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Supplement of

Wintertime aerosol dominated by solid-fuel-burning emissions across Ireland: insight into the spatial and chemical variation in submicron aerosol

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Table S1. Ionization efficiencies (IE) and relative ionization efficiencies (RIE) obtained through the calibration with ammonium nitrate and ammonium sulfate.

	Dublin	Carnsore Point	Birr	Mace Head
IE	3.24e-11	3.28e-11	3.24e-11	4.47e-11
RIE (sulfate)	0.96	0.74	0.96	0.56
RIE (ammonium)	7.58	6.74	7.58	4.76

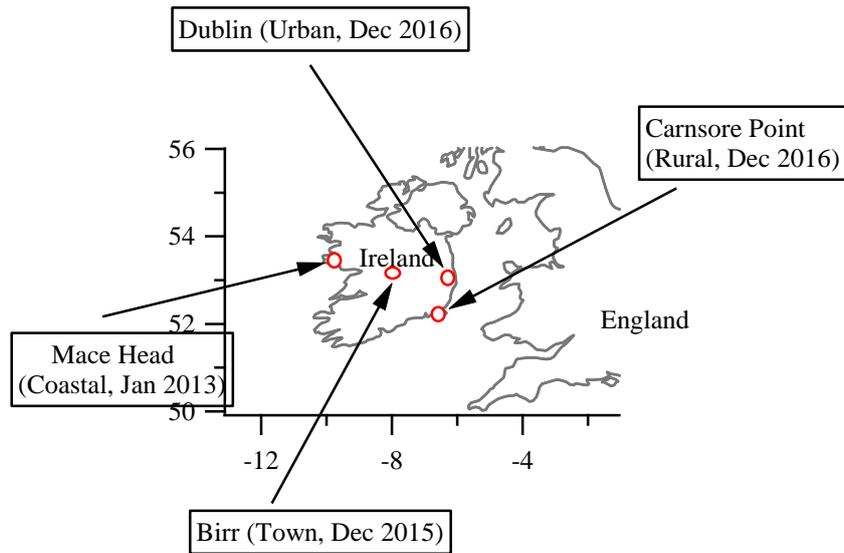


Figure S1. The location of the four measurement sites in Ireland: an urban background site in Dublin; a site in the midland town of Birr; a rural site at Carnsore Point on the east coast and a coastal site at Mace Head on the west coast. The x-axis is the latitude and the y-axis is the longitude. The measurements at the Dublin site and Carnsore Point were conducted simultaneously in December 2016. The measurement at the Birr site was conducted in December 2015 and was in January 2013 for Mace Head.

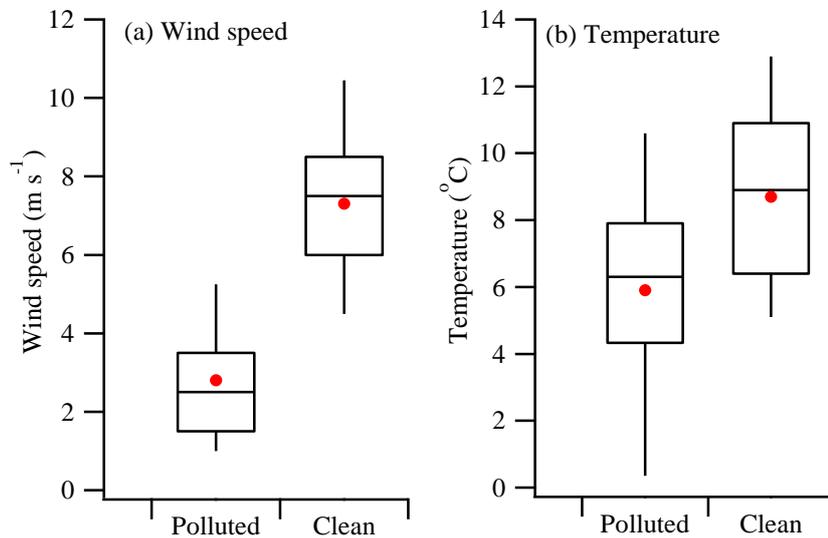


Figure S2. Wind speed (a) and temperature (b) during polluted and clean periods in Dublin December 2016. The boxes represent the 25th percentile (lower edge), median (solid line), mean (cube mark), and 75th percentile (higher edge). The whiskers represent the 5th and 95th percentiles.

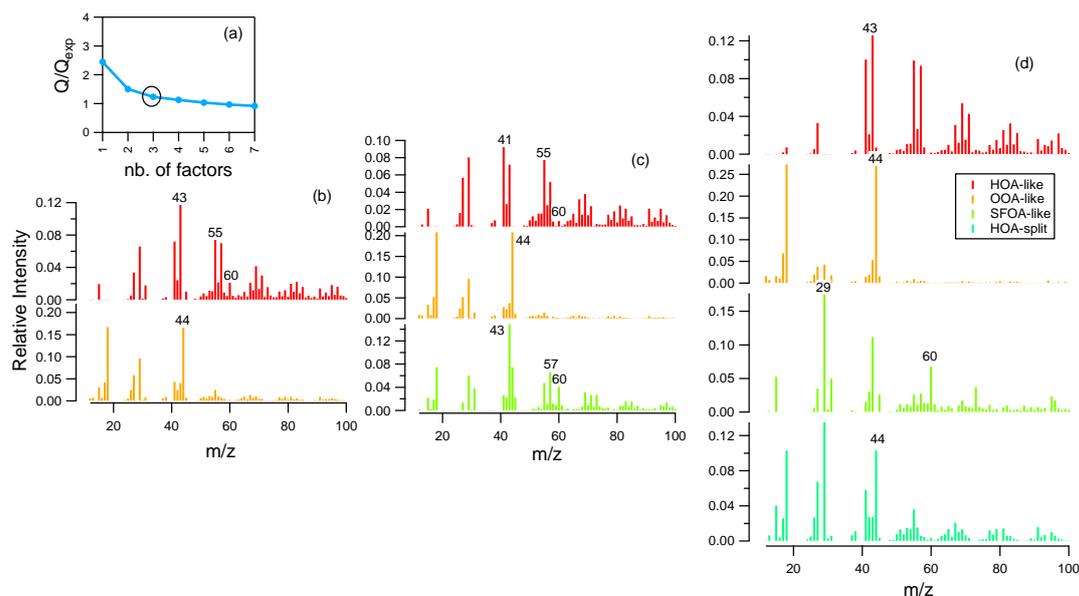


Figure S3. Q/Q_{exp} as a function of number (nb.) of factors (a) for the free PMF in Dublin December 2016. Free PMF with 2-4 factor solutions (b-d). There was a slight drop in the Q/Q_{exp} value from 2 to 3 factors, and Q/Q_{exp} was 1.2 at the 3-factor solution. However, no strong change in Q/Q_{exp} was observed by further increasing the number of factors. The 4-factor solutions led to the splitting of factors as indicated by a good time series correlation between Factor 1 and Factor 4 ($R=0.88$).

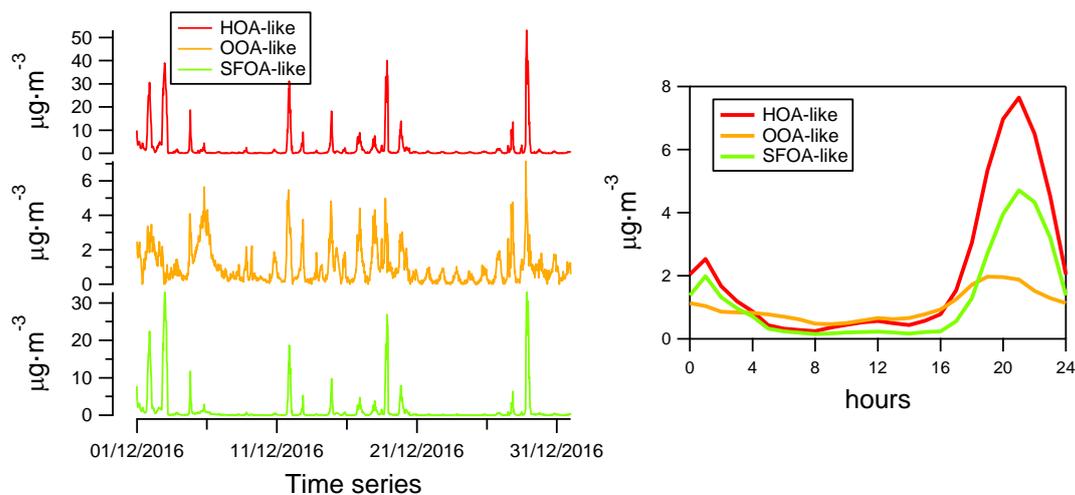


Figure S4. The time series (left) and diurnal pattern (right) of the free PMF 3-factor solution in Dublin December 2016.

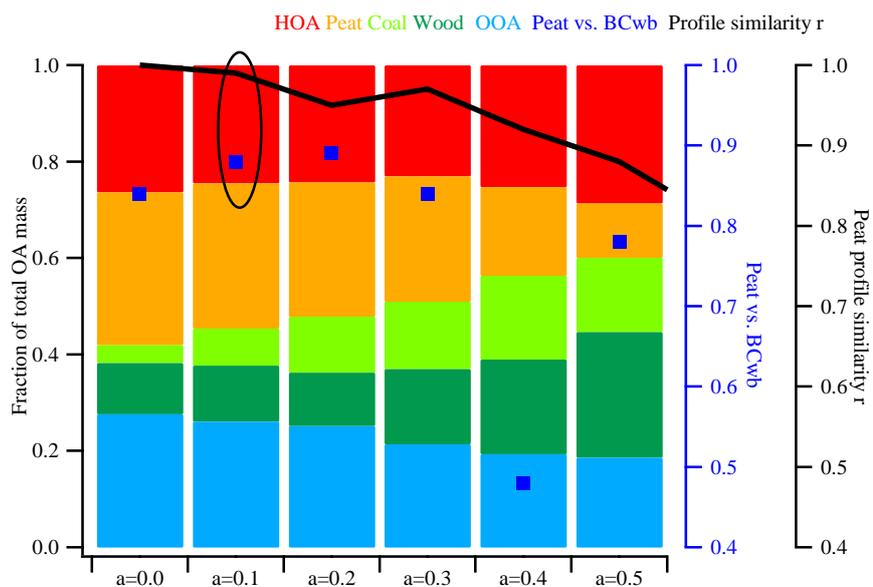


Figure S5. Relative fraction of the resolved factors (left axis), as well as the time series correlation coefficient r of peat versus BB_{wb} and profile similarity between the resolved factor and reference profile (right axis) as a function of a value for the ME-2 solutions in Dublin December 2016. The best a value approach is highlighted by a black circle.

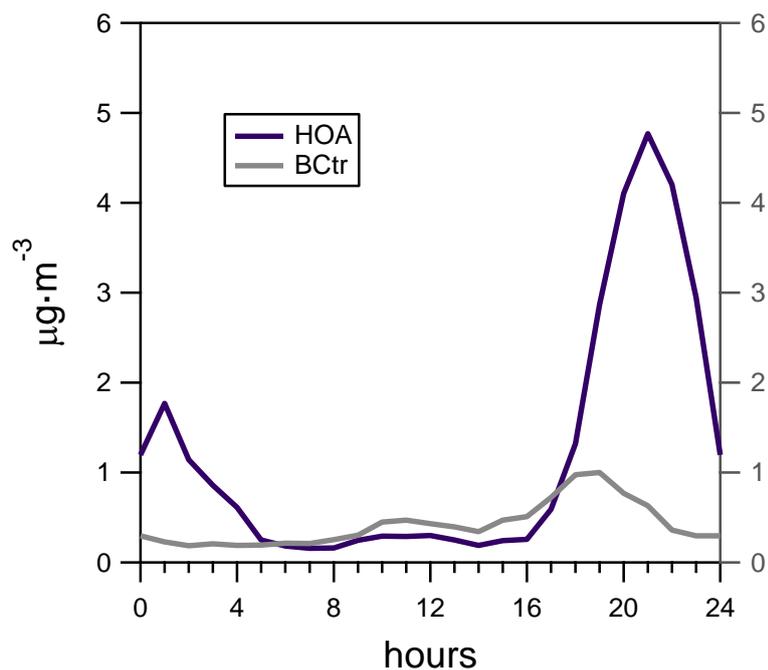


Figure S6. The diurnal pattern of HOA and BCtr in Dublin over the entire period in December 2016.

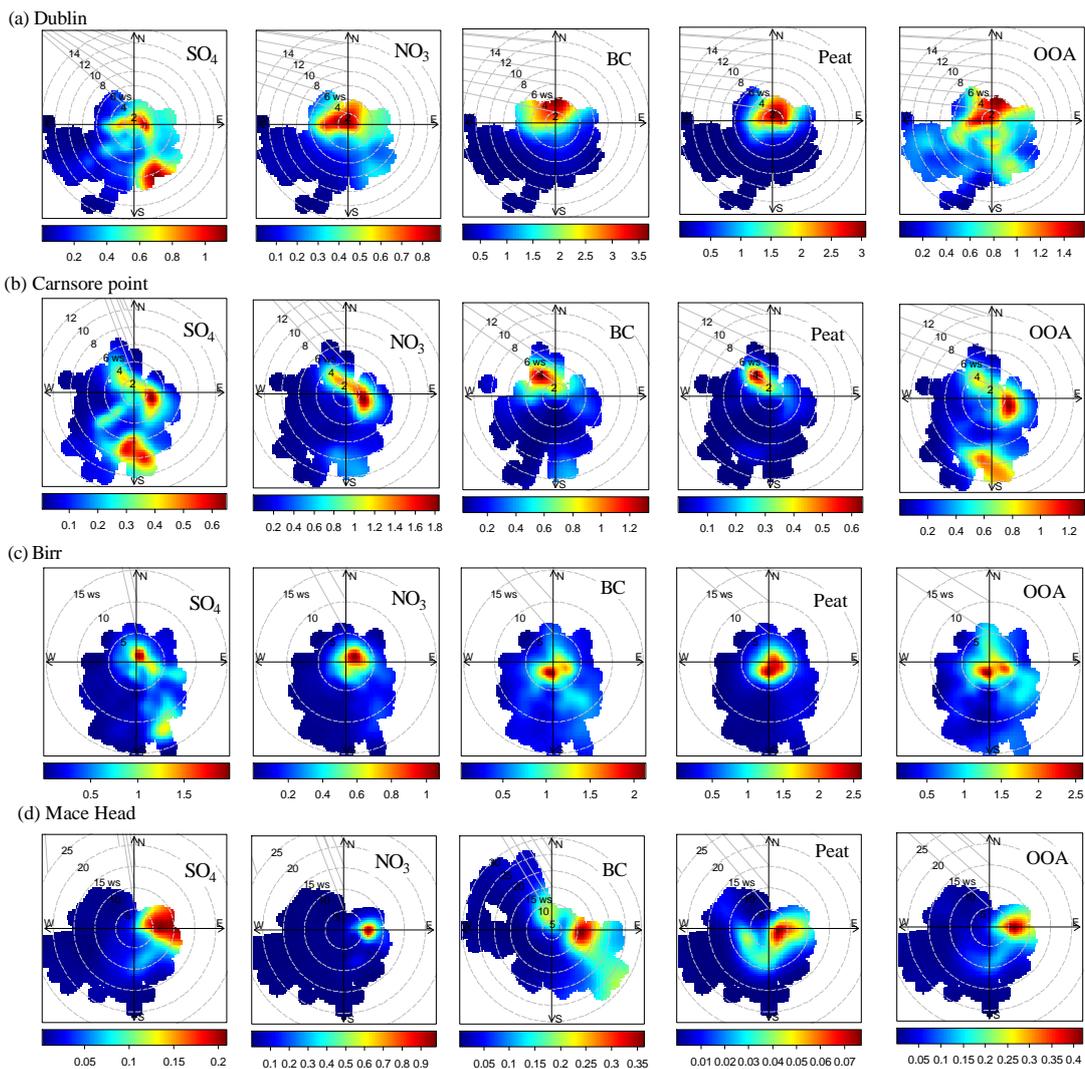


Figure S7. Wind rose of sulfate (SO_4), Nitrate (NO_3), black carbon (BC), Peat, and oxygenated organic aerosol (OOA) factor in (a) Dublin; (b) Carnsore Point; (c) Birr; and (d) Mace Head. The plots are color-coded based on concentration of each species in $\mu\text{g m}^{-3}$. Wind rose plots were generated with OpenAir software in R (Carslaw and Ropkins, 2012).

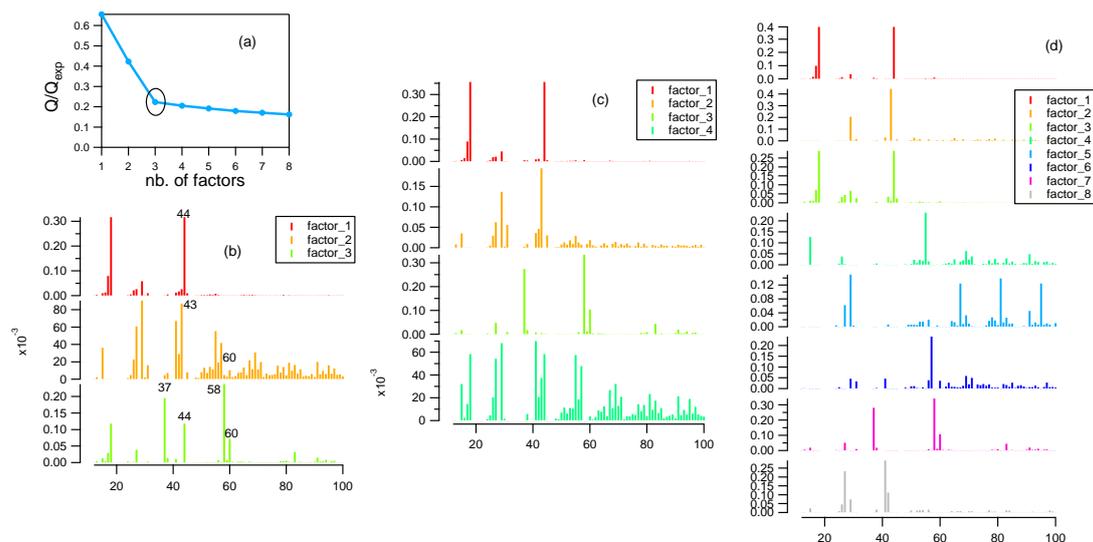


Figure S8. Q/Q_{exp} as a function of number (nb.) of factors (a) for the free PMF solutions at Carnsore Point in December 2016; free PMF with 3-4 factor solutions (b-c); and 8-factor solution (d). There was a large drop in the Q/Q_{exp} value from 2 to 3 factors, and Q/Q_{exp} was 0.2 at the 3-factor solution. However, no strong change in Q/Q_{exp} was observed by further increasing the number of factors. The 4-factor solution led to the splitting of factors and no more meaningful factors could be identified

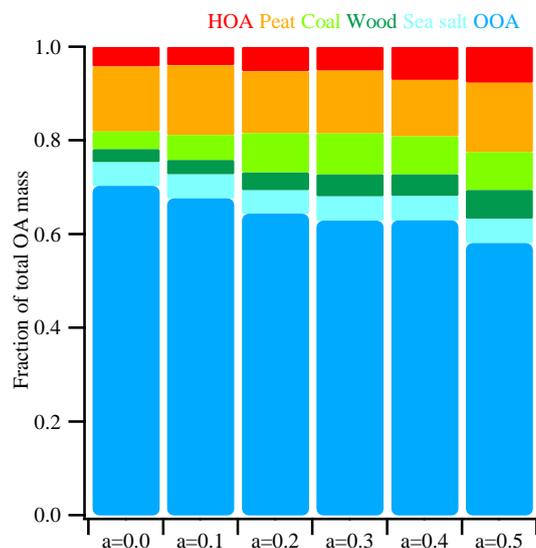


Figure S9. The relative fraction of HOA, peat, coal, wood, sea salt, and OOA factors as a function of a values for the ME-2 solutions at Carnsore Point in December 2016.

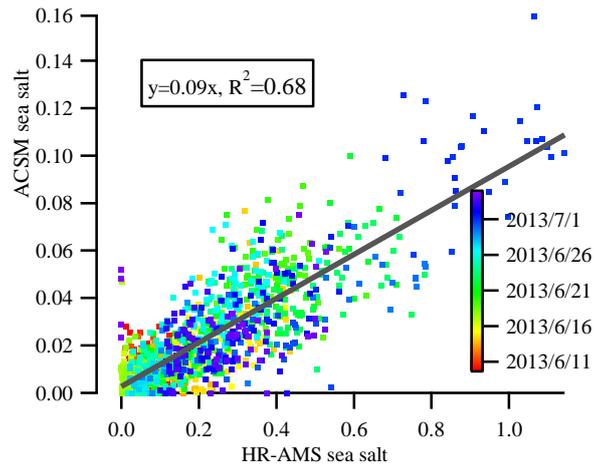


Figure S10. Comparison of the time series of sea salt between the one resolved by ACSM-PMF and the standard HR-AMS at Mace Head during summer 2013. The slope is 0.09. The summer data is shown here because the winter HR-AMS data was not available.

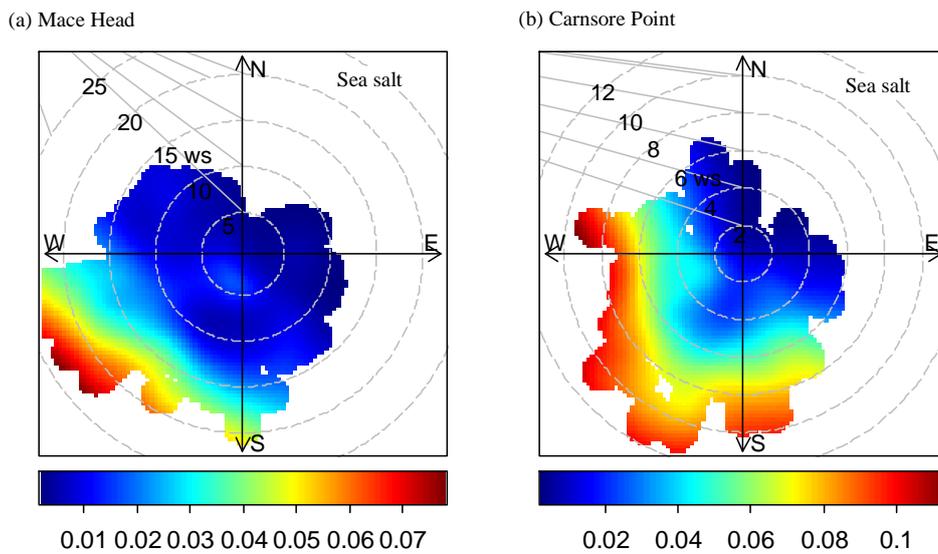


Figure S11. Wind rose of sea salt factor at Mace Head (a) and Carnsore Point (b). The plots are color-coded based on concentration of each species in $\mu\text{g m}^{-3}$. Wind rose plots were generated with OpenAir software in R (Carslaw and Ropkins, 2012).

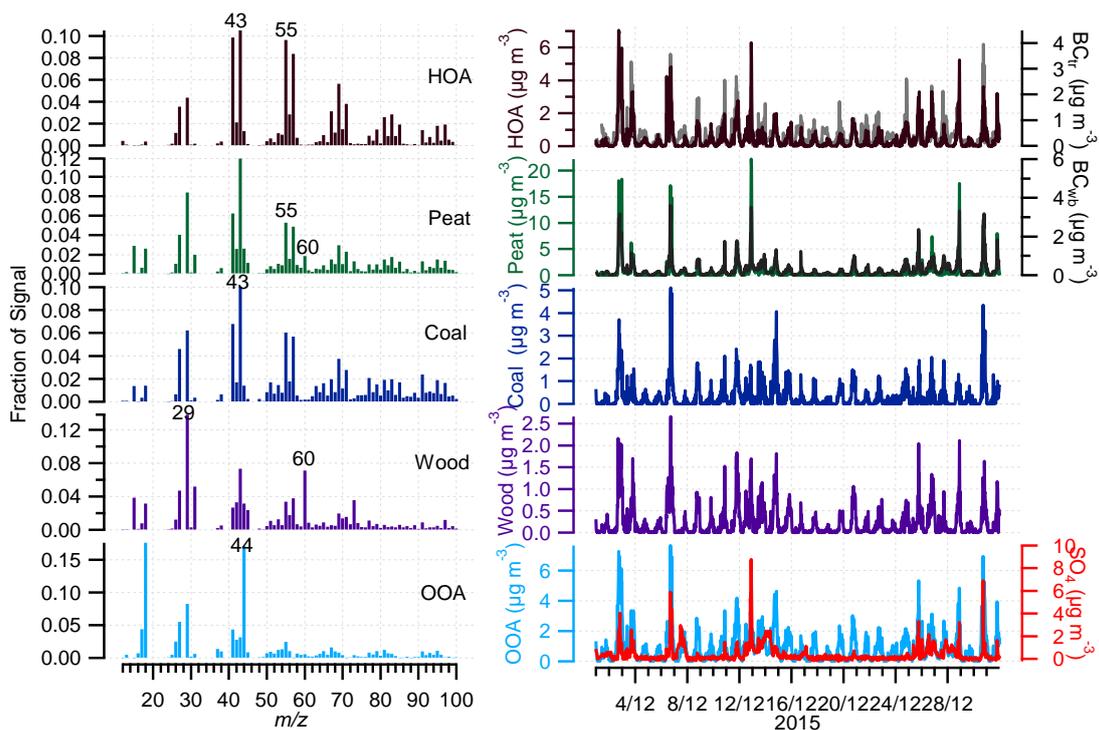


Figure S12. Profiles and time series of HOA, peat, coal, wood, and oxygenated OA (OOA) at the midland town site in Birr December 2015. The time series of BC_{tr} , BC_{wb} , and sulfate (SO_4) were also included to support OA source apportionment.

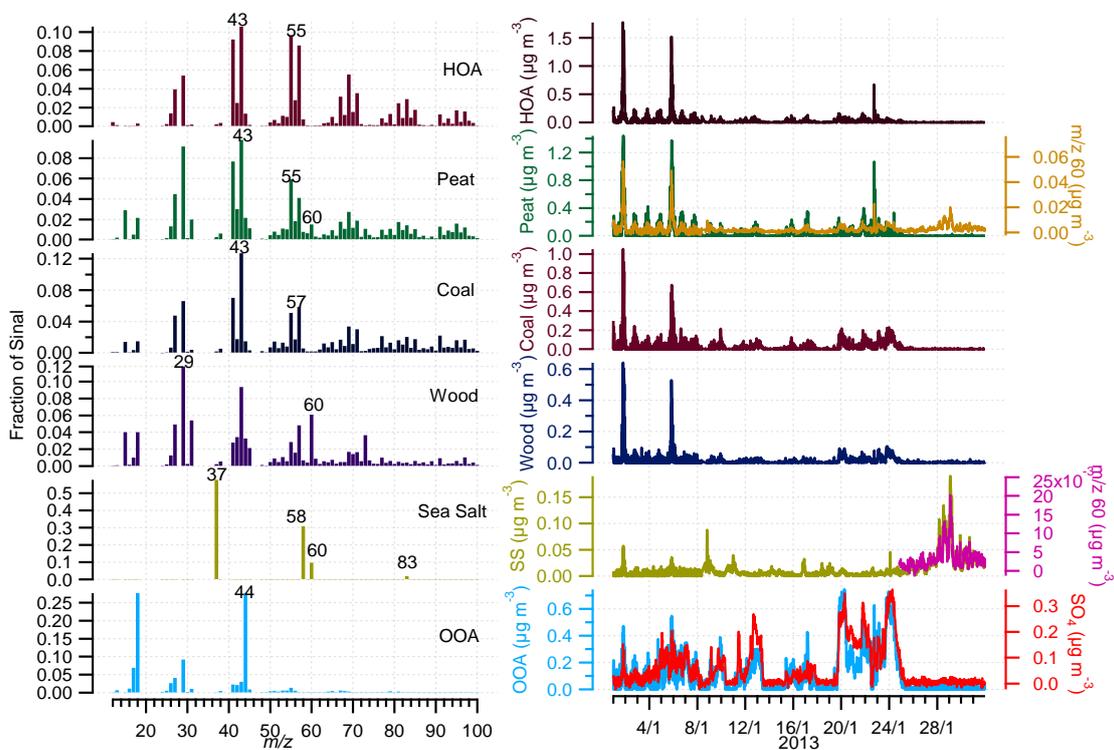


Figure S13. Profiles and time series of HOA, peat, coal, wood, sea salt, and oxygenated OA (OOA) at the coastal site in Birr January 2013.

site in Mace Head January 2013. The time series of sulfate (SO_4) and m/z 60 were also included to support OA source apportionment. The time series of m/z 60 show a better correlation with sea salt than peat on 26-31 Jan.

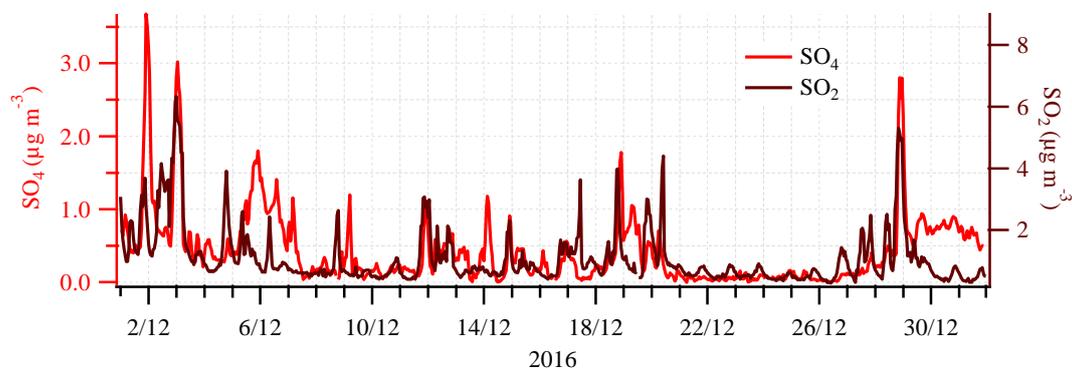


Figure S14. Time series of sulfate (SO_4) and its precursor gas of SO_2 in Dublin in December 2016.

References:

Carslaw, D. C., and Ropkins, K.: Openair-an R package for air quality data analysis, *Environmental Modelling & Software*, 27, 52-61, 2012.