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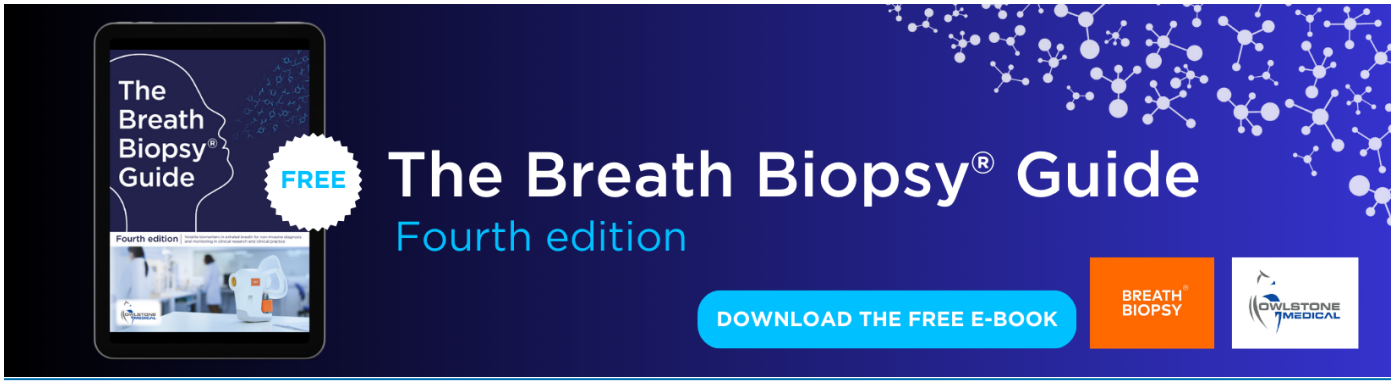

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On the contribution of organics to the North East Atlantic aerosol number concentration

Jakub Bialek[1], Manuel Dall'Osto[1,2], Ciaran Monahan[1], David Beddows[3] and Colin O'Dowd[1,4]

[1] School of Physics and Centre for Climate and Air Pollution Studies, Ryan Institute, National University of Ireland Galway, University Road, Galway, Ireland
[2] Institute for Environmental Assessment and Water Research (IDAEA-CSIC), Barcelona, Spain
[3] National Centre for Atmospheric Science, Division of Environmental Health and Risk Management, School of Geography, Earth and Environmental Sciences, University of Birmingham, Edgbaston, Birmingham B15 2TT, UK

E-mail: colin.odowd@nuigalway.ie

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Abstract
k-means statistical-cluster analysis of submicron aerosol size distributions is combined with coincident humidity tandem differential mobility analyser data, leading to five unique aerosol categories for hygroscopic growth factors (HGFs): low sea-salt background marine, high sea-salt background marine, coastal nucleation, open ocean nucleation and anthropogenically influenced scenarios. When considering only marine conditions, and generic aerosol species associated with this environment (e.g. non-sea-salt sulfate, sea-salt, partly soluble organic matter and water insoluble organic matter), the two-year annual average contribution to aerosol number concentration from the different generic species was made up as follows: 46% (30–54%) of partially modified ammonium sulfate particles; 23% (11–40%) of partially modified sea-salt; and the remaining 31% (25–35%) contribution attributed to two distinct organic species as evidenced by different, but low, HGFs. The analysis reveals that on annual timescales, ~30% of the submicron marine aerosol number concentration is sourced from predominantly organic aerosol while 60% of the anthropogenic aerosol number is predominantly organic. Coastal nucleation events show the highest contribution of the lowest HGF mode (1.19), although this contribution is more likely to be influenced by inorganic iodine oxides. While organic mass internally mixed with inorganic salts will lower the activation potential of these mixed aerosol types, thereby potentially reducing the concentration of cloud condensation nuclei (CCN), pure organic water soluble particles are still likely to be activated into cloud droplets, thereby increasing the concentration of CCN. A combination of dynamics and aerosol concentrations will determine which effect will prevail under given conditions.

Keywords: marine aerosol, organic aerosol, cloud condensation nuclei, aerosol hygroscopicity
S Online supplementary data available from stacks.iop.org/ERL/7/044013/mmedia

1. Introduction

The hygroscopic growth behaviour of aerosol particles is one of the important parameters controlling the direct and indirect climate effects because, due to water uptake,

[4] Author to whom any correspondence should be addressed.

aerosol particles change their size and their optical properties (Tang 1996) with scattering efficiency typically increasing with increasing particle size. The major proportion of hygroscopic particle growth is usually related to inorganic species such as ammonium nitrate, ammonium sulfate and sodium chloride—common species whose hygroscopic growth behaviour is known relatively accurately (Tang and Munkelwitz 1994). For example, the hygroscopic growth factor (HGF, defined here as the ratio the particle diameter exposed to 90% relative humidity to that at 40% relative humidity) for pure sea-salt is 2.2 while that for pure ammonium sulfate is 1.8 for accumulation mode sizes (specifically, 165 nm diameter). In contrast, hydrocarbon-like organic species possess a HGF close to 1, while oxygenated organic species possess a HGF of the order of 1.2–1.4 (Liu and Wang 2010). Organics often contribute substantially to fine aerosol mass and potentially impact on hygroscopicity (Kanakidou *et al* 2005, O'Dowd *et al* 2004); however, their contribution to the number population and distribution is not often quantified, particularly through measurements. HGF analysis, in addition to elucidation of aerosol hygroscopicity, can also elucidate both the state of mixing of the aerosol population (i.e. whether the aerosol is internally mixed or externally mixed), and the contribution of generic aerosol types to the number concentration. Number concentration is important particularly in terms of influencing cloud droplet number concentration and the indirect radiative effect of aerosols on cloud microphysics—specifically, all else being equal, an increase/decrease in the number of CCN results in a concomitant increase/decrease in droplet concentration, a decrease/increase in effective radius, and ultimately, an increase/decrease in cloud albedo (Twomey 1974). At a given saturation, the CCN concentration is determined by the number concentration of particles of a given hygroscopicity and state of mixing. Chemical composition drives hygroscopicity and therefore, the concentration of particles possessing a particular hygroscopic grouping can elucidate the contribution of particular aerosol chemical type to the aerosol and CCN population. Given that even the organic aerosol in clean marine air with low hygroscopicity can be efficient CCN (in contrast to polluted air) (Ovadnevaite *et al* 2011a), hygroscopic growth factor measurements provide a useful approach towards quantification of the relative contribution of different aerosol species, or types, to the CCN population.

This study deployed a humidity tandem differential mobility analyser (H-TDMA) to quantify the relative contribution of predominantly organic and predominantly inorganic aerosol in marine and continental air over the North East Atlantic. Previous studies, using bulk chemical analysis, have demonstrated a strong seasonal cycle in organic enrichment in clean marine aerosol, with peak enrichment occurring during summer (O'Dowd *et al* 2004). The fractional organic mass increased with decreasing particle size. More elaborate on-line mass spectrometry used for chemical characterization of primary organic plumes point to primary organic sea-spray number concentrations exceeding 300 cm^{-3} (Ovadnevaite *et al* 2011b) under certain conditions. In the latter study, the primary organic spray was characterized by HGFs of the order of 1.2 or less (Ovadnevaite *et al* 2011a).

Prior to the aforementioned study, typical HGFs reported for marine aerosols were more characteristic of predominantly inorganic (sulfate and sea-salt) aerosol species (for example, over the western North Pacific (Maßling *et al* 2007)); over the eastern North Pacific (Berg *et al* 1998); along with other oceanic regions Swietlicki *et al* 2000, Zhou *et al* 2001, Maßling *et al* 2003, Tomlinson *et al* 2007, Good *et al* 2010). These studies were campaign based which, consequently, may have missed periods of high biological activity if present. In contrast, analysis of continental aerosol sampled at the same location (Mace Head) revealed multiple HGF modes including low hygroscopicity modes at 1.1 and 1.2 and were also associated with high organic mass fractions (Dall'Osto *et al* 2010). The longest time series for HGF in marine air exists for Mace Head where measurements started in June 2008. Two years of HGF data, covering 2009–10, are analysed in the present work.

2. Experimental details

The Mace Head Atmospheric Research Station is located in Connemara, County Galway on the Atlantic Ocean coastline of Ireland at 53°19′36″ N, 9° 54′14″ W and offers a clean sector from 190° through to 300°. Meteorological records show that, on average, over 60% of the air masses arriving at the station are from the clean sector (Jennings *et al* 2003, O'Connor *et al* 2008). Air is sampled at 10 m height from a main air inlet positioned at 80–120 m from coastline depending on tide height (available at: www.macehead.org). Size distributions were sampled using a Thermo Systems Inc. scanning mobility particle sizer (SMPS) operating 10 min size distribution scans between 20 and 500 nm (Wang and Flagan 1990) and a nano-SMPS covering sizes 3–20 nm.

Hygroscopic properties of aerosol were measured using a H-TDMA, as described in Nilsson *et al* (2009), and Rader and McMurry (1986). A higher HGF indicates more hygroscopic particles resulting from a higher affinity for water. The Mace Head H-TDMA incorporates a dry DMA (RH < 40% dried with Nafion^{TM} dyer), Gore-Tex^{TM} humidifier and a second DMA placed in a temperature controlled box for stabilization of the relative humidity. In the humidifier, the aerosol sample is humidified to 90% relative humidity. Particle diameter is given as a mobility diameter—for volume equivalent diameter one should apply a correction factor of 1.08 to the generally cubic sea salt mode. HGFs are determined for dry size particles of 35, 50, 75, 110 and 165 nm. An inversion algorithm is used to retrieve the HGF probability distribution function (PDF), corrected for multiple charges, inherent broadening of measured distribution and for humidity variations in the second DMA, following Gysel *et al* (2009). The standard deviation (σ) of the HGF–PDF is sensitive to small values of total particle concentration (n_{total}) with an accuracy of ±80%, ±50%, ±10% and ±5% for n_{total} 10, 50, 1000 and 5000, respectively. In the marine environment, such noise occurs sometimes during 'clean sector' advection with relatively low wind speed (below 5 m s^{-1}). Additionally,

the sensitivity of σ to small n_{total} decreases with increasing breadth of the HGF–PDF, which, in the marine environment, is often broad due to the external mixing of sea-salt, organic compounds and inorganic salts. Overall, the mean HGF σ, due to counting statistics is between 5 and 10% of the HGF in the marine environment.

3. Data analysis

Following the study of Dall'Osto *et al* (2011), one-hour average SMPS size distributions were analysed by using *k*-means cluster analysis (Beddows *et al* 2009). The size distributions were normalized by their vector length and cluster analysed. The use of cluster analysis was justified in this work using a cluster tendency test, providing a calculated Hopkins index of 0.20, implying the presence of cluster structures in the dataset. The *k*-means method minimizes the sum of the squared distances between all points and the cluster centre and identifies homogeneous groups by minimizing the clustering error (defined as the sum of the squared-Euclidean distances between each dataset point and the corresponding cluster centre). The clustering reduces the complexity of the dataset and this simplification allows easier characterization according to temporal and spatial trends. In order to choose the optimum number of clusters the Dunn-index (DI) was used, which aims to identify dense and well-separated clusters. The DI is defined as the ratio between the minimal inter-cluster distance to maximal intra-cluster distance. Since the method's internal criteria seeks clusters with high intra-cluster similarity and low inter-cluster similarity, algorithms that produce clusters with high DI are more desirable. In other words, for Dunn's index, it was required to find clustering which maximizes this index. The Dunn-index for the results of the *k*-mean cluster analysis for different cluster numbers showed a clear maximum for 15 clusters for the 2009 dataset and 11 clusters for the 2010 dataset, which were further grouped into five generic categories. There was 72% data coverage for the year 2009 and 81% for 2010 amounting to more than 10 000 h over coincident H-TDMA and SMPS measurements. Following Dall'Osto *et al* (2011), the 15 (for 2009) and 11 (for 2010) clusters were further grouped into five more generic aerosol types found in the region. This final re-grouping was fundamentally based on size distribution similarities, as per the primary clustering techniques outlined above and in more detail in the supplementary information section (available at stacks.iop.org/ERL/7/044013/mmedia), and also on similarities in air mass categorization and meteorological parameters. The combined set of parameters allowed the distinction between two typical background marine distributions, one with low sea-salt concentrations and the other with high sea-salt concentrations; another marine background distribution type but occurring under open ocean new particle production and growth events (O'Dowd *et al* 2010); another with size distribution and associated aerosol properties typical of anthropogenically influenced aerosol; and finally, an aerosol type characterized by the presence of a strong nucleation mode resulting from coastal new particle production events. As outlined in Dall'Osto *et al* (2011), the background clean marine clusters occur predominantly in maritime tropical, polar and Arctic air masses; anthropogenic air masses are predominantly polar continental or polar continental marine; open ocean new particle production events are also predominantly in maritime tropical, polar and Arctic air masses as typically are coastal nucleation events.

4. Results and discussion

The clustering procedure identified the following five categories: (1) high sea-salt background marine (occurring for 9% of the data coverage period over the two years) typical of background clean marine size distributions with secondary mode visible in the larger accumulation mode sizes (300–400 nm diameter) during winter months; (2) low sea-salt background marine (occurring for 25% of the data coverage period) with a clear bimodality in the submicron size range; (3) open ocean nucleation (occurring for 23% of the period), characterized by a dominant Aitken mode between 15 and 50 nm with a spring seasonality peaking in May and corresponding to the North East Atlantic high biological activity period (O'Dowd *et al* 2010, Dall'Osto *et al* 2012); (4) anthropogenically influenced (occurring for 28% of the period), characterized by size distributions generally monomodal in submicron sizes; (5) coastal nucleation (occurring for 15% of the period), with a clear and dominant nucleation mode at sizes less than 10 nm.

The total particle number concentrations (average $\pm$ 1σ) for diameters $D > 10$ nm (N_{10}) and $D > 3$ nm (N_3) for high sea-salt background marine were 353 ± 159 cm^{-3} and 993 ± 248 cm^{-3}; for low sea-salt background marine were 680 ± 306 cm^{-3} and 2100 ± 525 cm^{-3}; for anthropogenically modified were 1443 ± 505 cm^{-3} and 2076 ± 519 cm^{-3}; for open ocean nucleation were 2039 ± 714 cm^{-3} and 8068 ± 2017 cm^{-3}; and for coastal nucleation cases were 5852 ± 2048 cm^{-3} and $19\,195 \pm 4800$ cm^{-3}, respectively. Given that the nucleation mode can overwhelm number concentrations, and that particles less than 35 nm in size are unlikely to be activated as cloud condensation nuclei, the contribution of different aerosol species, as derived from the H-TDMA analysis, is presented in terms of contribution of HGF modes at sizes larger than 35 nm, while accepting there is an interpolation uncertainty over the five H-TDMA sizes, from 35 to 165 nm. Consequently, the concentration of particles larger than 35 nm (N_{35}) diameter is also reported for the five clusters and the relative concentration of different HGF modes to total number concentration, discussed later, refers to N_{35}. Specifically, for high sea-salt background marine $N_{35} = 374 \pm 112$ cm^{-3}, for low sea-salt background marine $N_{35} = 352 \pm 106$ cm^{-3}, for open ocean nucleation $N_{35} = 665 \pm 200$ cm^{-3}, coastal nucleation $N_{35} = 701 \pm 210$ cm^{-3} and for anthropogenically influenced, $N_{35} = 1650 \pm 555$ cm^{-3}.

Associated with the size distribution clusters, there were distinct HGF–PDFs which can be characterized by different combinations of HGF categories. Specifically, four HGF modes were found, although with slightly different HGF bins compared to those reported by Swietlicki *et al* (2000): 1–1.2

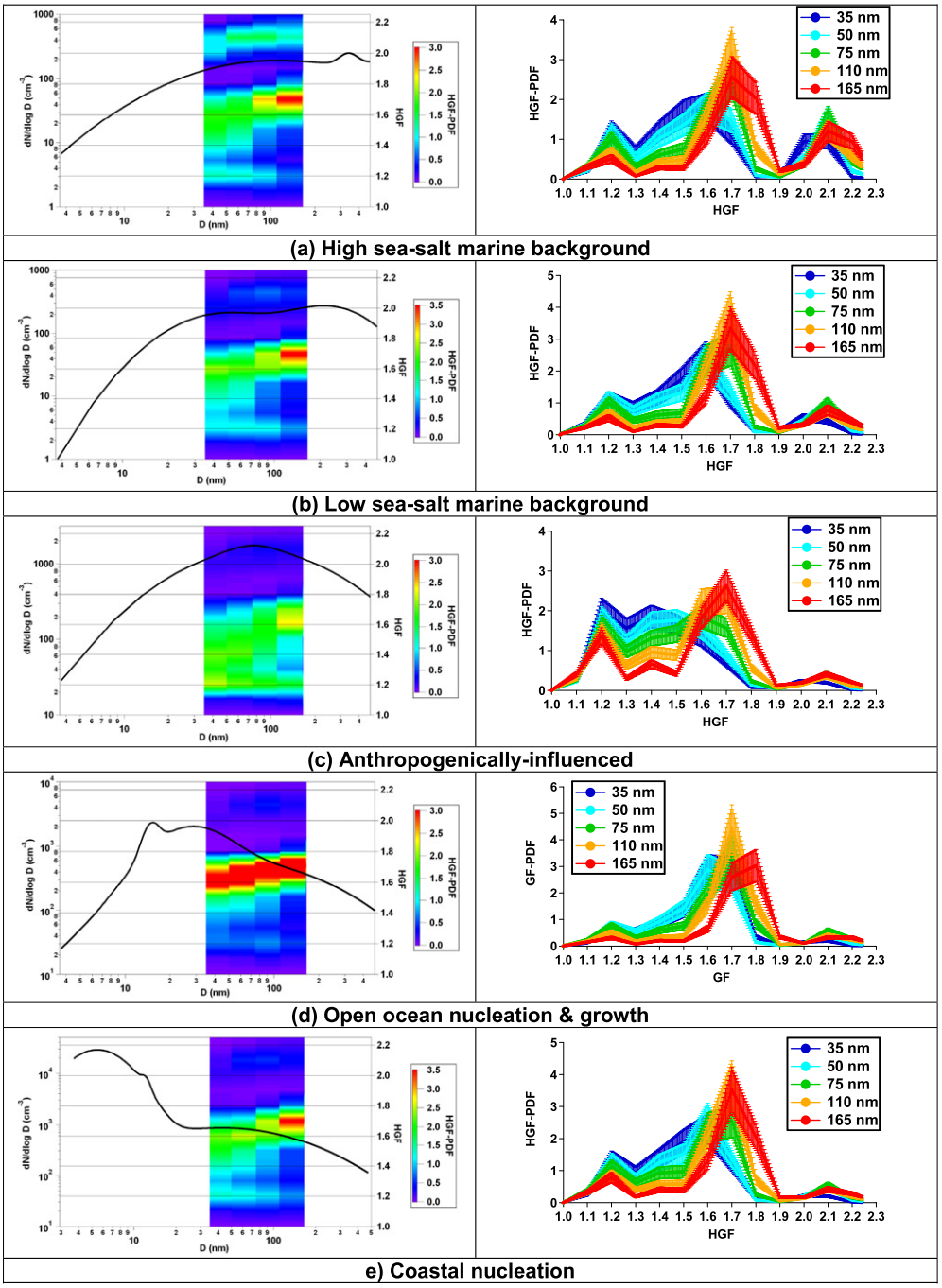

Figure 1. Left panel: clustered size distributions (±1 standard deviation) and coincident H-TDMA derived and interpolated contour map of HGF–PDF. Right panel: size-selected HGF–PDF as a function of size for clustered categories.

for nearly hydrophobic (NH), 1.3–1.5 for low hygroscopicity (LH), 1.6–1.8 for mainly hygroscopic (MH) and 2–2.2 for sea-salt (SS). The relative contributions of each mode to the HGF–PDF varies strongly with the origin of the air masses (i.e. aerosol category).

The five size distribution categories and associated HGF–PDFs are shown in figure 1. For the high sea-salt background marine category, typical of the cleanest marine winter conditions, the two main modes correspond to MH and SS for sizes 75 nm and larger while the contributions from NH and LH modes become more important for 35 and 50 nm sizes. Examination of the interpolated HGF contour plot reveals a three-mode external mixture with HGF-1.2 dominating at lower sizes, 2.0–2.1 dominating at larger sizes and the most dominant HGF mode increasing from 1.4 for smaller sizes to 1.7 at larger sizes. The SS mode is still seen for particles down to 35 nm, while the NH mode is also persistent for 165 nm sizes. It should be noted that pure ammonium sulfate or sea-salt HGFs are not seen in this case, or any case for that matter, suggesting that these predominantly inorganic aerosols are always influenced to

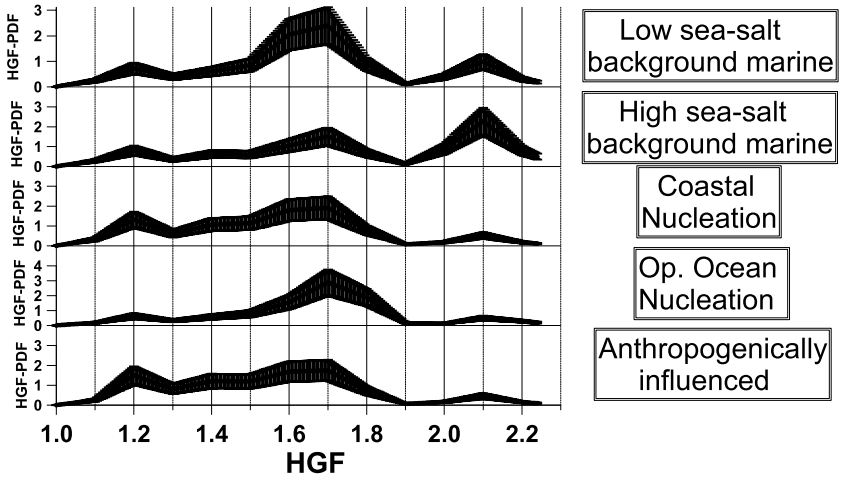

Figure 2. HGF–PDFs for clustered categories averaged over the five H-TDMA sizes of 35, 50, 75, 110 and 165 nm.

Table 1. Relative contributions of different HGF modes to total number concentrations of analysed sizes. Nearly hydrophobic (NH), low hygroscopicity (LH), mainly hygroscopic (MH) and sea-salt (SS)—see figure SI 4 available at stacks.iop.org/ERL/7/044013/mmedia.

	Hygroscopic growth factor mode			
Cluster category	NH mode	LH mode	MH mode	SS mode
High sea-salt background marine	$1.18 \pm 0.14(13\%)$	$1.48 \pm 0.5(12\%)$	$1.69 \pm 0.17(35\%)$	$2.10 \pm 0.18(40\%)$
Low sea-salt background marine	$1.19 \pm 0.18(11\%)$	$1.42 \pm 0.25(19\%)$	$1.69 \pm 0.23(52\%)$	$2.10 \pm 0.18(17\%)$
Anthropogenic	$1.19 \pm 0.13(15\%)$	$1.42 \pm 0.24(45\%)$	$1.66 \pm 0.24(30\%)$	$2.10 \pm 0.17(10\%)$
Coastal nucleation	$1.19 \pm 0.15(19\%)$	$1.42 \pm 0.20(20\%)$	$1.65 \pm 0.24(50\%)$	$2.10 \pm 0.17(10\%)$
Open ocean nucleation	$1.19 \pm 0.19(9\%)$	$1.52 \pm 0.31(26\%)$	$1.71 \pm 0.21(54\%)$	$2.10 \pm 0.18(11\%)$

some degree by organics. For the low sea-salt background marine case, a similar pattern is seen as in the high sea-salt case, except the SS mode is much diminished. The gradual increase in HGF with increasing particle size is seen for the dominant HGF mode.

For the anthropogenically influenced category, the SS mode is the least pronounced and a much broader distribution of HGFs is evident. The larger, 110 and 165 nm, particles PDF is characterized by a bimodal distribution with peaks below 1.2 and between 1.6 and 1.7, whereas the smallest sized particles, 35 and 50 nm, possess a PDF with a peak at 1.2 and a broad peak between 1.35 and 1.6. Overall, there is a systematic lowering in PDF towards low HGFs in comparison to marine conditions. For the open ocean nucleation category, the interpolated contour plot reveals one dominant MH mode, although increasing in HGF from 1.6 for 35 nm particles to 1.8 for 165 nm particles; however, two less pronounced modes are also seen at 1.2 and 2.1 HGFs. For the coastal nucleation category, there is again a dominant MH mode ranging from HGF 1.5 to 1.7, increasing with particle size, and notable NH and LH modes. Next to the continental case, the coastal category has the lowest HGF distribution, although this is thought to be more likely due to condensation of iodine oxides rather than organics (Väkevä *et al* 2002).

HGF–PDFs were averaged over the five different aerosol sizes and the resultant PDFs are illustrated in figure 2 for the five cluster categories. The distinction between the high sea-salt background marine and low sea-salt background marine category is evident. Again, no pure sea-salt or pure sulfate modes are evident, suggesting a degree of internal mixing between organics and the inorganic salts, and two clear, predominantly organic, modes are evident at 1.2 and 1.4. The HGF–PDFs were fitted with log-normal distributions (see figure SI 4 available at stacks.iop.org/ERL/7/044013/mmedia) to calculate the relative contribution of each of the four dominant modes to the total (normalized) number concentration and the results are presented in table 1.

For the high sea-salt background marine case, 40% of the number concentrations is contributed by predominantly-sea-salt particles (HGF = 2.1), 35% by predominantly sulfate particles (HGF = 1.69) and the remaining 25% by predominantly organic particles with hygroscopicity ranging from 1.19 to 1.42. For the low sea-salt background marine case, the sea-salt contribution decreases to 17%, the sulfate contribution increases to 52% while the organic contribution increases to 30%. The open ocean nucleation case is dominated also by sulfate aerosol (54%), with the next most important contribution (26%) coming from a HGF mode of 1.52. The coastal nucleation case is again dominated by sulfates (50%), with 39% from the two lowest HGF modes. Finally, for the continental case, the largest contribution of 45% comes from the 1.43 HGF mode, with 15% coming from the 1.19 HGF mode and 30% and 10% from sulfate and sea-salt, respectively. These results point to an important role for organics in terms of their contribution. In particular,while organic mass internally mixed with inorganic salts will lower the activation potential of these mixed aerosol types, thereby potentially reducing the concentration of CCN, pure organic

water soluble particles are still likely to be activated into cloud droplets, thereby increasing the concentration of CCN. Which effect will dominate will depend on an interplay between the overall CCN concentration and dynamics.

5. Conclusion

Two years of aerosol size distributions are analysed and characterized in terms of *k*-mean cluster analysis leading to five generic classifications. These five categories were labelled high sea-salt background marine, low sea-salt marine, open ocean nucleation, coastal nucleation and, anthropogenically influenced and for each one, the average HGF–PDFs were obtained. Over the five categories, four broad modes of HGFs were encountered to different degrees: 1.19 (NH), 1.42–1.52 (LH), 1.65–1.7 (MH) and 2.1 (SS). In all but the anthropogenically influenced case and high sea-salt background marine case, the MH mode dominated number concentration (52–54%) and a general trend of increasing HGFs was observed for increasing sizes associated with this mode. In the high sea-salt background marine case, sea-salt dominated the number concentration with a 40% contribution. Two predominantly organic HGF modes were observed with HGFs of 1.19 for the LH mode and between 1.42 and 1.52 for the LH mode. The NH mode contributed between 9% and 19% to total number concentration, the highest occurring for the coastal case, while the LH mode contributed 12% to 45% with the highest occurring from the anthropogenically influenced case. Combining both organic HGF modes, predominantly organic particles contribute between 25 and 30% to general background marine number concentration, 35% for open ocean nucleation cases, and 60% for anthropogenically influenced cases. For the coastal nucleation case, where low HGFs are thought to be significantly influenced by iodine oxidize rather than organics, the contribution of the two lower HGF modes was 49%. In summary, over yearly timescales, aerosol particles of predominantly organic composition contribute ∼30% to total submicron marine number concentration population and ∼60% to the continental number concentration population. While organic mass internally mixed with inorganic salts will lower the activation potential of these mixed aerosol types, thereby potentially reducing the concentration of CCN, pure organic water soluble particles are still likely to be activated into cloud droplets, thereby increasing the concentration of CCN.

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