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LETTER

Changing supersites: assessing the impact of the southern UK EMEP supersite relocation on measured atmospheric composition

H L Walker 1,2, M R Heal 1, C F Braban 1, S Ritchie 3, C Conolly 3, A Sanocka 1, U Dragosits 1 and M M Twigg 1

1 Natural Environment Research Council, Centre for Ecology & Hydrology, Penicuik, United Kingdom
2 School of Chemistry, The University of Edinburgh, United Kingdom
3 Ricardo Energy & Environment, Harwell, United Kingdom

E-mail: hwalker55@ceh.ac.uk

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Abstract

In January 2016 the United Kingdom’s southern European Monitoring and Evaluation Programme (EMEP) level-2 air pollution monitoring ‘supersite’ was relocated from Harwell, Oxfordshire to Chilbolton Observatory, Hampshire. As no co-location study was undertaken, this work retrospectively investigates whether the supersite relocation has led to discontinuities in the time series of concentrations of commonly studied gaseous pollutants (NOx, NH3, SO2 and O3) and particulate matter (PM2.5 and PM10). Two years of measurements pre- and post-relocation (2014–15 and 2016–17 respectively) were analysed in conjunction with meteorological variables and local emission data. The deweather package was applied to the concatenated time series to minimise the influence of meteorology. Similar average concentrations of PM2.5, PM10, SO2 and O3 were observed, but there were substantial differences in that of NOx and NH3 (increase by factors of ∼1.6 and ∼3, respectively). The considerably higher NH3 concentrations at Chilbolton are attributed to the close proximity of mixed farmland, in particular to a strong south-westerly source contributing to ∼50% of the annual average. NOx and PM concentrations in easterly winds arriving at Chilbolton are ∼2.7 and ∼1.5 times larger than at Harwell, from sources including the M3 motorway and Greater London. Westerly concentrations of NOx remain similar, therefore despite a higher frequency of westerly wind, annual mean concentrations are larger. Lower concentrations of PM arriving from the west result in similar annual averages. The secondary inorganic and black carbon components of PM were broadly similar between the sites. The differences in average NOx and NH3 at Chilbolton must be taken into account when considering long-term regional trends based on the southern UK supersite data.

1. Introduction

Atmospheric pollution has a significant influence on human and ecosystem health. Inhalation of ozone (O3) and particulate matter has been linked to cardiovascular and respiratory diseases (WHO 2006, 2013). Deposition of acidic gases causes acidification of terrestrial and aquatic ecosystems, and nitrogen deposition (e.g. from nitrogen oxides, NOx, or ammonia, NH3) leads to eutrophication (Sutton et al 2011, RoTAP 2012).

Monitoring of air pollutant concentrations is important for quantifying these effects and their spatio-temporal trends (Fagerli and Aas 2008, Malley et al 2016). In Europe, the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP, www.emep.int) has the aim of providing member states with this quantitative information (Tørseth et al 2012). Measurements within EMEP are made at rural sites representative of the surrounding area (Spangl et al 2007, Joly and Peuch 2012) and adhere to prescribed sampling methods and siting criteria detailed by the Chemical
Coordinating Centre (EMEP-CCC) to ensure comparability (Kuhlbusch et al 2014). Spatially distributed sites provide data for evaluation of regional models (Fagerli and Aas 2008, Cape 2009, Malley et al 2014), whilst hourly measurements for 40+ years allow characterisation of emission patterns and separation of long-term trends from interannual variability (Tørseth et al 2012). Such data help establish achievable mitigation strategies (Pope and Wu 2014) and track impacts of implemented measures.

The UK has been a member of EMEP since its inception in 1979, and currently operates two level-2 ‘supersites’ (UNECE 2004a, 2004b) at Auchencorth Moss in southern Scotland, and Chilbolton Observatory in southern England (Defra 2018a, 2018b; figure 1) measuring a wide range of pollutants. The southern supersite, Chilbolton, opened in January 2016 following the closure of the previous site (Harwell, figure 1) in December 2015, which had been active for several decades. The aim of this study is to investigate whether the move from Harwell to Chilbolton led to a temporal change in the chemical climate recorded by the southern rural UK supersite. Given the extensive instrumentation, duplicate measurements could not be run for any overlap period. An analysis is presented of measurements in the years directly pre- and post-relocation (2014–15 and 2016–17 respectively), focusing on nitrogen oxides (NOx), ammonia (NH3), sulphur dioxide (SO2), particulate matter <10 and 2.5 μm in diameter (PM10 and PM2.5, respectively) and ozone (O3).
Table 1. Annual mean concentrations (with 95% confidence intervals) and annual percentage data capture of measurements for the investigated species at Harwell (2014–15), and Chilbolton (2016–17).

| Species | Instrument | Harwell 2014 | Harwell 2015 | Chilbolton 2016 | Chilbolton 2017 |
|---------|------------|--------------|--------------|----------------|----------------|
| SO₂     | Monitor for Aerosols and Gases in Air (MARGA) | 0.245 ± 0.028 (94%) | 0.150 ± 0.0027 (70%) | 0.157 ± 0.0044 (58%) | 0.131 ± 0.0032 (79%) |
| HONO    |            | 0.464 ± 0.0068 (94%) | 0.340 ± 0.0049 (70%) | 0.494 ± 0.011 (58%) | 0.454 ± 0.008 (79%) |
| HNO₃    |            | 0.159 ± 0.0029 (94%) | 0.138 ± 0.0032 (68%) | 0.159 ± 0.0050 (57%) | 0.162 ± 0.0037 (80%) |
| NH₃     |            | 1.96 ± 0.049 (95%) | 2.05 ± 0.044 (70%) | 5.88 ± 0.22 (54%) | 6.23 ± 0.18 (82%) |

2. Method

2.1. Site details
The Harwell supersite is predominantly surrounded by agricultural land near Didcot, Oxfordshire (lat: 51.571°, lon: −1.325°, altitude: 126 m), 70 km west of London and 20 km south of Oxford (figure 1). The closest minor road is ~400 m to the west and a dual-carriageway (A34) ran ~2 km to the east. The town of Didcot is ~6.4 km to the north-east. Approximately 8 km in the same direction was a coal and oil fired power station ("Didcot A"), formerly a nearby major point source of NO₃ and SO₂ (Abdalmogith and Harrison 2006, Vieno et al. 2010), that closed on 22nd March 2013 and was demolished in 2016. "Didcot B", a natural gas power plant on the same site, remains operational. Measurement of atmospheric pollutants at Harwell started in 1976 (UKEAP 2015) prior to its incorporation as an EMEP level-2 supersite (UNECE 2004b). Operations ceased on 31st December 2015.

The Chilbolton Observatory site is located ~50 km south of Harwell in an agricultural (mainly arable) landscape, ~200 m south-east of the edge of Chilbolton village, Hampshire (lat: 51.150°, lon: −1.438°, altitude: 78 m) and 100 km south-west of London (Defra 2018b). Two single-carriageway main roads run ~1 km to the west (A3057), and ~3 km to the south (A30) (figure 1). The site began monitoring as an EMEP level-2 supersite on 11th January 2016, after instrument relocation from Harwell.

2.2. Measurement data
The full suite of measurements at the two sites is summarised in UK Eutrophying and Acidifying Pollutants (UKEAP) network reports (UK-AIR Library 2018), a subset of which are investigated in this work (table 1, and supplementary information figure S1 is available online at stacks.iop.org/ERC/1/041001/mmedia). Concentration data were downloaded from the UK Department for Environment, Food and Rural Affairs (Defra) online data repository (UK-AIR Data Selector 2018).

Data are evaluated here for the two years either side of site relocation (Harwell 2014–15 and Chilbolton 2016–17, respectively). Data capture statistics are given in table 1. O₃, NOₓ, SO₂, PM₁₀, and PM₂.₅ measurements derive from the UK Automatic Urban and Rural Network (AURN). Instrumentation selection, calibration and data ratification follow EU Air Quality Directives (2008/50/EC), and data are archived as hourly averages (UK-AIR Library 2018). Hourly measurements of NH₃ and other trace gases, together with water-soluble ions within PM₂.₅ and PM₁₀, are provided by the Monitor for Aerosols and Gases in Air (MARGA) instrument (EMEP 2007, Steiger et al. 2018) with data quality assurance processes as described in Twigg et al. (2015).

Hourly meteorological data comprise on-site measurements at Harwell and Chilbolton (Wrench). Data were also downloaded for the meteorological station at Benson, Oxfordshire (lat: 51.616°, lon: −1.096°, altitude: 57 m), marked on figure 1 (a), from the NOAA Integrated Surface Database using the worldmet package (Carlswal 2018, NOAA 2018) for the full time period being considered. These data were used to validate the use of concatenated meteorological time series from the two supersites (see supplementary information).

2.3. Meteorological detrending
To examine for evidence of a step-change in concentration coincident with site relocation, the deweather function (Carlswal 2017) was applied to concatenated Harwell and Chilbolton datasets (2014–17) in a technique

| Species | Instrument | Harwell 2014 | Harwell 2015 | Chilbolton 2016 | Chilbolton 2017 |
|---------|------------|--------------|--------------|----------------|----------------|
| O₃      | UV absorption | 56.0 ± 0.45 (99%) | 57.1 ± 0.42 (98%) | 48.9 ± 0.53 (97%) | 51.5 ± 0.48 (98%) |
| NO₂     | Chemiluminescence | 10.5 ± 0.30 (97%) | 9.20 ± 0.22 (97%) | 18.5 ± 0.49 (91%) | 13.8 ± 0.35 (97%) |
| NO₂     |            | 7.99 ± 0.20 (97%) | 7.68 ± 0.16 (97%) | 14.3 ± 0.25 (91%) | 11.2 ± 0.19 (97%) |
| PM₁₀    | TEOM-FDMS | 14.3 ± 0.27 (74%) | 15.1 ± 0.21 (89%) | 14.9 ± 0.25 (85%) | 13.3 ± 0.19 (98%) |
| PM₂.₅   |            | 9.06 ± 0.19 (96%) | 8.35 ± 0.17 (96%) | 9.45 ± 0.19 (93%) | 7.38 ± 0.17 (98%) |
| SO₂     | UV fluorescence | 1.01 ± 0.054 (98%) | 0.939 ± 0.013 (93%) | 0.876 ± 0.017 (64%) | 0.763 ± 0.013 (89%) |
known as ‘meteorological normalisation’ (Grange et al 2018). This accounts for non-linear and complex relationships between predictors, such as meteorological or temporal variables (Carslaw and Taylor 2009), allowing changes in time series not directly caused by these predictors to be identified. Models are built using a stochastic process that results in reduced variance of the final model (Friedman 2002), but consequently, a slightly different model is produced with each run (Elith et al 2008). A set of 10 identically-built deweather models were performed for each pollutant time series, using meteorological predictor variables measured at the same site as the pollutants. Further details of the model, including comparisons using different meteorological data, are given in the supplementary information (section S2 and figure S3).

2.4. Emission inventories

Annual emission estimates for NH₃, NOₓ, SO₂, PM₂.₅ and PM₁₀ were obtained from the UK National Atmospheric Emission Inventory (NAEI, http://naei.beis.gov.uk/) for both locations. Individual species emissions were taken from the 2015 inventory and were aggregated over the 15 km × 15 km area surrounding each site shown in figure 1. The areas were defined according to the gridded agricultural sector NH₃ emissions which have a spatial resolution of 5 km × 5 km, but for all other pollutants the underlying resolution was 1 km × 1 km.

3. Results and discussion

3.1. Overview

Table 1 summarises the annual mean concentration and corresponding 95% confidence interval for each pollutant for each year. Chilbolton MARGA measurements have the lowest data capture (54%–58%) since these measurements did not commence until 11th February 2016, and instrument issues led to missing data between 9th July and 1st September 2016. Between August 2014 and September 2015 a plume from the volcanic eruption at Holuhraun, Iceland, passed over the UK and is observed in the SO₂ time series (figure S1). Similarly in Spring 2014, elevated PM concentrations were caused by a combination of Saharan dust and ammonium nitrate formed from European emissions (Vieno et al 2016) (figure S1). Other PM episodes are also apparent between 2014 and 2017 (figure S1), typically accumulating in low wind speeds and lasting no more than a few days (Defra 2015, 2016, 2017).

3.2. NOₓ

The average 2016 and 2017 concentration of NOₓ at Chilbolton was approximately 1.6 times greater than the average 2014 and 2015 concentration at Harwell (annual means of 18.5 and 13.8 μg m⁻³ cf 10.5 and 9.20 μg m⁻³ respectively, table 1). The NOₓ deweather time series also shows an abrupt increase coincident in timing with the relocation of the measurement site (figure 2(a)).

In contrast to the greater average NOₓ concentrations at Chilbolton, the total local NOₓ emissions integrated over the 15 km × 15 km area around Harwell are more than 5 times greater than from the same-size area around Chilbolton (figure 3). The NOₓ emissions close to Harwell are dominated by specific sources including Didcot town, Didcot B power station in the north-east, and high traffic flow on the dual-carriageway (A34) running north-south 2 km to the east. Across the 225 individual 1 km × 1 km grid squares within the area surrounding Harwell, 47% of total local emissions are contributed by the grid square with the highest emissions, whilst the 25 highest emission grid squares contribute 84%. Similar analysis at Chilbolton shows less dominance of local emissions from the single highest (6%) and 25 highest (61%, including part of the A303, ~8 km north) grid squares.

These differences in NOₓ source configurations are illustrated by pollution wind roses (figure 4). The aforementioned sources north of Harwell generate the most polluted air transported to the site but do not contribute significantly to the annual average than other directions. Considering NOₓ to have an atmospheric lifetime of ∼4–6 h in the mid-latitudes (Beirle et al 2011) and an average wind speed of ∼15 km h⁻¹ for southern England (figure S2; Wrench, NOAA 2018), the London pollution plume could plausibly be observed at both sites. It appears Harwell is less influenced by London (~100° bearing) than Chilbolton (~70°), for which this wind direction provides a dominant contribution to the annual average. However, the M3 motorway also contributes NOₓ to this wind direction, stretching for ~56 km between Chilbolton and London. Traffic along the length of the M3 (~95 km) and other major roads are the probable reason why a larger average concentration of NOₓ (>2.2 times higher) is observed in easterly (0°–180°) than in westerly winds (181°–360°, table 2). More frequent westerly winds during 2017 led to a reduced annual average compared with 2016 (13.8 cf 18.5 μg m⁻³).

The importance of wind direction to NOₓ concentrations at both sites is reflected in the relative importance of variables used in the deweather models (table S2). Wind direction had the largest mean relative importance of
19.1%, closely followed by ambient temperature (18.7%) and (long-term) trend (18.4%). Unsurprisingly, predicted concentrations of NOx are elevated at lower ambient temperatures (<10 °C) and during colder months (October-February), as these conditions limit the dispersion of emissions (AQEG 2004), and there is a higher demand for domestic heating. The importance of the trend variable is a consequence of the abrupt increase in NOx at the time of site relocation in the concatenated deweather time series (figure 2(a)). This supports the interpretation of a real difference in annual mean NOx concentration between Harwell and Chilbolton.

3.3. NH3
The annual mean NH3 concentrations at Chilbolton in 2016 and 2017 (5.88 and 6.23 μg m⁻³, respectively, table 1) are about 3 times higher than at Harwell in 2014 and 2015 (1.96 and 2.05 μg m⁻³, respectively), the greatest difference of all pollutants investigated. Although data capture at Chilbolton in 2016 was rather low (58%), and most missing data occurred during the summer (figure S1) when NH3 concentrations are typically larger (Tang et al 2018), therefore the annual average in 2016 is likely biased low by the missing data.

Application of the deweather model to the concatenated time series of NH3 concentrations (figure 2(b)) confirms a distinct increase in mid-February 2016, coincident with commissioning of the MARGA at Chilbolton. Trend is the dominant variable in the deweather model (45.5 ± 0.6%, figure S4 and table S2). The raw time series shows elevated springtime concentrations at Harwell (figure S1), consistent with typical livestock manure and synthetic fertiliser applications observed in NW European springtime (Vieno et al 2016). These springtime peaks are mirrored in the predicted concentrations of NH3 during 2014 and 2015, and reflected by the partial dependency of the week-of-year variable predicting increased concentrations during Julian weeks 5–20 (figure S4). Following site relocation the deweather time series showed NH3 concentrations remaining high, with large and infrequent peaks that demonstrate little seasonality. Despite this, maximum hourly NH3 at Chilbolton in 2016 and 2017 were during spring (126 μg m⁻³ on 22nd March 2016 and 94.4 μg m⁻³ on 24th May 2017).
Ammonia has a short atmospheric lifetime and high spatial heterogeneity in its sources (Vogt et al 2013, Dammers et al 2017), so receptor concentrations are significantly influenced by local emissions. The contrasting concentrations at Harwell and Chilbolton are not reflected in the total NAEI estimates of local NH$_3$ emissions, which show similar averages over the 15 km $\times$ 15 km area surrounding each site (figure 5). However, the total at Harwell includes contribution from a disproportionately large 1 km $\times$ 1 km grid square over the site of Didcot B power station, over 3.4 times greater than any other grid square surrounding either site. When emissions from only the agricultural sector are considered, the greatest emissions are associated with the 5 km $\times$ 5 km grid square also containing the site, suggesting the presence of a nearby agricultural source or sources.

Polar plots of NH$_3$ concentration as a function of wind speed and direction for both sites (figure 5) reveal an obvious dominant local source to the south-west of Chilbolton. Figure 6 demonstrates the difference in concentration between this source and background NH$_3$ at Chilbolton in 2016, by dividing prevailing wind into four direction sectors: east (E, 30°–180°), south-west (SW, 180°–250°), north-west (NW, 250°–350°) and north (N, 350°–30°). The N sector appears to reflect background NH$_3$ levels, with a mean concentration of 2.2 $\mu$g m$^{-3}$, comparable to average NH$_3$ concentrations measured at Harwell across all wind directions (figure 5). Mean concentrations in E, NW and SW sectors are higher (3.1, 3.4 and 7.1 $\mu$g m$^{-3}$, respectively) and demonstrate an inverse relationship with wind speed, suggesting nearby sources at Chilbolton. Approximately 1.5 km from the

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**Figure 3.** The top panel shows annual emissions maps of NO$_x$ and NH$_3$ from the National Atmospheric Emissions Inventory database (NAEI, https://naei.beis.gov.uk/) for the 15 km $\times$ 15 km area surrounding Harwell and Chilbolton sites. Underlying data area at 5 km $\times$ 5 km grid resolution for agricultural NH$_3$, displayed at 1 km $\times$ 1 km resolution. The bar plot in the lower panel displays the total emissions of NO$_x$, NH$_3$, SO$_2$, PM$_{2.5}$ and PM$_{10}$ for the same area around both sites, whilst the black point indicates the emission from the component grid square with the maximum emission across the contributing grid squares.
Figure 4. Pollution wind roses for NO$_x$ and PM$_{2.5}$ concentrations at Harwell in 2015 and at Chilbolton in 2016, depicting the contribution of 5 μg m$^{-3}$ concentration bins in each wind sector to the annual mean concentration. Wind and pollutant concentration measurements were co-located and the underlying time resolution was hourly for both. The maximum hourly concentration in each year is the upper-limit value of the red segment in each legend.

Table 2. Annual mean concentrations of hourly NO$_x$ measurements for easterly (0°–180°) and westerly (181°–360°) wind directions. The remaining percentage of data (% NA) includes hours where NO$_x$ concentration or wind direction were not recorded.

| Site and year | Mean NO$_x$ concentration/μg m$^{-3}$ | Percentage of total annual wind/% |
|---------------|--------------------------------------|----------------------------------|
|               | Westerly | Easterly | Westerly | Easterly | NA  |
| Harwell 2014  | 11.1     | 10.0     | 39       | 59       | 2   |
| Harwell 2015  | 10.5     | 8.60     | 34       | 55       | 11  |
| Chilbolton 2016 | 12.0     | 28.6     | 54       | 35       | 11  |
| Chilbolton 2017 | 10.2     | 22.6     | 69       | 28       | 3   |

Figure 5. Polar plots of hourly NH$_3$ concentration (MARGA) as a function of hourly wind speed and direction at Harwell 2015 (left) and Chilbolton 2016 (right). Wind and NH$_3$ concentration measurements were co-located.
before and after site relocation: twice each year at Harwell, 3 times at Chilbolton in 2016 and once in 2017.

Hourly SO₂ concentrations at both sites are determined by two different measurement methodologies, but there was poor agreement between them (figure S5). This is anticipated, as the limit of detection (LOD) of theUV fluorescence analyser (1.5 ppb, ∼4 μg m⁻³) is approximately two orders of magnitude greater than that of the MARGA (0.04 μg m⁻³ (Makkonen et al. 2012)). The aim of the AURN SO₂ analysers is to detect pollution events (Twigg et al. 2016) where concentrations could exceed national/EU limit values (hourly and daily means of 350 and 125 μg m⁻³), whereas the MARGA is designed to monitor changes in background concentrations of SO₂. Extending the time series of SO₂ measurements at Harwell back to 2012 shows that there was a large reduction in concentrations in early 2013 (figure 7), coincident with the closure of Didcot A coal and oil power station, which whilst operational was one of the largest SO₂ emission sources in the UK (Vieno et al. 2010). Following this event there are no discernible changes in SO₂ concentration, including through the site relocation, apart from a singular peak in August 2014 which can be attributed to an Icelandic volcanic eruption (Twigg et al. 2016). Whilst the emission inventory estimates greater local SO₂ emissions around Harwell than around Chilbolton (figure 3), these are overwhelmingly dominated at Harwell by Didcot B power station. The relative infrequency of north-easterly wind to the site, coupled with the elevated source of these emissions, resulted in little influence of Didcot B on ambient measured concentrations. Consequently, there is effectively no change in measured SO₂ arising from the site relocation.

3.4. SO₂

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3.5. Particulate matter

Data capture was high for total PM₁₀ and PM₂.₅ (table 1), but poorer for the individual species concentrations. Annual mean concentrations of PM₁₀ and PM₂.₅ were similar at Harwell and Chilbolton (2014 and 2015 average PM₁₀ = 14.7 μg m⁻³, 2016 and 2017 average PM₁₀ = 14.1 μg m⁻³; equivalent data for PM₂.₅ = 8.8 and 8.4 μg m⁻³), indicating no substantive change in long-term mean PM concentration at the time of site relocation. There was also no significant difference in the number of PM₁₀ daily mean exceedances of 50 μg m⁻³ before and after site relocation: twice each year at Harwell, 3 times at Chilbolton in 2016 and once in 2017.

The deweather models for both PM₂.₅ and PM₁₀ (figures 2(c) and (d)) show temporal variability, but no evidence of a significant step-change in concentration at the time of site relocation. The model predicted lower concentrations of PM₁₀ in June 2016 because measured values in this period 12th June—31st July 2016 (mean of 7.6 μg m⁻³) were substantially lower than the mean for the rest of the time series (14.4 μg m⁻³). There is a similar effect in the modelled PM₂.₅ time series. Model disagreement in the latter part of the PM₂.₅ deweather time series is presumed to be due to the presence of a large measurement value (186.8 μg m⁻³, 27th April 2017),
almost twice any other hourly PM$_{2.5}$ concentration. The stochastic nature of the deweather function resulted in some models being built using this data point, while others were not.

The annual NAEP emissions integrated over the 15 km $\times$ 15 km area surrounding each site are substantially greater around Harwell than Chilbolton for both PM$_{10}$ and PM$_{2.5}$ (figure 3). Locations of high PM$_{2.5}$ emissions within the area surrounding Harwell include those with substantial NO$_x$ emissions (Didcot town, Didcot B power station and the A34). The 25 1 km$^2$ grid squares with highest PM emissions around Harwell contribute 63% to the total, whereas the equivalent at Chilbolton contribute 54%. As with NO$_x$, an increase in PM$_{2.5}$ concentrations is observed in the prevailing easterly wind at Chilbolton as compared to Harwell (13.2 in 2016 cf 8.1 $\mu$g m$^{-3}$ in 2015, table S3), however westerly winds have lower concentrations (6.8 cf 9.6 $\mu$g m$^{-3}$), which accounts for the comparable annual averages pre- and post-relocation. The same conclusions apply for PM$_{10}$ (figure S6).

Data capture for MARGA measurements were poorer than for TEOM-FDMS measurements (table 1). Nevertheless for all secondary inorganic aerosol (SIA) ions analysed, the data capture rate was sufficient for comparisons of annual measurements (all years >58%). Anthropogenically-derived NH$_4^+$, NO$_3^-$ and SO$_4^{2-}$ dominate PM$_{2.5}$ at both sites, contributing an average of 58% to total measured mass at Harwell (2014–15) and 59% at Chilbolton (2016–17). The calculated SIA ion balance for PM$_{2.5}$ (figure 8) shows good correlation for all years, despite a few events of exception. More acidic aerosols are observed in 2014, with the greatest excess in September where concentrations of SO$_4^{2-}$ were significantly elevated from the Holuhraun eruption plume passing over the UK (Twigg et al 2016). Both years at Chilbolton exhibit a large excess of basic aerosol on days reaching high NH$_4^+$ concentrations (8.64 and 22.6 $\mu$g m$^{-3}$ on 22nd March 2016 and 11th March 2017 respectively; table 3). In general, all years appear to have a slight bias towards basic aerosol and are comparable to observations at the northern UK EMEP supersite (Auchencorth Moss; Twigg et al 2015). Similar observations apply to the coarse fraction (PM$_{\text{coarse}}$ = PM$_{10}$–PM$_{2.5}$), where SIA contributes 15% to total aerosol mass at both Harwell (2014–15) and Chilbolton (2016–17). Annual average black carbon measurements (Aethalometer, Black Carbon Network) are comparable at ~0.40 $\mu$g m$^{-3}$ between Harwell (2014–15) and Chilbolton (2016–17). Given the comparable total PM at both sites, this indicates that there is also a similar contribution of secondary organic aerosols (SOA) in both fractions.

### 3.6. Ozone

Data capture for hourly O$_3$ concentrations was high (>98% for each year, table 1). Annual mean concentrations of O$_3$ were somewhat lower after the site relocation, 48.9 and 51.5 $\mu$g m$^{-3}$ for 2016 and 2017 respectively at Chilbolton, compared with 56.0 and 57.1 $\mu$g m$^{-3}$ for 2014 and 2015 at Harwell (table 1). However, the deweather model time series for O$_3$ data (figure 2(e)) does not show evidence of a step–change associated with the site relocation (the modelled decrease in mid-2016 is coincident with the similar observation for PM). The slightly lower annual mean background O$_3$ at Chilbolton than at Harwell may be due to interannual variability in O$_3$, but the lower background concentration is also consistent with an inverse concentration relationship with the unambiguously higher background NO$_x$ at Chilbolton.
4. Conclusions

The relocation of the southern UK EMEP supersite from Harwell to Chilbolton in January 2016 has not resulted in discontinuities in average measured concentrations of PM$_{2.5}$, PM$_{10}$, SO$_2$ and O$_3$ (based on two-year pre- and post-relocation time comparisons), but has led to substantial increases in average concentrations of NO$_x$ and NH$_3$, by a factor of $\sim 1.6$ and $\sim 3$, respectively.

Concentrations of NO$_x$ and PM in easterly wind arriving at Chilbolton are $\sim 2.7$ and $\sim 1.5$ times larger than at Harwell, from common sources including the M3 motorway and Greater London. Prevailing winds from the west contribute similar NO$_x$ concentrations at both sites, therefore despite a higher frequency of westerly wind at Chilbolton, the larger easterly concentrations result in larger annual means. Westerly winds carry lower concentrations of PM to Chilbolton than Harwell, thereby resulting in similar annual averages. Measurements show no substantive difference in the contribution of secondary inorganic aerosols (SIA) and black carbon to the
total PM mass between the two site locations, although more NH$_4^+$ events are observed in the SIA at Chilbolton. Background concentrations of NH$_3$ at both sites reflect the presence of mixed farmland; however the contribution of very strong local sources to the south-west of Chilbolton cause the large increase in annual average between sites.

In conclusion, when considering long-term regional trends based on the southern UK supersite data, the increase in NO$_x$ and NH$_3$ at Chilbolton must be taken into account and the Harwell and Chilbolton datasets should be treated separately.

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**ORCID iDs**

H L Walker [https://orcid.org/0000-0002-1531-4147](https://orcid.org/0000-0002-1531-4147)
M R Heal [https://orcid.org/0000-0001-5539-7293](https://orcid.org/0000-0001-5539-7293)
C F Braban [https://orcid.org/0000-0003-4275-0152](https://orcid.org/0000-0003-4275-0152)
U Dragosits [https://orcid.org/0000-0002-9283-6467](https://orcid.org/0000-0002-9283-6467)
M M Twigg [https://orcid.org/0000-0002-5462-3348](https://orcid.org/0000-0002-5462-3348)

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