

Formation of highly oxygenated organic aerosol in the atmosphere: Insights from the Finokalia Aerosol Measurement Experiments

Lea Hildebrandt,¹ Evangelia Kostenidou,² Nikos Mihalopoulos,³ Douglas R. Worsnop,⁴ Neil M. Donahue,¹ and Spyros N. Pandis^{1,2}

Received 20 August 2010; revised 22 September 2010; accepted 27 September 2010; published 3 December 2010.

[1] Aged organic aerosol (OA) was measured at a remote coastal site on the island of Crete, Greece during the Finokalia Aerosol Measurement Experiments (FAME-08 and FAME-09), which were part of the EUCAARI intensive campaigns. Quadrupole aerosol mass spectrometers (Q-AMSS) were employed to measure the size-resolved chemical composition of non-refractory submicron aerosol (NR-PM₁), and to estimate the extent of oxidation of the OA. The experiments provide unique insights into ambient oxidation of aerosol by measuring at the same site but under different photochemical conditions. NR-PM₁ concentrations were about a factor of three lower during FAME-09 (winter) than during FAME-08 (summer). The OA sampled was significantly less oxidized and more variable in composition during the winter than during the early summer. Lower OH concentrations in the winter were the main difference between the two campaigns, suggesting that atmospheric formation of highly oxygenated OA is associated with homogeneous photochemical aging. **Citation:** Hildebrandt, L., E. Kostenidou, N. Mihalopoulos, D. R. Worsnop, N. M. Donahue, and S. N. Pandis (2010), Formation of highly oxygenated organic aerosol in the atmosphere: Insights from the Finokalia Aerosol Measurement Experiments, *Geophys. Res. Lett.*, 37, L23801, doi:10.1029/2010GL045193.

1. Introduction

[2] Submicron atmospheric aerosols have a highly uncertain effect on climate [*Intergovernmental Panel on Climate Change*, 2007], and they adversely affect human health [*Pope and Dockery*, 2006]. Organic aerosol (OA) comprises 20–90% of the global PM₁ mass [*Kanakidou et al.*, 2005], but its formation and evolution remain poorly understood [*Jimenez et al.*, 2009]. After formation or emission, OA can re-evaporate and be further oxidized (“aged”) and transported in the atmosphere [*Robinson et al.*, 2007]. Air-quality models tend to under-predict the concentrations of OA in the atmosphere, especially in the summer when photochemical activity is high [*Volkamer et al.*, 2006; *Goldstein and Galbally*, 2007; *Karydis et al.*, 2007] suggesting that we do not under-

stand the aging of OA and formation of secondary OA (SOA) well.

[3] The complex OA composition prevents speciation on a molecular basis, let alone tracking the chemical evolution of its molecular components. However, aerosol mass spectrometry and factor analysis enable differentiation among several types of OA, mostly distinguished by their oxygen content and thermal volatility [*Jimenez et al.*, 2009]. The resulting OA components include hydrocarbon-like OA (HOA), biomass burning OA (BBOA), and oxygenated OA (OOA), a more oxidized component which dominates the total OA mass in most locations [*Zhang et al.*, 2007]. OOA can frequently be further separated into highly oxygenated OA, now referred to as low-volatility OOA (LV-OOA) and less oxygenated OA, referred to as semi-volatile OOA (SV-OOA) [*Jimenez et al.*, 2009; *Ng et al.*, 2010].

[4] Oxidation reactions after initial OA formation can affect the concentration and composition of OA. Most fresh SOA formed from gaseous precursors in laboratory experiments resembles SV-OOA [*Ng et al.*, 2010]. After additional OH oxidation, the aged SOA can start to resemble LV-OOA. Primary OA formed in laboratory experiments, such as OA from diesel exhaust or woodsmoke, initially resembles HOA or BBOA, but upon aging can resemble SV-OOA [*Jimenez et al.*, 2009]. Thus, SV-OOA can be created from essentially all OA precursors in the laboratory, and source strengths and mechanisms of this moderately oxidized OA can be tested on laboratory timescales. Illuminating the formation of LV-OOA is more challenging. Typical laboratory experiments only access the first few generations of oxidation [*Kroll et al.*, 2009], and hence this highly oxidized OA cannot easily be created in laboratory experiments, especially if the experiments aim to approximate atmospheric concentration levels. Studying aged ambient OA is also difficult since it is usually mixed with fresher OA, especially if the ambient measurements are conducted close to sources. Thus, we need to conduct well-situated ambient measurements to explore the formation of highly oxygenated OA.

[5] The Finokalia Aerosol Measurement Experiments (FAMES) were designed to study ambient OA of different photochemical ages. Finokalia is on the island of Crete, and it is an ideal location to study the aging of OA. The station is isolated and often downwind of European sources [*Koulouri et al.*, 2008] and therefore allows the study of OA from Europe, as well as marine and Saharan aerosol. Also, the photochemical conditions are conducive to high levels of oxidation, especially in the summer [*Berresheim et al.*, 2003].

[6] We conducted two campaigns at this site to measure OA under different conditions. FAME-08 was in the early summer (May 8–June 5, 2008) under strong oxidizing conditions. FAME-09 was in the winter (February 25–March 25,

¹Center for Atmospheric Particle Studies, Carnegie Mellon University, Pittsburgh, Pennsylvania, USA.

²Institute of Chemical Engineering and High Temperature Chemical Processes, Foundation of Research and Technology, Patra, Greece.

³Environmental Chemical Processes Laboratory, University of Crete, Heraklion, Greece.

⁴Aerosol and Cloud Chemistry, Aerodyne Research Inc., Billerica, Massachusetts, USA.

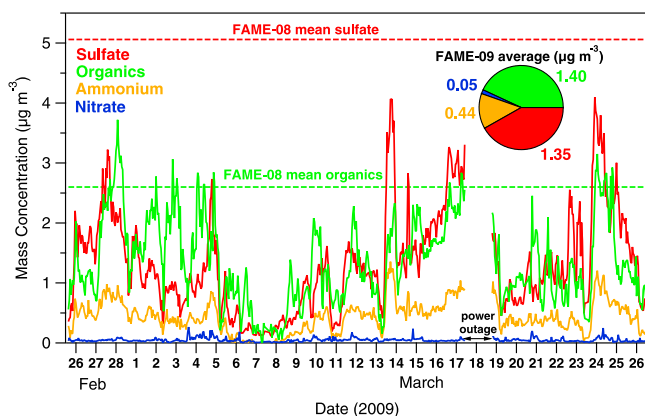


Figure 1. Time series of total mass concentrations of NR-PM₁ components and average concentrations (inset) measured by the AMS during FAME-09, as well as mean values of total OA and sulfate concentrations during FAME-08 (horizontal dashed lines).

2009) under milder oxidizing conditions. The OA measured during FAME-08 has been characterized in detail in a previous publication [Hildebrandt *et al.*, 2010]. The OA observed during FAME-08 was highly oxygenated, resembling LV-OOA throughout the campaign, regardless of the source region. The hypothesis motivating FAME-09 was that wintertime OA would be less oxygenated and more variable in composition than summertime OA.

2. Experimental Procedure

2.1. Measurement Campaign

[7] Measurements were conducted at the Finokalia Station of the Environmental Chemical Processes Laboratory of the University of Crete [Hildebrandt *et al.*, 2010] as part of the EUCAARI intensive campaigns [Kulmala *et al.*, 2009]. Detailed results from FAME-08 have been published [Hildebrandt *et al.*, 2010; Lee *et al.*, 2010; Pikridas *et al.*, 2010], and details on FAME-09 will be presented elsewhere. Here, we focus on the comparison of OA sampled during FAME-08 and FAME-09.

2.2. Instrumentation and Methods

[8] The size-resolved submicron aerosol composition was measured using Q-AMSS from Aerodyne Research, Inc. [Canagaratna *et al.*, 2007]. The particles were sampled at ambient relative humidity. The particle number distribution was monitored using scanning mobility particle sizers (SMPSs) from TSI. A number of other measurements were performed during these campaigns, including PM composition from filter samples and aerosol light scattering and absorption (nephelometers, aethalometers). Meteorological data are also available.

2.3. Q-AMS Measurements

[9] The Q-AMSS alternated operation between mass spectrum (MS) scanning mode and particle time-of-flight (pToF) mode every fifteen seconds. The sample averaging time was set at three minutes, and further averaging was performed in post-analysis. Data were analyzed using a standard AMS fragmentation table and batch table [Allan *et al.*, 2004], with a few modifications. These modifications, as well as details on

the calibration of the AMS, the OA detection limit and uncertainties in the AMS measurements and analysis are discussed in the auxiliary material.¹ In summary, the average collection efficiency of the AMS was 0.85 during FAME-08 and 0.6 during FAME-09, estimated from comparison to SMPS distributions and confirmed by comparison to filter measurements. For the analysis of the organic composition (Section 3.2) we did not include data when $C_{OA} < 0.3 \mu\text{g m}^{-3}$, a reasonable estimate for the OA detection limit in the Q-AMS [e.g., Drewnick *et al.*, 2009]. Importantly, the differences observed in the organic composition cannot be explained by the expected uncertainty and variability in AMS measurements.

[10] We focus on the OA mass spectra from the two measurement campaigns. The fragments at mass-to-charge ratio (m/z) 44 mostly correspond to the CO_2^+ ion and can therefore be used as a semi-empirical measure of the extent of oxidation of the OA. The organic mass fraction at m/z 44, $f_{44} = [m/z\ 44]/C_{OA}$, where C_{OA} is the total mass concentration of the organic aerosol, can be used to estimate the oxygen to carbon ratio, $(\text{O:C})_{44}$, and the organic mass to organic carbon ratio, $(\text{OM:OC})_{44}$, of the OA [Aiken *et al.*, 2008].

3. Results

3.1. Bulk Chemical Composition

[11] Figure 1 shows 1-h averages of the chemical composition of dry NR-PM₁ measured by the AMS during FAME-09, the campaign average contributions of the different aerosol components to the total aerosol mass, and the mean values of organics and sulfate during FAME-08. During March 5–7 the area experienced a Saharan dust event with PM₁₀ concentrations exceeding $1000 \mu\text{g m}^{-3}$. NR-PM₁ concentrations were extremely low during this time period, presumably because the larger Saharan particles scavenge smaller particles by coagulation and low-volatility vapors by condensation. The campaign-average total dry NR-PM₁ concentration measured by the AMS was $3.2 \mu\text{g m}^{-3}$, a factor of three lower than during FAME-08; average total OA was $1.4 \mu\text{g m}^{-3}$, a factor of two lower than during FAME-08 [Hildebrandt *et al.*, 2010]. The bulk composition also differed: during FAME-08 OA comprised 28% of dry NR-PM₁ and the ratio of OA to AMS-Sulfate ($\text{OA}:\text{SO}_4^{2-}$) was approximately 0.5, during FAME-09 OA comprised 43% of dry NR-PM₁ and $\text{OA}:\text{SO}_4^{2-}$ was unity.

3.2. Organic Composition

[12] Figure 2 shows 2-h averages of f_{43} , f_{44} and f_{57} as a function of time during FAME-09. Figure 2 also shows the mean values of f_{43} and f_{44} , as well as the 10th and 90th percentiles of f_{44} during FAME-08. Almost all values of f_{44} during FAME-09 are below the 10th percentile of the FAME-08 values; the OA was significantly less oxidized in the winter (FAME-09) than the early summer (FAME-08). The average f_{44} of 18.2% observed during FAME-08 corresponds to an estimated $(\text{O:C})_{44}$ of 0.8 and $(\text{OM:OC})_{44}$ of 2.2. In comparison, the average f_{44} of 10.6% observed during FAME-09 corresponds to an estimated $(\text{O:C})_{44}$ of 0.5 and $(\text{OM:OC})_{44}$ of 1.8. The organic aerosol was not only less oxidized but also much more variable in composition during FAME-09.

¹Auxiliary materials are available in the HTML. doi:10.1029/2010GI045193.

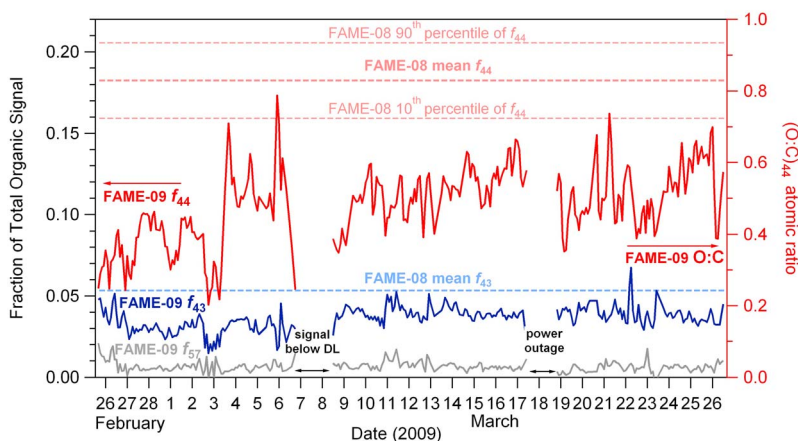


Figure 2. Time series of f_{43} , f_{44} and f_{57} (left vertical axis) and estimated $(\text{O:C})_{44}$ (right vertical axis) during FAME-09 and FAME-08 mean values. The organic aerosol was more variable in composition and on average significantly less oxidized in the winter (FAME-09) than the early summer (FAME-08).

This variability is another indication that the OA during FAME-08 was more aged than during FAME-09. As during FAME-08, low f_{57} (0.6% on average) suggests little or no contribution from fresh, non-oxidized OA, consistent with the remoteness of the site.

4. Discussion

[13] A meaningful comparison of the OA composition during FAME-08 and FAME-09 requires an understanding of the similarities and differences in the aerosol sources and ambient conditions during the campaigns. Based on the footprint residence time plots from the FLEXPART model [Pikridas *et al.*, 2010], the source regions of the aerosol sampled over the course of the two study periods were comparable. In particular, during FAME-09, the source region was categorized as marine for 61% of the campaign (38% in FAME-2008), as Saharan for 7% of the campaign (9% in FAME-08) and as continental for 32% of the campaign (53% in FAME-08). Based on the somewhat higher abundance of marine and lower abundance of continental air masses during FAME-09, we would expect OA to be more oxidized (aged) during FAME-09 than during FAME-08; however, we observe the opposite. We also stress that, during FAME-08, the aerosol from all sources was consistently highly oxygenated. Thus the lower oxidation and increased variability in composition of the OA during FAME-09 were likely not due to different source regions.

[14] Since the two campaigns were conducted during different seasons, there were differences in the emissions. Biogenic VOC emissions are temperature dependent and higher in the summer than in the winter. Based on emission inventories [Visschedijk *et al.*, 2007], total isoprene emissions in Europe were on average nine times higher during FAME-08 than during FAME-09, and total terpene emissions were six times higher. The isoprene and terpene emissions in Greece had similar ratios (C. Pilinis, unpublished results, 2010). However, preliminary model results using the PMCAMx-2008 model [Murphy and Pandis, 2009] suggest that the contribution of biogenic SOA to total OA sampled at Finokalia were similar during the two campaigns (about 50%) (C. Fountoukis, unpublished results, 2010). Thus, the lower

biogenic VOC emissions during the winter cannot explain the difference in OA composition between the two campaigns. Differences in the emissions of other compounds were smaller. For example, total European CO, NO and NO₂ emissions were on average 10% higher during FAME-09 than during FAME-08, SO₂ emissions were 26% higher, and aromatic VOC emissions were similar (C. Pilinis, unpublished results, 2010). Therefore, the campaign differences were probably not mainly due to different emissions, nor different source regions, but rather due to different meteorological and photochemical conditions.

[15] Photochemical conditions were milder during FAME-09. We approximate ambient OH concentrations using measures of UV intensity (approximated by the photolysis rate of NO₂, j_{NO_2}) and O₃ concentrations. The UV intensity during FAME-09 was lower and more variable than during FAME-08 (Table S1 of the auxiliary material). Using the correlations developed by Berresheim *et al.* [2003], the approximate mean midday OH concentrations were $12 \times 10^6 \text{ cm}^{-3}$ during FAME-08 and $5.5 \times 10^6 \text{ cm}^{-3}$ during FAME-09. The Berresheim correlations are derived from summer data and hence they can only provide an upper limit for FAME-09 (the slant path is higher in the winter) and a better estimate for FAME-08. Ozone concentrations were lower in the winter (FAME-09 average = 44.6 ppb) than in the summer (FAME-08 average = 53.2 ppb). Since O₃ is a source of OH, we infer that OH concentrations were on average at least a factor of two (and possibly much more) lower during FAME-09. The significantly lower OH concentrations could explain more variable composition and lower average extent of oxidation of OA in the winter. Considering that ambient conditions were more humid during FAME-09 (mean RH = 73%) than during FAME-08 (mean RH = 60%), and that f_{44} was not significantly correlated with inorganic acidity (FAME-08 $R^2 = 0.065$, FAME-09 $R^2 = 0.015$), it is unlikely that aqueous or heterogeneous processing played a dominant role in the formation of highly oxygenated OA during FAME-08. Furthermore, the time scale of heterogeneous OH oxidation is approximately two weeks, using the simple mass balance suggested by Robinson *et al.* [2006], the FAME-08 average particle size (300 nm) and diurnal-average OH concentration ($2 \times 10^6 \text{ cm}^{-3}$). Thus, heterogeneous aging appears too slow,

and most of the oxidation was probably homogeneous. We estimate that the OH exposure needed for the OA to reach the highly oxidized state observed during FAME-08 is approximately 4×10^{11} molecules cm^{-3} s [Hildebrandt et al., 2010], which is about ten times less than the exposure required for heterogeneous production of LV-OOA [Jimenez et al., 2009]. The average ambient temperature was about 9 K higher during FAME-08 than during FAME-09 (Table S1). Using the volatility distribution from FAME-08 [Lee et al., 2010], we estimate that there was about 30% more OA with effective saturation concentration $\leq 10 \mu\text{g m}^{-3}$ in the gas phase during FAME-08, which is expected to accelerate the gas-phase oxidation.

[16] In order to further explore difference in the meteorological conditions during the two campaigns, we used data from the European Centre for Medium-Range Weather Forecasts (<http://www.ecmwf.int/research/era/do/get/era-interim>). When considering only the grid cell containing the field site (Figure S1), average low cloud cover was 27 times higher during FAME-09 than during FAME-08 (Table S1). The average low cloud cover in a much larger domain including most of Europe was more than twice as high during FAME-09 (Table S1 and Figure S2). Thus, photochemical conditions were milder during FAME-09 – not just at the field site, but also during transport to the site.

[17] The difference in $\text{OA}:\text{SO}_4^{2-}$ during the two study periods could also be due to the different photochemical conditions, which can affect the concentrations of SO_4^{2-} and OA. Previous work has shown that OH oxidation of SO_2 is the main source of submicron SO_4^{2-} in the area [Mihalopoulos et al., 2007]. During FAME-08, SO_2 concentrations and the mass ratio of SO_2 to sulfate were low (average $\text{SO}_2:\text{SO}_4^{2-}$ was 0.06), suggesting that SO_2 was almost completely oxidized to sulfate before it reached the measurement site. This was not the case under the milder oxidizing conditions during FAME-09, when the average $\text{SO}_2:\text{SO}_4^{2-}$ was 0.7. Furthermore, the higher extent of oxidation of the FAME-08 OA (section 3.2) suggests that fragmentation reactions were probably more significant during FAME-08 [Kroll et al., 2009]. Fragmentation reactions and increased evaporation due to higher temperatures can decrease the concentration of OA and therefore cause a lower $\text{OA}:\text{SO}_4^{2-}$ during FAME-08.

5. Implications

[18] Organic aerosol was less oxidized and its composition was more variable under milder photochemical conditions in the winter (FAME-09) than it was in the summer (FAME-08). Since the main difference between the OA sampled during FAME-09 and FAME-08 is the more extreme photochemical conditions in the summer, we hypothesize that homogeneous photochemical aging drives the transformation from younger and more variable OA (FAME-09) to older, more uniform and highly oxygenated OA (FAME-08).

[19] The range and mean values of the summer and winter OA support the hypothesis that the OOA factors emerging from PMF data analysis do not represent distinct aerosol populations but rather the lower and upper limits bounding the range of variation in degree of oxidation observed during a given campaign. Ng et al. [2010] showed that the two OOA factors from various field campaigns show progressively rising amounts of oxidation correlating with expected photochemical age. Hildebrandt et al. [2010] showed that the FAME-08

(summer) factors *both* fell well within the historical LV-OOA range and that the factors showed a clear diurnal variation consistent with daytime oxidation. In the winter (FAME-09) we see much greater variability and lower mean degree of oxidation with much lower mean photochemical age.

[20] **Acknowledgments.** The authors gratefully acknowledge Petri Titta and Ari Laaksonen, University of Kuopio, for loaning their Q-AMS during FAME-09, as well as Claudia Mohr, Peter F. DeCarlo, Andre S. H. Prevot and Urs Baltensperger, Paul Scherrer Institut, for loaning and assisting with their Q-AMS during FAME-08. This research was supported by the US National Science Foundation (ATM-0732598) and the FP6 project EUCAARI – European Integrated Project on Aerosol Cloud Climate and Air Quality Interactions. Lea Hildebrandt was supported by a National Science Foundation Graduate Research Fellowship.

References

- Aiken, A. C., et al. (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, doi:10.1021/es703009q.
- Allan, J. D., et al. (2004), A generalised method for the extraction of chemically resolved mass spectra from aerodyne aerosol mass spectrometer data, *J. Aerosol Sci.*, *35*, 909–922, doi:10.1016/j.jaerosci.2004.02.007.
- Berresheim, H., et al. (2003), OH in the coastal boundary layer of Crete during MINOS: Measurements and relationship with ozone photolysis, *Atmos. Chem. Phys.*, *3*, 639–649, doi:10.5194/acp-3-639-2003.
- Canagaratna, M. R., et al. (2007), Chemical and microphysical characterization of ambient aerosols with the aerodyne aerosol mass spectrometer, *Mass Spectrom. Rev.*, *26*, 185–222, doi:10.1002/mas.20115.
- Drewnick, F., et al. (2009), Aerosol quantification with the Aerodyne Aerosol Mass Spectrometer: Detection limits and ionizer background effects, *Atmos. Meas. Tech.*, *2*, 33–46, doi:10.5194/amt-2-33-2009.
- Goldstein, A. H., and I. E. Galbally (2007), Known and unexplored organic constituents in the Earth's atmosphere, *Environ. Sci. Technol.*, *41*, 1514–1521, doi:10.1021/es072476p.
- Hildebrandt, L., et al. (2010), Aged organic aerosol in the eastern Mediterranean: The Finokalia Aerosol Measurement Experiment—2008, *Atmos. Chem. Phys.*, *10*, 4167–4186, doi:10.5194/acp-10-4167-2010.
- Intergovernmental Panel on Climate Change (2007), *Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change*, edited by S. Solomon et al., Cambridge Univ. Press, Cambridge, U. K.
- Jimenez, J. L., et al. (2009), Evolution of organic aerosol in the atmosphere, *Science*, *326*, 1525–1529, doi:10.1126/science.1180353.
- Kanakidou, M., et al. (2005), Organic aerosol and global climate modeling: A review, *Atmos. Chem. Phys.*, *5*, 1053–1123, doi:10.5194/acp-5-1053-2005.
- Karydis, V. A., A. P. Tsimpidi, and S. N. Pandis (2007), Evaluation of a three-dimensional chemical transport model (PMCAMx) in the eastern United States for all four seasons, *J. Geophys. Res.*, *112*, D14211, doi:10.1029/2006JD007890.
- Koulouri, E., et al. (2008), Chemical composition and sources of fine and coarse aerosol particles in the eastern Mediterranean, *Atmos. Environ.*, *42*(26), 6542–6550, doi:10.1016/j.atmosenv.2008.04.010.
- Kroll, J. H., et al. (2009), Measurement of fragmentation and functionalization pathways in the heterogeneous oxidation of oxidized organic aerosol, *Phys. Chem. Chem. Phys.*, *11*, 8005–8014, doi:10.1039/b905289e.
- Kulmala, M., et al. (2009), Introduction: European Integrated Project on Aerosol Cloud Climate and Air Quality Interactions (EUCAARI)—Integrating aerosol research from nano to global scales, *Atmos. Chem. Phys.*, *9*, 2825–2841, doi:10.5194/acp-9-2825-2009.
- Lee, B.-H., et al. (2010), Volatility of organic aerosol sampled during FAME-2008, *Atmos. Chem. Phys. Discuss.*, *10*, 17,435–17,466, doi:10.5194/acpd-10-17435-2010.
- Mihalopoulos, N., et al. (2007), Formation of particulate sulfur species (sulfate and methanesulfonate) during summer over the Eastern Mediterranean: A modelling approach, *Atmos. Environ.*, *41*, 6860–6871, doi:10.1016/j.atmosenv.2007.04.039.
- Murphy, B. N., and S. N. Pandis (2009), Simulating the formation of semi-volatile primary and secondary organic aerosol in a regional chemical transport model, *Environ. Sci. Technol.*, *43*(13), 4722–4728, doi:10.1021/es803168a.
- Ng, N. L., et al. (2010), Organic aerosol components observed in worldwide datasets from aerosol mass spectrometry, *Atmos. Chem. Phys.*, *10*, 4625–4641, doi:10.5194/acp-10-4625-2010.

- Pikridas, M., et al. (2010), The Finokalia Aerosol Measurement Experiments–2008 (FAME-08): An Overview, *Atmos. Chem. Phys.*, *10*, 6793–6806, doi:10.5194/acp-10-6793-2010.
- Pope, C. A., and D. W. Dockery (2006), Health effects of fine particulate air pollution: Lines that connect, *J. Air Waste Manage.*, *56*, 709–742.
- Robinson, A. L., N. M. Donahue, and W. F. Rogge (2006), Photochemical oxidation and changes in molecular composition of organic aerosol in the regional context, *J. Geophys. Res.*, *111*, D03302, doi:10.1029/2005JD006265.
- Robinson, A. L., et al. (2007), Rethinking organic aerosols: Semivolatile emissions and photochemical aging, *Science*, *315*, 1259–1262, doi:10.1126/science.1133061.
- Visschedijk, A. J. H., et al. (2007), *A High Resolution Gridded European Emission Database for the EU Integrated Project GEMS*, Organ. for Appl. Sci. Res., Delft, Netherlands.
- Volkamer, R., J. L. Jimenez, F. San Martini, K. Dzepina, Q. Zhang, D. Salcedo, L. T. Molina, D. R. Worsnop, and M. J. Molina (2006), Secondary organic aerosol formation from anthropogenic air pollution: Rapid and higher than expected, *Geophys. Res. Lett.*, *33*, L17811, doi:10.1029/2006GL026899.
- Zhang, Q., et al. (2007), Ubiquity and dominance of oxygenated species in organic aerosols in anthropogenically-influenced Northern Hemisphere mid-latitudes, *Geophys. Res. Lett.*, *34*, L13801, doi:10.1029/2007GL029979.
-
- N. M. Donahue, L. Hildebrandt, and S. N. Pandis, Center for Atmospheric Particle Studies, Carnegie Mellon University, Doherty Hall, 5000 Forbes Ave., Pittsburgh, PA 15213, USA. (spyros@andrew.cmu.edu)
- E. Kostenidou, Institute of Chemical Engineering and High Temperature Chemical Processes, Foundation of Research and Technology, PO Box 1414, GR-26504 Patra, Greece.
- N. Mihalopoulos, Environmental Chemical Processes Laboratory, University of Crete, PO Box 2208, GR-71003 Heraklion, Greece.
- D. R. Worsnop, Aerosol and Cloud Chemistry, Aerodyne Research Inc., 45 Manning Rd., Billerica, MA 01821, USA.