Assessing the Dynamics of Organic Aerosols over the North Atlantic Ocean

Jérôme Kasparian1, Christel Hassler2,3, Bas Ibelings2,3, Nicolas Berti1, Sébastien Bigorre4, Violeta Djambazova5, Elena Gascon-Diez2, Grégory Giuliani2, Raphaël Houlmann1, Denis Kiselev1, Pierric de Laborie2, Anh-Dao Le2,3, Thibaud Magouroux1, Tristan Neri2, Daniel Palomino2,3, Stéfanie Pfändler2, Nicolas Ray2, Gustavo Sousa2, Davide Staedler1,5, Federico Tettamanti1, Jean-Pierre Wolf1 & Martin Beniston1,2,3

The influence of aerosols on climate is highly dependent on the particle size distribution, concentration, and composition. In particular, the latter influences their ability to act as cloud condensation nuclei, whereby they impact cloud coverage and precipitation. Here, we simultaneously measured the concentration of aerosols from sea spray over the North Atlantic on board the exhaust-free solar-powered vessel “PlanetSolar”, and the sea surface physico-chemical parameters. We identified organic-bearing particles based on individual particle fluorescence spectra. Organic-bearing aerosols display specific spatio-temporal distributions as compared to total aerosols. We propose an empirical parameterization of the organic-bearing particle concentration, with a dependence on water salinity and sea-surface temperature only. We also show that a very rich mixture of organic aerosols is emitted from the sea surface. Such data will certainly contribute to providing further insight into the influence of aerosols on cloud formation, and be used as input for the improved modeling of aerosols and their role in global climate processes.

The direct and indirect radiative forcing of the atmosphere by aerosols is recognized as a major source of uncertainty in global climate modeling1, as highlighted by the recent IPCC reports2. This effect is highly dependent on the particle size and composition3. Besides the terrestrial aerosols of anthropogenic origin (linked to fossil-fuel combustion, industrial processes, transport...) or those of natural origin (e.g., sand, pollen, bacteria, as well as particles emitted by erosion, fires, volcanoes...), it is now recognized that sea spray aerosols (SSA) are also key players in the climate system. In particular, they are now considered to be the main contributor to light scattering in the marine boundary layer and possibly the most important natural aerosol component capable of influencing the Earth’s radiative balance4,5.

The production of SSA is closely linked to air bubbles generated in the ocean by breaking waves, which then rise to the surface and burst into hundreds of droplets over a broad size range, from the nm scale to tens of micrometers6,7. The process efficiency (i.e., the emission flux varying from 10^4–10^6 particles/m^2/s) is primarily linked to wind speed, which modulates the amplitude of oceanic waves and to sea surface temperatures (SST)8.

Oceans are one of the largest reservoirs of organic carbon on Earth, with a wide variety of substances9. Besides sea-salt, the composition of the aerosol particles is complex4,10, as it comprises sulfur compounds that can be found in the form of dimethyl sulfide (DMS) in secondary particles11, polysaccharides, proteins, amino acids, fatty acids, humic substances, gels, micro-organisms (e.g., phytoplankton or bacteria) and their fragments, as well as vesicles12. These active species, together with sulfates13,14,15 can act as cloud condensation nuclei (CCN) and ice crystal nucleating agents in clouds16. They therefore contribute indirectly to the planetary radiative balance, in addition to their direct effect via light scattering.

1Université de Genève, Group of Applied Physics, Chemin de Pinchat 22, CH1211 Geneva 4, Switzerland. 2Université de Genève, Institute for Environmental Sciences, 66 Boulevard Carl Vogt, CH 1211 Geneva 4, Switzerland. 3Université de Genève, Department F.-A. Forel for Environmental and Aquatic Sciences, 66 Boulevard Carl Vogt, CH 1211 Geneva 4, Switzerland. 4Woods Hole Oceanic Institute, 86 Water St, Woods Hole, MA 02543, USA. 5TIBIO SagL, via alla Valle I I, 6949, Switzerland. Correspondence and requests for materials should be addressed to J.K. (email: jerome.kasparian@unige.ch)
Numerous laboratory studies and marine measurement campaigns have been carried out to identify the relationship between ocean surface characteristics, biological activity and the composition of SSA. No definitive conclusions have so far been reached, due to the complexity of the physico-chemical processes involved, including the aging of aerosol particles. For example, the use of Chlorophyll $a$ (Chl $a$) maps retrieved by satellite imagery over the ocean, as a proxy for the concentration of organic aerosols concentrations, is still the subject of intense scientific debate.

In this paper, we report the results of a large-scale measurement campaign performed over 5,000 nautical miles in the North Atlantic Ocean (from Miami on June 8th to London on August 30th, 2013), focusing on the northern part of the campaign, namely the Boston – Halifax– St John’s route (July 4th to August 1st, 2013, See Table S1). This segment of the journey covered a wide range of weather and ocean conditions, with inversions into both cold- and warm-water masses on the margins of the Gulf Stream. The use of the world-largest solar-powered vessel, the Swiss “Tûranor PlanetSolar”\(^{19}\), which does not produce exhaust fumes, significantly limited contamination of the measurements. Together with the aerosol particle size, a single-particle fluorescence spectrometer (GAP-SPFS, named after ref. 20), specifically developed for the expedition, continuously characterized the composition of individual particles in real time and in-situ. In particular, it allowed to specifically identify organic-bearing particles.

We show that the abundances of total and organic-bearing aerosols, respectively, display different spatio-temporal distributions, and can be modelled by different empirical relationships as a function of the physico-chemical parameters of the ocean and the atmosphere (temperature, wind, salinity). Surprisingly, the organic-bearing particle concentration can be parameterized with a dependence on water salinity and sea-surface temperature only. Furthermore, the fluorescent spectra of individual particles reveal that a very complex mixture of organic aerosols is emitted simultaneously. Such data will provide new insight into the dynamics of organic aerosols and their influence of aerosols on cloud formation.

### Results and Discussion

#### Sea spray particle concentration and size distribution

The total aerosol concentrations in the 250 nm–32 μm diameter range varied between $2 \times 10^8$ and $5 \times 10^{10}$ m$^{-3}$, with an average of $1.1 \times 10^{10}$ m$^{-3}$ and a median of $1 \times 10^{10}$ m$^{-3}$ (Fig. 1a). Considering a particle lifetime of 5 days\(^4\) and a mixing layer thickness of 1 km, and integrating over the measured size distribution (Fig. 1c), these concentrations yield a global emission flux of 1,000 Tg aerosols/year. Assuming 5% carbon content as proposed by\(^4\), this flux corresponds to 50 Tg C/year. Such values are consistent with typical global estimations relying on satellite-based measurements and global model simulations, that yield values ranging from 2 to 300 Tg C/year\(^{21,22,23}\).

The concentration of particles up to 10 μm was in particular positively correlated with air and sea surface temperature (Fig. 1b; See also Table S3 and Figure S3), while the shape of the distribution seems less affected (Fig. 1c). This evolution might be related to the influence of temperature on the process of bubble fragmentation as suggested from laboratory experiments\(^24,25\). However, methods for simulating actual wave breaking usually require large infrastructures with wave paddle generators or large wind tunnels, which was not the case of the said laboratory experiments. Further experiments are therefore needed to confirm this assumption.

Also, Fig. 1a displays sharp transitions in the particle number concentration, illustrating the local dynamics induced by the contrast between water masses and weather conditions. Furthermore, coastal regions (close to the stopovers in Boston, Halifax and St John’s) are associated with both higher total particle concentration and smaller particles, with a narrower size distribution.

Obviously, particle transport and possible import of terrestrial aerosols have to be considered. However, several observations allow us to expect a moderate influence of land and aerosol transport in our measurement, except close to the stopovers: (i) the fast decay of the number concentration as a function of the distance from the coast; (ii), the lack of correlation between the wind direction and our measurements, and (iii) the dominant wind direction (mostly from the South, i.e., from the open ocean), that was aiming at land all over the considered period, except for 36 hours. We can therefore expect that the data evaluated as “micrometer sized sea spray related” in this paper are not significantly altered by terrestrial imports. Note that, moreover, freely flowing aerosol particles are directly aspirated from the atmosphere, so that the true size of the particles/droplets is measured rather than their dry size.

Figure 2 displays the concentration of particles in the 1–2.5 μm size range. This size corresponds to the most important mode of the sea-spray aerosols mass size distribution\(^26,29\), and has the largest direct contribution to the radiative transfer\(^4\). The abundance of aerosols is positively correlated with wind speed, illustrating the influence of the latter on white-cap formation and nebulization via the bursting, at the ocean surface, of bubbles originating from wave breaking\(^26,28,30–35\). Temperature also modulates the emission of particles from the ocean surface, by affecting the viscosity of water\(^4\). The models describing these processes primarily rely on laboratory experiments and/or large-scale ocean monitoring\(^26,28,30–35\). We propose a complementary characterization of aerosol concentration and size distribution, in the form of a continuous series of local measurements of aerosols close to the water surface in a wide variety of environmental conditions.

As shown in Fig. 2, the particle concentration remains small below a windspeed of 3 m/s, and rises beyond. This limit is consistent with the typical threshold for generating white caps\(^36,37\). We fitted our experimental data with the model of Gong et al\(^31\), that estimates the flux of particles as a function of the wind at 10 m altitude $u_{10}$.

$$\frac{dF}{d r_{80}} = 1.373 r_{41}^{3.41} r_{80}^{-A} (1 + 0.05 r_{80}^{3.45}) \times 10^{1.460} \exp(-B^2)$$

(1)
where \( r_{80} \) is the radius at a Relative Humidity (RH) of 80% \((r_{80} = 2r_{dry})\), \( A = 4.7(1 + \Theta r_{dry})^{-0.017r_{dry}^{3.44}} \), \( B = 1 - \log_{10}(r_{80})/0.433 \), and \( \Theta = 30 \). We then applied the temperature correction factor proposed by Jaegle\(^8\), which yields the flux:

\[
\frac{dF}{dr_{80}} = 1.373u_{10}^{3.44}r_{80}^{-4}(1 + 0.057r_{80}^{0.45}) \times 10^{1.607e^{-(-0.3)}} \times (0.3 + 0.17 - 0.0076T^{-2} + 0.00021T^{3}) \]

\( (2) \)

where \( T \) is the sea-surface temperature in °C. The dispersion of the data in the wind-temperature plane results in the spread of the model points in Fig. 2. The fit shows the consistency of this model and its temperature correction with our data.
The significant correlations between aerosols in the 1–10 μm range and local environmental parameters like wind speed and temperature (See Table S3) further suggest that these particles are mainly produced locally, as discussed above.

**Organic particles.** Organic particles were individually identified by the single-particle fluorescence spectrometer (GAP-SPFS, See Supplementary Information) among aerosol particles larger than 1 μm, based on their fluorescence spectrum. While the sensitivity range of the GAP-SPFS restricts the results to the larger particles, we point out that the contribution of the smaller particle sizes to the total aerosol mass is minimal, so that our results can be expected to be representative of the aerosol mass concentration.

This identification relies on the fact that UV-excited fluorescence originates almost exclusively from organic compounds, mainly from aromatic rings. Although the total concentration of fluorescent aerosols locally peaks at 20'000 particles m⁻³, the averaged value over the entire expedition is 355 m⁻³ and the corresponding median is 122 m⁻³. The concentration of fluorescent particles therefore displays much larger relative variations than the total aerosol concentration. Furthermore, their spatio-temporal distributions are different. The percentage of fluorescent aerosols is negatively correlated with water temperature \( R = -0.16, p = 1.5 \times 10^{-3} \) and salinity \( R = -0.46, p < 10^{-4} \) (Fig. 3b and Table S3). Although these correlations are relatively small, the associated \( p \) values, much smaller than 0.01, denote extremely high statistical significance. In contrast, the total aerosol concentration displays the opposite dependence (Figure S3): Warm, salty waters such as those of the Gulf Stream are associated with a higher aerosol concentration as well as a lower organic load as compared to the cold(er) water masses outside of the Gulf Stream. This decoupling is comparable to previously reported observations suggesting different origins and behaviors for organic and non-organic aerosols. The influence of factors like the concentration of organic matter in the seawater, or phytoplankton blooms is likely to contribute to this decoupling.

In particular, different water masses, characterized by their temperature and salinity, exhibit very different forms of biological activity. Cold sub-polar water is usually associated with greater nutrient concentration relative to the Gulf Stream, favoring the development of bacteria as well as phytoplankton. Accordingly, the relative abundance of fluorescing (organic-bearing) particles is also positively correlated with the surface concentration of Chlorophyll a (Fig. 3a,c, Table S3). Although small \( R = 0.135 \) this correlation is well significant statistically \( (p = 0.006) \).

Finally, the relative abundance of organic-bearing aerosols is highly correlated \( R = -0.36, p < 10^{-4} \) with the mass fraction of particles of 1 μm diameter and less, as measured by the optical aerosol sizer (Fig. 3d). This may be understood by considering the different emission dynamics of different particle types. Organic-bearing particles are mainly film drops. In contrast, larger particles are mostly composed of sea salt emitted both as film and jet drops.

Based on the time-series recorded during the expedition, we sought an empirical model capable of describing the fraction of fluorescing (organic) particles as a function of the physico-chemical parameters. We performed an analysis of variance (ANOVA), considering sea-surface temperature, salinity, Chl a concentration, wind, and relative humidity. As displayed in Fig. 4, up to 35% of the variance of our data can be explained by the following two-parameter model:

\[
F = \exp(45.29 - 1.3256 \times S - 1.6972 \times T + 0.0485 \times S \times T)
\]  

where \( F \) is the fraction of organic-bearing particles among the particles larger than 1 μm, in %, \( S \) the salinity in PSU, and \( T \) the sea-surface temperature in °C. Remarkably, already 25% of the variance is explained by the terms where salinity \( S \) is present. Conversely, considering the Chl a concentration, wind speed, and relative humidity did not improve the model.

Salinity is hardly accessible by remote sensing, and only two satellites offer this measurement, namely the Aquarius/SAC-D and SMOS. Therefore, we also sought an alternative empirical model relying only on

---

**Figure 2. Influence of wind on particle abundance.** Hourly-averaged particle concentration in the 1–2.5 μm size range. Data are compared with the empirical wind cap model of Gong et al. applied to the real wind measurements. The boxplots depict the magnitude of the fluctuations induced by the consideration of the Jaegle temperature correction (See text for details).
information that is more easily accessible by remote sensing. The best model can explain up to 17% of the data variance and reads:

\[ F = \exp(6.62 - 0.585 \times T + 0.0112 \times T^2) \] (4)

Although these empirical models are derived for summertime in the north-Atlantic ocean only, they suggest that water mass characteristics are relevant in predicting the organic load of the aerosols. In contrast, the local Chl \( a \) concentration appears less relevant in this regard. Such observations are in line with the reduced correlation between the dynamics of micro-organisms (e.g., plankton blooms) and the organic contents of aerosols\(^{18}\), that may be attributed to the delay between the blooms and the organic enrichment of aerosols\(^{18}\).

The high contribution of salinity to Equation (3) may seem surprising, as it is not expected to influence the biological activity, nor the nebulization mechanism. We may speculate that salinity plays here the role of a tracer of the water masses\(^{44}\). Its domination over sea-surface temperature in that regard could be related to the larger short-term variability of temperature, especially due to the interactions with the atmosphere. Furthermore, as the

Figure 3. Organic aerosols. (a) Fluorescent particles as a fraction of the total of the single-particle counts by the fluorescence spectrometer compared to satellite-measured Chlorophyll \( a \) (background map, data from AQUA MODIS\(^{26}\)), between Boston and St John's. Map created with QGIS 2.12\(^{27}\). (b–d) Influence of (b) surface water temperature and salinity, (c) Chl \( a \), and (d) the mass fraction of particles below 1 \( \mu m \), on the relative abundance of fluorescent particles. Data represent hourly averages for each parameter during our expedition. \( R, p \), and \( N \) denote Pearson's correlation coefficient, \( p \) value and number of samples used for calculating the correlation, respectively. The dotted lines in (c) and (d) display the linear fits yielding the correlation coefficients displayed on the graphs.
deviate from outside the eddy. Overall relationships between environmental parameters with both inorganic and organic aerosols significantly.

These fluorophores dominate the reference spectra of the organisms measured by the GAP-SPFS (Figure S5). Besides the vitamin B6-related pyridoxamine, vitamin B2-related compounds like riboflavin, flavin mononucleotide (FMN) and flavin adenine dinucleotide (FAD) are a dominant feature for the majority of the analysed particles (in particular clusters 1 and 2) is a broad range from 1–30 μm, which allows detection of microorganisms and water sprays in this size range, but not organics from the particles in the Aitken or accumulation modes.

Nevertheless, the variability of the fraction of organics-bearing particles displayed in Fig. 4 is quite large, due to the effect of the local variability in environmental conditions, which add to the global trends. Such events include (i) the crossing of the cold water mass of the Labrador Current, south of Halifax (See Fig. 3a), where the fraction of organic-bearing aerosols can represent up to 20% of the total particles, (ii) three fog episodes (Figs 1a, 3a, and Table S1) where the total particle number concentration significantly rises without any discernible impact on the organic particle concentration, and (iii) the intersection of three eddies by the solar-powered ship (See Table S1 and Figure S6). In particular, a cold-core eddy was characterized by significantly lower temperature and salinity, and higher dissolved oxygen and Chl a concentration. Simultaneously, the concentration of particles increased in the 3–4 μm range and decreased for sub-μm particles. Therefore, the overall relationships between environmental parameters with both inorganic and organic aerosols significantly deviate from outside the eddy.

Organic (fluorescent) aerosol particle speciation. To gain a qualitative insight into the organic particles, we grouped the individual particles into clusters of similar fluorescence spectra as measured by the GAP-SPFS. Figure 5 displays the spectra of the nine most abundant clusters, which individually account for at least 7.5% of the fluorescent particles collected at sea. Together, these clusters account for 85% of the fluorescent particles. It should be noted that spectra could only be measured for particle sizes in the coarse mode, in the size range from 1–30 μm, which allows detection of microorganisms and water sprays in this size range, but not organics from the particles in the Aitken or accumulation modes.

The nine identified families of spectra are well distinguishable, and were observed consistently throughout the entire campaign. A direct assessment of the composition of each aerosol particle based on these characteristic spectra is often impossible, since each aerosol particle is constituted of a complex mixture of fluorescent components. A dominant feature for the majority of the analysed particles (in particular clusters 1 and 2) is a broad blue emission peaking around 420 nm, while excited at 337 nm. This emission is often related to the fluorescence of coenzymes like hydrogenated Nicotinamide adenine dinucleotide (phosphate) (NAD(P)H) and pyridoxamine, as well as cellulose, chitin and pteridine compounds. These fluorophores dominate the reference spectra of the organisms measured by the GAP-SPFS (Figure S5). Besides the vitamin B6-related pyridoxamine, vitamin B2-related compounds like riboflavin, flavin mononucleotide (FMN) and flavin adenine dinucleotide (FAD) are

**Figure 4. Empirical models for the fraction of organic aerosols.** (A) Model including salinity (Equation (3)). (B) Model restricted to satellite-accessible parameters (Equation (4)).
often responsible for broad and weaker features around 550 nm, as observed in clusters 2, 4, 6 and 7. The fluorescence of aromatic amino acids (tryptophan, tyrosine, phenyl-alanine) is not accessible here, since they do not significantly absorb at 337 nm.

The broad fluorescence spectra, characteristic of clusters 6 and 9 are reminiscent of spectra from humic-like substances (HULIS, See also Figure S5), which comprise more than 100 chemical species. They constitute an essential component of natural organic matter, both in soil and in water. These biogenically and terregeneous-derived, heterogeneous organic substances constitute less than 3% in the open ocean, whereas they represent 50–80% of the total dissolved organic matter in freshwaters. In accordance with this, few HULIS were observed, and indeed these represent just 0.2% of the fluorescent aerosols. Most of the HULIS particles were detected close to the coast, which is consistent with the terrestrial origin of these aerosols.

The temporal series of all main clusters except HULIS are highly correlated (Figure S4), showing that the corresponding fluorescing (organic-bearing) species “cocktails” behave similarly in time, space, and for various atmospheric and oceanic conditions. Such observation is consistent with the fact that organic carbon displays similar properties in aerosols at various locations. Despite the relatively constant concentration of total dissolved organic carbon in the North Atlantic (50–80 μM), recent analyses highlighted the fact that several thousand different organic compounds are simultaneously present in surface waters, some of which can be produced and transformed as a result of biological activity. As for the aerosols, organic compounds of different molecular sizes are present in solution, ranging from low to high molecular weight and colloidal material, contributing to the size continuum between dissolved and particulate organic matter. Our results confirm that, at least in the North-Atlantic and in Summer, a homogeneous “cocktail” containing the entire spectrum of dissolved organic compounds, including micro-organisms, is emitted simultaneously and constitutes about 2–5% of the total aerosol mass.

In order to better assess the composition of self-referenced fluorescence clusters, we are currently developing a new dedicated instrument, which will selectively sort particles exhibiting the same spectrum. This selective sampling will allow further laboratory chemical analyses in the laboratory and enable a look-up table for the future campaigns to be created.

**Conclusion**

In summary, this study enabled to characterize the contrasted spatial and temporal dynamics of total and organic marine aerosols, respectively. We propose an empirical parameterization of the fraction of organic aerosols, based exclusively on the salinity and temperature of the water masses encountered during the measurement campaign. Such relationships may provide a deeper insight into the mechanisms of particle formation. It also offers a unique opportunity to estimate the total and organic aerosol load over the ocean from easily-measured ocean and atmosphere physico-chemical parameters. The distinction between particle types with different direct

**Figure 5. Organic particle speciation.** Fluorescence spectra of the main self-identified particle clusters, ordered by decreasing abundances.
References
1. Spada, M., Jorba, O., Prather, K. A. & Bates, T. S. Modelling and evaluation of the global sea-salt aerosol distribution: sensitivity to emission schemes and resolution emission schemes and resolution effects at coastal/orographic sites. Atmos. Chem. Phys. 13, 11957–11965 (2013).
2. Stocker, T. E., Qin, D. et al. IPCC Fifth Assessment Report. In Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change (Cambridge University Press, 2013).
3. Seinfeld, J. H. & Pandis, S. Atmospheric Chemistry and Physics—From Air Pollution to Climate Change, 2nd edition (Wiley-Interscience, 2006).
4. Quinn, P. K., Collins, D. R., Grassman, V. H., Prather, K. A. & Bates, T. S. Chemistry and Related Properties of Freshly Emitted Sea Spray Aerosol. Chem. Rev. 115, 4383–4399, doi:10.1021/cr500713g (2015).
5. de Leeuw, G. et al. Production flux of sea spray aerosol. Rev. Geophys. 49, RG2001, doi:10.1029/2010RG000349 (2011).
6. Norris, S. J. et al. Near-surface measurements of sea spray aerosol production over whitecaps in the open ocean. Ocean science 9, 133, doi:10.5194/os-9-133-2013 (2013).
7. Keene, W. C. et al. Chemical and physical characteristics of nascent aerosols produced by bursting bubbles at a model air-sea interface. J. Geophys. Res. 112, D21302, doi:10.1029/2007JD008864 (2007).
8. Jæger, J., Quinn, P. K., Bates, T. S., Alexander, B. & Lin, J. T. Global distribution of sea salt aerosols: new constraints from in-situ and remote sensing observations. Atmos. Chem. Phys. 11, 3137, doi:10.5194/acp-11-3137-2011 (2011).
9. Hedges, J. I. Global Biogeochmical cycles: progress and problems. Mar. Chem. 59, 67, doi:10.1016/S0304-4203(92)80096-S (1992).
10. Ganit, B. & Meshkidze, N. The physical and chemical characteristics of marine primary organic aerosol: a review. Atmos. Chem. Phys. 13, 3979, doi:10.5194/acp-13-3979-2013 (2013).
11. Russell, L. M., Pandis, S. N. & Seinfeld, J. H. Aerosol Production and Growth in the Marine Boundary-Layer. J. Geophys. Res. 99(D10), 20989–21003, doi:10.1029/94JD01932 (1994).
12. Biller, S. J. et al. Bacterial Vessicles in Marine Ecosystems. Science 343, 183, doi:10.1126/science.1243457 (2014).
13. Modini, R. L. et al. Primary marine aerosol-cloud interactions off the coast of California. J. Geophys. Res. 119, 4137, doi:10.1002/2013JD021006 (2014).
14. O'Dowd, C. D., Smith, M. H., Consterdine, I. E. & Lowe, J. A. Marine aerosol, sea-salt, and the marine sulphur cycle: a short review. Atmos. Chem. Phys. 13, 3977, doi:10.5194/acp-13-3977-2013 (2013).
15. Wilson, T. W. et al. A Marine Biogenic Source of Atmospheric Ice-Nucleating Particles. Nature 525, 234, doi:10.1038/nature14986 (2015).
16. Quinn, P. K. et al. Contribution of sea surface carbon pool to organic matter emission in sea spray aerosol. Nature Geoscience 7, 228, doi:10.1038/ngeo2092 (2014).
17. O'Dowd, C. et al. Connecting marine productivity to sea-spray via nanoscale biological processes: Phytoplankton Dance or Death Discot Scientific Reports 5, 14883, doi:10.1038/srep14883 (2015).
18. Planet Solar Deepwater Expedition. http://www.planetsolar.org/deepwater/ (Date of access: 10/01/2017) (2013).
19. Pinncick, R. G. et al. Fluorescence spectra and elastic scattering characteristics of atmospheric aerosol in Las Cruces, New Mexico, USA: Variability of concentrations and possible constituents and sources of particles in various spectral clusters. Atmos. Environ. 65, 195–204, doi:10.1016/j.atmosenv.2012.09.020 (2013).
20. Vignati, E. et al. Global scale emission and distribution of sea-spray aerosol: Sea-salt and organic enrichment. Atmos. Chem. Phys. 44, 670–677, doi:10.1002/2010JD014346 (2010).
21. Gantt, B. et al. Wind speed dependent size-resolved parameterization for the organic mass fraction of sea spray aerosol. Atmos. Chem. Phys. 11, 8777–8790, doi:10.5194/acp-11-8777-2011 (2011).
22. Long, M. S., Keene, W. C., Kieber, D. J., Erickson, D. J. & Maring, H. A. State-based source function for size- and composition-resolved marine aerosol production. Atmos. Chem. Phys. 11, 1203–1216, doi:10.5194/acp-11-1203-2011 (2011).
23. Sellegrini, K., O'Dowd, C. D., Yoon, Y. J., Jennings, S. G. & de Leeuw, G. Surfactants and submicron sea spray generation. J. Geophys. Res. 111, D22215, doi:10.1029/2005JD006658 (2006).
24. Salter, M. E., Nilsen, E. D., Butler, A. & Bilde, M. On the seawater temperature dependence of the sea spray aerosol generated by a continuous plunging jet. J. Geophys. Res.: Atmos. 119, 9052–9072, doi:10.1002/2013JD021376 (2014).
25. A. S. Goddard Space Flight Center; Ocean Ecology Laboratory, Ocean Biology Processing Group. SeaWiFS Ocean Color Data. doi:10.5067/ORBVIEW-2/SEAWiFS_OC.2014.0 (2014).
26. QGIS Development Team. QGIS Geographic Information System. Open Source Geospatial Foundation Project. http://www.qgis.org (Date of access: 06/10/2016).
27. Clarke, A. D., Owens, S. R. & Zhou, J. C. An ultraline sea-salt flux from breaking waves: Implications for cloud condensation nuclei in the remote marine atmosphere. J. Geophys. Res.: Atmos. 111, D06202, doi:10.1029/2005JD006565 (2006).
28. Fuentes, E., Cowie, H., Green, D., de Leeuw, G. & McFiggans, G. Laboratory-generated primary marine aerosol via bubble-bursting and atomization. Atmos. Meas. Tech. 3, 141–162, doi:10.5194/amt-3-141-2010 (2010).
29. Spada, M., Jorba, O., Pérez García-Pando, C., Janic, Z. & Baldasano, J. M. Modeling and evaluation of the global sea-salt aerosol distribution: sensitivity to size-resolved and sea-surface temperature dependent emission schemes. Atmos. Chem. Phys. 13, 11735–11755, doi:10.5194/acp-13-11735-2013 (2013).
30. Gong, S. L. A parameterization of sea-salt aerosol source function for sub and super-micron particles. Global Biogeochem. Cy. 17, 1997, doi:10.1029/2003GB002079 (2003).
31. Monahan, E. C., Spiel, D. E. & Davidson, K. L. A model of marine aerosol generation via whitecaps and wave disruption, in: Oceanic Whitecaps (edited by: Monahan, E. C. & Mac Niocaill, G.), 167–174, (Springer, 1986).
32. Smith, M. H., Park, P. M. & Consterdine, J. E. Marine aerosol concentrations and estimated fluxes over the sea. Q. J. Roy. Meteor. Soc., 119, 898–924, doi:10.1002/qj.59711951211 (1993).
33. Hoppel, W. A., Frick, G. M. & Fitzgerald, J. W. Surface source function for sea-salt aerosol and aerosol dry deposition to the ocean surface. J. Geophys. Res. 107, D4382, doi:10.1029/2001JD002014 (2002).
E.G.D., D.K., A.D.L., T.M., T.N., D.P., S., G.S. performed the experiment and acquired the data; G.G., P.L., N.R., M.B., J.P.W., J.K., C.H., B.I. conceived the experiment; J.K., CH, A.D.L., D.K., R.H. set it up; N.B., S.B., V.D., Moore, C. M. et al. Processes and patterns of oceanic nutrient limitations. Nature Geoscience 6, 701–710, doi: 10.1038/ngeo1765 (2013).

Acknowledgements
We are grateful to the key commitment of the MS “Tûranor PlanetSolar” crew in the success of the expedition: G. d’Albouville, B. Delbot, V. Brunet, H. Buratti, A. Mikkelsen, A. Simon. We appreciated the experimental support from M. Moret and S. Herrmelin, logistical help from C. Yvon, and assistance by D. Raboud at the University of Geneva. We acknowledge the assistance by N. Gall at MEOPAR, Halifax, P. Goulpié and R. Bros de Puechredon at PlanetSolar SA, and Idronaut SA for lending instruments. We gratefully acknowledge the financial support by the H. Dudley Wright and the Henri Moser Foundations, the Rector’s Office and the Institute for Environmental Sciences at the University of Geneva, as well as a generous anonymous donator. We also acknowledge funding by the Swiss National Science Foundation via the NCCR MUST Project.

Author Contributions
M.B., J.P.W., J.K., C.H., B.I. conceived the experiment; J.K., Ch. A.D.L., D.K., R.H. set it up; N.B., S.B., V.D., E.G.D., D.K., A.D.L., T.M., T.N., D.P., S., G.S. performed the experiment and acquired the data; G.G., P.L., N.R., M.B., J.K., M.B., J.P.W. supervised the expedition trajectory; C.H., J.K., A.D.L., D.S., F.T. analyzed the results, J.K., C.H., B.I., J.P.W., M.B. wrote the manuscript. All authors were given the opportunity to review the paper before submission.
Additional Information
Supplementary information accompanies this paper at http://www.nature.com/srep

Competing Interests: The authors declare no competing financial interests.

How to cite this article: Kasparian, J. et al. Assessing the Dynamics of Organic Aerosols over the North Atlantic Ocean. Sci. Rep. 7, 45476; doi: 10.1038/srep45476 (2017).

Publisher's note: Springer Nature remains neutral with regard to jurisdictional claims in published maps and institutional affiliations.

This work is licensed under a Creative Commons Attribution 4.0 International License. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in the credit line; if the material is not included under the Creative Commons license, users will need to obtain permission from the license holder to reproduce the material. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/

© The Author(s) 2017