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**Submission of**

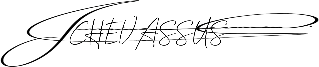
“Marine Organic Aerosols at Mace Head: Effects from Phytoplankton and Source Region Variability” by Emmanuel Chevassus et al.

**Dear Editor,**

Please find ‘Marine Organic Aerosols at Mace Head: Effects from Phytoplankton and Source Region Variability, by Emmanuel Chevassus et al., submitted for consideration for publication in *Atmospheric Chemistry and Physics* (ACP)*.* This study presents the first source apportionment from high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) measurements at Mace Head. Resolved organic aerosols sources were marked by prevailing pristine maritime conditions with balanced mass contributions from both primary and secondary sources. We introduce the use of a new metric “effective transfer entropy” to predict how secondary aerosols originate from their reactants – This analysis establishes that oxidised SOA are a blend formed from open ocean air masses aging through ozonolysis and local Peat-OA oxidation. We also suggest that the Greenland summertime blocking might be a yet unexplored source of secondary aerosols. Conversely our transfer entropy approach also demonstrates that other open Ocean sources, namely, primary organic aerosols (PMOA) and methanesulphonic acid (MSA-OA) are fully exempt of anthropogenic influences. We trust that this work is appropriate for publication by ACP as it demonstrates that reactants leading to secondary formation can be predicted but also provides a characterisation of marine sources building up on previously identified tracers. Notably that PMOA reflects phytoplankton extracellular metabolic processes largely shaped by abacterial processes whereas MSA-OA is marked by stress enzymes. The manuscript introduces a novel approach for calculating lags between phytoplankton activity and aerosol formation. Traditionally, chlorophyll-a has been used as a tracer for phytoplankton, here we improve on this approach by retrieving instead specific phytoplankton taxa such as diatoms, coccolithophores, cyanobacteria, and green algae from the NASA Ocean Biogeochemical Model (NOBM). The study concludes that MSA-OA forms quickly after coccolithophore blooms (1-2 days), while diatoms contributions occur more slowly (9 days). In contrast, PMOA is more directly influenced by diatoms (5 days), but also by more diverse groups, namely chlorophytes (10 days), and cyanobacteria (11 days). This manuscript has not been published and is not under consideration for publication elsewhere. The authors of this paper have had no prior contact with a ACP Editorial Board Member concerning any of the work described in this manuscript. We have no conflicts of interest to disclose, and we would welcome some of the following experts in atmospheric science to review our report: Yangmei Zhang, State Key Laboratory of Severe Weather/Key Laboratory of Atmospheric Chemistry of China, Chinese Academy of Meteorological Sciences, Beijing, 100081, PR China, [ymzhang@cma.gov.cn](mailto:ymzhang@cma.gov.cn); Karine Sellegri, LAMP, Laboratoire de Météorologie Physique (UMR 6016 Université Clermont Auvergne, CNRS), Aubière, France , [karine.sellegri@uca.fr](mailto:karine.sellegri@uca.fr); Julia Schmale, School of Architecture, Civil and Environmental Engineering, École Polytechnique Fédérale de Lausanne, Switzerland, [Julia.schmale@epfl.ch](mailto:Julia.schmale@epfl.ch); Matteo Rinaldi, Institute of Atmospheric Sciences and Climate, Italian National Research Council (CNR-ISAC), Bologna, Italy, [m.rinaldi@isac.cnr.it](mailto:m.rinaldi@isac.cnr.it); Laurent Poulain, Leibniz Institute for Tropospheric Research, Leipzig, 04318, Germany, [poulain@tropos.de](mailto:poulain@tropos.de); Ingeborg E. Nielsen, Department of Environmental Science, Aarhus University, Roskilde, 4000, Roskilde, Denmark Arctic Research Centre, Aarhus University, Aarhus, 8000, Aarhus, Denmark, ien@envs.au.dk ; Maija Peltola, Institute for Atmospheric and Earth System Research (INAR) / Physics, Faculty of Science, University of Helsinki, 00014, Finland, [maija.peltola@helsinki.fi](mailto:maija.peltola@helsinki.fi) .In addition, we suggest the following Editorial Board Member: Samara Carbone, Federal University of Uberlandia, Agrarian Sciences Institute Environmental and Sanitary Engineering an, Brazil, [samara.carbone@gmail.com](mailto:samara.carbone@gmail.com)

We thank you for your time and consideration.

Sincerely,



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SCOPE: This study enhances the understanding of aerosol-phytoplankton interactions and their implications for atmospheric processes over the North-East Atlantic. We provide the first source apportionment of organic aerosols at Mace Head using HR-ToF-AMS data and introduce transfer entropy as a novel approach to trace secondary aerosol origins. We show that oxidised SOA are formed from a blend of open ocean air masses aging through ozonolysis and local peat burning derived OA oxidation. In contrast, this method also confirms that other open ocean sources, such as primary organic aerosols (PMOA) and methanesulphonic acid (MSA-OA), remain unaffected by anthropogenic influences. PMOA reflects phytoplankton extracellular metabolic processes, largely shaped by abacterial activity, while MSA-OA is linked to stress enzyme markers. We improve upon traditional chlorophyll-a lags estimate studies by instead using specific phytoplankton taxa, including diatoms, coccolithophores, cyanobacteria, and green algae, using the NASA Ocean Biogeochemical Model (NOBM). MSA-OA forms rapidly after coccolithophore blooms (1-2 days), while PMOA is influenced by diatoms (5 days) and other groups like chlorophytes (10 days) and cyanobacteria (11 days).

Short Summary: This study presents the first source apportionment of organic aerosols at Mace Head using high-resolution time-of-flight aerosol mass spectrometry. Transfer entropy is introduced as a novel method to trace secondary organic aerosol origins, revealing that aged organics form from both open ocean air masses and local peat burning emissions. Methanesulphonic acid (MSA-OA) and primary marine organic aerosols (PMOA) makeup mirrors phytoplankton activity. Using phytoplankton taxa data from the NASA Ocean Biogeochemical Model, this study finds that MSA-OA forms rapidly after coccolithophore blooms (1-2 days), while diatoms contributions occur later (9 days). PMOA on the other hand is influenced by diatoms (5 days) but also by chlorophytes (10 days) and cyanobacteria (11 days).

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