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## **Clean Air for London (CLEARFLO) Final Campaign Summary**

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## **Clean Air for London (CLEARFLO) Final Campaign Summary: Detling, UK**

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## Summary

This field campaign funded the participation of scientists from seven different research groups and operated over thirty instruments during the Winter Intensive Operating Period (January-February 2012) of the Clean Air for London (ClearfLo) campaign. The campaign took place at a rural site in Detling, UK, 45 kilometers southeast of central London. The primary science questions for the ClearfLo winter IOP (intensive operational periods) were: 1) “what is the urban increment of particulate matter (PM) and other pollutants in the greater London area?” and 2) “what is the contribution of solid fuel use for home heating to wintertime PM?” An additional motivation for the Detling measurements was the question of whether coatings on black carbon particles enhance absorption.

The following four key accomplishments have been identified thus far:

1. Chemical, physical, and optical characterizations of PM have been identified from local and regional sources as outlined in Figures 1, 2, 3 and 5.
2. Measurements have been obtained of urban increments in particulate matter and gases in London, as illustrated in Figure 4.
3. Optical properties and chemical composition measurements from coatings on black carbon containing particles indicates absorption enhancement.
4. The first deployment of a chemical ionization instrument (multiple orifice volatilization impactor chemical ionization time of flight mass spectrometer [MOVI-CI-TOFMS]) was made in order to measure both particle-phase and gas-phase nitrated aromatics, as shown in Figure 5.

Analysis of the large data set acquired in Detling is ongoing and will yield further key accomplishments as time goes on. Measurements of urban and rural aerosol properties will contribute to improved models of regional aerosol emissions and atmospheric aging and removal. Measurements of absorption enhancement caused by black carbon coatings will contribute to improved models of the direct radiative properties of PM.

## Acronyms and Abbreviations

|               |   |
|---------------|---|
| AMF           | ARM Mobile Facility   |
| ANL           | Argonne National Laboratory   |
| ARI           | Aerodyne Research, Inc.   |
| ARM           | Atmospheric Radiation Measurement   |
| BC            | black carbon  |
| CalNex/CARES  | California Nexus/Carbonaceous Aerosols and Radiative Effects Study                            |
| ClearfLo      | Clean Air for London  |
| GVAX          | Ganges Valley Aerosol Experiment  |
| HR-ToF-AMS    | high resolution time of flight aerosol mass spectrometer                                      |
| IOP           | intensive operational period  |
| LANL          | Los Alamos National Laboratory  |
| MAAP          | multi-angle absorption photometer   |
| MOVI-CI-TOFMS | multiple orifice volatilization impactor chemical ionization time of flight mass spectrometer |
| NA            | nitrated aromatics  |
| NERC          | UK Natural Environment Research Council   |
| NK            | North Kensington, UK  |
| PASS          | photo-acoustic soot spectrometer  |
| PM            | particulate matter  |
| PM1           | particulate matter (less than one micron in diameter)   |
| PTR-MS        | proton transfer reaction mass spectrometer  |
| PSI           | Paul Scherrer Institute   |
| SOA           | secondary organic aerosol   |
| SP-AMS        | soot particle aerosol mass spectrometer   |
| UV            | ultra violet  |

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## 1.0 Background

This field campaign was originally funded as a supersite in Pantnagar, India. The purpose of the location was to complement the cloud and aerosol measurements being collected by the Atmospheric Radiation Measurement (ARM) Climate Research Facility's ARM Mobile Facility (AMF) already situated at the Nainital Observatory during the Ganges Valley Aerosol Experiment (GVAX). However, the Pantnagar site campaign was cancelled in late November, 2011, shortly before the scheduled shipment of instruments to India. Instead, the field campaign was approved to participate in the Clean Air for London (ClearfLo) campaign in January and February, 2012. ClearfLo was a large, multidisciplinary study of the London urban atmosphere; aimed at understanding the relationships between surface meteorology, gas-phase composition, and particulate matter. To assess these relationships, measurements were obtained at three separate locations: a city street site, a city background site (away from local traffic sources), and a rural location (which sampled the outflow from the London megacity). In addition to year-long measurements, two IOPs occurred during January-February and July-August, 2012. The winter time studies focused on gas and particle emissions from solid fuels used for domestic heating, as well as other sources of local and regional air pollution. The entire ClearfLo field campaign was coordinated by the UK National Centre for Atmospheric Science with support from the UK Natural Environment Research Council (NERC).

The Detling, UK, site was a multi-institutional effort that occurred in a rural location, 45 km southeast of central London. It deployed a suite of instruments that measured particulate chemical composition, (including organic, inorganic, black carbon, optical, and hygroscopic properties), and calculated size distributions, gas-phase oxidant precursors, and aerosol precursors. The Detling site was partially supported by NERC funding. A particular focus of this study was to understand how black carbon particles are processed. Results from the CalNex/CARES campaign in 2010 suggested that current scientific understandings of the morphology of coated black carbon particles is incomplete, so this study attempted to provide more insight by serving as another test case.

Douglas Worsnop, Leah Williams, and Scott Herndon from Aerodyne Research, Inc. (ARI) provided instruments to monitor aerosol chemical composition, (including organics, inorganics, and black carbon), extinction, and single-scattering albedo. ARI also deployed instruments to obtain state-of-the-art measurements of trace gas species. Nga Lee Ng from the Georgia Institute of Technology provided high resolution real-time measurements of non-refractory PM<sub>1</sub> (particulate matter less than 1 micron in diameter) chemical composition elements. Joel Thornton at the University of Washington deployed a recently developed, high-sensitivity mass spectrometer system (MOVI-CI-TOFMS) capable of near real-time measurements of molecular level gas and particle-phase organics composition. W Berk Knighton from Montana State University deployed a proton transfer reaction mass spectrometer (PTR-MS) to measure gas-phase volatile organic compounds. Manvendra Dubey from Los Alamos National Laboratory (LANL) provided measurements of in-situ aerosol optical properties and size distributions. Andre Prévôt from the Paul Scherrer Institute (PSI) in Switzerland made filter and rotating drum impactor measurements of carbonaceous and elemental particles, as an unfunded collaborator. Richard Coulter from Argonne National Laboratory (ANL) provided remote sensing and meteorology measurements. Table 1 provides a complete list of instruments that successfully collected data at the Detling site.



**Table 1.** Instrument List for ClearfLo Detling, UK Site.

| Institute / Investigator | Instrument                     | Species/Parameters Measured  |
|--------------------------|--------------------------------|--|
| ARI / S. Herndon         | 2B Tech                        | O <sub>3</sub>   |
| ARI / S. Herndon         | Thermo 42i                     | NO   |
| ARI / S. Herndon         | ARI CAPS                       | NO <sub>2</sub>  |
| ARI / S. Herndon         | Licor                          | CO <sub>2</sub>  |
| ARI / S. Herndon         | ARI QCL                        | CO, N <sub>2</sub> O   |
| ARI / S. Herndon         | ARI QCL                        | NO <sub>2</sub>  |
| CEH / D. Famulari        | ARI mini-QCL                   | N <sub>2</sub> O, CO <sub>2</sub> , H <sub>2</sub> O   |
| LANL / M. Dubey          | Picarro CRDS                   | CH <sub>4</sub> , CO <sub>2</sub> , H <sub>2</sub> O   |
| ARI / S. Herndon         | GC-FID                         | Gas-phase VOC  |
| MSU / W. B. Knighton     | PTR-MS                         | Gas-phase VOC  |
| UW / C. Mohr             | ARI MOVI-CI-TOFMS              | Gas and particle-phase organic acids   |
| GIT / N. L. Ng           | ARI HR-ToF-AMS                 | Non-refractory, submicron particulate (SO <sub>4</sub> =/NO <sub>3</sub> -/Cl-/NH <sub>4</sub> +/Organics) |
| ARI / L. Williams        | ARI SP-AMS                     | Non-refractory submicron particulate + black carbon  |
| PSI / S. Visser          | 3-stage Rotating Drum Impactor | Trace elements in PM <sub>1.0</sub> , PM <sub>2.5</sub> and PM <sub>10</sub>                               |
| PSI / P. Zotter          | Hi Vol Sampler                 | <sup>14</sup> C in TC, EC/OC concentrations  |
| LANL / A Aiken           | DMT SP-2                       | Black carbon number, mass loading and coating thicknesses  |
| PSI / P Zotter           | Magee AE31 Aethalometer        | 7 wavelength aerosol optical absorption and equivalent black carbon  |
| ARI / L Williams         | Thermo MAAP                    | Aerosol optical absorption and equivalent black carbon   |
| LANL / A Aiken           | DMT PASS-3                     | 3 wavelength aerosol optical absorption, scattering  |
| ARI / P Massoli          | ARI CAPS PMex 630              | Aerosol extinction at 630 nm   |
| LANL / A Aiken           | ARI CAPS PMex 450              | Aerosol extinction at 450 nm   |
| LANL / A Aiken           | TSI Laser Particle Sizer 3340  | Particle size and number (0.07-10 micrometer)  |
| LANL / A Aiken           | TSI SMPS                       | Particle size and number (8-600 nm)  |
| ARI / L Williams         | Portamet                       | T, RH, wind speed and direction  |
| ANL / R Coulter          | Vaisala WXT520 Weather Station | T, RH, wind speed and direction, precipitation   |
| ANL / R Coulter          | Micro Pulse Lidar              | Cloud base, aerosol extinction via backscatter   |
| ANL / R Coulter          | Pyranometer SPN-1              | Total, diffuse radiation   |
| ANL / R Coulter          | SODAR                          | Wind field up to 400 m   |
| ANL / R Coulter          | MFRSR                          | Radiance, 7 wavelengths  |

## 2.0 Notable Events or Highlights

The four key accomplishments listed in the executive summary are discussed in more detail in section 4.0. Overall, the campaign was successful. The majority of deployed instruments collected usable data over 90% of the time.

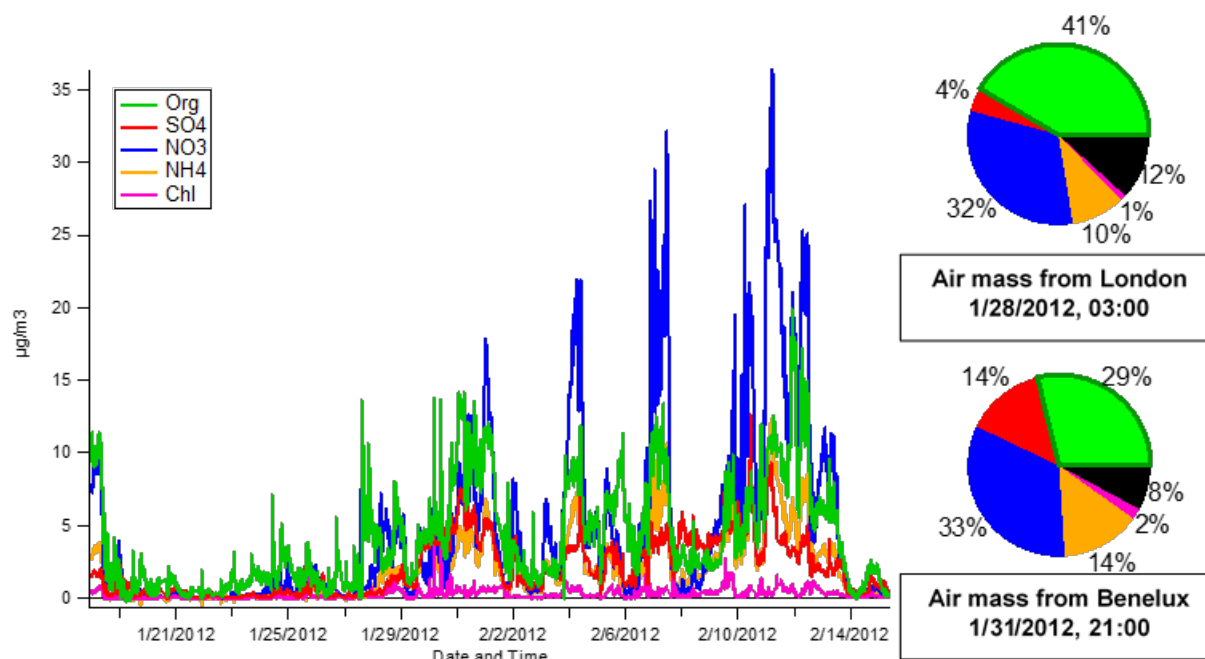
## 3.0 Lessons Learned

The major issue with this campaign was the sudden change in field site location. The abrupt termination of the GVAX-Pantnagar site was unfortunate and wasted a significant amount of time spent in preparation for India. Hopefully, the political groundwork will be performed more carefully before any deployments to foreign countries are arranged in the future. On the other hand, the opportunity to deploy to ClearfLo, instead, was very much appreciated. The staff members at the Field Operations and Deployment office at LANL, particularly Amon Haruta, were very helpful in arranging the instrument shipments to a new location, on very short notice.

A second issue was the lack of any useful data from several instruments (the cloud condensation nuclei counter borrowed from Brookhaven National Laboratory, and the sunset semi-continuous OC/EC and medium volume filter sampler borrowed from the University of Wisconsin). No one on site possessed first-hand experience in operating these three particular instruments and there was no opportunity to use them prior to the field campaign. Thus, the lesson learned is to test all instruments in the lab before deploying them in the field.

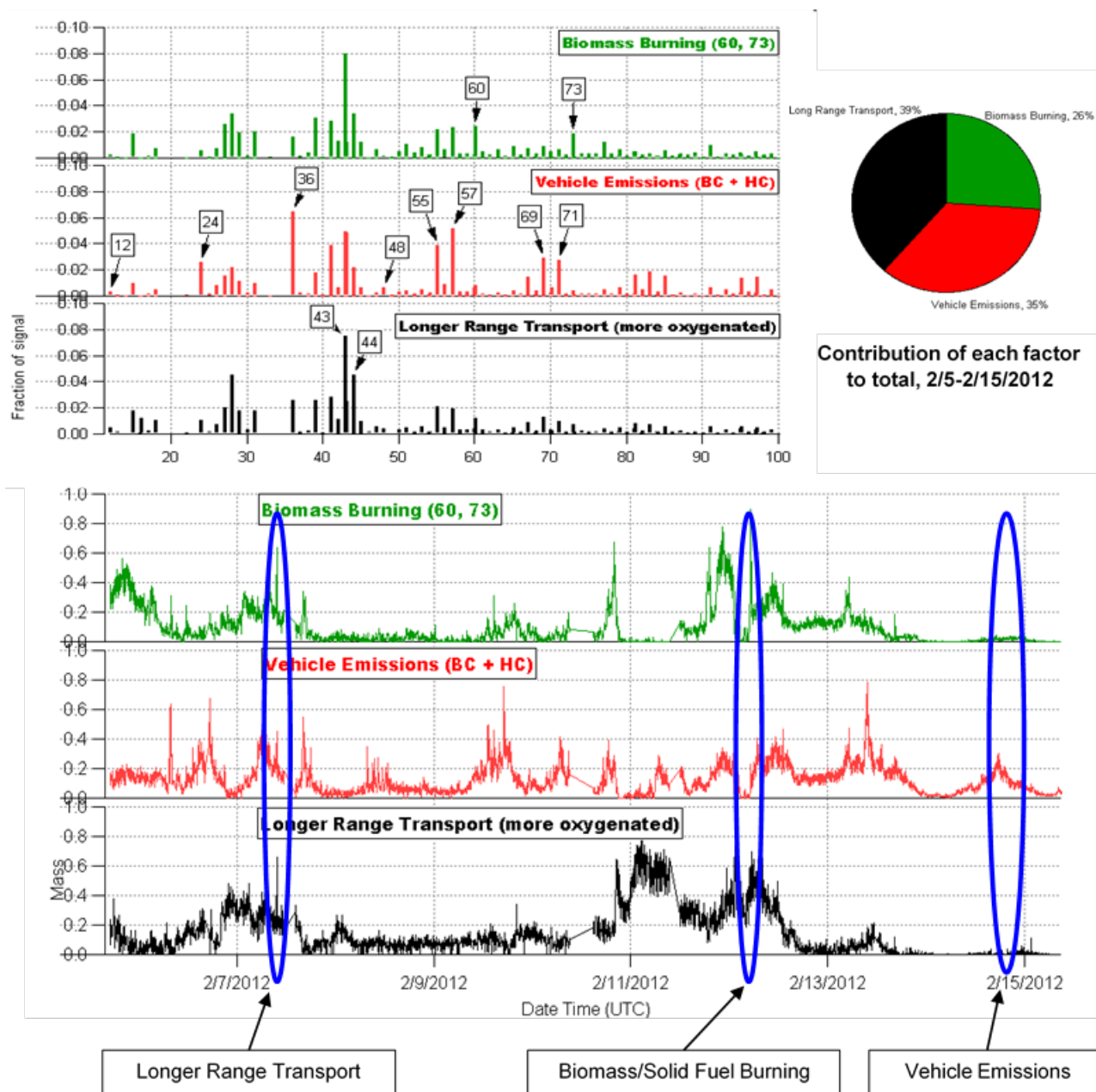
## 4.0 Results

This section discusses the four key accomplishments at Detling, which are listed in the executive summary. The first was the chemical, physical, and optical characterization of PM from local and regional sources. The high resolution time of flight aerosol mass spectrometer (HR-ToF-AMS) measured the chemical composition and size distribution of non-refractory PM<sub>1</sub>. One highlight was the observation of high nitrate levels during time periods when back-trajectories illustrated that air masses originated from polluted regions of northern Europe or the London metropolitan area. Figure 1 shows the data for the HR-ToF-AMS. The left panel demonstrates the time series for mass loading, according to chemical species; each of which is differentiated by a separate color. Green indicates organics, red indicates sulfate, blue indicates nitrate, yellow indicates ammonia, and pink indicates chloride. Variations in mass loading correlate with back-trajectories that illustrate differences in origination sources for sampled air masses. The fractional composition is shown on the right for an air mass arriving from London on January 28, 2012, and for an air mass arriving from northern Europe (Benelux region) on January 31, 2012. The pie charts include the HR-ToF-AMS measurements of non-refractory PM<sub>1</sub> and the measurement of black carbon obtained from the multi-angle absorption photometer (MAAP). The high nitrate levels are consistent with observations from the Mexico City metropolitan area during MILAGRO/MCMA 2006.



**Figure 1.** *Left panel:* HR-ToF-AMS time series for non-refractory particle mass loading of organics (green), nitrate (blue), sulfate (red), ammonia (yellow) and chloride (pink). *Right panel:* Fractional composition of PM1 from HR-ToF-AMS (same colors as left panel) plus MAAP for black carbon (black) for air mass from London (top) and Benelux (bottom).

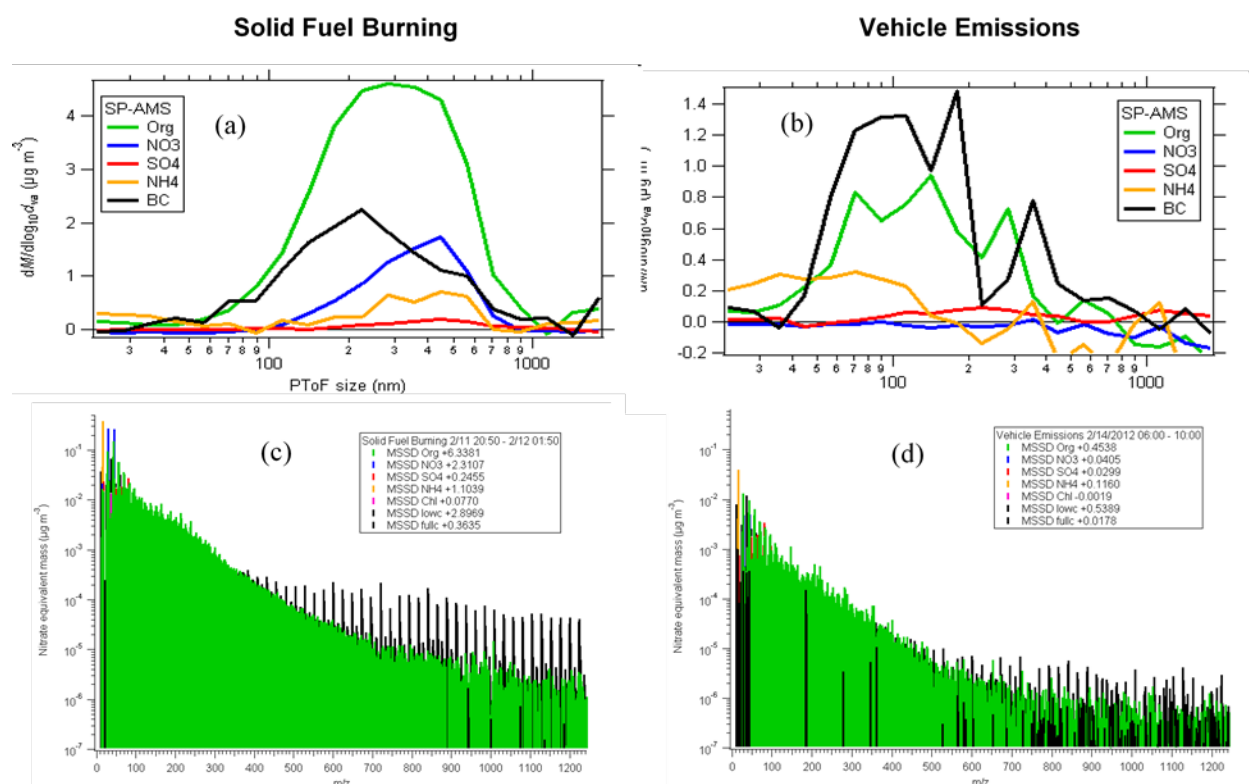
A second highlight of this data analysis was the separation of organic coatings on black carbon particles into three chemical factors associated with vehicle emissions, wood burning, and long range transport. The soot particle aerosol mass spectrometer (SP-AMS) deployed in Detling had both a thermal vaporizer to detect non-refractory PM1 (same vaporization method as the HR-ToF-AMS), and a laser vaporization module to identify black carbon. During the last ten days of the campaign (February 5 through 15, 2012), the thermal vaporizer was removed from the SP-AMS. Thus, in this configuration, the SP-AMS detected only black carbon containing particles. The laser vaporization process, followed by electron impact ionization, provided quantitative measurements of both black carbon and any non-refractory particle coatings. The bottom panel of Figure 2 shows the time series for the three positive matrix factorization elements. These time trends can be compared with air mass back-trajectories and co-located measurements of gas-phase species and aerosol optical properties to identify their sources. It is clear from the time trends that most time periods have contributions from all three factors. Thus, source apportionment should be attributed with caution.



**Figure 2.** *Top left:* Mass spectra of three factors derived by positive matrix factorization of the organic coatings on black carbon containing particles measured with the SP-AMS. *Top right:* contribution of each factor to total non-refractory coating on BC particles. *Bottom panel:* Time series of each factor. Circled regions indicate time periods dominated by one of the factors for comparison with other measurements.

Figure 3 compares the chemically resolved size distributions and mass spectra for black carbon containing particles and associated non-refractory coatings. They highlight two time periods; the first dominated by solid fuel burning (panels a and c) and the second dominated by vehicle emissions (panels b and d). The solid burning time period was associated with stagnant winds, a size distribution peaking at 200 to 300 nm, and the influx of particles containing high levels of organic and black carbon elements. Also, note the presence of fullerenes. This may be a chemical fingerprint for solid fuel/biomass-burning, which

will assist with source appointment. The vehicle emission time period, conversely, was associated with morning rush hour. Winds originated from the southeast and blew across a large roadway adjacent to the sampling site. The size distribution peaked at less than 100 nm and the chemical composition was dominated by black carbon. In this mass spectrum, the high  $m/z$  fullerene peaks are much smaller, relative to the noise level.

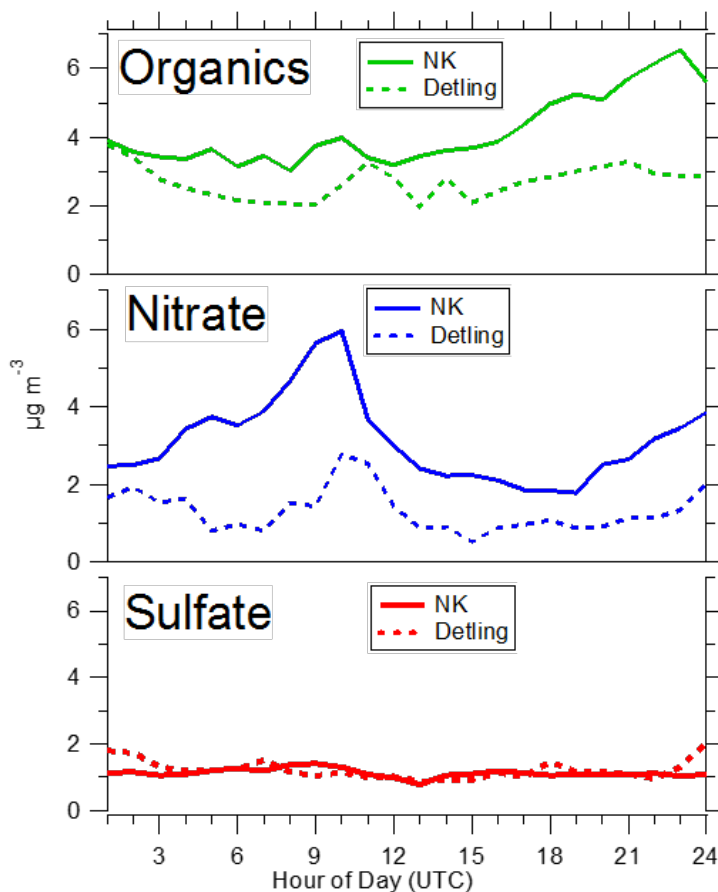


**Figure 3.** Chemically resolved size distributions and mass spectra for black carbon containing PM<sub>1</sub> using the SP-AMS for a time period dominated by solid fuel burning (panels a and c) and a time period dominated by vehicle emissions (panels b and d).

The second key accomplishment was the observation of an urban incremental increase in PM mass loading between London and Detling. Because Detling is located about 45 km southeast of downtown London, it represents the rural background. Figure 4 compares the hourly average organic mass loading (top panel), nitrate mass loading (center panel), and sulfate mass loading (bottom panel) between the urban location at North Kensington (NK) in London and the rural site at Detling. The sulfate mass loading is the same at the two locations and shows no diurnal variation, as is typical of a regional pollutant. In contrast, both organics and nitrate are higher at NK than at Detling and show variation throughout the day, as is typical for PM derived from local sources. The nitrate peak in the morning is due to contributions from rush hour traffic, while the organics peak at night is due to contributions from home heating using solid fuels.

More detailed analysis is underway to compare the PM composition at Detling with that measured in London. For time periods when air masses originated in Europe, passed over Detling and then London, this comparison will address how much of the urban PM originated in London and how much was

regionally influenced. For time periods when air masses passed over London and then over Detling, this comparison will address how urban aerosol was processed and how urban PM contributed to regional air quality.

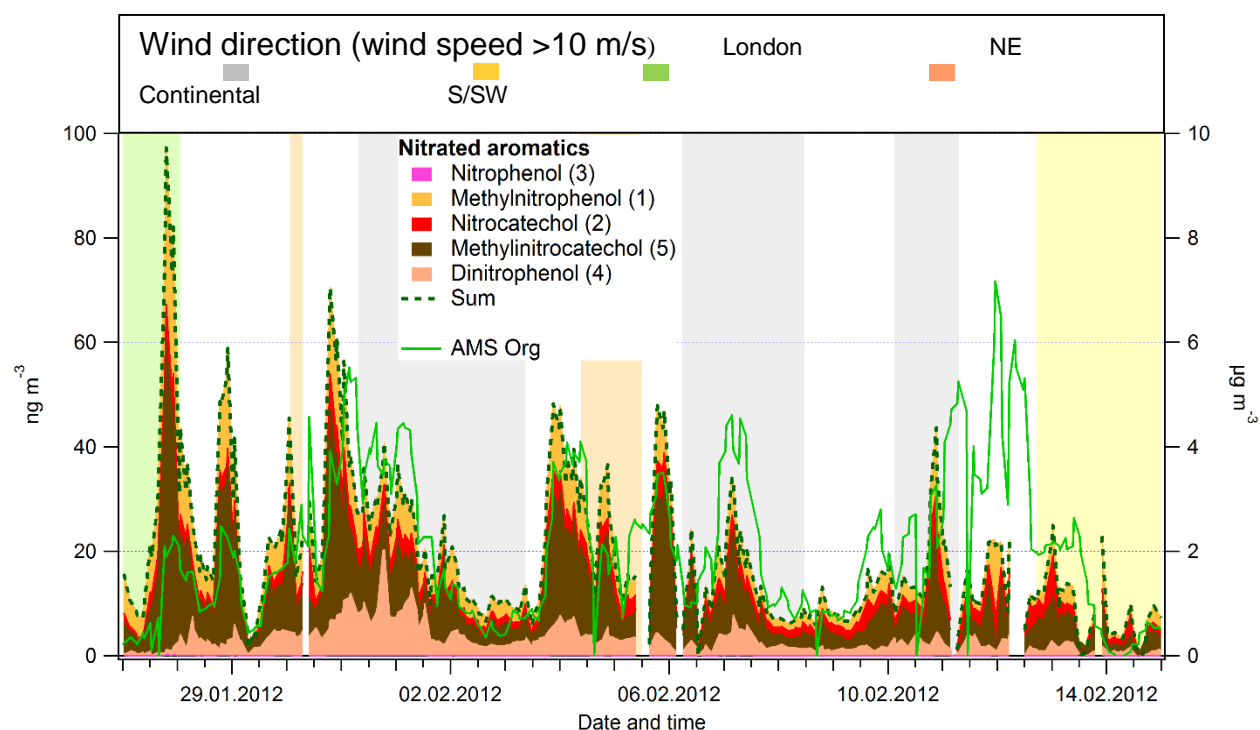


**Figure 4.** *Top panel:* Organic mass loading at North Kensington (NK) and Detling. *Center panel:* Nitrate mass loading at NK and Detling. *Bottom panel:* Sulfate mass loading at NK and Detling.

The third key accomplishment was the observation of absorption enhancement from coatings on black carbon containing particles. Many of the instruments were located downstream of a thermal denuder; which removes volatile material from ambient PM. The thermal denuder data contributed to the SP-AMS measurements of black carbon coatings and the photo-acoustic soot spectrometer (PASS) measurements of optical absorption, which investigated the effect of coatings on optical properties. In particular, absorption enhancement was searched for in this data set to compare with data sets from biomass-burning plumes (where absorption enhancement was previously observed). It was also compared with data from CalNex/CARES (where absorption enhancement was not observed). Certain time periods have been found to be correlated with significant absorption enhancement, and the amount of coating material. A manuscript detailing these findings has been submitted to *Nature*.

The fourth key accomplishment was the first field deployment of a chemical ionization mass spectrometer; designed to measure both gas-phase and particle-phase organics. In the near-ultraviolet (UV) range of the electromagnetic spectrum, light-absorbing organic matter, termed “brown carbon,” can

be an important absorbing component of ambient PM. Brown carbon appears to be especially significant in biomass/biofuel combustion emissions, yet specific light-absorbing compounds remain largely unidentified. Among the high number of poorly constrained brown carbon compounds are nitrated aromatics (NA). Figure 5 shows measurements of five different NA compounds in the particle-phase, by the name of MOVI-CI-TOFMS. These first online measurements in ambient air show that while NA may comprise a small fraction of total OA in Detling ( $\sim 2\%$ ), they are a potentially significant contributor to light absorption in the near-UV by brown carbon (accounting for  $6 \pm 3\%$  of the brown carbon absorption). Thus, NA can potentially affect atmospheric radiative transfer and consequently, air quality and climate.



**Figure 5.** Time series of particulate nitrated phenols (stacked) as identified by the MOVI-CI-TOFMS during the ClearfLo campaign in Detling and total organic PM1 measured by the HR-ToF-AMS (green line). Vertical colored regions indicate wind direction for wind speeds  $>10$  m/s.

## 5.0 Public Outreach

An overview of the ClearfLo field campaign, press coverage, and resulting list of publications can be found at: <http://www.clearflo.ac.uk>.

## 6.0 Clear Air for London (CLEARFLO), Detling, UK Publications

### 6.1 Journal Articles/Manuscripts

- Mohr, C, FD Lopez-Hilfiker, P Zotter, ASH Prévôt, L Xu, NL Ng, SC Herndon, LR Williams, JP Franklin, MS Zahniser, DR Worsnop, WB Knighton, AC Aiken, KJ Gorkowski, MK Dubey, JD Allan, and JA Thornton. 2013. "Contribution of nitrated phenols to wood burning brown carbon light absorption in Detling, UK during winter time." *Environmental Science Technology* 47(12): 6316–6324, [doi:10.1021/es400683v](https://doi.org/10.1021/es400683v).
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- Visser, S, JG Slowik, M Furger, P Zotter, N Bukowiecki, F Canonaco, U Flechsig, K Appel, DC Green, AH Tremper, DE Young, PI Williams, JD Allan, H Coe, SC Herndon, LR Williams, C Mohr, L Xu, NL Ng, A Detournay, E Nemitz, JF Barlow, CH Halios, ZL Fleming, U Baltensperger and ASH Prévôt. 2015. "Advanced source apportionment of size-resolved trace elements at multiple sites in London during winter." *Atmospheric Chemistry and Physics Discussions* 15: 11291-11309, [doi:10.5194/acp-15-11291-2015](https://doi.org/10.5194/acp-15-11291-2015).
- Xu, L, L Williams, P Chhabra, E Fortner, B Knighton, S Herndon, D Worsnop, D Young, J Allan, and NL Ng. 2015. "Characterization of submicron aerosol aging in Detling, UK during winter time by HR-ToF-AMS coupled with a thermal denuder." In preparation.



Williams, LR, S Herndon, J Jayne, A Freedman, B Brooks, J Franklin, P Massoli, E Fortner, P Chhabra, M Zahniser, H Stark, T Onasch, DR Worsnop, F Lopez-Hilfiker, C Mohr, J Thornton, NL Ng, L Xu, M Kollman, B Knighton, M Dubey, A Aiken, K Gorkowski, T Martin, R Coulter, S Visser, M Furger, P Zotter, and A Prévôt. 2015. "Overview of ClearfLo Detling Site (January to February, 2012)." In preparation.

## 6.2 Meeting Abstracts/Presentations/Posters

Seven oral presentations in breakout session and five posters. 2012. Presented at the ASR Science Meeting. Arlington, Virginia.

Massoli, P, A Aiken, K Gorkowski, S Herndon, E Fortner, J Jayne, J Franklin, W Brooks, P Chhabra, T Onasch, L Williams, M Dubey, D Worsnop and A Freedman. 2012. "Optical characterization of aerosols at a rural site in southeast England during the winter ClearfLo IOP." Presented at the European Aerosol Conference. Granada, Spain.

Williams, LR, S Herndon, J Jayne, A Freedman, B Brooks, J Franklin, P Massoli, E Fortner, P Chhabra, M Zahniser, H Stark, T Onasch, DR Worsnop, F Lopez-Hilfiker, C Mohr, J Thornton, NL Ng, L Xu, M Kollman, WB Knighton, M Dubey, A Aiken, K Gorkowski, T Martin and R Coulter. 2012. "Overview of ClearfLo Detling site: Study of aerosol sources and processing at a rural site southeast of London." Presented at the European Aerosol Conference. Granada, Spain.

Williams, L, S Herndon, J Jayne, A Freedman, W Brooks, J Franklin, P Massoli, E Fortner, P Chhabra, M Zahniser, H Stark, T Onasch, DR Worsnop, F Lopez-Hilfiker, C Mohr, JA Thornton, NL Ng, L Xu, M Kollman, B Knighton, M Dubey, A Aiken, K Gorkowski, T Martin, and R Coulter. 2012. "Overview of ClearfLo Detling site: Study of aerosol sources and processing at a rural site southeast of London." Presented at the American Association for Aerosol Research Annual Meeting. Minneapolis, Minnesota.

Ng, NL, L Xu, M Kollman, J Jayne, S Herndon, W Brooks, L Williams, P Massoli, E Fortner, P Chhabra, T Onasch, and D Worsnop. 2012. "Aerosol Composition at a Rural Site Southeast of London Measured by High Resolution Mass Spectrometry." Presented at the American Association for Aerosol Research Annual Meeting. Minneapolis, Minnesota.

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