Analysis of New Particle Formation (NPF) Events at Nearby Rural, Urban Background and Urban Roadside Sites

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ABSTRACT

New Particle Formation (NPF) events have different patterns of development depending on the conditions of the area in which they occur. In this study, NPF events occurring at three sites of differing characteristics (rural Harwell (HAR), urban background North Kensington (NK), urban roadside Marylebone Road (MR), London, UK) were studied (seven years of data). The different atmospheric conditions in each study area not only have an effect on the frequency of the events, but also affect their development. The frequency of NPF events is similar at the rural and urban background locations (about 7% of days), with a high proportion of events occurring at both sites on the same day (45%). The frequency of NPF events at the urban roadside site is slightly less (6% of days), and higher particle growth rates (average 5.5 nm h^{-1} at MR compared to 3.4 nm h^{-1} and 4.2 nm h^{-1} at HAR and NK respectively) must result from rapid gas to particle conversion of traffic-generated pollutants. A general pattern is found in which the condensation sink increases with the degree of pollution of the site, but this is counteracted by increased particle growth rates at the more polluted location. A key finding of this study is that the role of the urban environment leads to an increment of 20% in $N_{16\text{-}20nm}$ in the urban background compared to that of the rural area in NPF events occurring at both sites. The relationship of the origin of incoming air masses is also considered and an association of regional events with cleaner air masses is found. Due to lower availability of condensable species, NPF events that are associated with cleaner atmospheric conditions have lower growth rates of the newly formed particles. The decisive effect of the condensation sink in the development of NPF events and the survivability of the newly formed

particles is underlined, and influences the overall contribution of NPF events to the number of ultrafine particles in an area. The other key factor identified by this study is the important role that urban pollution plays in new particle formation events.

1. INTRODUCTION

Ultrafine particles (particles with diameter smaller than 100 nm) typically make the greatest contribution in the total particle count, especially in urban environments (Németh et al., 2018), but a very small contribution to total volume and mass (Harrison et al., 2000). Research studies have indicated that ultrafine particles can cause pulmonary inflammation and may contribute to cardiovascular disease (Oberdörster, 2000) and have increased possibility to penetrate the brain and central nervous system (Politis et al., 2008) compared to fine and coarser particles. Since some studies report that toxicity per unit mass increases as particle size decreases (Penttinen et al., 2001; MacNee et al., 2003; Davidson et al., 2005); it is considered possible that particle number concentrations may be a better predictor of health effects than mass concentrations (Harrison et al., 2000; Atkinson et al., 2010; Kelly et al., 2012; Samoli et al., 2016). Additionally, NPF events have an impact on climate (Makkonen et al., 2012) either by increasing the number of cloud condensation nuclei (Spracklen et al., 2008; Merikanto et al., 2009; Dameto de España et al., 2017; Kalkavouras et al., 2017), or directly affecting the optical properties of the atmosphere (Seinfeld and Pandis, 2012).

The sources of ultrafine particles in urban areas can either be primary particles or emission sources from traffic (Shi et al., 1999; Harrison et al., 2000), airports (Masiol et al., 2017) and other combustion related processes (Keuken et al., 2015; Kecorius et al., 2016), or by new particle formation (NPF) from gaseous precursors. New particle formation as described by Kulmala et al.

(2014), is the process of production of low-volatility vapours, clustering of these vapours, nucleation, activation of the clusters with a second group of vapours and condensational growth to larger sizes. This process can occur both locally or on a larger scale; in the latter case the events are characterized as regional. Regional events have been found to take place in a scale of hundreds of kilometres (Németh and Salma, 2014; Shen et al., 2018), without being affected by air mass advection (Salma et al., 2016). NPF is one of the main contributors of particles in the atmosphere (Spracklen et al., 2010; Kulmala et al., 2016; Rahman et al., 2017) and this relative contribution increases moving from a kerbside to a rural area (Ma and Birmili, 2015). While NPF events in rural and remote areas have been widely studied for many years (O'Dowd et al., 2002; Dal Maso et al., 2005; Ehn et al., 2010; Dall'Osto et al., 2017; Kalkavouras et al., 2017), in urban areas intensive studies have started mainly in recent years (Jeong et al., 2010; Minguillón et al., 2015; Peng et al., 2017; Németh et al., 2018). Early studies in Birmingham, UK highlighted the connection of NPF events with solar radiation (Shi et al., 2001) and a low condensation sink (Alam et al., 2003), a measure of pre-existing aerosol loading (Dal Maso et al., 2002). The importance of a low condensation sink was further underlined by later studies, as being one of the most influential variables in the occurrence of NPF in all types of environment (Wehner et al., 2007; Park, Yum and Kim, 2015; Pikridas et al., 2015). An important contributor to many NPF pathways is SO_2 (Woo et al., 2001; Berndt et al., 2006; Laaksonen et al., 2008), which in the presence of solar radiation forms H_2SO_4, often the main component of the initial clusters (Kuang et al., 2008; Kulmala et al., 2013; Bianchi et al., 2016; Kirkby et al., 2016). Dall'Osto et al. (2013) pointed out that the role of SO_2 is less significant in urban areas compared to rural and background areas. SO_2 concentration variability in urban areas was found to have a small impact on the frequency of NPF events (Alam et al., 2003; Jeong et al., 2010), though it can have an effect on the number of particles formed (Charron et al., 2007). Furthermore, Dall'Osto et al. (2018) in their research at 24 sites in Europe, pointed out the different role SO_2 seems to play depending on its concentration, and that of other species. Jayaratne et al. (2017) however found that in the heavily polluted environment of Beijing, China, NPF events were more probable in sulphur rich conditions rather than sulphur poor. Apart from its role in the initial formation of the clusters, H_2SO_4 seems to participate in the early stages of growth of the newly formed clusters (Kulmala et al., 2005; Iida et al., 2008; Xiao et al., 2015). In later stages of growth, low or extremely low volatility organic compounds (O'Dowd et al., 2002; Laaksonen et al., 2008; Metzger et al., 2010; Kulmala et al., 2013; Tröstl et al., 2016; Dall'Osto et al., 2018) appear to be more important, while the role of ammonium nitrate in particle growth is also considered (Zhang et al., 2017). While in rural areas the organic compounds are mainly of biogenic origin (Riccobono et al., 2014; Kirkby et al., 2016), in urban areas they mainly originate from combustion processes (Robinson et al., 2007; Gentner et al., 2012). Many comparative studies have reported higher growth rates in urban areas compared to background sites (Wehner et al., 2007; Jeong et al., 2010; Salma, et al., 2016; Wang et al., 2017), as well as greater particle formation rates (Salma, et al., 2016; Nieminen et al., 2018) and a higher frequency of NPF events (Peng et al., 2017), which was attributed to the higher concentration of condensable species. Salma et al. (2014) however reported fewer NPF events in the city centre of Budapest compared to the urban background, due to the higher condensation sink. Due to the complexity of the conditions and mechanisms within an urban area (Harrison, 2017), NPF events are harder to study and factors to be attributed. Increased concentrations of particles in the size range 1.3 – 3 nm were measured at a kerbside site when downwind from the road, following the trends in traffic-related nucleation mode particles, associating them with traffic emissions and thus not resulting from homogeneous nucleation mechanisms (Rönkkö et al., 2017; Hietikko et al., 2018), and studies in Barcelona, Spain (Dall'Osto et al., 2012; Brines et al., 2014) and Leicester, U.K. (Hama et al., 2017), attributed a larger portion of nucleation mode particles to vehicular emissions compared to photochemically induced nucleation. As the condensation sink is higher within an urban environment, NPF events are less favoured. Their occurrence is attributed to either ineffective scavenging or the higher growth rate of the newly formed particles (Kulmala et al., 2017), when sufficient concentrations of precursors are present in the atmosphere (Fiedler et al., 2005), as particle formation was found to take place on both event and non-event days (Riipinen et al., 2007).

In this study, NPF events in three areas of different land use in the southern U.K. are analyzed. Studies for NPF events have been conducted in the past for Harwell, Oxfordshire (Charron et al., 2007; 2008) and the effect of NPF upon particle size distributions was also considered for N. Kensington, London (Beddows et al., 2015). A combined study including all three sites has also been conducted, but in the aspect of ultrafine particle variation (Von Bismarck-Osten et al., 2013). The present study is the first to use a combined long term database for all three sites, focusing on the trends and conditions of NPF events at these sites, as well as the first which identifies NPF events at the highly trafficked Marylebone Road site, as up to this point ultrafine particles were attributed only to traffic (Charron and Harrison, 2003; Dall'Osto et al., 2011). As in this study a rural and an urban background area are studied alongside a kerbside site in the city of London in close proximity, the conditions and development of NPF events in a mid-latitude European region are discussed in relation to the influence of different local environments.

2. DATA AND METHODS

2.1 Site Description and Data Availability

This study analysed NPF events in three areas in the southern United Kingdom (Fig. 1). Harwell in Oxfordshire, is located about 80 km west of the greater London area. The site is in the grounds of the Harwell Science Centre in Oxfordshire (51° 34' 15'' N, 1° 19' 31'' W) and is representative of a rural background area; a detailed description of the site was given by Charron et al. (2013). North Kensington is a suburban area in the western side of London, U.K, 4.5 km west of Marylebone Road. The site is located in the grounds of Sion Manning School (51° 31' 15'' N, 0° 12' 48'' W) and is representative of the urban background of London. A detailed description of the site was given by Bigi and Harrison (2010). Marylebone Road is located in the centre of London, U.K. The site is located on the kerbside of Marylebone road (51° 31' 21'' N; 0° 9' 16'' W), a very busy arterial route within a street canyon. A more detailed description of the area can be found in Charron and Harrison (2003).

At all three sites, seven years (2009 – 2015) of particle number size distributions in the range of 16.6 – 604 nm have been measured and recorded as 15-minute averages, using a Scanning Mobility Particle Sizer (SMPS), comprised by an Electrostatic Classifier (EC, TSI model 3080) and a condensation Particle Counter (CPC, TSI Model 3775), operated on behalf of the Department for Environment, Food and Rural Affairs (DEFRA) in the U.K. At all sites the inlet air is dried, and operation is in accord with the EUSAAR/ACTRIS protocol (Wiedensohler et al., 2012). These 15-minute measurements were averaged to an hourly resolution. In Harwell there were 46930 hours of available SMPS data (76.5% coverage), in N. Kensington 51059 (83.3% coverage) and at Marylebone Road 45562 (74.3% coverage). Detailed data availability is found in Table S1. A free-standing CPC (TSI model 3022A) also operated alongside for most of the years of the survey and was used to give an estimate of particles in the 7-16.6 nm range by difference from the SMPS.

Additionally, air pollutants and other aerosol chemical composition data were extracted from the DEFRA website (https://uk-air.defra.gov.uk/). Meteorological data for Harwell and Heathrow airport (used for N. Kensington and Marylebone road) were available from the Met Office, while solar radiation data from Benson station (for Harwell) and Heathrow airport (for N. Kensington and Marylebone Road), were extracted from the Centre for Environmental Data Analysis (CEDA) site (http://www.ceda.ac.uk). Back trajectory data calculated using the HYSPLIT model (Draxler and Hess, 1998), were extracted by the NOAA Air Resources Laboratory (https://ready.arl.noaa.gov/READYtransp.php) and were processed using the Openair package for R (Carslaw and Ropkins, 2012).

2.2 Methods

2.2.1 NPF events selection

The identification of the NPF event days was made by visual inspection of SMPS data, supplemented with the use of CPC data to confirm the formation of a new mode of particles, using the criteria set by Dal Maso et al. (2005). NPF events are considered when a distinctly new mode of particles which appears in the size distribution at nucleation mode size, prevails for some hours and shows signs of growth. Using these criteria, NPF events are classified into two classes, I and II depending on the confidence level. Class I events are further classified to Ia and Ib, with class Ia containing very clear and strong particle formation events, while Ib contains less clear events. In this study the events of class Ia only are considered as being the most suitable for analysing case studies of NPF events (Figure S1). At this point it should be mentioned that due to the particle size range available, NPF events in which new formed particles failed to grow beyond 16.6 nm (if any) could not be identified. Though such rare occasions were identified using the CPC data, bursts of new particles in the size range < 16.6 nm that did not appear in the SMPS dataset were ignored as their development was unknown. High time resolution data for gaseous pollutants and aerosol constituents was used to identify pollution events affecting particle concentrations and these were removed from the data analysis. This analysis took account of the fact that nanoparticle emissions from Heathrow Airport affect size distributions at London sites (Harrison et al., 2018), and such primary emission influences were not included as NPF events.

2.2.2 Calculation of the condensation sink and growth rate

For the calculation of the condensation sink the method proposed in Kulmala et al. (2001) was used in which the condensation sink is calculated as

$$\mathbf{CS = 4\pi D \sum \beta_M\, r\, N} \qquad (1)$$

where r is the radius of the particles and N is the number concentration of the particles. D is the diffusion coefficient calculated (for T = 293 K and P = 1013.25 mbar) according to Polling et al. (2000):

$$\mathbf{D_{vap} = 0.00143 \cdot T^{1}.75\ \frac{\sqrt{M_{air}^{-1} + M_{vap}^{-1}}}{P\left(D_{x,air}^{\frac{1}{3}} + D_{x,vap}^{\frac{1}{3}}\right)^{2}}} \qquad (2)$$

where P is air pressure, M is the molar mass and D_x is the diffusion volume for air and H_2SO_4. β_M is the Fuchs correction factor calculated as (Fuchs et al., 1971):

$$\beta_M = \frac{1 + K_n}{1 + \left(\frac{4}{3a} + 0.377\right)K_n + \frac{4}{3a}K_n{}^2} \qquad (3)$$

where K_n is the relation of the particle diameter and the mean free path of the gas λ_m, called the Knudsen number.

The growth rate of the particles on nucleation event days was also calculated as proposed by Kulmala et al. 2012, using the formula

$$GR = \frac{D_{P_2} - D_{P_1}}{t_2 - t_1} \qquad (4)$$

for the size range 16.6 – 50 nm. The number of points taken depended on the development of the event and were considered from the start of the event until a) growth stopped, b) GMD reached 50 nm or c) the day ended.

2.2.3 Calculation of the urban increment (U.I.)

The urban increment is defined as the ratio of the number concentration of particles below 20 nm for event days to the average (for the period April – October, when the majority of the events take place) for North Kensington to that at Harwell. This provides with a measure of the new particles formed in each area in comparison to the average conditions, and is calculated by

$$\text{U.I.} = \frac{\mathbf{NK_{Nuc\ Max} - NK_{Bg}}}{\mathbf{HW_{Nuc\ Max} - HW_{Bg}}} \quad (5)$$

where $NK_{Nuc\ Max}$ is the maximum concentration of particles below 20 nm found in the diurnal cycle on event days (found at 13:00) and NK_{Bg} is the average mean concentration at the same time (same for Harwell in the denominator).

2.2.4 Calculation of nucleation strength factor (NSF) and the P parameter

The Nucleation Strength Factor (NSF) was proposed as a measure of the effect nucleation events have in the composition of ultrafine particles in an area. Two factors were proposed. First is the NSF_{NUC}. This is calculated as

$$\mathbf{NSF_{NUC}} = \frac{\left(\frac{\mathbf{N}_{(\text{smallest size available}-100)}}{\mathbf{N}_{(100-\text{largest size available})}}\right)_{\text{nucleation days}}}{\left(\frac{\mathbf{N}_{(\text{smallest size available}-100)}}{\mathbf{N}_{(100-\text{largest size available})}}\right)_{\text{non-nucleation days}}} \quad (6)$$

and provides of a measure of the concentration increment on nucleation days exclusively caused by new particle formation (NPF). The second factor is NSF_{GEN} calculated as

$$\mathbf{NSF_{GEN}} = \frac{\left(\frac{\mathbf{N_{smallest\ size\ available-100}}}{\mathbf{N_{100-largest\ size\ available}}}\right)_{\mathbf{all\ days}}}{\left(\frac{\mathbf{N_{smallest\ size\ available-100}}}{\mathbf{N_{100-largest\ size\ available}}}\right)_{\mathbf{non-nucleation\ days}}} \quad (7)$$

and gives a measure of the overall contribution of NPF on a longer span (Salma et al. 2017).

The dimensionless survival parameter P, as proposed in Kulmala et al. (2017), was calculated as

$$\mathbf{P} = \frac{\mathbf{CS'}}{\mathbf{GR'}}$$

where CS' = CS/(10^{-4} s^{-1}) and GR' = GR/(1 nm $hour^{-1}$). CS and GR values used were calculated with the methods mentioned at 2.2.2. An increased P parameter is an indication that a smaller percentage of newly formed particles will survive to greater sizes. Hence this is the inverse of particle survivability, and values of P<50 are typically required for NPF in clean or moderately polluted environments, although higher values of P are observed in highly polluted atmospheres (Kulmala et al, 2017).

3. RESULTS AND DISCUSSION

3.1 NPF Events in the Background Areas

3.1.1 Conditions and trends of NPF events

The number of NPF event days for each site per year, those that took place simultaneously at both urban and rural background sites, as well as those events that took place at all three sites simultaneously appear in Table 1. Given that overall data recovery was in the range of 74-83%, results from individual years are unreliable, but the seven-year runs should average out most of the effects of incomplete data recovery. The number of events is similar for Harwell and N. Kensington, with a frequency of about 7% of all days with data. There is a clear seasonal variation favouring summer and spring (Figure 2) for both areas of the study. A similar pattern of variation was found for N. Kensington by Beddows et al. (2015). In general, higher solar radiation, lower relative humidity, low cloud cover and higher pressure conditions, lower concentrations of pollutants as well as lower condensation sink are found when NPF events took place in all areas (Figure S2), as was also reported by Charron et al. (2007) for Harwell. While SO_2 is one of the main factors for NPF events to occur, concentrations are lower when events take place. This is indicative that SO_2 concentrations in these areas are sufficient for events to take place, and higher concentrations are likely to be associated with higher pollution and a higher condensation sink. The proxy for [H_2SO_4] was calculated for the background sites using the method outlined in Petäjä et al., (2009) and was found to be higher on event days for both background sites (results not included). This indicates the possible positive effect of increased concentrations of H_2SO_4 in the occurrence of NPF events as well as, since SO_2 concentrations were found lower, the increased role of either the solar radiation (via the formation of OH radical) or the reduced condensation sink to its formation. For the case of gaseous ammonia (results not included) for Harwell where data was available, as there was no distinct variation found between event and non-event days, but as the concentration of ammonia in the U.K. is in the range of few ppb (Sutton et al., 1995), it is sufficient according to ternary nucleation theory (Korhonen et al., 1999) for NPF events not to be limited by ammonia. The average growth rate for Harwell was found to be 3.4 nm h^{-1}, within the range given by Charron et al. (2007) and higher at N. Kensington at 4.2 nm h^{-1}, a trend found for all seasons (Figure 3). The increased growth rate in the urban area can be related to the greater presence of organic matter and other condensable species. In both areas NPF events had higher growth rates in summer than in spring, as was also found in previous studies (Kulmala et al., 2004; Nieminen et al., 2018). This may be associated with the higher concentration of organic compounds emitted by trees during summer (Riipinen et al., 2007), or faster oxidation rates due to higher concentrations of hydroxyl radical and ozone (Harrison et al., 2006).

About 45% of the events took place simultaneously in both background areas. These events are characterized as regional, as NPF took place on a larger scale, regardless of the local conditions of the given area. In this case, meteorological conditions were even clearer, indicative of the greater dependence of regional events on synoptic conditions rather than local. While most chemical constituents were also lower in concentration during regional events, different patterns were found for organic compounds and sulphate for each background area. In Harwell sulphate was higher during regional events, while in N. Kensington organic compounds were higher during regional events. This may be indicative of the variable role that specific chemical species have in condensational nanoparticle growth (Yue et al., 2010). In all cases though, the concentrations of these species were lower compared to the average conditions. Despite these differences, the growth rate of particles was found to be higher for local events in N. Kensington (4.4 nm h^{-1}) compared to regional events (3.9 nm h^{-1}), though within the margin of uncertainty. In Harwell, no difference was found in the growth rate between regional and local events.

3.1.2 Variability of the origin of the air masses on NPF events

As both background sites are relatively close to each other (about 80 km) and had similar number of event days, a combined clustering of back trajectories for the event days (only) in these two areas was attempted. This would provide an insight into the origin of air masses for local and regional events, as well as the conditions for these air masses. The data for local N. Kensington events and both local and regional events in Harwell were clustered together and the results along with the characteristics of the air mass clusters are found in Figure S3.

Cluster C3, which is placed between C2 and C4 among those originating from the Atlantic Ocean, has the highest percentage for both area specific and regional events. Specifically, for regional events the percentage is over 35%, much higher compared to all other, showing a clear “preference” of regional events for cleaner and faster moving air masses from mid-latitudes of the Atlantic Ocean. This “preference” explains the lower production and growth rate of the new particles found for regional events, compared to local ones, as air masses from this area have lower organic carbon and SO_2 concentrations. Cluster C5, originating straight from the north but representing air masses that have crossed the Irish Sea and have not extensively gone over land presents a similar case. These cold and clean air masses are associated with a low growth rate and survivability of the newly formed particles. Local events for both sites apart from those in Cluster C3 are highly associated with Clusters C1 and C2. C1, which contains slow and polluted air masses, presents the highest growth rate and as a result high particle survivability, as given by the P parameter (see later). On the other hand, C2 which consists of warm and moist air masses from lower latitudes is the least common for regional events and presents high growth rate and survival probability of the particles. Apart from the weak relation found with particulate organic carbon concentrations and growth rate (Figure S3), there appears to be an inverse relation between the temperature and survivability of the particles. Warmer air masses seem to be related to higher particle survival probability, which may be attributable to greater growth rates as temperature increases (Yli-Juuti et al., 2011).

3.1.3 Urban increment and particle development

The urban environment, depending on the conditions, may have a positive or negative effect in the number of the particles formed and their consequent survival and growth. Both Harwell and N. Kensington are in background areas, rural and urban respectively. As a result, while the concentrations of pollutants are higher in N. Kensington than Harwell, their effect is smaller compared to that of Marylebone Road. A comparison of the particles smaller than 20 nm, gives insight into the formation and survival of the newly formed particles in the initial stages. Calculating the urban increment (equation 5) using the two background sites showed around 20% more particles of size 16 - 20 nm in N. Kensington than Harwell for event days, an increment that is even stronger when solely local events are considered (Figure 4). As the sizes of the particles in the calculation are relatively large and due to the higher condensation sink found in N. Kensington, this increment is expected to be larger for smaller size particles. A possible explanation for this result may be the greater concentration of organic compounds which is observed in N. Kensington, as discussed earlier, which leads to more rapid formation of secondary condensable species that enhances the nucleation process in the more polluted area.

Considering the local events, most of the pollutant concentration data available appear to be higher which is reflected in the condensation sink as well. The role of the polluted background appears to be decisive in the further growth of the newly formed particles, especially for Harwell. This, at both sites causes the number of particles of greater size to be smaller for the later hours in the days of local events (Figure S4). Another possible reason for this difference in the larger size ranges can be the higher concentration of organic content on the days of regional events at N. Kensington (as discussed earlier). On the other hand, for Harwell all hydrocarbons with available data are lower throughout the day (apart from ethane) during regional events. Unlike N. Kensington, at Harwell particles smaller than 20 nm as well as the growth rate of the newly formed particles are almost the same for regional and local events.

The calculation of the increment in Marylebone Road provided negative results; particles smaller than 20 nm were less abundant on event days compared to the average, throughout the day. This is due to the fact that Marylebone road is heavily affected by traffic pollution and on average, conditions do not promote NPF events due to the high condensation sink, unless clear conditions prevail, which are also associated with a low particle load.

3.2 NPF Events at Marylebone Road

For many years, NPF events were thought not to take place in heavily polluted urban areas, as the effect of the increased condensation sink was considered crucial in suppressing the formation and growth of new particles. Recent long term analyses have shown this is not the case and nowadays an increasing number of studies confirm the occurrence of NPF events in urban areas. In this study, for the same period of seven years as for the two background areas, NPF events were found to occur for 6.1% of days at Marylebone Road, lower than in the background areas. Though, due to the particle size range available there cannot be a definitive answer to whether the formation of the particles takes place in the specific locality of the sampling site, due to the observed increase in particle concentrations in the range 7 – 16 nm (provided by the CPC data) and the increased growth rates found in urban areas in general, it can be assumed that the formation takes place either in the area of the measuring site or in its close vicinity, while the growth of the particles persists in the area for several hours, despite the high condensation sink. Seasonal variation is similar to that at the background sites, but day of the week variation is stronger at Marylebone Road further favouring weekends (Figure S5), as on these days traffic intensity is lower.

In general, similar conditions found to affect NPF events at the background sites are also found at Marylebone Road, despite a much larger condensation sink. (Figure S2). As a result, less particles of size smaller than 20 nm were found on NPF event days than the average for the site, as the sum of background particles plus those formed on these days were less than that on an average day. The growth rate of the newly formed particles (5.5 nm h^{-1}), is higher than that of the background sites which is in agreement with the findings in the study of the background areas on the possible role of the condensable species, the concentrations of which are even greater at the urban kerbside. About 15% of NPF event days at Marylebone Road presented particle shrinkage after the initial growth; the study of these cases though is outside of the context of the present work. At Marylebone Road, the number of NPF days which were common with the background sites was fewer, as local conditions (high condensation sink) are detrimental to the occurrence of NPF events and thus the days of regional events including Marylebone Road were separately studied for this site. The regional event days that were common for all three sites were 37 (31% of events at Marylebone Road) (Table 1). As with the other two areas, the growth rate is higher during local events, but the conditions are mixed, with lower concentrations of sulphate and organic compounds but higher SO_2, NOx and elemental carbon. The relationship with higher wind speed (mainly western) (Figure S6), solar radiation (which results in greater H_2SO_4 formation) and lower relative humidity, indicate the stronger relation of the regional events with synoptic conditions than the local events in the heavily polluted environment of Marylebone Road.

3.3 Connection of NPF Events with Incoming Air Masses

3.3.1 Air mass back trajectory clustering and connection with NPF events

The origin of the air masses plays a very important role in the occurrence of NPF events, as shown in Section 3.1.2. Air masses of different origins have different characteristics. Back trajectories provide excellent insight into the source of the air masses. Air mass back trajectories were calculated both for all days and for NPF event days for each site separately, with the aim of complementing the analysis in Section 3.1.2 which addressed only the event days. The additional analysis gives a view of the frequency of NPF events within different air mass types. The initial air mass back trajectory clustering ended up with an optimal solution of 9 clusters of different air masses. As many of these clusters had similar characteristics and origin, solutions with fewer clusters were attempted. As the number of clusters was decreasing clusters became a mixture of different origins, thus making the distinction of different sources harder. As a result, the method chosen was to merge clusters of similar origin and characteristics, which kept the detail of the large number of clusters and made the separation of the different origins more distinct.

The resulting four merged clusters (Figure 5), using the characterisation proposed by McIntosh et al. (1969) are:

- An **Arctic** cluster, which originates mainly from the northerly sector. It occurs about 10% of the time and consists of cold air masses, which either passed over northern parts of the U.K. or through the Irish Sea.

- A **Tropical** cluster, which originates from the central Atlantic. It occurs 25% of the time and contains warmer air masses. A small percentage of this cluster contains masses that have passed over countries south of the U.K. Even though these days were more polluted, the clustering method was unable to clearly distinguish these days as it does not take into account particle numbers or composition, even when the 9-cluster solution was applied.
- A **Polar** cluster, which originates from the north Atlantic. It is the most common type of air mass arriving in the areas of study and occurs about 40% of the time bringing fast moving, "clean" air masses with increased marine components (Cl, Na, Mg) from the west. This cluster also contains airmasses that have passed through Ireland, though an effect on particle size and chemical composition is not distinct.
- A **Continental** cluster, which originates from the east. It occurs about 25% of the time and consists mainly of slow moving air masses, originating from the London area (for the background areas) and/or continental Europe. It has higher concentrations of most pollutants as well as the highest condensation sink.

The occurrence of each air mass class for average and event days for Harwell and London (both sites) can also be found in Figure 5, while their main characteristics for each site can be found in Table S2. Though in this case the air mass grouping for each site was done in a different analysis, the resulting groups are almost identical in their characteristics and frequency, as the sites are close to each other.

The Polar cluster is the one prevailing on both average and event days. This consists of clean fast-moving air masses originating mainly from mid and high latitudes of the Atlantic, and this cluster presents favourable conditions for NPF events. The association of NPF events with air masses from the mid-Atlantic at N. Kensington was also found by Beddows et al. (2015). Cool Arctic air masses on average are not clean as they may have passed over the northern U.K. The event days associated with this air mass type have the lowest concentrations of the pollutants within available data for all areas. The increased percentage of events with this air mass at all sites indicates that lower temperatures, in a clear atmosphere with sufficient solar radiation are favourable for NPF events as found in previous studies (Napari et al., 2002; Jeong et al., 2010; Kirkby et al., 2011). A similar trend of increased probability with polar and arctic maritime air masses was also found for Hyytiälä, Finland by Nilsson et al. (2001). Tropical air masses have a lower probability for NPF events, which is associated with the fact that a number of these days are associated with air masses which have passed from continental areas south of the U.K. (France, Spain etc.). Specifically for Marylebone Road the NPF probability is a lot lower (11% versus 17% for N. Kensington and 20% for Harwell). This is due to the fact that these air masses are more related to southerly winds which, in Marylebone Road are associated with a street canyon vortex which causes higher pollutant concentrations at this site. Finally, the Continental cluster presents the lowest probability for NPF events. The air masses in this group originate from continental Europe and for the background areas in most cases have passed over the London region as well. This results in both a higher condensation sink and concentration of pollutants, which limits the number of days with favourable conditions for NPF events. Growth rate for all sites though appears to be higher for air masses originating from more polluted areas (Figure 6), which appear to enhance the growth process due to containing a higher concentration of condensable species (after oxidation).

3.4 Nucleation Strength Factor (NSF)

The NSF (equations 6 and 7) is used to describe the effect nucleation events have on the number of particles at a site. The values of NSF for each site and for seasons spring and summer are shown in Table 2. The decrease of the contribution of NPF events to particle number, moving from the rural area to the kerbside was also found in previous studies (Salma et al., 2014; 2017). This is explained by the increased contribution to the particle number concentrations of other sources, mainly combustion in the urban environment, compared to rural areas. Apart from this trend, in the background areas the increase of $N_{16\text{-}100}$ was greater in spring than summer. This effect seems stronger in the urban background area compared to the rural, as in that area the variability of $N_{16\text{-}100}$ is greater for event days compared to that of the rural area. On the other hand, the contribution of NPF events in the longer span, as is illustrated by the NSF_{GEN} appears to favour summer for all areas, showing the increased formation and survivability of particles in this season.

For Marylebone Road the result for the increase of the $N_{16\text{-}100}$ is greater in summer than in spring, in contrast to what was found for the background sites. This is due to the fact that in summer the traffic intensity is decreased, giving the contribution from NPF events a stronger effect compared to the other sources. The very small increase found on NPF events in Marylebone Road, with a factor of just 1.26, a lot lower than that found in the urban area of Seoul, South Korea (Park et al., 2015), is indicative of the reduced effect of NPF events in an area which is heavily affected by traffic, as also pointed out by Von Bismarck-Osten et al. (2013) in their study on particle composition in Marylebone Road.

3.5 The Survival Parameter P

The average values of the P parameter for each of the areas of this study are 10.5 for Harwell, 15.8 for N. Kensington and 28.9 for Marylebone Road. The values found put Marylebone Road to the upper end of heavily polluted areas in Europe, North Kensington to the same level as many other urban areas in Europe, while Harwell had somehow higher values compared to other rural background areas in Europe, as calculated by Kulmala et al. (2017). The seasonal, air mass origin and local versus regional variations can be found in Figure 7 (winter is excluded due to very low number of events). While the increasing trend of the P parameter as we move from rural background to kerbside was expected, it can be seen that there is a clear seasonal pattern in all three areas, with summer having the lowest P parameter (greatest survivability) compared to the other two seasons. This is associated with the higher growth rate found in summer for all areas of this study, as the differences in the condensation sink on event days are negligible between seasons. The case is similar for regional and local events. The result per air mass origin is related to the different conditions and parameters of each incoming air mass in each area. For example, the higher P parameter for Tropical air masses at Marylebone Road, is associated with the higher condensation sink found for this kind of air masses, due to the street canyon effect which is specific for Marylebone Road for southerly wind directions with which these air masses are mainly related, while the higher values for the rather clean Arctic air masses for the other two areas are associated with the lower growth rates found for this kind of air mass in these areas. The more polluted Continental air masses seem to have a different effect for rural and urban areas. Their higher condensation sinks and concentrations of pollutants have a negative effect on P-values for the rural site and a positive effect at the urban sites. The exact opposite is found for the cleaner air masses of the Polar cluster, which appear to result in reduced P-values of the newly formed particles at the urban sites. This is related to the lower condensation sink associated with this air mass type.

4. CONCLUSIONS

Seven years of data from three distinct areas (regional background, urban background, kerbside) in the southern U.K. were analysed and the conditions associated with NPF events were studied. NPF events were found to occur on about 7% of days at background sites and less at the kerbside site. The conditions on event days for all three areas were similar, with clear atmospheric conditions and a lower condensation sink. While the condensation sink appears to be the most important factor limiting NPF events at the kerbside site, SO_2 was found to have smaller concentrations on event days for all areas, which indicates that either on average it is in sufficient concentrations for NPF events to occur, or that other variables that participate in the production mechanism of H_2SO_4 are more important. The growth rate of the newly formed particles increases from the rural site to the kerbside and is greater in summer compared to other seasons for all three sites. Almost half of the NPF events at the rural and urban background sites were found to happen simultaneously. In these cases, the atmospheric conditions were cleaner, which resulted in slower growth rates. While most of the chemical species available were at lower concentrations in regional events, a difference in the behaviour with respect to sulphate and organic compounds was found between the two background site types.

The prevailing origin of air masses in the southern U.K. is from mid and high latitudes of the Atlantic Ocean. These fast-moving air masses present an increased probability for NPF to occur. The case is similar for the cooler and cleaner arctic air masses, while air masses from the tropics and continental Europe, having greater pollutant content, have decreased NPF probability, but a higher growth rate of particles when NPF events occurred. Regional events appear to be more associated with cleaner air masses, presenting a smaller growth rate and condensation sink compared to local events. The difference in growth rate is probably related to the greater content of condensable species; a positive relation of particle survival probability with temperature was also found.

Comparing the background areas in this study, particles of 16-20 nm were found to be about 20% greater in concentration (above long-term average) on NPF event days at the urban background site compared with the rural site. This is associated with a higher abundance of condensable species in the urban environment, which enhances the nucleation and growth process. This effect though is limited as particle size increases and NPF events have a greater effect on the overall $N_{<100\ nm}$ in the rural areas, compared to urban, as calculated by the NSF. The effect becomes even smaller at the kerbside as the number of background particles emitted by traffic is a lot greater.

The occurrence of NPF events at the highly polluted Marylebone Road site is at first sight surprising given the elevated condensation sink. This must be counteracted by an abundance of condensable material, which is surprising given the generally modest rate of atmospheric oxidation processes in comparison to residence times in a street canyon (Harrison, 2017). However, Giorio et al. (2015), using Aerosol Time-of-Flight Mass Spectrometry, reported rapid chemical processes within the Marylebone Road street canyon leading to production of secondary particulate matter from road traffic emissions. They postulated that this resulted from very local gas to particle conversion from vehicle-emitted pollutants. Condensation of such reaction products upon pre-existing particles could explain the enhanced particle growth rates observed at Marylebone Road (Figure 3).

Finally, particle survival probability was found to decrease moving from rural to urban areas. While formation and initial growth of new particles is increased in urban areas, their survivability reduces as their size increases. The probability of particles to survive to greater sizes was found to be increased in summer for all areas, which is also explained by the higher growth rate. The probability is also different depending upon the origin of the air masses and is related to conditions specific for each area.

In the present work, the effects of atmospheric conditions upon the NPF process are studied. NPF is a complex process, highly affected by meteorological conditions (local and synoptic), the chemical composition as well as the pre-existing conditions in an area. For this reason, the study of NPF events in one area cannot provide safe assumptions for other areas, as the mixture of conditions found in different places is unique and alters the occurrence and development of NPF events. Thus, more studies on the conditions and the trends in NPF events should be conducted to better understand the effect of the numerous variables that affect those processes.

DATA AVAILABILITY

Data supporting this publication are openly available from the UBIRA eData repository at https://doi.org/10.25500/edata.bham.00000307.

AUTHOR CONTRIBUTIONS

This study was conceived by MD and RMH who also contributed to the final manuscript. The data analysis was carried out by DB with guidance from DCSB, and DB also prepared the first draft of the manuscript. FDP provided advice on the analysis.

COMPETING INTERESTS

The authors have no conflict of interests.

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TABLE LEGENDS:

Table 1: Number of NPF events per site.

Table 2: Annual and seasonal NSF for all areas of study.

FIGURE LEGENDS:

Figure 1: Map of the measuring stations.

Figure 2: Number of NPF events per season for all seven years of the present study (Winter – DJF; Spring – MAM; Summer – JJA; Autumn – SON) at Harwell (rural), N. Kensington (urban background) and Marylebone Road (urban roadside).

Figure 3: Growth rate per season at the three sites.

Figure 4: Diurnal variation of $N_{16\text{-}20nm}$ at each site: annual average and NPF event days.

Figure 5: Map and frequency of incoming air mass origin – average and for NPF events per site.

Figure 6: Growth rate per incoming air mass at each of the sites.

Figure 7: Survival parameter P (a) per season, (b) for regional and local events (for Marylebone Road) is regional for all 3 sites and (c) by incoming air mass origin.

Table 1: Number of NPF events per site.

	Harwell	**N. Kensington**	**Marylebone Road**	**Regional (Background sites)***	**Regional (All 3 sites)****
2009	9	0	4	0	0
2010	29	22	22	11	9
2011	15	10	23	4	1
2012	8	28	12	3	0
2013	25	23	27	13	11
2014	29	34	13	18	6
2015	25	22	18	11	10
Overall	140	139	119	60	37

* Refers to events occurring simultaneously at Harwell and N. Kensington
** Refers to events which occur simultaneously at all three sites

Table 2: Annual and seasonal NSF for all areas of study.

	Harwell	**N. Kensington**	**Marylebone Road**
NSF_{NUC} (Spring)	2.04	2.03	1.20
NSF_{NUC} (Summer)	2.01	1.72	1.26
NSF_{NUC} (Year)	2.25	1.86	1.26
NSF_{GEN} (Spring)	1.10	1.07	1.02
NSF_{GEN} (Summer)	1.18	1.11	1.01
NSF_{GEN} (Year)	1.10	1.06	1.02

Figure 1: Map of the measuring stations.

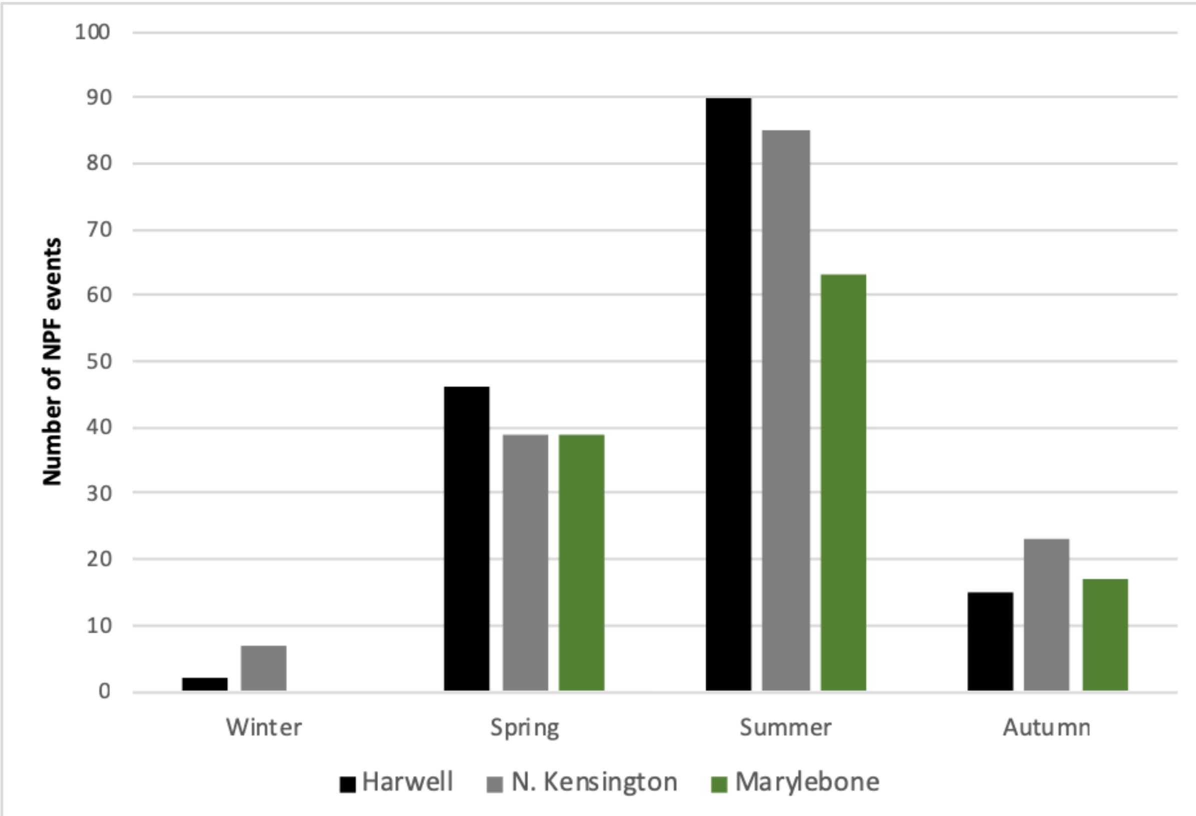


Figure 2: Number of NPF events per season for all seven years of the present study (Winter – DJF; Spring – MAM; Summer – JJA; Autumn – SON) at Harwell (rural), N.Kensington (urban background) and Marylebone Road (urban roadside).

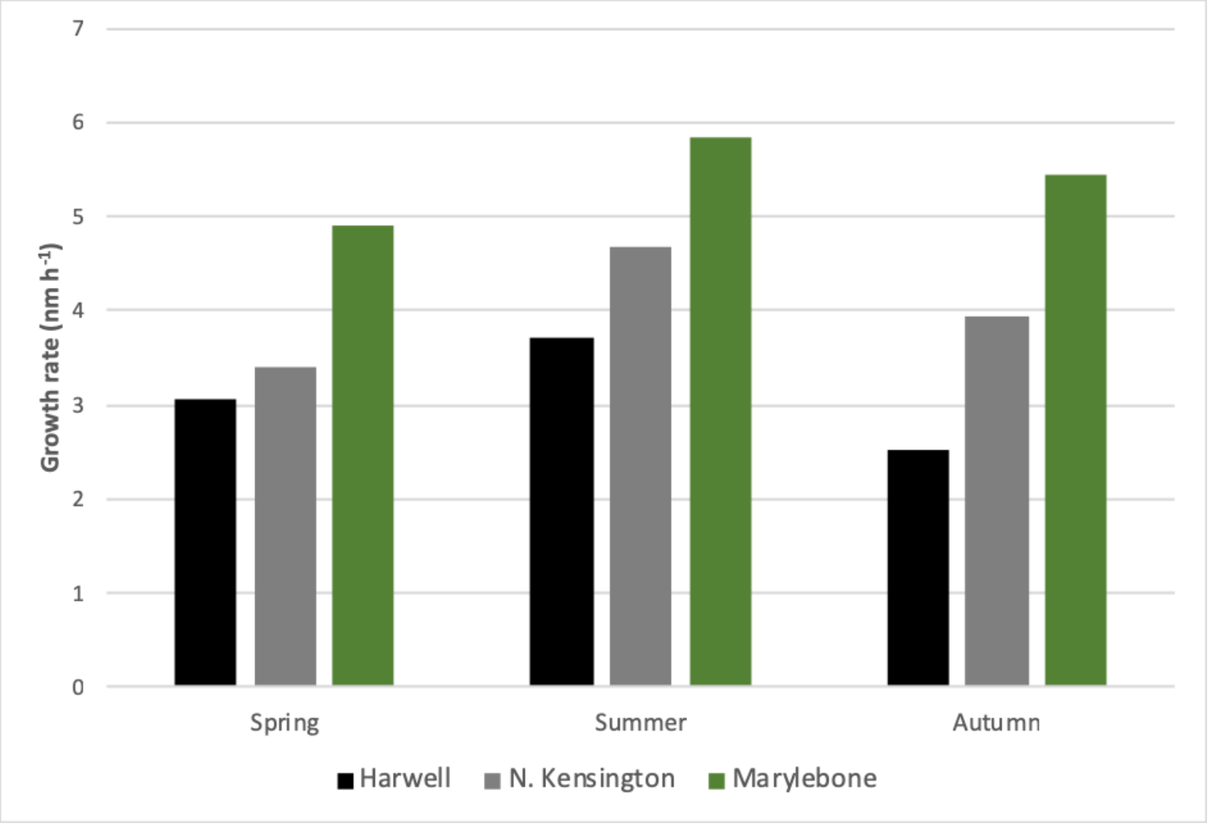


Figure 3: Growth rate per season at the three sites.

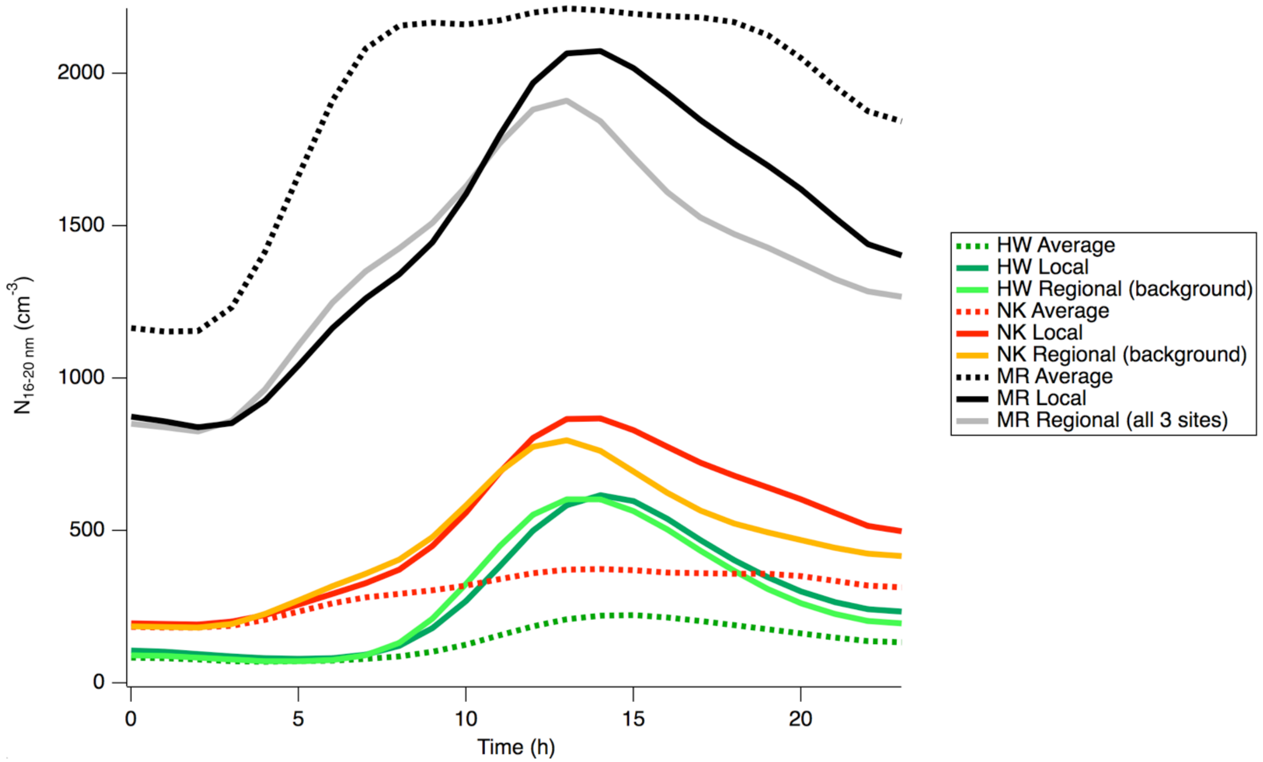


Figure 4: Diurnal variation of $N_{16\text{-}20nm}$ at each site: annual average and NPF event days.

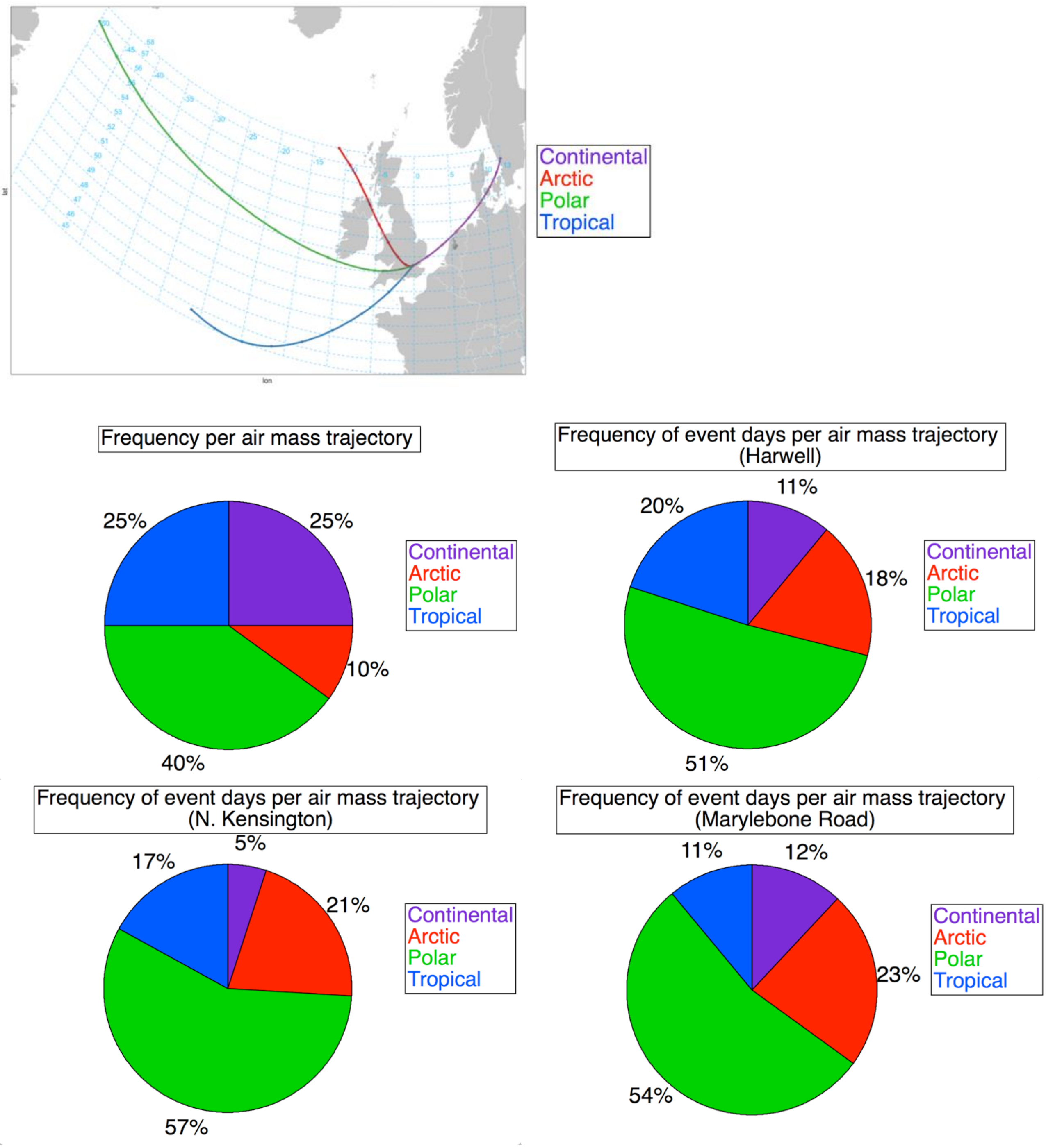


Figure 5: Map and frequency of incoming air mass origin – average and for NPF events per site.

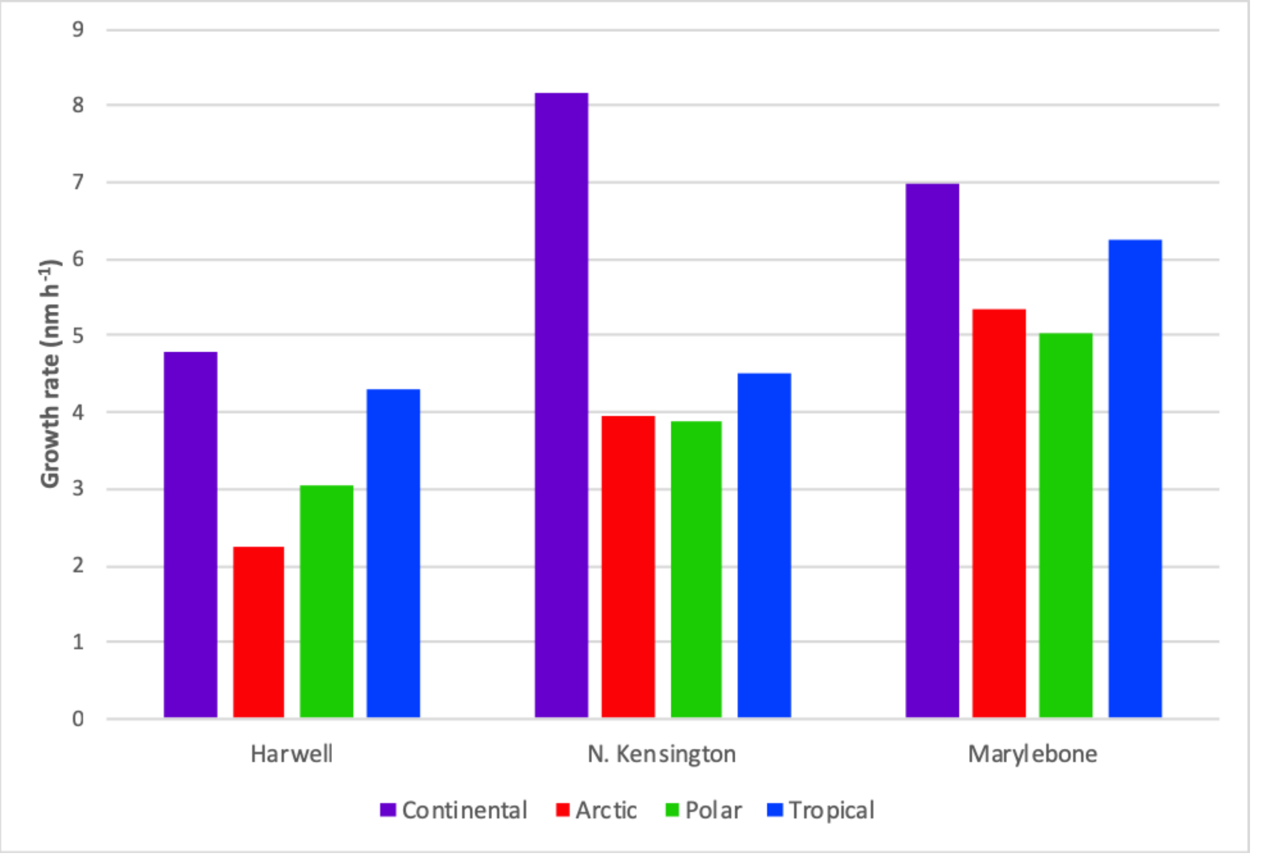
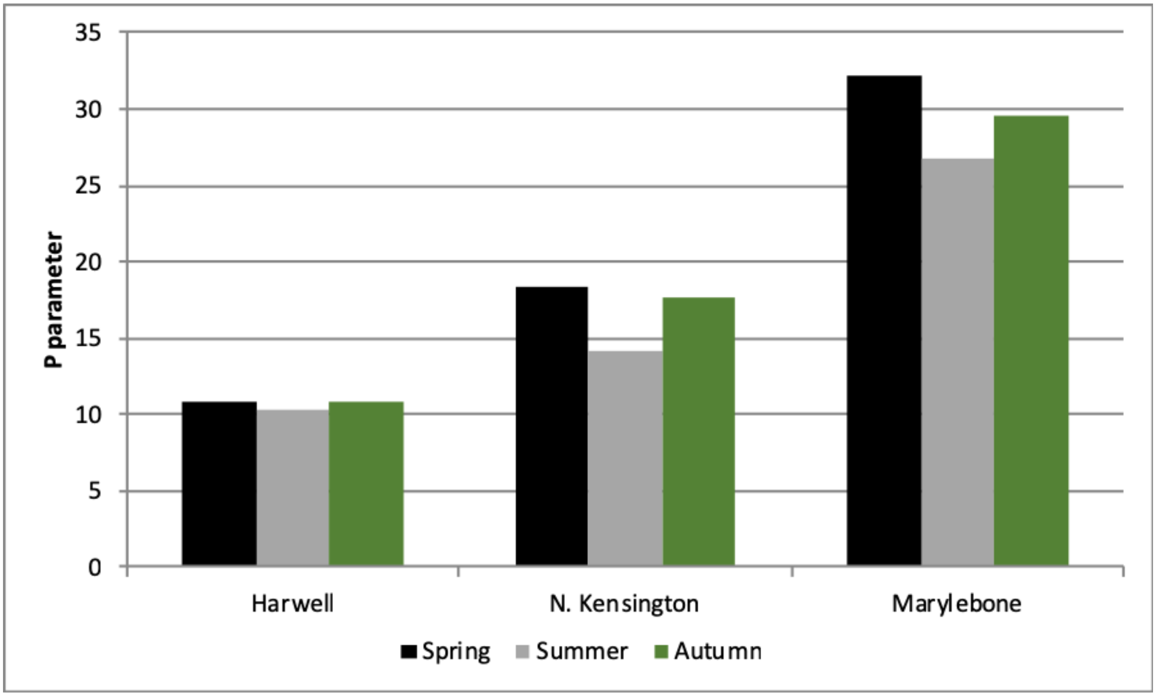
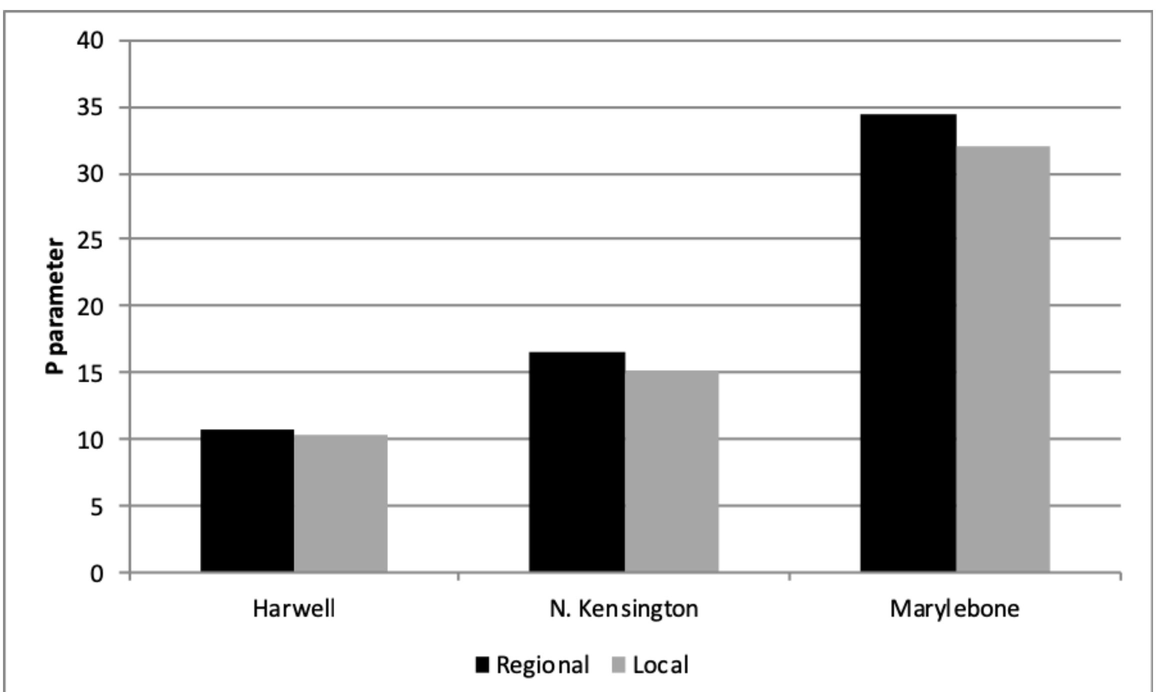


Figure 6: Growth rate per incoming air mass origin at each of the sites.

(a)


35
30
25
20
15
10
5
0
P parameter
Harwell
N. Kensington
Marylebone
Spring
Summer
Autumn

(b)


40
35
30
25
20
15
10
5
0
P parameter
Harwell
N. Kensington
Marylebone
Regional
Local

(c)

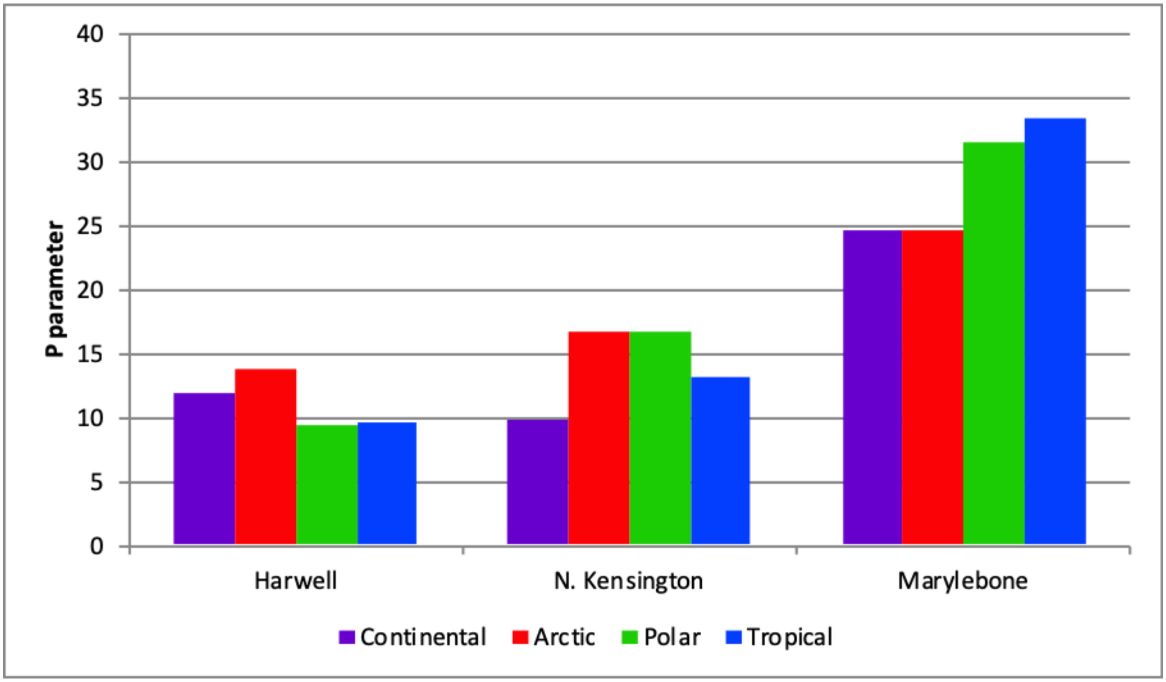


Figure 7: Survival parameter P (a) per season, (b) for regional and local events (for Marylebone Road regional is for all 3 sites) and (c) by incoming air mass origin.