1115 ± 874   |
| **Oxoacids**             |             |         |              |
| Pyruvic, Pyr             | 3.20–205    | 17.6    | 30.1 ± 47.9  |
| **ω-Oxocids**            |             |         |              |
| Glyoxal, ωC\(_2\)       | 9.85–348    | 87.1    | 95.9 ± 78.7  |
| 3-Oxopropanoic, ωC\(_3\) | 0.96–54.5  | 7.04    | 10.2 ± 10.7  |
| 4-Oxobutanoic, ωC\(_4\) | 5.19–102    | 24.2    | 28.4 ± 21.1  |
| 5-Oxopentanoic, ωC\(_5\) | 1.13–24.0  | 5.84    | 7.61 ± 5.45  |
| 7-Oxooctanoic, ωC\(_7\) | 0.21–34.6   | 6.97    | 9.27 ± 9.38  |
| 8-Oxodecanoic, ωC\(_8\) | n.d.–27.6   | 2.82    | 4.87 ± 6.90  |
| 9-Oxononanoic, ωC\(_9\) | n.d.–12.4   | 2.52    | 3.01 ± 3.51  |
| **Total oxoacids**       | 27.0–694    | 162     | 189 ± 165    |
| **α-Dicarboxyls**        |             |         |              |
| Glyoxal, Gly             | 2.09–35.7   | 13.7    | 15.9 ± 10.6  |
| Methylglyoxal, mGly      | 3.95–76.8   | 19.0    | 22.1 ± 15.3  |
| **Total α-dicarboxylics**| 6.04–131    | 32.9    | 38.0 ± 29.6  |
| **Fatty acids**          |             |         |              |
| C\(_14\)                 | 5.58–3808   | 47.7    | 221 ± 753    |
| C\(_16\)                 | 22.7–5263   | 146     | 412 ± 1031   |
| C\(_17\)                 | 8.70–307    | 33.8    | 52.6 ± 63.0  |
| C\(_18\)                 | 6.18–1778   | 47.0    | 134 ± 350    |
| C\(_18:1\)               | 5.75–3686   | 58.8    | 220 ± 727    |
| C\(_20\)                 | n.d.–98.7   | 4.90    | 9.06 ± 19.3  |
| C\(_22\)                 | n.d.–15.8   | 0.00    | 0.75 ± 3.15  |
| C\(_24\)                 | n.d.–91.3   | 2.39    | 9.23 ± 19.9  |
| C\(_24\)                 | n.d.–34.0   | 0.00    | 1.89 ± 6.90  |
| **Total fatty acids**    | 54.4–15240  | 338     | 1072 ± 2992  |
| OC                       | 5.58–50.1   | 13.0    | 17.1 ± 10.2  |
| EC                       | 2.19–14.2   | 5.25    | 5.87 ± 3.14  |

* SD, standard deviation;  
  * n.d., not detectable
Figure 1. Molecular distributions of diacids, oxoacids, α-dicarbonyls, and fatty acids in PM$_{2.5}$ in Guiyang, Southwest China.

Figure 2. Temporal variations of (a) total diacids, oxoacids, α-dicarbonyls and fatty acids; (b) OC and EC and (c) K$^+$, NH$_4^+$, NO$_3^-$, and SO$_4^{2-}$ ions in PM$_{2.5}$ in Guiyang, Southwest China.
cities (904 ng m\(^{-3}\)) (Ho et al 2007). These comparisons suggest that Guiyang aerosols should have been significantly contributed from other sources rather than from fossil fuel combustion and biomass burning.

### 3.4. Temporal and diurnal variations

Although, on average, most of the species were dominant in daytime (figure 1), none of the species and carbonaceous components showed a clear diurnal trend during the campaign (figure 2). However, total diacids, oxoacids and α-dicarbonyls as well as OC were drastically increased by a factor of 2 ~ 3 on December 15 daytime followed by a small decrease in the nighttime sample on that day (figure 2(a)). EC also showed a sharp increase in daytime on December 15, similar to that of OC. Interestingly, concentrations of total fatty acids were found to be sharply increased by a factor of about 10 and 60 in December 15 day- and night-time samples, respectively, compared to that in December 14 samples (figure 2(b)). On the contrary, inorganic ions showed neither diurnal trend during the campaign nor the peak values in the December 15 samples (figure 2(c)).

### 3.5. Origins and secondary formation/transformations

#### 3.5.1. Biogenic emissions

It is well known that EC emits directly into the atmosphere from fossil fuel combustion and biomass burning and used as a good tracer for primary combustion generated-carbon (Andreae 1983, Yu et al 2004). While K\(^+\) emits from biomass burning but not from fossil fuel combustion and hence, it can be used as a marker for biomass burning (Andreae 1983). Similarly, NO\(_3^-\) and SO\(_4^{2-}\) are secondarily produced in the atmosphere from NO\(_x\) and SO\(_2\) generally emitted from combustion sources (Wang et al 2016). Hence, the most and third most abundances of SO\(_4^{2-}\) and NO\(_3^-\), respectively (table S1), suggest that the contributions from anthropogenic emissions and secondary formation were high to PM\(_{2.5}\) in Guiyang. Conversely, OC showed a very good linear relationship with EC (figure 3(a)), which implies that they both should have been derived from common combustion source(s), potentially fossil fuel combustion and biomass burning. However, both OC and EC did not show any relation either with K\(^+\) and sum of NO\(_3^-\) and SO\(_4^{2-}\) (figures 3(b), (c)). Therefore, it is unlikely that either fossil fuel combustion or biomass burning are the major sources of carbonaceous aerosols in Guiyang. Similarly, the second most abundance of Na\(^+\), a tracer for sea-salt, suggests that the sea-salt contribution should also be high, but it is unlikely because Guiyang is located in central Southwest China and far away from the sea shore.

Recently, Pavuluri et al (2015a) reported that dimethyl sulfide (DMS) emission from higher plants and its subsequent oxidation processes in the atmosphere is a major source of SO\(_4^{2-}\) in growing season. Na\(^+\) can also be...
significantly derived from microbial (e.g., fungi) activities in soil because the emitted spores and hyphae are enriched with several elements including Na (Burford et al 2003). Therefore, we presume that the biogenic emission of VOCs including DMS and their subsequent oxidation in the atmosphere as well as the microbial activities are dominant sources of the PM$_{2.5}$ in Guiyang, although we do not preclude the significant contributions from anthropogenic sources. In fact, the fourth most abundance of Ca$^{2+}$ (table S1) indicates a substantial contribution of soil dust as well to the PM$_{2.5}$ in Guiyang.

Ph and t-Ph, which originate from fossil fuel combustion and/or plastic burning, were higher than that of C$_3$ diacid that is derived from both anthropogenic and biogenic emissions followed by secondary processes (Fu et al 2010, Kawamura and Pavuluri 2010) (table 1; figure 1). However, similar to OC and EC, total diacids, oxoacids and α-dicarboxyls did not showed any covariance with K$^+$ (figure 2). Both C$_4$ and Ph diacids, which are considered to be mainly derived from fossil fuel combustion (Kawamura et al 1996), were also did not showed any linear relation with NO$_3^-$ and SO$_4^{2-}$ (not shown here). Such insignificant relations with the marker ions again indicate that OA in Guiyang should have been mainly derived from other sources rather than the emissions from fossil fuel combustion and biomass burning.

Interestingly, concentrations of C$_9$ diacid, which is produced by photochemical oxidation of unsaturated fatty acids (e.g., C$_{18:1}$) emitted from marine and terrestrial biota and/or biomass burning (Kawamura et al 1996), were also close to those of C$_3$ diacid (table 1; figure 2). Furthermore, fatty acids showed even-carbon-numbered predominance with a maximum at C$_{16}$. The carbon preference index (CPI, the ratio of sums of even- to odd-carbon numbered species) of the measured fatty acids ranged from 1.82–41.3 (ave. 12.0 ± 9.06). Such molecular distributions and CPI values suggest that they were most likely derived from terrestrial higher plants (Fu et al 2008). It is likely because the dense forest vegetation in Guiyang (Xiao et al 2012) that could emit high amounts of volatile organics and fatty acids, resulting in significant SOA by secondary processes in the atmosphere.

However, the temporal variations of EC, which does not derive from biogenic emissions and even from soil dust, and Ph and t-Ph acids were similar to those of OC, and sub-totals of diacids and related compounds. Such similarities suggest that the carbonaceous aerosols should have been significantly derived from combustion sources, excluding the biomass and fossil fuel combustion, in addition to biogenic emissions.

### 3.5.2. Cooking emissions

Recently, it has been reported that cooking emissions contribute 10 ~ 35% of total OA loading in urban areas (Rogge et al 1991, Allan et al 2010, Crippa et al 2013, Lee et al 2015). Lee et al (2015) found that contribution from cooking emissions in the commercial and shopping area of Mong kok in Hong Kong even dominated to primary OA, but their contribution to secondary OA was not fully understood. Recently, Liu et al (2018a) found from chamber experiments on heated cooking oils that they significantly contribute to both POA and SOA and the cooking emissions have been suggested to be an important source of OA in urban areas. It has also been reported that emissions of n-alkanes, n-alkenals and fatty acids are significant from cooking with seed oils (Schauer et al 2002) and meat charbroiling (Louvaris et al 2017). All these organics can produce SOA, particularly diacids and related compounds, upon photochemical oxidation in the atmosphere (Kawamura et al 1996). Therefore, the consistency in the covariance of total fatty acids with diacids, oxoacids α-dicarboxyls as well as OC imply that the OA in Guiyang region should have been significantly derived from cooking activities as well. Further we presume that EC should have also been derived from cooking activities in Guiyang, although we do not preclude the emission from fossil fuel combustion.

### 3.5.3. Municipal waste burning

As noted earlier, concentrations of diacids and related compounds and OC and EC, but not those of marker ions; K$^+$, NH$_4^+$, NO$_3^-$, and SO$_4^{2-}$, in December 15 samples were raised drastically (figure 2). The rise was more pronounced in the case of fatty acids and t-Ph acid compared to that in other species and components (figure 2). The absence of such rise in the concentrations of inorganic markers imply that there is no sudden input from biomass burning and/or fossil fuel combustion. It is also unlikely that biogenic emissions and subsequent secondary processes cause such a sharp rise for just one day during the campaign. Furthermore, to the best of our knowledge, there was no any unusual cooking activity in the vicinity of the sampling location. Therefore, the carbonaceous aerosols (including EC) must have been derived from another combustion source, potentially municipal waste burning (Kawamura and Pavuluri 2010).

It has been reported that seasonality of Ph acid (summer > winter) is opposite to that of the primary organics emitted from coal combustion and biomass burning in China (Wang et al 2007). Based on the vertical measurements of diacids at the coastal and inland sites over China, Zhang et al (2016) proposed that Ph acid can be directly emitted from evaporation of plasticizers widely used in China at higher temperatures, in addition to secondary formation from naphthalene. Kawamura and Pavuluri (2010) found very high loading (range, 7.6–158 ng m$^{-3}$) of t-Ph acid in the tropical Indian aerosols collected from Chennai and attributed for municipal waste burning. It has also been reported that t-Ph acid is abundant in smoke particles emitted from
open burning of new plastic bags (907 ng mg$^{-1}$) roadside litter (5033 ng mg$^{-1}$) and landfill trash 176 ng mg$^{-1}$) (Simoneit et al. 2005).

Concentrations of Ph and t-Ph acids increased from 70.2 and 45.0 ng m$^{-3}$ in 14 December nighttime sample to 279 and 296 ng m$^{-3}$ and 204 and 533 ng m$^{-3}$ in 15 December day- and night-time samples, respectively (figure 2(a)). Similarly, concentrations of all the measured fatty acids, particularly C$_{14}$, C$_{18}$ and C$_{18:1}$, (not shown here) and their sum (figure 2(a)) were highly increased in December 15 samples. Furthermore, total fatty acids and t-Ph acid were well covaried, particularly on December 15 and during the following several days as well (figure 2(a)). Total fatty acids showed a very good linear relationship ($r^2 = 0.80$) with t-Ph irrespective of the inclusion or exclusion of the outliers in the observed data set (figure S2). It implies that they should have been mainly generated from similar type of sources and/or processes.

As described above, t-Ph acid can be abundantly derived from landfill trash burning (Simoneit et al. 2005, Kawamura and Pavuluri 2010). In fact, most of the municipal waste is disposed into landfills in many cities including Guiyang in China without regulations, and the local residents often set light to the garbage that could emit lot of organic gases and carbonaceous particles including EC. Interestingly, both OC and EC showed very good linear relations with C$_6$ diacid (figure 3(e)) that must have generated from food and other biological materials waste burning, whereas moderate correlations with both C$_{16}$ and t-Ph acids (figures 3(d), (f)). These results reveal that an unnoticed sudden municipal waste burning in the vicinity of Guiyang urban area should have caused such a drastic enhancement in the carbonaceous aerosol loading on 15 December during the campaign. Further carbonaceous aerosols should have been extensively derived from the burning of biological material (e.g., food) rather than the plastics present in the municipal waste.

Figure 4. Linear relations between mass ratios of selected diacids and the relative abundance of C$_2$ diacid in PM$_{2.5}$ in Guiyang, Southwest China.
3.5.4. Secondary formation and transformations
Although diacids did not show a clear diurnal pattern during the campaign, their averages were higher in daytime than in nighttime, except for M, mM, i-Ph and t-Ph acids (figure 1). Such higher concentrations of most of diacids in daytime can be attributed to enhanced in situ photochemical production from their precursors due to high ambient temperature and solar radiation in daytime. The relative abundance (Cf%) of C2 diacid, an end product of chain oxidation reactions via ωC2 acid and C2 and kC3 diacids, has been proposed as a proxy for aging of OA (Kawamura and Sakaguchi 1999). Interestingly, as can be seen from figure 4, the scatter plots between Cf% and the mass ratios of C2 to ωC2, C4 and C6 acids and to C4/C9 ratios showed positive relations but no relation was found between Cf% and C2/kC3 (not shown here). Such relations imply that the secondary processing of OA was significant in the Guiyang atmosphere at local scale but the OA was not aged. Further the linear relations between C2%/ and C2/C9 and C4/C9 indicates that the photochemical breakdown of all diacids derived from biological materials (including those from municipal waste burning) to their lower homologues was significant in Guiyang.

Similarly, the mass ratios of M/F has also been considered as a proxy for aging (Kawamura and Sakaguchi 1999). The M/F ratios were also showed a significant diurnal pattern; on average 3.01 in daytime and 5.66 in nighttime, in Guiyang. They were almost twice compared to those (1.5 in summer and 3.0 in winter) reported in 14 Chinese cities, which have been considered to be derived from fossil fuel combustion (Ho et al 2007), and those (1.8 in daytime and 2.7 in nighttime) reported in biomass burning aerosols from Brazil (Kundu et al 2010). The former again indicates the enhanced in situ photochemical formation of OA (Kawamura and Sakaguchi 1999) in daytime, while the latter further supports that the contributions from fossil fuel combustion and biomass burning emissions are minor in Guiyang.

4. Conclusion
Our results imply that the emission of gaseous organic compounds, e.g., unsaturated fatty acids, from higher plants and cooking activities and municipal waste burning and their subsequent secondary processes are the major sources of organic aerosols in southwest China. The results obtained also suggest that the cooking and municipal waste burning emissions significantly contributes to EC. Further this study implies that municipal waste burning is a large source of carbonaceous aerosols including more oxygenated species in the urban atmosphere. Hence, it is highly necessary to further investigate the importance of different types of municipal waste burning to the carbonaceous aerosol loading and their impacts on the Earth’s climate system and air quality.

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ORCID iDs
C M Pavuluri @ https://orcid.org/0000-0002-2955-474X
K Kawamura @ https://orcid.org/0000-0003-1190-3726
P Q Fu @ https://orcid.org/0000-0001-6249-2280

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