**Unveiling the Complexity of Primary and Secondary Marine Organic Aerosols at Mace Head**

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# **2. Abstract**

Secondary organic aerosols (SOA) are recognized as a significant component of organic aerosol (OA) over oceans, yet, their chemical composition, especially in coastal and open ocean contexts, remain elusive due to the overall scarcity of high-resolution online data. In this study, positive matrix factorization (PMF) was performed on OA mass spectra obtained from high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) measurements. The focus was on a summertime period marked by enhanced biological productivity where pristine conditions prevailed for 60.4% of the time, while clean conditions were dominant for 30.5% of the measurement period. Four OA factors were deconvolved by the source apportionment model. The analysis revealed primary marine organic aerosols (PMOA) as the predominant submicron OA at Mace Head (MHD), accounting for 42% of the total resolved mass. This was trailed by more oxidized oxygenated organic aerosols (MO-OOA) at 32%, methanesulphonic acid organic aerosols (MSA-OA) at 17%, and locally emitted peat-derived organic aerosols (Peat-OA) at 9% of the aggregate OA mass. The total mass concentrations of POA and SOA were overall equal and highly prevalent in the marine boundary layer consistently with previous findings. Our data support distinctive aerosol sources across the North Atlantic Ocean. We observed aged polar air masses flowing from Greenland to MHD along with concomitant ozone production. MSA-OA on the other hand originated from the highly biologically productive Iceland basin, possibly associated with emissions from phytoplankton like coccolithophores and diatoms, while PMOA was primarily sourced from North Atlantic pelagic regions dominated by chlorophytes and cyanobacteria instead. These results stress the necessity for highly-time resolved data to grasp aerosol composition and underlying sources in view of changing ocean-climate interactions and biogenic influences.

**Keywords:** Submicron Marine Aerosols, Secondary Organic Aerosols, HR-ToF-AMS, Positive Matrix Factorization

# **1 Introduction**

The marine environment plays a critical role in regulating climate (Cochran *et al*. 2017). However, aerosols in the atmospheric marine boundary layer (MBL) remain a significant source of uncertainty in radiative forcing estimates (Y. Zhang *et al*. 2014; Meskhidze *et al*. 2019), primarily due to limited knowledge about aerosol mass, chemical composition, and particle number distributions (Carslaw *et al*. 2017). Along with this, the origin of marine organic aerosols, specifically whether primary or secondary, requires further investigation.

Primary Marine Organic Aerosols (PMOA) consist of sea spray aerosols, produced by bursting bubbles, film, jet, and spume drops that carry sea salt particles enriched in biogenic organic aerosols (C. D. O’Dowd *et al*. 2004; Facchini *et al*. 2008) and typically exhibit various sources and sinks (Kieber *et al*. 2016; Beaupré *et al*. 2019).

In contrast to PMOA, marine Secondary Organics Aerosols (SOA) arise from new particle formation involving gas-to-particle conversion (Zheng *et al*. 2021; Peltola *et al*. 2022) as well as condensation and oxidation reactions of precursors such as volatile organic compounds (Kroll *et al*. 2018; Hallquist *et al*. 2009). SOA formation occurs through various processes such as homogeneous, heterogeneous and multiple phase reactions (McNeill 2015; Marais *et al*. 2016) as well photochemistry reactions (Brüggemann, Hayeck, and George 2018). While various SOA molecular classes have been identified, the complexity of SOA, which consist of thousands of multifunctional compounds (Goldstein and Galbally 2007) including high molecular weight species and oligomers from diverse sources underscores the pressing need for continued exploration. All of this can now be partly described thanks to continuous widespread progresses in aerosol mass spectrometry (DeCarlo *et al*. 2006; Laskin, and Nizkorodov 2012). The present study focuses on source apportionment, aiming to delineate the chemical composition of secondary aerosols in pristine marine settings, particularly distinguishing between SOA and POA sources.

Both these SOAs and POAs (Mayer *et al*. 2020) serve as cloud condensation nuclei (CCN), impacting cloud albedo and lifetime, leading to uncertainties in global chemistry-climate models (Bellouin *et al*. 2020). Radiative transfer implications from such interactions range from -2.65 to -0.07 Wm-2, contrasting with CO2 radiative forcing estimate of 1.83±0.18 Wm-2 (Etminan *et al*. 2016). In pristine environments, SOA nucleation events significantly shape CCN concentrations, altering cloud radiative forcing (Liu and Matsui 2022). In the ongoing debate over whether primary sea spray (Xu *et al*. 2021) or secondary aerosols (Quinn *et al*. 2017; Mayer *et al*. 2020) exert a dominant influence on cloud formation in pristine environments, our study at Mace Head Atmospheric Research Station, through employing advanced online aerosol physicochemical techniques, aims at precisely distinguishing between aerosol sources and quantifying elemental ratios which might serve as a proxy for ulterior parametrisations (Han *et al*. 2022; Li *et al*. 2023).

Complementing this, previous literature shows that phytoplankton activity is related to emissions of organic and sulphate aerosol mass precursors (O’Dowd *et al*. 2015; Sanchez *et al*. 2021). However, the sensitivity of aerosol composition (and thus CCN) to ocean biology is currently unaccounted for in climate models (Sellegri *et al*. 2021). In this study, we compare aerosols’ geographic sources with NASA Ocean Biogeochemical Model (NOBM) taxonomic group simulations (Rousseaux, Hirata, and Gregg 2013). This multi-faceted approach allows us to place the local measurements at Mace Head into the broader context of ocean-atmosphere interactions and explore the potential influences of changing marine ecosystem dynamics on atmospheric aerosol loading over the North Atlantic region.

# **2. Materials and Methods**

## **2.1 Site Description**

Mace Head (MHD) research station is located on the west coast of Ireland (53.33°N, 9.90°W) on a peninsula that overlooks tidal areas over which North Atlantic Ocean air masses advect. These air masses, originating from a nominal clean sector (between 190° and 300°; Grigas *et al*. 2017) are predominantly shaped by south-westerly winds from the Atlantic Ocean, ushered by the polar jet's low-pressure areas. Importantly, these air mases are mostly untouched by anthropogenic influences, with over 60% of air masses over Ireland's west coast being classified as pristine marine (Grigas *et al*. 2017; Sanchez *et al*. 2022). However, around 40% exhibit varying degrees of anthropogenic influences, particularly during periods of continental outflow under high-pressure regimes (Jennings *et al*. 2003).

The research was strategically conducted during summertime (August 2015), a period characterized by heightened biological activity to hone in on the interplay between atmospheric dynamics and biological processes in marine settings.

## **2.2 In-situ measurements**

Ambient submicron aerosol major species were monitored using an Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) equipped with a standard vaporizer. The instrument overall working principles have been extensively described in the literature (DeCarlo *et al*. 2006; Canagaratna *et al*. 2007). The HR-ToF-AMS runs were performed on a 5mins time resolution basis on the single-reflection highly sensitive V mode configuration (mass resolution up to 3000 m/Δm) while detection limits were estimated based on the approach described by Drewnick *et al* (2009). The vaporizer operated at a nominal temperature of 650°C. Ionisation efficiency (IE), particle velocity and inlet flow were determined following standard methods while applying standard RIE. The particle transmission and detection efficiency expressed as the collection efficiency (CE; Huffman *et al*. 2005) was corrected for detection losses due to vaporizer particles bounce off by applying the composition-dependent collection efficiency (Middlebrook *et al*. 2012) using a value of 1 as the lower limit of CE as a typically applied for clean marine air masses (Ovadnevaite *et al*. 2012).

The AMS data were analysed using SQUIRREL (SeQUential Igor data RetRiEvaL) v1.65B and PIKA (Peak Integration by Key Analysis) v1.25B software packages. Sea salt was estimated based on a scaling factor of 51 of the common sea salt ion NaCl+ (m/z 57.96) (Ovadnevaite *et al*. 2012) while MSA was quantified by upscaling the CH3SO2 ion (Ovadnevaite *et al*., 2014). Influences from MSA on SO4 and OA were accounted as follows:

An improved Ambient (I-A) method was adopted for the mass spectra elemental ratio analysis of O:C, H C, N:C, S:C, and the OM: OC (organic matter to organic carbon) ratio (Canagaratna *et al*. 2015). High-resolution analysis was performed on each ion in the mass range 12–130 m/z with ion fitting applied to difference between open and closed spectra. Based on their elemental composition (C, O, H, N, S), ions were then grouped into chemical families: Cx, CxHy, CxHyOz (z = 1), CxHyOz (z > 1), CxHyNw (w = 1), CxHyNw (w >1), CxSj, HyOz, NwHy, NwOz, SjOz, and CxSi where the indices x, y, z, w, j represent the number of C, H, O, N, S atoms, respectively.

The concentration of equivalent black carbon (eBC) was measured by a multi-angle absorption photometer (MAAP, Thermo Fisher Scientific model 5012). The MAAP operated at a flow rate of 10 L min−1 and a 5 min time resolution. The transmittance and reflectance of eBC-containing particles were measured by the MAAP at two different angles to derive optical absorbance as mentioned in (Xu *et al*. 2020).

Finally, ozone was measured with an UV O3 spectrometer (Model 8810, Monitor Labs San Diego, CA), the raw voltage output was converted to concentration values based on Automatic Urban/Rural Network (AURN) calibration audits (see Derwent *et al*. 2018) and meteorological data were continuously recorded at the station (including rainfall, solar radiation, wind speed, wind direction, temperature, relative humidity and pressure).

## **2.3 Source apportionment**

The HR organic mass spectra was deconvolved using the Positive Matrix Factorization (PMF; Paatero and Tapper 1994; Paatero 1999) source-receptor model to investigate the various source contributions to OA. A major advantage of using HR data over unit mass resolution is the distinct quantification of multiple ions sharing the same nominal mass, thereby allowing for a more exact characterisation of the temporal fluctuations of different ion families (e.g., CxHy+, CxHyOz+). The information richness in the HR-ToF-AMS datasets, as a result of the improved chemical resolution, is advantageous for restricting the PMF solutions, minimizing rotational ambiguity and results in more reliable solutions and a larger number of interpretable OA factors.

The IGOR PRO Source Finder (SoFi v6.8.1) toolkit (F. Canonaco *et al*. 2013) was used to run the PMF algorithm. Solutions were assessed across 2 to 12 factors using the unconstrained factors rotational Fpeak tool. Factors were explored for Fpeaks (rotations) between −1 and 1 (0.1 steps). A final solution consisting of 4 factors was retained as the optimal solution based on its Q/Qexp ratio value (1.46), being at least 3% above the minimum Q/Qexp ratio (as recommended by Q. Zhang *et al*. 2011). Following previous studies recommendations (Canonaco *et al*. 2021), the solutions were also investigated in regard to key diagnostic plots, diurnal profiles, correlations with meaningful external tracer time series and reference mass spectra from the aerosol mass spectrometer database (Ulbrich *et al*. 2009).

## **2.4 Air masses trajectory analysis**

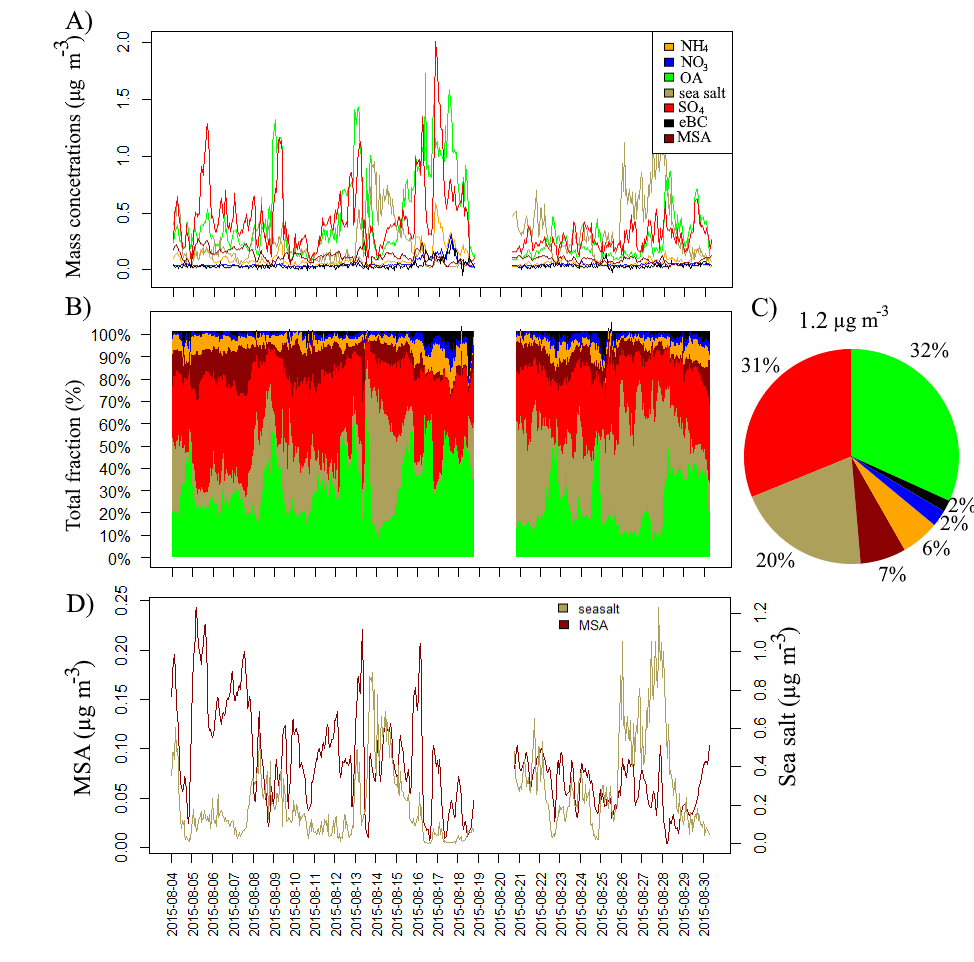
Air masses back trajectories analysis was performed using the Hybrid Single Particle Lagrangian Integrated Trajectory (Stein *et al*. 2015). Meteorological data were accessed from the Global Data Assimilation System (GDAS) archived by NOAA Air Resources Laboratory. HYSPLIT was used to calculate 72 hours back trajectories every 3 hours with starting height set to 100m. To investigate potential source regions leading to total particle mass concentrations from each resolved source, the back trajectories were gridded to 1° × 1° grid cells and linked to particle concentrations using trajectory source contribution functions. While common source contribution functions assume that trajectories centrelines estimates represent good estimates, we focused on the Simplified Quantiative Transport Bias Analysis (STQBA) approach which considers plumes transport bias along air mass trajectories.

Additionally, the fraction of time spent over the ocean, within the Marine Boundary Layer (MBL; altitude <BLH), in the Marine Free Troposphere (MFT; altitude >BLH) and over land in the planetary boundary layer (PBL; altitude < BLH) was quantified. In this study, BLH was determined from the fifth generation ECMWF (European Centre for Medium-Range Weather Forecasts) atmospheric reanalysis (ERA5) dataset based on the bulk Richardson number (Guo *et al*. 2021). Other datasets included NASA Ocean Biogeochemical Model (NOBM) taxonomic group simulations (Rousseaux, Hirata, and Gregg 2013) which allowed for *coccolithophores*, *diatoms*, *chlorophytes* and *cyanobacteria* geographic repartition estimates as well as lags calculations similarly to O’Dowd *et al* (2015). Finally, to delineate lands from oceans, the R package *rnaturalearth* was used to obtain a high-resolution land mask for Ireland allowing for identification of purely marine air masses (no advection over land for at least 3 days prior to being sampled at MHD).

# **3. Results**

## **3.1 Submicron aerosol chemical composition overview**

The mass concentration time series of organic aerosols (OA), Methane sulphonic acid (MSA), sulfate (SO42-), Nitrate (NO3-), ammonium (NH4+) and sea salt measured by the HR-ToF-AMS as well as eBC from MAAP measurements is shown on Figure 1. The average chemical composition was dominated by OA (32%), followed by SO42− (31%), sea salt(20%), MSA (7%), NH4+ (6%), NO3− (2%) and eBC (2%) (Figure 1).



**Figure 1: A) OA, SO4, sea salt, MSA, NH4, NO3 and eBC mass concentrations time series - μgm-3 B) Relative contributions to total PM1 C) Pie plot of total contributions to total PM1 D) shows MSA and sea salt.**

The total average bulk submicron aerosol mass was 1.2 μg m− 3 over the entire measurement period, these high SO4 and OA relative contributions and overall low concentrations are common for coastal sites during summertime in the marine boundary layer as reported over the North & South Atlantic Ocean (Ovadnevaite *et al*., 2014; Huang *et al*., 2018) as well as in the Artic (Willis *et al*., 2017; Nielsen *et al*., 2019). MSA in particular showed mass concentrations in the range of those previously reported at Mace Head (Ovadnevaite *et al*. 2011) and more diverse locations such as the central Arctic (Dada *et al*. 2022) and the Atlantic Ocean from 53° N to 53° S where average mass concentrations of 0.04 ± 0.03 µg m-3 (Huang *et al*. 2018) were reported.

Average bulk total OA mass concentration was 0.36 ± 0.32 μg m-3 with equivalent daytime and nighttime concentrations (Kruskal-Wallis p-value>0.05). Building upon this, the low mass concentrations of NH4+, NO3 and corresponding N:C ratio of 0.006 ± 0.002, indicate a limited presence of amino acids (below detection limit) from usual sources such as the North Atlantic oligotrophic gyre, ornithogenic emissions (i.e., birds), phytoplankton, bacteria, or in situ atmospheric processes (Schmale *et al*., 2013; Van Pinxteren *et al*., 2022). Furthermore, the observed average SO42- mass concentration was ~ 0.4 ± 0.3 µg m-3, along with a S/C ratio of 0.009 ± 0.002. This S/C value closely aligns with the 0.013 ratio value typically reported for MSA aerosols (Crippa *et al*. 2013).

Following pollution thresholds established for the North-East Atlantic (Grigas *et al*., 2017), pristine conditions (eBC levels below 0.015 μg m−3) were observed during 60.4% of the measurement period. Clean conditions (eBC levels between 0.015 and 0.05 μg m−3) prevailed 30.5% of the time, and moderately polluted conditions (eBC levels between 0.05 and 0.3 μg m−3) occurred for 9.1% of the time with a significant pollution event spanning from August 17th to 19th 2015 onwards.

Likewise, CO mixing ratios were below 100 ppb for over 70% of the time, similarly to other pristine sites (Zhao *et al*. 2022). Winds were westerly advecting through the clean sector (190-300°) prevailing for over 78% of the time. Finally mean wind speed was 6.6 m/s with 23% of the time below the whitecap threshold of 4 m/s and 36% of the time under conditions for mixed surf and spray plumes (O'Dowd *et al*., 2014) hinting at strong sea spray influences.

The average OM/OC value was 2.10 ± 0.14, aligning with the value of 1.9 previously reported for clean marine polar air masses at MHD (Ovadnevaite *et al*. 2014) .Additionally, AMS derived OM/OC values in the high Arctic (Nielsen *et al*. 2019) also fall within the range of 1.96 to 2.42 for PMOA and MO-OOA respectively, here median OM/OC value was 2.42 with minimum and maximum OM/OC values of 1.71 and 2.42 respectively). This indicates the presence of both POA (i.e., saturated hydrocarbons, unsaturated hydrocarbons and cycloalkanes) as well oxygenated SOA formed with photochemical processing during long range transport (Aiken *et al*. 2008; Simon *et al*. 2011).

To get a better sense of the main aerosol’s sources, total marine boundary layer (MBL), marine free troposphere (MFT) and planetary boundary layer (PBL) respective contributions are shown on Figure S3-A. Overall, the measurement period was dominated by marine boundary layer influences (91% of the time), with minimal marine free troposphere influences (8%) and extremely low land-influences from the planetary boundary layer (1%) further hinting at pristine marine conditions.

Finally, four OA plumes were then isolated with ‘plume1’ identified as “MSA-influenced” (Figure S3-C), ‘plume3’ as pollution event and ‘plume4’ as ‘ozone driven’. Overall, given the varying lengths of these identified periods, correlations with external tracers and apparent different geographic sources (Trajectories for each of these peaks are represented on Figure S3)—Source apportionment is then conducted, and periods of interest are then further refined to elucidate how organics (whether POA and SOA) behave in regard with various conditions and their respective sources.

## **3.2 Source apportionment**

To accurately classify and categorize the diverse sources of organic aerosols present at Mace Head, source apportionment was performed utilizing the Positive Matrix Factorization (PMF) method on the organic mass spectrum, which ranged from m/z 12.00 to m/z 130.08. The resulting four-factor solution, as depicted in Figure 2 and 3, yielded a Q/Qexp ratio of 1.38 and accounted for up to 90% of the total measured organic aerosol mass.

Based on the PMF analysis and upon considering consistency with physical and chemical properties, a four-factor solution was determined as the optimal representation as opposed to solutions with a different number of factors (Figure S1):

Methane Sulphonic Acid Organic Aerosols (MSA-OA): Representing approximately 17.2% of the total delineated PMF solutions, MSA displayed a distinct m/z fragment at m/z 78.98 (CH3SO2+) accounting for 36.3% of its total mass spectra signal intensity. The identification of specific methane sulphonic acid tracer ions further substantiated its origin. More details on all factors are provided below in sections 3.2.1-3.2.4.

More Oxidised Organic Aerosols (MO-OOA): Making up about 31.8% of the total elucidated PMF solutions, this factor exhibited prominent m/z fragments at m/z 27.99 and m/z 43.99 and showed significant correlations (Figure S4) with reference mass spectra for MO-OOA (R = 0.97), SV-OOA (R = 0.76), and was interpreted as MO-OOA after examining elemental ratios and correlations.

Primary Marine Organic Aerosols (PMOA): Comprising roughly 42.2% of the total resolved PMF solutions. this This factor exhibited m/z fragments similar to MO-OOA (Schmale *et al*. 2013), but with higher contributions from aliphatics (CxHy) such as alkanes, alkenes, alkynes, cycloalkanes and functional derivatives such as alcohols (CxHyOz, where z=1) as established in earlier studies (Ovadnevaite, Ceburnis, *et al*. 2011; Crippa *et al*. 2013)

Peat-OA, accounting for approximately 8.8% of the total PMF solutions, was characterised by saturated hydrocarbons (CnH2n+1), unsaturated hydrocarbons (CnH2n−1) and cycloalkanes (CnH2n) ion series. This factor was identified as Peat-OA owing to its good correlation (R=0.75) with the Peat-OA reference mass spectra (Lin *et al*. 2017). Additionally, its mass spectra signature such as the dominance of m/z 43 by C3H7+ rather than C2H3O+, facilitated the distinction of peat emissions over wood emissions.

**A screenshot of a computer screen

Description automatically generated**

**Figure 2. Factors mass spectra (MSA-OA in red, MO-OOA in green, PMOA in blue, Peat-OA in black) and associated improved ambient OM:OC, O:C and H:C ratios (Canagaratna *et al*. 2015).**

**A screenshot of a computer screen

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**Figure 3. Factors time series (MSA-OA in red, MO-OOA in green, PMOA in blue, Peat-OA in black) and associated relative contribution time series and pie chart for the whole period**

### **3.2.1 More Oxidised Oxygenated Organic Aerosols (MO-OOA)**

MO-OOA main contributing ions were associated with oxygenated compounds belonging to the COOH functional group, reflecting pronounced fragmentation of mono- and dicarboxylic acids into fragments with multiple oxygen atoms (Duplissy *et al*. 2011)

Specifically, CxHyOz (z > 1) ions family (m/z 43.98, m/z 44.99, m/z 71.01…) accounted for 63.1% of the total mass spectra intensity, followed by CxHyOz (z = 1) ions family (m/z 27.99, m/z 43.02, m/z 42.01…) contributing 13.68% to MO-OOA, adding up to a total carboxyls contribution of 76.8%. Additionally, CO+ accounted for 25% of MO-OOA intensity, aligning with values reported for remote oceans carboxylic acids (Russell *et al*. 2010).

In contrast, CxHy (aliphatics) ions family (m/z 13.00, m/z 15.02, m/z 16.03…) contributed only 13.7% to MO-OOA total mass spectra intensity. Nitrogen-containing ions fragments constituted a very low portion of the signal (0.8%), similarly to previous remote ocean measurements (Ovadnevaite, Ceburnis, *et al*. 2011).

MO-OOA mass spectra correlated well (Figure S4) with the laboratory reference mass spectra for MO-OOA (R=0.96) and “SV-OOA” (R=0.61) as well as field measurement (Hu *et al*. 2016) of more oxidised organic aerosols (R=0.98). The weak contribution from C2H3O+ (3.16%) which has been reported to be predominantly due to non-acid oxygenates (Ng *et al*., 2011a) suggests a considerable prevalence of aging/oxidation during transport over the North-East Atlantic Ocean. This is also further confirmed by the low m/z 43:44 ratio of 0.12 hinting to MO-OOA rather than less oxidised species (Ng *et al*. 2011). This factor respective O:C ratio and H:C ratio of 0.78 and 1.17 further agree with MO-OOA reported at other similar locations (Figure S6).

Here, MO-OOA had a strong contribution from CO2+ (25.7%) which is assumed to originate mainly from acids or acid-derived compounds (Duplissy *et al*. 2011; Ng *et al*. 2011) that are known to be mostly water-soluble (Decesari *et al*. 2007) such as organic acids (e.g., mesotartaric acid, meso-erythritol, tartaric acid, oxalic acid) (Cui *et al*. 2022) formed from oligomerization of small α-dicarbonyls (e.g., glyoxal) (Cui *et al*. 2022).

MO-OOA formation through ozonolysis was postulated based on a robust hourly averaged correlation (R=0.67) of MO-OOA to O3 across the entire observational period (Figure S4). The *effective transfer entropy* test (Behrendt *et al*. 2019) revealed the non-linear dynamics between O3 and MO-OOA, indicating O3 as a significant reactant in the formation of MO-OOA from its precursors (p-value<0.05). This aligns with studies showing O3 to be the oxidation driver during summertime in the marine environment (Ovadnevaite *et al*., 2011), where unsaturated aliphatic chains (C=C double bonds) react with ozone to form oxidised compounds (Decesari *et al*, 2011).

### **3.2.2 Methanesulphonic Acid Organic Aerosols (MSA-OA)**

The mass profile of MSA-OA revealed that two oxygenated carbon families CHO (sum of CxHyOk and CxHyOw where k = 1 and w > 1) dominated 53.4% of the total mass spectra fraction followed by aliphatics (pure Hydrocarbon‐like, CxHy+) whose fraction accounted for 33.3%. MSA-OA was further clearly identified owing to its substantial contribution from the CxSy+ family (6%) over other sources, this is in line with the CxSy+ contribution (7%) for MSA-OA also reported by Huang *et al*. (2018). Similarly, to results reported by (Schmale *et al*. (2013), the correlation coefficient with the AMS database MSA-OA laboratory reference spectrum (Figure S4) was rather moderate (R=0.28), although this factor spectra still allowed for the precise identification of characteristic MSA ions at m/z 44.98 (CHS+), 47.00 (CH3S+), 64.97 (HSO2+), 77.98 (CH2SO2+), 77.99 (CH3SO2+), and 95.99 (CH4SO3+). MSA-OA O:C and H:C ratios were 0.66 and 1.39 respectively, close to values reported by Loh *et al*. (2022). The excellent correlation (R=0.82) between this factor and the CxSy+ family (Figure S4) further highlights the sulphur predominance for MSA-OA as opposed to other factors. Furthermore, MSA-OA CxHy family features a typical CH3+ ion at m/z 15.02 that is absent from other factors. Likewise, the CxHyOw (w=1) family featured the tracer ion CH3O+ which is a heat stress related marker (Moschos *et al*. 2022). Additionally, CH2O+, C6H4O+ and C5H3O+ relative intensities were also dominant for the MSA-OA factor. Finally, the CxSy+ fragment family was dominated by CHS+, CH2S+, CH3S+, CH2SO+, CH3SO+, CH2SO2+, CH3SO2+ and CH4SO3+ which are common MSA ions found in the literature (Moschos *et al*. 2022). Overall, the CxHy+ and CxSy+ fragment ions families indicate a clear MSA fragmentation pattern with a characteristic high CH3+ relative intensity typical for marine SOA in line with recent findings (Huang *et al*. 2018; Moschos *et al*. 2022).

Additionally, MSA-OA correlated moderately (Figure S4) with particulate sulphate (R=0.51) which is expected since dimethyl sulphide, released by phytoplankton, can be oxidized to MSA or to sulphur dioxide and then to sulphuric acid, leading to their partitioning into the particulate phase (Mungall *et al*, 2018).

### **3.2.3 Primary Marine Organic Aerosols (PMOA)**

High-resolution mass spectrum of this factor revealed that two CHO oxygenated carbon families (sum of CxHyOk and CxHyOw where k = 1 and w > 1) dominated 61.5% of the total mass spectra followed by aliphatics (pure Hydrocarbon‐like, CxHy) whose fraction accounted for 36.2% of the total mass spectra signal aligning with previous findings reported by Ovadnevaite *et al* (2011);

The CxHy mass spectra family was dominated by ion series CnH2n‐3 (m/z 39.02, 53.03, 67.05 etc…) indicating dienes, alkynes, and cycloalkenes contributions, which is further confirmed by the presence of CnH2n‐1 ions series (m/z 27.02, 41.04, 55.05 etc…) while the CnH2n+1 family (m/z 43.05, 57.07 etc... n>2) indicative for anthropogenically influenced refined hydrocarbons was absent from this factor mass spectra. The biogenic origin of this factor was also indicated by its low correlation with eBC (R=0.17). Moreover, the CxHyOw (w=1) family featured ions series (m/z 55.02, 69.03, 83.05, etc…) related to alkenyl groups, diunsaturates, cyclic alcohols, and ethers. All of this is consistent with previous reports of water-insoluble organics being formed in sea spray (Ovadnevaite *et al*. 2011; O’Dowd *et al*. 2004). Additionally, this factor mass spectrum closely resembled (R=0.99) marine organic aerosols (MOA) mass spectra (Ovadnevaite, O’Dowd, *et al*. 2011; Crippa *et al*. 2013) and was therefore designated as such (Figure S4).

The prevalence of oxygenated components as well as the absence of hydrocarbon-like m/z (e.g., C4H7+ and C4H9+), resulted in a high O:C ratio of 0.66 and an O/C ratio of 1.16 respectively, close to literature O:C values (Figure S6). The high H:C ratio (1.16) we report reflects unsaturated hydrocarbons with double or triple bonds (Ovadnevaite *et al*., 2011). This is consistent with prior atmospheric measurements that have shown that PMOA containing a large fraction of unsaturated organic hydrocarbons are dominated by insoluble organic colloids and aggregates (Facchini *et al*. 2008; Rinaldi *et al*. 2020) which points to PMOA being composed of microgels derived from algae rather than exopolymers produced from bacteria (Bigg and Leck 2008; Bates *et al*. 2012).

Pure carbohydrate fragments (m/z 56.03, 60.02, 61.03, 73.03, 85.03) only accounted for about 1.26% of the total PMOA aerosols mass suggesting that PMOA likely did not originate from ordinary bacterial cell fragments, which comprise approximately 55% proteins and amino acids and 10% carbohydrates (Schmale *et al*., 2013). Contributions from glycogen; m/z 55.01 (1.36%), mannitol; m/z 56.02 (0.4%) and polysaccharide species; m/z 97.02 (0.7%) (Glicker *et al*. 2022) tracer ions were also relatively poor. Contributions of alkenes and CxHyOw (w=1) to PMOA would thus point to plankton bloom as a suitable source for PMOA (Ovadnevaite *et al*. 2011).

### **3.2.4 Peat Related Organic Aerosols (Peat-OA)**

Peat-OA mass spectra was largely dominated by saturated Alkanes-(76.92%) and Alkenes- which is typical for peat. Likewise, the presence of aromatic ion series at m/z 77.03 (C6H5+) and m/z 91.0.5 (C7H7+) (Cubison *et al*. 2011) and the ratio between m/z 55.05 (C4H7+) and m/z 57.07 (C4H9+) of 1.74 as well as the ratio between m/z 43.05 (C3H7+) and m/z 44.01 (C2H3O+) of 1.03 all allow for the clear distinction of peat burning over other sources (Lin *et al*. 2017). Peat-OA was freshly emitted as evidence by the pollution wind rose (Figure S2 E, F) and concurrent increase along with eBC (R=0.33) which indicate that both were locally co-emitted within the PBL (Figure S2).

This specifically shows with the very low contributions for the tropylium aromatic ring signal ion at C2H4O2+ (m/z 60), m/z 91.05 (C7H7+), C9H7+ (m/z 115.06) which are usually associated with extensive aging (Yazdani *et al*., 2022). Likewise, the high signal for C4H9+ at m/z 57.0.7, C7H11+ at m/z 95.09 (Ahern *et al*. 2019) and the total absence of CO2+ signal is also consistent with primary combustion sources of fossil origin (Nielsen *et al*. 2019). Finally, this factor mass spectrum correlated well (R=0.75) with previous measurements of Peat-OA in Galway city (Lin *et al*. 2017) (Figure S4).

## **3.3 Elemental ratios -Van Krevelen diagram**

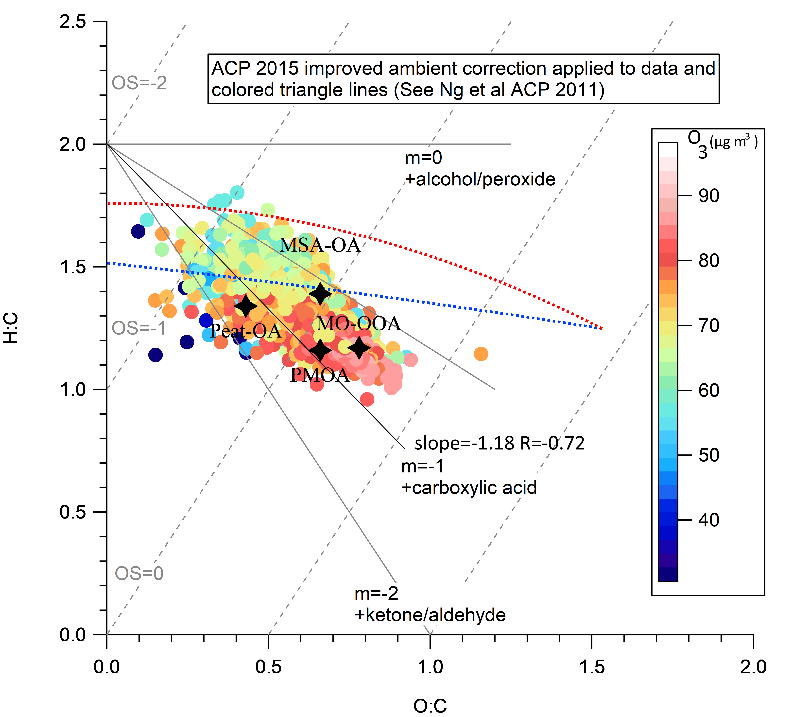
The Krevelen diagram (Heald *et al*., 2010) shows discernible patterns for the PMF factors (Figure 4). The slope of a fitted line with intercept forced to 2 (O: C = 0 and H: C = 2; Head *et al*., 2010) was used to infer the OA composition and chemical processes involved. A slope of -1 indicates the integration of carboxylic acids without fragmentation or alcohol + carbonyl addition, while a slope of -0.5 suggests the inclusion of carboxylic acids with fragmentation, and a slope of 0 signifies the addition of alcohols/peroxides (Heald *et al*. 2010; Ng *et al*. 2011).

The bulk OA slope was -1.18 spanning over values of -1.8 to 0.8 in the carbon oxidation state space. This shows that higher levels of oxidation involving the generation of carboxylic acids and the subsequent breakdown of the carbon backbone were prevalent over the measurement period which is consistent with the marked presence of SOA (Heald *et al*.2010, Ng *et al*., 2011). Furthermore, the correlation coefficient of -0.8 between H:C and O:C values highlights continuous SOA formation along the O3 gradient as opposed to successive events from different oxidants as would be otherwise evidenced by a positive association (Gu *et al*. 2023). The O:C ratios for MO-OOA, PMOA and MSA-OA all fall within the range of 0.64–1.15 reported for diverse OOA factors from previous studies (Aiken *et al*., 2008; Jimenez *et al*., 2009). All factors had H:C values lower than 2 which indicate that they all contained unsaturated carbons capable of reacting with O3.

MSA-OA (O:C=0.66, H:C=1.9) was then examined by colouring the VK scatter plot (Figure S7) with the MSA-OA/SO4 ratio, a proxy for biological marine sources contributions from DMS (Chen *et al*., 2021). We note values ranging from 0.001 (ubiquitous anthropogenic influences) to 0.354 (significant contribution from biological marine sources) with an average value of 0.102 in line with pristine conditions (Huang *et al*., 2018). Figure S7 revealed that high MSA-to-sulphate ratio were consistent with molecules such as C2-C12 saturated diacids and C10 and C5 keto-acids (levulinic and pinonic acid respectively) and inconsistent with C8-C30 saturated alcohols, C8-C32 saturated acids, C4-C12 carbohydrates (trehalose, erythritol, arabitol, mannitol, sucrose, galactose, glucose, fructose etc…) similarly to results reported for summertime Arctic aerosols (Wilis *et al*., 2017). However, as opposed to Artic aerosols, H:C ratios being higher, we report no association with C4 unsaturated diacids (e.g maleic and fumaric acid).

The PMOA (O:C=0.66, H:C=1.16) factor position on the Van Krevelen diagram indicate an abundance of aliphatic hydrocarbons, alcohols, fatty acids and esters. PMOA lies in the lower right corner of the VK plot (less oxidised more saturated) and hints at the presence of Humic-like molecules, characterized by polar carbonyl (keto and carboxyl) functional groups alongside hydrophobic aliphatic chains which are hypothesized to be enriched in PMOA (Radoman *et al*. 2022).

Finally, Peat-OA (O:C=0.43, H:C=1.34) position was consistent with lignin-like compounds (H/C = 0.6–1.5, O/C = 0.1–0.6) which have been largely associated with terrestrial origin OA (Jang *et al*. 2022).



**Figure 4. Relationship between the ToF-AMS estimated hydrogen-to-carbon (H/C) and oxygen-to-carbon (O/C) ratios of organic species [Canagaratna *et al*., 2015] colored by O3 mixing raito, all observations above ToF-AMS detection limits are shown for the entire period. Grey lines represent the ambient range of O/C and H/C observed by Ng *et al*. [2011] while dashed line represent the average carbon oxidation state (OSc ≈ 2 × O : C − H : C) (2011) superimposed on the Van Krevelen diagram (Ng *et al*. 2011, Kroll *et al*., 2011). Elemental composition of C8–C30 saturated alcohols, C8–C32 saturated acids, C2 –C11 saturated diacids, C4 unsaturated diacid (maleic and fumaric acid), C4 –C12 carbohydrates (e.g., trehalose, erythritol, arabitol, mannitol, sucrose, galactose, glucose, and fructose), and C5 and C10 ketoacids (levulinic and pinonic acid, respectively) are shown for reference (Willis *et al*., 2017).**

## **3.4 Air masses and source apportionment**

**Figure 5. Simplified Quantitative Transport Bias Analysis (SQTBA) -Gaussian Air masses dispersion for PMF sources (PMOA, MO-OOA, MSA-OA) and O3**

MO-OOA (Figure 8-A) strongest sources can be traced back northward along a cyclonic gradual crescent shape spreading from Greenland Seas South of Cape Farewell (See Figure S9 for Ocean areas toponomy). This culminates further with air masses sources spanning over the East Greenland Current (Denmark Strait) upwards to the Iceland Sea south of Jan Mayen. MO-OOA is otherwise ubiquitous and shows contributions over the Newfoundland, Labrador and Iceland basins as well as other areas. O3 (Figure 5-B) shared similar origin as MO-OOA further confirming its role in MO-OOA formation. Overall, we observe aged polar air masses eventually flowing from Greenland to MHD. The sustained blockade and aging of air masses over Greenland is known and attributed to summertime high-pressure systems surrounding this region influenced by Arctic amplification (Pettersen *et al*. 2022; Preece *et al*. 2023) where Irminger current also acts as a hotspot for turbulent eddies and heat transport which might contribute to aerosol nucleation (Semper *et al*. 2022). Here the presence of a blocking anticyclone transition (Figure S9) leading to reduced cloud cover and warm air advection might ultimately have contributed to an increase in aged SOA at the southern tip of Greenland possibly owing to its orography.

A screenshot of a computer screen

Description automatically generated

**Figure 6. Time averaged maps (0.67 x 1.25 deg) over 2015-Aug, Region 59W, 37N, 34E, 82N of dominant phytoplankton groups from NOBM Model data (Gregg and Rousseaux 2017; Buchard *et al*. 2017) and corresponding lagged crossed correlations for MSA-OA (red) and PMOA (blue) against A) Diatoms B) Coccolithophores, C) Chlorophytes and D) Cyanobacteria :blue and red shaded areas correspond to maximum significant crossed-correlations extracted from the autocorrelation function (ACF) 95% criteria.**

MSA-OA (Figure 5-C) main sources include the Iceland basin and more specifically the Iceland-Faroe Ridge. This is consistent with literature highlighting the diversity of eukaryotic phytoplankton in the Icelandic marine environment with the haptophyte coccolithophore *Emiliania huxleyi* being dominant during summertime (Cerfonteyn *et al*. 2023) owing to nutrients transport by the North Atlantic Current acceleration (Oziel *et al*. 2020) and findings (O’Dowd *et al*. 2015: Mansour *et al*. 2023) indicating concomitant MSA concentrations uptick during summertime. MSA-OA also spans along the East Greenland Current (Denmark Strait) where wind-driven coastal upwelling (Håvik and Våge 2018) might result in increased DMS emissions (Edtbauer *et al*. 2020). Likewise, MSA-OA extend moderately over diverse regions such as the North-Western European Basin, the Newfoundland basin (where intense DMS fluxes have been reported; Bell *et al*. 2021) and the Labrador Sea.

PMOA (Figure 5-D) on the other hand strongly extends over the South of the Celtic Sea and West of the Bay of Biscay as well as West European basin waters and are otherwise diffuse all over the North Atlantic Ocean with moderate intensity hotspots over the Newfoundland basin (Davis strait) possibly owing to an inflated subpolar gyre (Hátún *et al*. 2016).

Examination of NOBM model data (Figure 6) further reveals distinct MSA-OA and PMOA patterns. MSA-OA overlap with coccolithophores dominated ecoregions as well as diatoms ones. Similarly, diatoms also seem to contribute to PMOA sources, which is in line with recent results hypothesising that diatoms have a greater atmospheric significance than other eukaryotes due to their observed enrichment in PMOA (Alsante *et al*, 2021) whereas association with coccolithophores appears much weaker than for MSA-OA. Another distinction lies in PMOA overlapping with chlorophytes (*flagellates*, *Phaeocystis* *spp*) over the Western European basin. This geographic area hosts more than 512 chlorophyte species (Narayanaswamy *et al*. 2010) with recent reports of chlorophytes being one of the key contributors to marine productivity (Landwehr *et al*. 2021), further research is warranted to fully understand their role along other phytoplankton in this region during summertime. Likewise, cyanobacteria (combination of *Synechococcus*, *Prochlorococcus*, and nitrogen fixers such as *Trichodesmium*) might also contribute to PMOA more sparsely, especially at lower latitudes in the North Atlantic Ocean as previously reported (Baer *et al*. 2017).

Here, calculated lagged correlations (Figure 6) further point at MSA-OA being directly associated with coccolithophores (with a lag of -1 day) as well as diatoms (lag of -9 days), however no significant correlations are observed for either cyanobacteria or chlorophytes. As opposed to MSA-OA, no association is observed between coccolithophores and PMOA (their autocorrelations are not statistically significantly different from zero). PMOA on the other hand are also associated with diatoms (lag of -5 days) and show unique associations with chlorophytes (lag of -10 days) as well as cyanobacteria (lag of-11 days).

Overall, association between OA enriched sea spray time series and phytoplankton groups remains controversial owing to a wide range of governing mechanisms as highlighted by previous studies using chl-α as a proxy to calculate cross correlation time lags over the North Atlantic which were found to vary between 8 days (Rinaldi *et al*. 2013) and 24 days (O’Dowd *et al*. 2015) depending on the period and length of measurements.

Late summer measurements (Mansour *et al*. 2020) show partially comparable lags to this study with a reported oceanic biological activity affecting aerosol properties within the order of 10-20 days. This delay roughly spans over the full blooming to decaying phase transitions of an algal bloom (Lehahn *et al*. 2014) and is linked to the release of SSA-transferable organic matter in surface seawater (O’Dowd *et al*. 2015).

Here, by focusing on the lagged correlations between PMF factors and specific phytoplankton groups rather than bulk-OA and chl-α, this study´s findings indicate that PMOA is formed on such timescale from cyanobacteria and chlorophytes owing to atmospheric transport from the Western European basin whereas overwhelming diatoms influence results in a much shorter lag of -5 days. Additionally, MSA-OA is rapidly produced from coccolithophores blooms in 1-2 days. This reflects stressed, senescent, grazed, or virus-infected phytoplankton releasing high quantities of DMSP which rapidly oxidises to form MSA-OA (Mansour *et al*. 2020).

Finally, the interpretation of diatoms´ role on either MSA-OA or PMOA remains ambiguous as the -5 days lag with PMOA could hint at lipase activity concurring to self-aggregation and formation of free fatty acids during bloom potentially followed by a post-bloom (lag of -9 days with MSA-OA) with significantly different taxa or simply advection from remote eco-regions further closer to the Arctic which have been reported to host rich MSA producing diatoms communities as opposed to more southerly latitudes (Becagli *et al*. 2016).

# **4. Conclusions**

This study leveraged high-resolution online aerosol mass spectrometry to investigate the chemical composition and sources of submicron organic aerosols in the remote marine environment of Mace Head, Ireland during summertime and marks the commencement of long-term source apportionment efforts at Mace Head. The results highlight the significant contribution of secondary organic aerosols (SOA), accounting for approximately 50% of the total submicron organic aerosol mass. Specifically, two distinct SOA factors were identified - more-oxidized oxygenated OA (MO-OOA) and methanesulfonic acid OA (MSA-OA).

Source apportionment using air mass trajectory analysis revealed distinctly different geographic origins for MO-OOA and MSA-OA. While MO-OOA originated from aged polar air masses transported from Greenland undergoing sustained ozonolysis, MSA-OA sources were concentrated around the biologically productive Iceland basin and Iceland-Faroe ridge regions, potentially linked to emissions from coccolithophores and diatoms. In contrast, PMOA did not seem associated with coccolithophores but rather with diatoms, cyanobacteria and chlorophytes.

Overall, this study demonstrates the complex aerosol chemistry and diverse geographic origins influencing SOA formation in the North East Atlantic marine environment. Our findings emphasize the need for further investigation to fully account for the various precursors and pathways contributing to SOA, given their significant impacts on aerosol-climate interactions.

*Competing interests.* The authors declare that they have no conflict of interest.

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*Data availability.* Data available upon request.

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