



## Review

# Advances in Organic Aerosol Characterization: From Complex to Simple

Stefania Gilardoni\*

*Institute of Atmospheric Sciences and Climate, National Research Council, 40129 Bologna, Italy*

**Keywords:** Organic aerosol; Aerosol mass spectrometry; Fourier-transform infrared spectrometry; Nuclear magnetic resonance; Organic functional groups.

Air pollution is the “world largest single environmental health risk” (WHO, 2016). According to the World Health Organization, every year ambient air pollution leads to more than 3 million premature deaths, and 84% of the global population is exposed to atmospheric aerosol levels higher than the limits set to protect human health (WHO, 2016). One of the most abundant, and still less characterized components of atmospheric aerosol is organic aerosol (OA). In particular, OA sources, formation mechanisms, atmospheric ageing, and effects on climate and human health are still subject of research.

OA is a mixture of thousands of chemical species, characterized by different physical, chemical, and toxicological properties. Such properties define the effects of OA, and particulate matter, on climate, air quality, and human health (Goldstein and Galbally, 2007). Traditional analytical techniques, based on the identification of single molecular species, describe only a limited fraction of OA mass, generally lower than 30% (Hallquist *et al.*, 2009). During the last decade the use of bulk OA spectrometric analysis introduced simplified descriptions of OA. Such approaches depict OA as formed by a few components with different degrees of oxidation (Aerosol mass spectrometry - AMS) (Zhang *et al.*, 2005), or as the sum of different organic functional groups, (Fourier transform infrared spectrometry - FTIR (Maria *et al.*, 2003), or proton nuclear magnetic resonance spectrometry - H-NMR (Decesari *et al.*, 2007)).

With the acronym AMS we refer here to high-resolution time of flight aerosol mass spectrometer (HR-ToF-AMS), soot photometer aerosol mass spectrometer (SP-AMS), and aerosol chemical speciation monitor (ACSM). The AMSs measure the chemical composition of submicron particles on-line, with a resolution of a few minutes (Jayne *et al.*, 2000; DeCarlo *et al.*, 2006). HR-ToF-AMS and ACSM measure non-refractory OA, which generally accounts for the largest fractions of carbonaceous matter, while SP-AMS allows the quantification of refractory material as well (Onasch *et*

*al.*, 2012). The OA mass measured by AMS correlates well with the organic carbon measured with thermo-optical method, such as online Sunset analyzer (Takegawa *et al.*, 2005; Takegawa *et al.*, 2009). The correlation slope reported in literature varies between 1.6 and 2, in agreement with theoretical values estimated from OA molecular composition (Turpin and Lim, 2001), and with organic mass to organic carbon ratio (OM/OC) measured by AMS and FTIR (Russell, 2003; Aiken *et al.*, 2008). Still, the accuracy of AMS OA measurements depends on the accuracy collection efficiency (CE) determination, which usually varies between 0.5 and 1, and depends on ammonium nitrate mass fraction, aerosol liquid water content (Middlebrook *et al.*, 2012), and organic aerosol composition (Docherty *et al.*, 2013). For such a reason efforts are now focusing on the development of new types of vaporizers, in order to achieved a constant collection efficiency equal to unity (Hu *et al.*, 2017).

Due to the high energy of mass spectrometry fragmentation, the information of original molecular formula is mostly lost. Nevertheless, multivariate statistical analysis of the mass spectra identifies OA components characterized by different oxygen content (Zhang *et al.*, 2005; Canagaratna *et al.*, 2007), which can be linked to primary and secondary OA components. In addition, the analysis of OA fragmentation mass spectra allows the indirect measure of OA elemental composition, and the OM/OC (Aiken *et al.*, 2008; Canagaratna *et al.*, 2015). The elemental analysis algorithms were developed based on the study of a large set of organic standards, representative of OA composition (Aiken *et al.*, 2007; Aiken *et al.*, 2008; Canagaratna *et al.*, 2015). Such algorithm takes into account neutral losses of fragment containing electronegative atoms (i.e., CO<sub>2</sub> and H<sub>2</sub>O), and the dependency of thermal decomposition mechanisms on the organic functional group composition. The accuracy of O:C and H:C determinations is 28% and 13%, respectively (Canagaratna *et al.*, 2015).

FT-IR analysis of bulk OA is performed by collecting infrared absorption spectra of particulate matter, or spectra of OA fractions isolated by organic solvent extraction. When particulate matter samples are analyzed directly, the totality of OA can be characterized. OA is described by FT-IR analysis as the sum of organic functional groups, i.e.,

\* Corresponding author.

E-mail address: s.gilardoni@isac.cnr.it

alkyl (C-H) bonds, carbonyl (C=O), carboxyl (C(OH)=O), amine (NH), hydroxyl (C-OH) groups, organic sulfate, and organic nitrate (Russell *et al.*, 2011; Ruggeri and Takahama, 2016). Time resolution is limited by the detection limit and ambient OA concentrations. It can range from less than 1 hour up to 24 hours (Gilardoni *et al.*, 2009). The calculation of OA mass based on FTIR assumes that all functional groups that compose OA are above the limit of quantification, and the mass fraction of functional groups transparent to IR radiation (i.e., tertiary carbon) is negligible. The comparison of FTIR and off-line thermal/optical organic carbon shows a Pearson correlation coefficient higher than 0.93 and errors smaller than 10% (Maria *et al.*, 2003; Weakley *et al.*, 2016). The OA concentration and OM/OC ratio measured by FTIR agree within 20% and 10% with AMS results, respectively (Russell *et al.*, 2009; Gilardoni *et al.*, 2014). The main limitations of FTIR are the potential evaporative losses of volatile OA during filter sampling, which could also explain the larger discrepancy with on-line OA (AMS) compared to off-line OA (thermal/optical).

H-NMR was developed for off-line analysis of water-soluble OA fraction. H-NMR is able to identify the following functional groups: aryl and alkyl C-H bonds, C=O from ketones, aldehydes, and carboxylic acids, C-O from ethers and alcohols, and O-C-O moieties (Decesari *et al.*, 2007). The time resolution ranges usually from a few hours up to 24 hours (Finessi *et al.*, 2012; Paglione *et al.*, 2014).

The ability of these bulk OA analysis to characterize a larger fraction of OA mass, compared to traditional techniques, promoted the investigation of OA sources, especially through receptor modeling. The analysis of AMS OA fragmentation spectra led to the identification of oxygenated organic aerosol (OOA), a proxy of secondary OA (SOA), which dominates over hydrocarbon like OA (a proxy of primary OA) in urban, rural, and remote areas (Zhang *et al.*, 2007; Crippa *et al.*, 2014). AMS and FT-IR analysis of aerosol collected in different continents identified the following main OA sources: fuel combustion, biogenic, biomass burning, cooking, and marine OA (Hawkins and Russell, 2010; Russell *et al.*, 2011; Mohr *et al.*, 2012; Corrigan *et al.*, 2013; Crippa *et al.*, 2013). The comparison between H-NMR reference and ambient spectra allowed the identification fresh and aged wood burning aerosol (Paglione *et al.*, 2014), primary and secondary marine OA (Decesari *et al.*, 2011), and biogenic SOA (Finessi *et al.*, 2012).

The information about the chemical structure of bulk OA was used to gain insights into SOA formation mechanisms. For example, FTIR and AMS studies identified the contribution of gas-phase ammonia in the formation of secondary organic nitrate (O'Brien *et al.*, 2013), gave insights into oligomerization chemical mechanisms involved in the formation of biogenic SOA (Russell *et al.*, 2011), and identified phenols as precursors of aqueous SOA (Sun *et al.*, 2010). The analysis of aqueous SOA with FT-IR and H-NMR indicated that different chemical mechanisms occur during processing in cloud and in wet aerosol particles (George *et al.*, 2015; Gilardoni *et al.*, 2016).

The ability to describe the complexity of the bulk OA with a limited set of variables helped the parameterization of OA atmospheric evolution, and climate properties (Jimenez *et al.*, 2009). The elemental ratios (i.e., O/C ratio and H/C ratio) from AMS studies were used to infer the OA compositional change with ageing (Ng *et al.*, 2010, 2011). Kroll *et al.* (2011) showed that the carbon oxidation state (derived from OA elemental composition) varies with the OA carbon number, and such variables together describe the chemical mechanisms behind OA atmospheric evolution. O/C ratio and oxidized organic functional groups were used to parameterize OA hygroscopicity and optical properties (Duplissy *et al.*, 2011; Lambe *et al.*, 2011; Kim *et al.*, 2014).

During the last decade, bulk OA spectrometry improved the ability of the scientific community to describe and predict spatial distribution, temporal evolution, and climate impact of OA. Still open questions remain, especially on the health outcome of OA. Deployment of FTIR and NMR is so far limited by the low measurement time resolution, and the OA sampling artifacts. Conversely, the high time resolved AMS measurements do not describe the OA chemical composition, which could be used to understand and eventually model aerosol toxicity. Further studies should be encouraged to deploy an integrated approach, to take advantage of the completeness, the link to chemical composition, and the simplicity of the different bulk OA spectrometric techniques.

## REFERENCES

- Aiken, A., Decarlo, P., Kroll, J., Worsnop, D., Huffman, J., Docherty, K., Ulbrich, I., Mohr, C., Kimmel, J., Sueper, D., Sun, Y., Zhang, Q., Trimborn, A., Northway, M., Ziemann, P., Canagaratna, M., Onasch, T., Alfarra, M., Prevot, A., Dommen, J., Duplissy, J., Metzger, A., Baltensperger, U. and Jimenez, J. (2008). O/C and OM/OC ratios of primary, secondary, and ambient organic aerosols with high-resolution time-of-flight aerosol mass spectrometry. *Environ. Sci. Technol.* 42: 4478–4485.
- Aiken, A.C., DeCarlo, P.F. and Jimenez, J.L. (2007). Elemental analysis of organic species with electron ionization high-resolution mass spectrometry. *Anal. Chem.* 79: 8350–8358.
- Canagaratna, M., Jayne, J., Jimenez, J., Allan, J., Alfarra, M., Zhang, Q., Onasch, T., Drewnick, F., Coe, H., Middlebrook, A., Delia, A., Williams, L., Trimborn, A., Northway, M., DeCarlo, P., Kolb, C., Davidovits, P. and Worsnop, D. (2007). Chemical and microphysical characterization of ambient aerosols with the aerodyne aerosol mass spectrometer. *Mass Spectrom. Rev.* 26: 185–222.
- Canagaratna, M.R., Jimenez, J.L., Kroll, J.H., Chen, Q., Kessler, S.H., Massoli, P., Ruiz, L.H., Fortner, E., Williams, L.R., Wilson, K.R., Surratt, J.D., Donahue, N.M., Jayne, J.T. and Worsnop, D.R. (2015). Elemental ratio measurements of organic compounds using aerosol mass spectrometry: Characterization, improved calibration, and implications. *Atmos. Chem. Phys.* 15: 253–272.

- Corrigan, A., Russell, L., Takahama, S., Aijala, M., Ehn, M., Junninen, H., Rinne, J., Petaja, T., Kulmala, M., Vogel, A., Hoffmann, T., Ebben, C., Geiger, F., Chhabra, P., Seinfeld, J., Worsnop, D., Song, W., Auld, J. and Williams, J. (2013). Biogenic and biomass burning organic aerosol in a boreal forest at Hyytiälä, Finland, during HUMPPA-COPEC 2010. *Atmos. Chem. Phys.* 13: 12233–12256.
- Crippa, M., DeCarlo, P., Slowik, J., Mohr, C., Heringa, M., Chirico, R., Poulain, L., Freutel, F., Sciare, J., Cozic, J., Di Marco, C., Elsasser, M., Nicolas, J., Marchand, N., Abidi, E., Wiedensohler, A., Drewnick, F., Schneider, J., Borrmann, S., Nemitz, E., Zimmermann, R., Jaffrezo, J., Prevot, A. and Baltensperger, U. (2013). Wintertime aerosol chemical composition and source apportionment of the organic fraction in the metropolitan area of Paris. *Atmos. Chem. Phys.* 13: 961–981.
- Crippa, M., Canonaco, F., Lanz, V.A., Äijälä, A., J.D., Carbone, S., Capes, G., Dall'Osto, M., Day, D.A., DeCarlo, P.F., Di Marco, C.F., Ehn, M., Eriksson, A., Freney, E., Hildebrandt Ruiz, L., Hillamo, R., Jimenez, J.L., Junninen, H., Kiendler-Scharr, A., Kortelainen, A.M., Kulmala, M., Mensah, A.A., Mohr, C., Nemitz, E., O'Dowd, C., Ovadnevaite, J., Pandis, S.N., Petäjä, T., Poulain, L., Saarikoski, S., Sellegri, K., Swietlicki, E., Tiitta, P., Worsnop, D.R., Baltensperger, U. and Prévôt, A.S.H. (2014). Organic aerosol components derived from 25 AMS data sets across Europe using a consistent ME-2 based source apportionment approach. *Atmos. Chem. Phys.* 14: 6159–6176.
- DeCarlo, P., Kimmel, J., Trimborn, A., Northway, M., Jayne, J., Aiken, A., Gonin, M., Fuhrer, K., Horvath, T., Docherty, K., Worsnop, D. and Jimenez, J. (2006). Field-deployable, high-resolution, time-of-flight aerosol mass spectrometer. *Anal. Chem.* 78: 8281–8289.
- Decesari, S., Mircea, M., Cavalli, F., Fuzzi, S., Moretti, F., Tagliavini, E. and Facchini, M. (2007). Source attribution of water-soluble organic aerosol by nuclear magnetic resonance spectroscopy. *Environ. Sci. Technol.* 41: 2479–2484.
- Decesari, S., Finessi, E., Rinaldi, M., Paglione, M., Fuzzi, S., Stephanou, E.G., Tziaras, T., Spyros, A., Ceburnis, D., O'Dowd, C., Dall'Osto, M., Harrison, R.M., Allan, J., Coe, H. and Facchini, M.C. (2011). Primary and secondary marine organic aerosols over the North Atlantic Ocean during the MAP experiment. *J. Geophys. Res.* 116: D22210.
- Docherty, K.S., Jaoui, M., Corse, E., Jimenez, J.L., Offenberg, J.H., Lewandowski, M. and Kleindienst, T.E. (2013). Collection efficiency of the aerosol mass spectrometer for chamber-generated secondary organic aerosols. *Aerosol Sci. Technol.* 47: 294–309.
- Duplissy, J., DeCarlo, P.F., Dommen, J., Alfarra, M.R., Metzger, A., Barmapadimos, I., Prevot, A.S.H., Weingartner, E., Tritscher, T., Gysel, M., Aiken, A.C., Jimenez, J.L., Canagaratna, M.R., Worsnop, D.R., Collins, D.R., Tomlinson, J. and Baltensperger, U. (2011). Relating hygroscopicity and composition of organic aerosol particulate matter. *Atmos. Chem. Phys.* 11: 1155–1165.
- Finessi, E., Decesari, S., Paglione, M., Giulianelli, L., Carbone, C., Gilardoni, S., Fuzzi, S., Saarikoski, S., Raatikainen, T., Hillamo, R., Allan, J., Mentel, T.F., Tiitta, P., Laaksonen, A., Petaja, T., Kulmala, M., Worsnop, D.R. and Facchini, M.C. (2012). Determination of the biogenic secondary organic aerosol fraction in the boreal forest by NMR spectroscopy. *Atmos. Chem. Phys.* 12: 941–959.
- George, K.M., Ruthenburg, T.C., Smith, J., Yu, L., Zhang, Q., Anastasio, C. and Dillner, A.M. (2015). FT-IR quantification of the carbonyl functional group in aqueous-phase secondary organic aerosol from phenols. *Atmos. Environ.* 100: 230–237.
- Gilardoni, S., Liu, S., Takahama, S., Russell, L.M., Allan, J.D., Steinbrecher, R., Jimenez, J.L., De Carlo, P.F., Dunlea, E.J. and Baumgardner, D. (2009). Characterization of organic ambient aerosol during MIRAGE 2006 on three platforms. *Atmos. Chem. Phys.* 9: 5417–5432.
- Gilardoni, S., Massoli, P., Giulianelli, L., Rinaldi, M., Paglione, M., Pollini, F., Lanconelli, C., Poluzzi, V., Carbone, S., Hillamo, R., Russell, L.M., Facchini, M.C. and Fuzzi, S. (2014). Fog scavenging of organic and inorganic aerosol in the Po Valley. *Atmos. Chem. Phys.* 14: 6967–6981.
- Gilardoni, S., Massoli, P., Paglione, M., Giulianelli, L., Carbone, C., Rinaldi, M., Decesari, S., Sandrini, S., Costabile, F., Gobbi, G.P., Pietrogrande, M.C., Visentin, M., Scotto, F., Fuzzi, S. and Facchini, M.C. (2016). Direct observation of aqueous secondary organic aerosol from biomass-burning emissions. *Proc. Natl. Acad. Sci. U.S.A.* 113: 10013–10018.
- Goldstein, A. and Galbally, I. (2007). Known and unexplored organic constituents in the earth's atmosphere. *Environ. Sci. Technol.* 41: 1514–1521.
- Hallquist, M., Wenger, J., Baltensperger, U., Rudich, Y., Simpson, D., Claeys, M., Dommen, J., Donahue, N., George, C., Goldstein, A., Hamilton, J., Herrmann, H., Hoffmann, T., Iinuma, Y., Jang, M., Jenkin, M., Jimenez, J., Kiendler-Scharr, A., Maenhaut, W., McFiggans, G., Mentel, T., Monod, A., Prevot, A., Seinfeld, J., Surratt, J., Szmigielski, R. and Wildt, J. (2009). The formation, properties and impact of secondary organic aerosol: current and emerging issues. *Atmos. Chem. Phys.* 9: 5155–5236.
- Hawkins, L.N. and Russell, L. (2010). Polysaccharides, proteins, and phytoplankton fragments: Four chemically distinct types of marine primary organic aerosol classified by single particle spectromicroscopy. *Adv. Meteorol.* 2010: 612132.
- Hu, W., Campuzano-Jost, P., Day, D.A., Croteau, P., Canagaratna, M.R., Jayne, J.T., Worsnop, D.R. and Jimenez, J.L. (2017). Evaluation of the new capture vaporizer for aerosol mass spectrometers (AMS) through field studies of inorganic species. *Aerosol Sci. Technol.* 51: 735–754.
- Jayne, J., Leard, D., Zhang, X., Davidovits, P., Smith, K., Kolb, C. and Worsnop, D. (2000). Development of an aerosol mass spectrometer for size and composition analysis of submicron particles. *Aerosol Sci. Technol.*

- 33: 49–70.
- Jimenez, J., Canagaratna, M., Donahue, N., Prevot, A., Zhang, Q., Kroll, J., DeCarlo, P., Allan, J., Coe, H., Ng, N., Aiken, A., Docherty, K., Ulbrich, I., Grieshop, A., Robinson, A., Duplissy, J., Smith, J., Wilson, K., Lanz, V., Hueglin, C., Sun, Y., Tian, J., Laaksonen, A., Raatikainen, T., Rautiainen, J., Vaattovaara, P., Ehn, M., Kulmala, M., Tomlinson, J., Collins, D., Cubison, M., Dunlea, E., Huffman, J., Onasch, T., Alfarra, M., Williams, P., Bower, K., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Salcedo, D., Cottrell, L., Griffin, R., Takami, A., Miyoshi, T., Hatakeyama, S., Shimono, A., Sun, J., Zhang, Y., Dzepina, K., Kimmel, J., Sueper, D., Jayne, J., Herndon, S., Trimborn, A., Williams, L., Wood, E., Middlebrook, A., Kolb, C., Baltensperger, U. and Worsnop, D. (2009). Evolution of organic aerosols in the atmosphere. *Science* 326: 1525–1529.
- Kim, H., Liu, S., Russell, L.M. and Paulson, S.E. (2014). Dependence of real refractive indices on O:C, H:C and mass fragments of secondary organic aerosol generated from ozonolysis and photooxidation of limonene and alpha-pinene. *Aerosol Sci. Technol.* 48: 498–507.
- Kroll, J., Donahue, N., Jimenez, J., Kessler, S., Canagaratna, M., Wilson, K., Altieri, K., Mazzoleni, L., Wozniak, A., Bluhm, H., Mysak, E., Smith, J., Kolb, C. and Worsnop, D. (2011). Carbon oxidation state as a metric for describing the chemistry of atmospheric organic aerosol. *Nat. Chem.* 3: 133–139.
- Lambe, A., Onasch, T., Massoli, P., Croasdale, D., Wright, J., Ahern, A., Williams, L., Worsnop, D., Brune, W. and Davidovits, P. (2011). Laboratory studies of the chemical composition and cloud condensation nuclei (CCN) activity of secondary organic aerosol (SOA) and oxidized primary organic aerosol (OPOA). *Atmos. Chem. Phys.* 11: 8913–8928.
- Maria, S.F., Russell, L.M., Turpin, B.J., Porcja, R.J., Campos, T.L., Weber, R.J. and Huebert, B.J. (2003). Source signatures of carbon monoxide and organic functional groups in Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) submicron aerosol types. *J. Geophys. Res.* 108: 8637.
- Middlebrook, A.M., Bahreini, R., Jimenez, J.L. and Canagaratna, M.R. (2012). Evaluation of composition-dependent collection efficiencies for the Aerodyne aerosol mass spectrometer using field data. *Aerosol Sci. Technol.* 46: 258–271.
- Mohr, C., DeCarlo, P.F., Heringa, M.F., Chirico, R., Slowik, J.G., Richter, R., Reche, C., Alastuey, A., Querol, X., Seco, R., Penuelas, J., Jimenez, J.L., Crippa, M., Zimmermann, R., Baltensperger, U. and Prevot, A.S.H. (2012). Identification and quantification of organic aerosol from cooking and other sources in Barcelona using aerosol mass spectrometer data. *Atmos. Chem. Phys.* 12: 1649–1665.
- Ng, N., Canagaratna, M., Zhang, Q., Jimenez, J., Tian, J., Ulbrich, I., Kroll, J., Docherty, K., Chhabra, P., Bahreini, R., Murphy, S., Seinfeld, J., Hildebrandt, L., Donahue, N., DeCarlo, P., Lanz, V., Prevot, A., Dinar, E., Rudich, Y. and Worsnop, D. (2010). Organic aerosol components observed in Northern Hemispheric datasets from Aerosol Mass Spectrometry. *Atmos. Chem. Phys.* 10: 4625–4641.
- Ng, N.L., Canagaratna, M.R., Jimenez, J.L., Chhabra, P.S., Seinfeld, J.H. and Worsnop, D.R. (2011). Changes in organic aerosol composition with aging inferred from aerosol mass spectra. *Atmos. Chem. Phys.* 11: 6465–6474.
- O'Brien, R.E., Nguyen, T.B., Laskin, A., Laskin, J., Hayes, P.L., Liu, S., Jimenez, J.L., Russell, L.M., Nizkorodov, S.A. and Goldstein, A.H. (2013). Probing molecular associations of field-collected and laboratory-generated SOA with nano-DESI high-resolution mass spectrometry. *J. Geophys. Res.* 118: 1042–1051.
- Onasch, T.B., Trimborn, A., Fortner, E.C., Jayne, J.T., Kok, G.L., Williams, L.R., Davidovits, P. and Worsnop, D.R. (2012). Soot particle aerosol mass spectrometer: Development, validation, and initial application. *Aerosol Sci. Technol.* 46: 804–817.
- Paglione, M., Saarikoski, S., Carbone, S., Hillamo, R., Facchini, M., Finessi, E., Giulianelli, L., Carbone, C., Fuzzi, S., Moretti, F., Tagliavini, E., Swietlicki, E., Stenstrom, K., Prevot, A., Massoli, P., Canagaratna, M., Worsnop, D. and Decesari, S. (2014). Primary and secondary biomass burning aerosols determined by proton nuclear magnetic resonance (<sup>1</sup>H-NMR) spectroscopy during the 2008 EUCAARI campaign in the Po Valley (Italy). *Atmos. Chem. Phys.* 14: 5089–5110.
- Ruggeri, G. and Takahama, S. (2016). Technical Note: Development of chemoinformatic tools to enumerate functional groups in molecules for organic aerosol characterization. *Atmos. Chem. Phys.* 16: 4401–4422.
- Russell, L., Bahadur, R., Hawkins, L., Allan, J., Baumgardner, D., Quinn, P. and Bates, T. (2009). Organic aerosol characterization by complementary measurements of chemical bonds and molecular fragments. *Atmos. Environ.* 43: 6100–6105.
- Russell, L., Bahadur, R. and Ziemann, P. (2011). Identifying organic aerosol sources by comparing functional group composition in chamber and atmospheric particles. *Proc. Natl. Acad. Sci. U.S.A.* 108: 3516–3521.
- Russell, L.M. (2003). Aerosol organic-mass to organic-carbon ratio measurements. *Environ. Sci. Technol.* 37: 2982–2987.
- Sun, Y.L., Zhang, Q., Anastasio, C. and Sun, J. (2010). Insights into secondary organic aerosol formed via aqueous-phase reactions of phenolic compounds based on high resolution mass spectrometry. *Atmos. Chem. Phys.* 10: 4809–4822.
- Takegawa, N., Miyazaki, Y., Kondo, Y., Komazaki, Y., Miyakawa, T., Jimenez, J.L., Jayne, J.T., Worsnop, D.R., Allan, J.D. and Weber, R.J. (2005). Characterization of an Aerodyne Aerosol Mass Spectrometer (AMS): Intercomparison with other aerosol instruments. *Aerosol Sci. Technol.* 39: 760–770.
- Takegawa, N., Miyakawa, T., Watanabe, M., Kondo, Y., Miyazaki, Y., Han, S., Zhao, Y., van Pinxteren, D., Brüggemann, E., Gnauk, T., Herrmann, H., Xiao, R., Deng, Z., Hu, M., Zhu, T. and Zhang, Y. (2009).

- Performance of an Aerodyne Aerosol Mass Spectrometer (AMS) during Intensive Campaigns in China in the Summer of 2006. *Aerosol Sci. Technol.* 43: 189–204.
- Turpin, B.J. and Lim, H.J. (2001). Species Contributions to PM<sub>2.5</sub> mass concentrations: revisiting common assumptions for estimating organic Mass. *Aerosol Sci. Technol.* 35: 602–610.
- Weakley, A.T., Takahama, S. and Dillner, A.M. (2016). Ambient aerosol composition by infrared spectroscopy and partial least-squares in the chemical speciation network: Organic carbon with functional group identification. *Aerosol Sci. Technol.* 50: 1096–1114.
- WHO (2016). Ambient Air Pollution: A Global Assessment of Exposure and Burden of Disease., World Health Organization.
- Zhang, Q., Alfarra, M.R., Worsnop, D.R., Allan, J.D., Coe, H., Canagaratna, M.R. and Jimenez, J.L. (2005). Deconvolution and quantification of hydrocarbon-like and oxygenated organic aerosols based on aerosol mass spectrometry. *Environ. Sci. Technol.* 39: 4938–4952.
- Zhang, Q., Jimenez, J., Canagaratna, M., Allan, J., Coe, H., Ulbrich, I., Alfarra, M., Takami, A., Middlebrook, A., Sun, Y., Dzepina, K., Dunlea, E., Docherty, K., DeCarlo, P., Salcedo, D., Onasch, T., Jayne, J., Miyoshi, T., Shimojo, A., Hatakeyama, S., Takegawa, N., Kondo, Y., Schneider, J., Drewnick, F., Borrmann, S., Weimer, S., Demerjian, K., Williams, P., Bower, K., Bahreini, R., Cottrell, L., Griffin, R., Rautiainen, J., Sun, J., Zhang, Y. and Worsnop, D. (2007). Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere midlatitudes. *Geophys. Res. Lett.* 34: L13801.

*Received for review, January 3, 2017*

*Revised, April 13, 2017*

*Accepted, April 13, 2017*