



## Supplement of

## Source apportionment of the organic aerosol over the Atlantic Ocean from $53^{\circ}$ N to $53^{\circ}$ S: significant contributions from marine emissions and long-range transport

Shan Huang et al.

Correspondence to: Laurent Poulain (poulain@tropos.de) and Shan Huang (shanhuang\_eci@jnu.edu.cn)

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Figure S1 The transmission efficiency as a function of particle volume equivalent diameter for: the Aerosol container inlet (blue dashed line) derived by the Particle Loss Calculator (von der Weiden et al., 2009), and AMS aerodynamic lens (red dashed line) shown as an average of of transmission efficiency curves in several studies (Bahreini et al., 2008; Jayne, 2000; Takegawa et al., 2009; Zhang et al., 2004). The measuring size range of the mobility particle size spectrometer is depicted.





Figure S2 Wind rose (on relative wind direction, RWD) of mass concentration of (a) organics and (b) eBC, as well as particle number concentration at size of (c) 15nm, and (d) 41nm. Data points are for CR4 at 20-min time resolution. (e) Location of Aerosol container and ship chimney, as well as the identified ship contamination range from 135° to 250° of RWD.



Figure S3 Example of CR3: unfiltered time series for mass concentration of organics (AMS)
and eBC (MAAP) and particle number concentration (Num. conc.) of 15 nm and 41 nm
particles, together with relative wind direction (RWD), relative wind velocity (RWV) and ship
speed.







Figure S5 Correlation between total particle mass concentration from AMS + MAAP (a) without sea salt and (b) with sea salt to that derived from particle number size distribution (PNSD), coloured by mass concentration of sea salt. Note that to derive the density of the particles the following densities have been used for individual species: 1.75 g cm<sup>-3</sup> for sulfate, nitrate and ammonium, 1.4 g cm<sup>-3</sup> for organics, 1.52 g cm<sup>-3</sup> for chloride, 1.77 g cm<sup>-3</sup> for eBC and 2.17 g cm<sup>-3</sup> for the sea salt. CE = 0.7 is chosen and applied.



2 Figure S6 Comparison between sea salt mass concentrations from filter measurements and AMS measurements during Polarstern cruises. The grey shadow presents the uncertainty range of the similar comparison at Mace Head, adapted from Ovadnevaite et al.(2012). Offline sea salt mass concentration is calculated according to the method from Bates et al. (2001) with the equation: sea salt ( $\mu g m^{-3}$ ) = Cl<sup>-</sup> ( $\mu g m^{-3}$ ) + Na<sup>+</sup> ( $\mu g m^{-3}$ ) × 1.47, the factor of 1.47 is the seawater ratio of  $(Na^{+} + K^{+} + Mg^{2+} + Ca^{2+} + SO_4^{2-} + HCO_3^{-})/Na^{+}$ . 



**Figure S7** Diagnostic plots: (a)  $Q/Q_{exp}$  ratio vs. number of factors, (b)  $Q/Q_{exp}$  vs. fPeak between -1 and 1 in step of 0.2 for 5-factor solution, (c) Pearson's correlation coefficient R for time series and mass spectra among 5 factors, (d)  $Q/Q_{exp}$  vs. seeds between 0 to 50 in step of 2, (e) variation of mass fraction of each factor as a function of fPeak, (f) variation of mass fraction of each factor as a function of seeds, (g) comparison of total measured mass and reconstructed mass, (h) sum of the residuals of the fit, (i)  $Q/Q_{exp}$  in time series, (j)  $Q/Q_{exp}$  for each m/z, and (k) scaled residuals for each m/z, with horizontal bars for median, boxes for interquartile and



Figure S8 Time series and mass spectra for OA components of (a, b) 4-factor solution, (c, d)
6-factor solution. (More explanation see Figure S9)



Figure S9 Correlation coefficients of time series and mass spectra for OA components of 4-factor solution and 6-factor solution comparing to 5-factor (selected) solution. The correlations among factors on time-series are shown in bars, while regarding mass spectra shown in cross with the same color code. Correlation coefficients reveal the evolution of the factors when the factor number changes: 4-factor solution already has stable MHOA-like, Comb-OOA-like, Anth-OOA-like and MOOA-like factors, but MNOA factor is not identified; 6-factor solution keeps stable MHOA-like, Comb-OOA-like, MNOA-like and MOOA-like factors, while Anth-OOA factor is separated into two parts. Note that the mass spectra of all factors show general resemblance due to high  $CO^+$  and  $CO_2^+$  ions. This results in higher  $R^2$  on mass spectra than on time-series. 

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Figure S10 Similar variation between water temperature and MNOA (also colored in water
temperature) during four cruises.

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Figure S11 Latitude distribution of MHOA mass concentration (left axis), comparing to the sea salt particle mass concentration (right axis). The MHOA is colored by wind speed (true wind speed). Red boxes mark the cases in which the MHOA and estimated sea salt show similar variation. Grey background indicates continental air masses, white one for marine air masses.



Figure S12 Density map of the maritime traffic with Polarstern cruise tracks (black lines). The
background snapshot was taken from <a href="https://www.marinetraffic.com/en/">https://www.marinetraffic.com/en/</a> on May 2014 and

- 4 assumed to be similar to the situation in 2011 and 2012.
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Figure S13 Fire maps obtained from an online database of MODIS satellite
(<u>http://rapidfire.sci.gsfc.nasa.gov/firemaps/</u>), colored by Comb-OOA mass concentration
during Polarstern cruises. The black arrows show the ship direction.

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