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Supplementary Information

Characterization of primary organic aerosol from domestic wood, peat, and coal burning in Ireland

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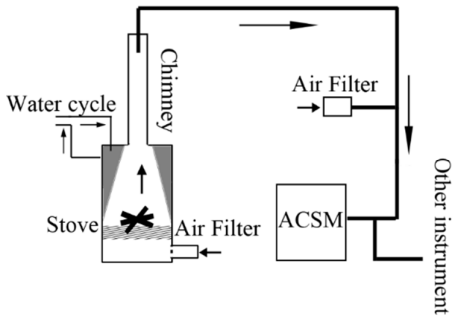
The following materials are included:

- Combustion and sampling system, Scheme S1
- Emission factors and caloric values for oil, peat, coal, and wood, Table S1
- Correlation coefficient (R^2) between ACSM profiles of different sources and PMF factors
- The households by the type of central heating (oil, peat, coal, and wood) from Central Statistics Office, 2011, Figure S1
- Relative fraction of ACSM measured species, Figure S2
- Mass spectra of each type of fuel under different states, Figure S3-5
- Relative difference of dry wood and smoky coal MS profile compared to peat at each m/z, Figure S6
- Time series and mass spectra of PMF solutions, Figure S7-9
- Relative contribution of the resolved factors and correlation between OOA and sulfate with different *a* values (0-0.2), Figure S10
- Back trajectory during the measurement period in Galway, Ireland, Figure S11

Summary: 13 pages, 1 scheme, 2 tables, and 11 figures

Figerprinting Setup and ACSM data analysis: A boiler stove is used for both home heating and the generation of hot water. The combustion chamber was built into a wall with the water pipe network inside the wall behind the chamber. The water pipes will take up part of the heat generated in the chamber, producing hot water for everyday use and also circulating through the central heating system warming up the house. The open fire chamber was directly connected to a chimney having no emission control. During each type of fuel sampling, new fuel was added to maintain the combustion which is always the case for the real application instead of waiting for its extinction and igniting a new burning. And each type of sampling fuel was continuously burned for at least 1 hour with a total use of fuel >5kg. ACSM measured the emission with ~1 min resolution and 1 h ACSM data was averaged to get the relative mass contribution and mass spectrum. The NR-PM_1 aerosols generated from the combustion of fuels were collected using a sampling line connected to the chimney. The sampling line was made of ordinary ½ inch copper pipe which extended approximately 10 cm inside the chimney flue. An automobile fuel filter was fitted 2 m downstream of the inlet, which was effective in trapping moisture and large particulate matter. This was followed by a gate valve to restrict the flow of smoke and allow dilution with clean air. The gate valve was adjusted to allow a dilution rate in the range of 80-160:1. The total length of copper line between the chimney and the mobile station was around 10 meters, which provided sufficient time for the aerosol to cool down to ambient temperature before ACSM measurement.

ACSM spectra analysis were performed using the standard ACSM analysis software (version: ACSM_local_1.5.12.0) provided by Aerodyne which is written within Wavemetrics Igor^{TM}. Collection efficiency (CE) in terms of the mass fraction of ammonium nitrate, particle acidity, and water content should be considered to account for sampling losses as suggested by Middlebrook et al (2012). However, in this study, CE-corrected NR-PM1 results in higher mass concentration than simultaneous PM_{10} measured by TEOM at several evening/night time peaks. Thus, the CE need to be further investigated. Here, we assumed a CE of 1, which provides a lower limit for ACSM-measured mass concentration. A CE of 1 was also used by Canonaco et al. (2013) in which they also found composition dependent CE would underestimate CE resulting in a higher CE-corrected PM_1 than collocated TEOM PM_{10}. However, changes in CE won't affect the relative contribution of all species since CE is applied to all measured specie



Scheme S1. Schematic of Irish residential solid fuel combustion and ACSM measurement system.

Table S1 Emissions factors from the CEPMEIP database (TNO,2001)[1,2,3]

Source	Emission factor Kg $PM_{2.5}$ TJ^{-1}	Net calorific value MJ kg^{-1}	KJ m^{-3}
Bituminous coal	30	27.84	
Sod peat	60	13.1	
Briquettes	60	18.55	
Petroleum coke	30	32.1	
Fuel oil	40	41.24	
Gas oil	5	43.31	
Kerosene	5	44.2	
LPG	0.2	47.16	
Natural gas	0.2		39334
Biomass (wood)	270	~16.00	

Table S2**.** Correlation coefficient (R^2) between ACSM profiles of different sources and PMF factors (dry wood (DW), wet wood (WW), dry raw peat (DP), wet raw peat (WP), peat briquettes (PB), bituminous (smoky) coal (SC), and ovoids (smokeless, based on anthracite) coal (SLC))

R^2	DW	WW	DP	WP	PB	SC	SLC	HOA[4]	BBOA[5]
DW	1	0.91	0.62	0.62	0.69	0.37	0.69	0.30	0.77
WW	0.91	1	0.47	0.48	0.54	0.32	0.54	0.21	0.57
DP	0.62	0.47	1	0.98	0.96	0.87	0.88	0.8	0.78
WP	0.62	0.48	0.98	1	0.99	0.81	0.89	0.82	0.84
PB	0.69	0.54	0.96	0.99	1	0.77	0.92	0.77	0.88
SC	0.37	0.32	0.87	0.81	0.77	1	0.78	0.76	0.51
SLC	0.69	0.54	0.88	0.89	0.92	0.78	1	0.64	0.83
HOA	0.30	0.21	0.80	0.82	0.77	0.76	0.64	1	0.58
BBOA	0.77	0.57	0.78	0.84	0.88	0.51	0.83	0.58	1

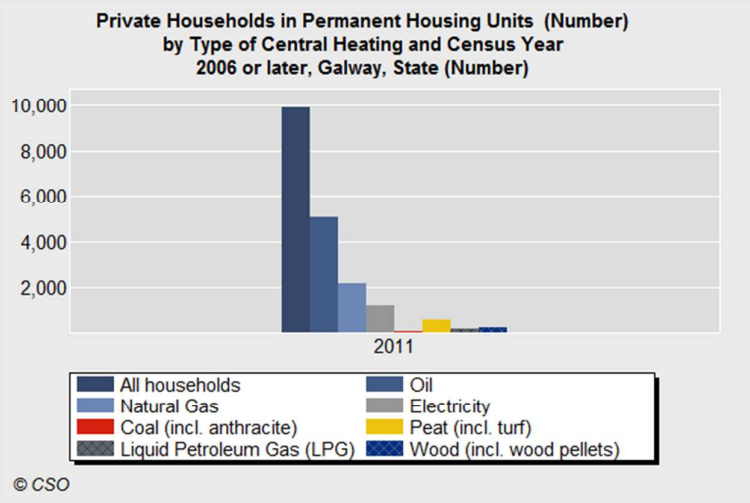


Figure S1. Private households by type of central heating in Galway (Image reprinted with permission from Central Statistics Office, 2011)[6].

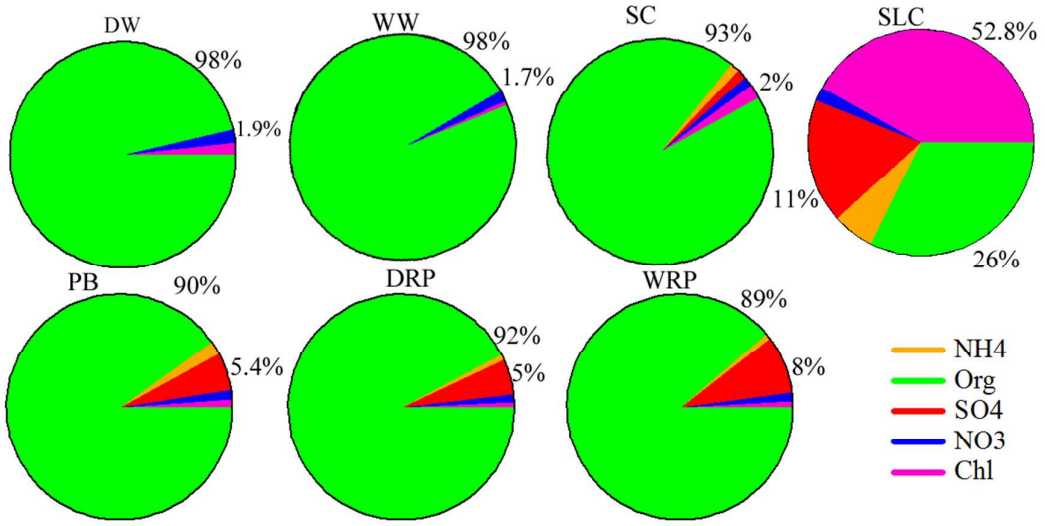


Figure S2. Chemical composition of non-refractory emissions (i.e. Organics, sulfate, nitrate, ammonium, and chloride) from burning dry wood (DW), wet wood (WW), dry raw peat (DRW), wet raw peat (WRP), peat briquettes (PB), smoky coal (SC), and smokeless coal (SLC) in a typical residential Irish stove. Peat is an accumulation of partially decayed vegetation, it contains more minerals (including sulfur) than fresh biomass (e.g. wood).

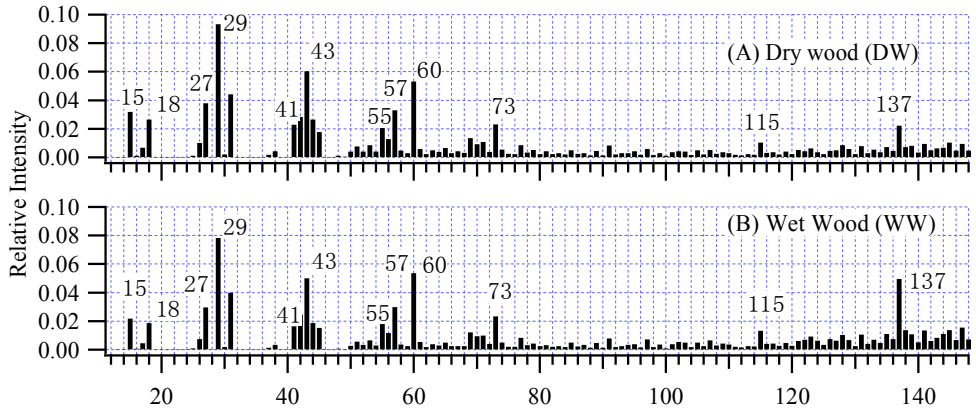


Figure S3. Average normalized mass spectra of measured organic aerosols from the combustion of (A) dry wood; (B) wet wood in a typical domestic Irish stove using an ACSM.

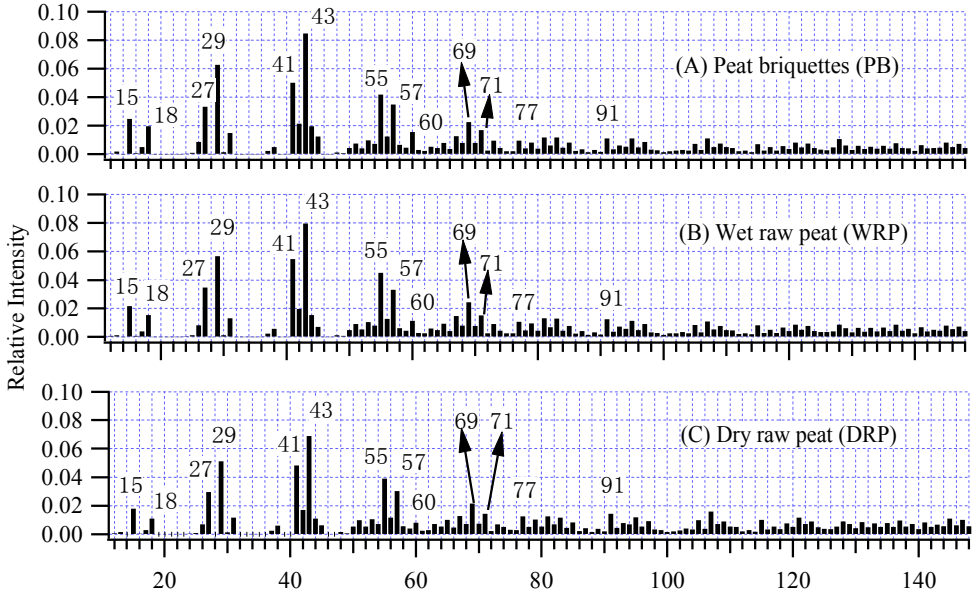


Figure S4. Average normalized mass spectra of measured organic aerosols from the combustion of (A) peat briquettes; (B) wet raw pet; (C) dry raw peat in a typical domestic Irish stove using an ACSM.

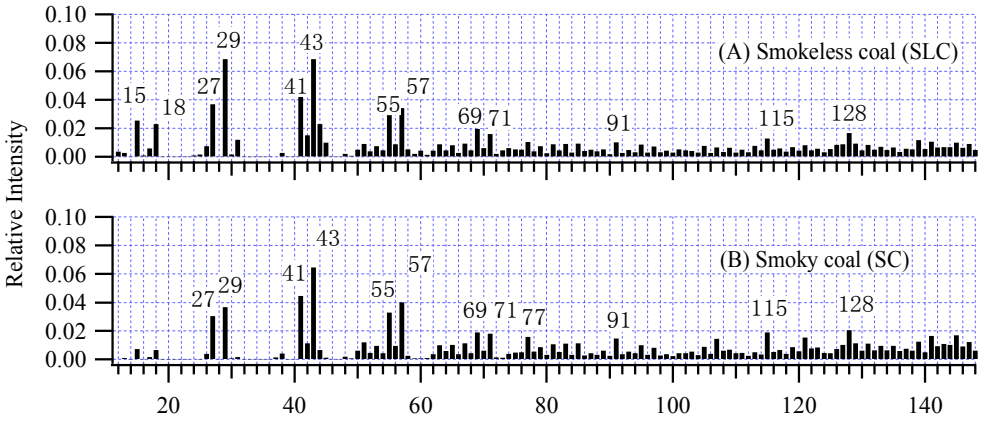


Figure S5. Average normalized mass spectra of measured organic aerosols from the combustion of (A) smokeless coal; (B) smoky coal in a typical domestic Irish stove using an ACSM.

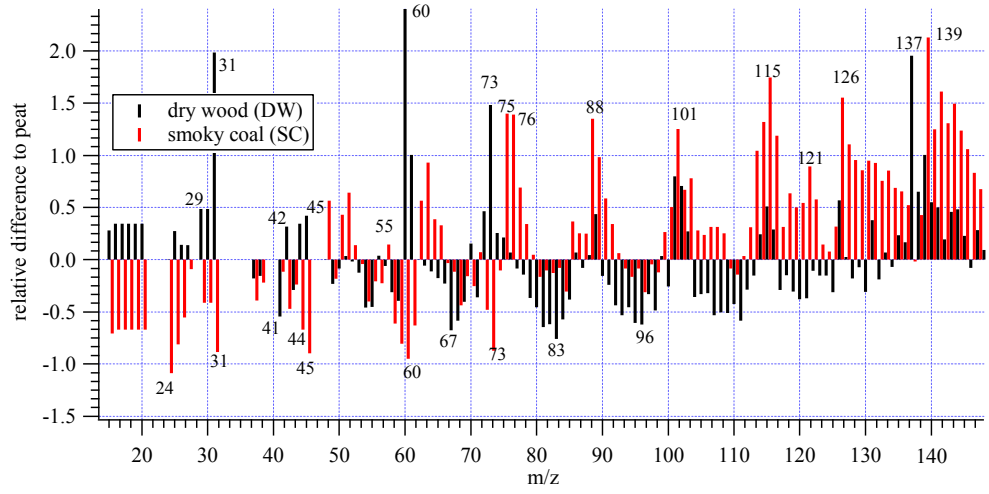


Figure S6. Relative difference of dry wood and smoky coal MS profile compared to peat at each m/z.

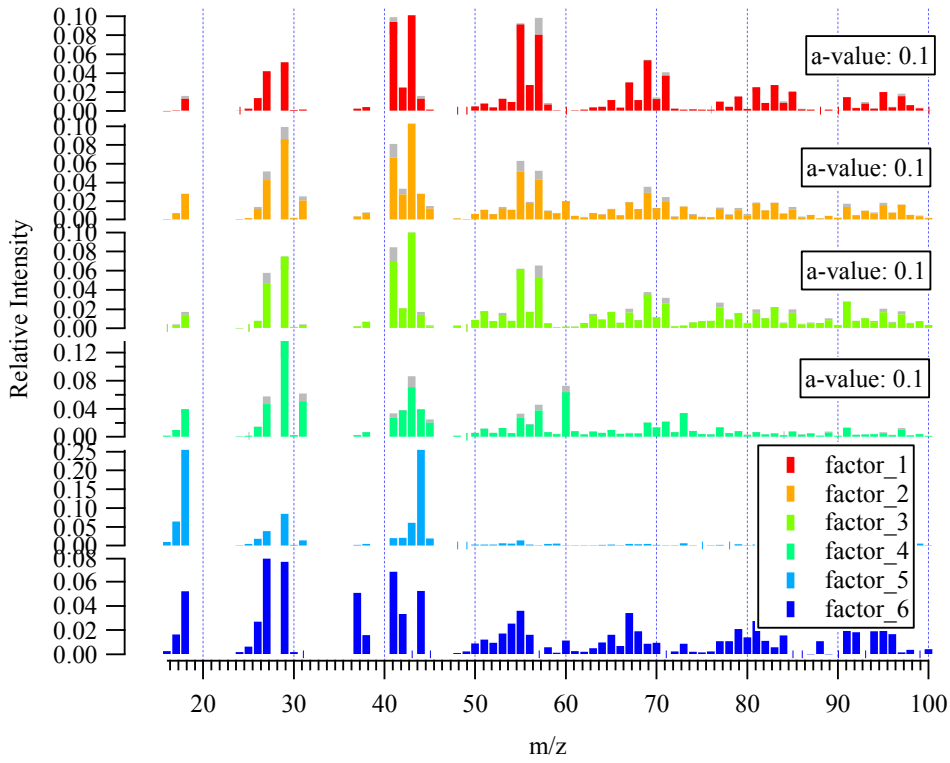


Figure S7. Factor profiles (mass spectra) of the 6-factor solution with four primary factors constrained and two additional left free. Factor 6 is not interpretable by comparing with the profiles in AMS database (http://cires1.colorado.edu/jimenez-group/AMSsd/). The α-value method within ME-2 was applied. Oil burning (factor 1) profile is from ambient data PMF-derived hydrocarbon-like organic aerosol (HOA) (Crippa et al. 2013)[4]. Peat (factor 2), coal (factor 3), and wood (factor 4) reference profiles are from fingerprinting experiments (Figure 1). Grey bar in the back represents reference profile employed.

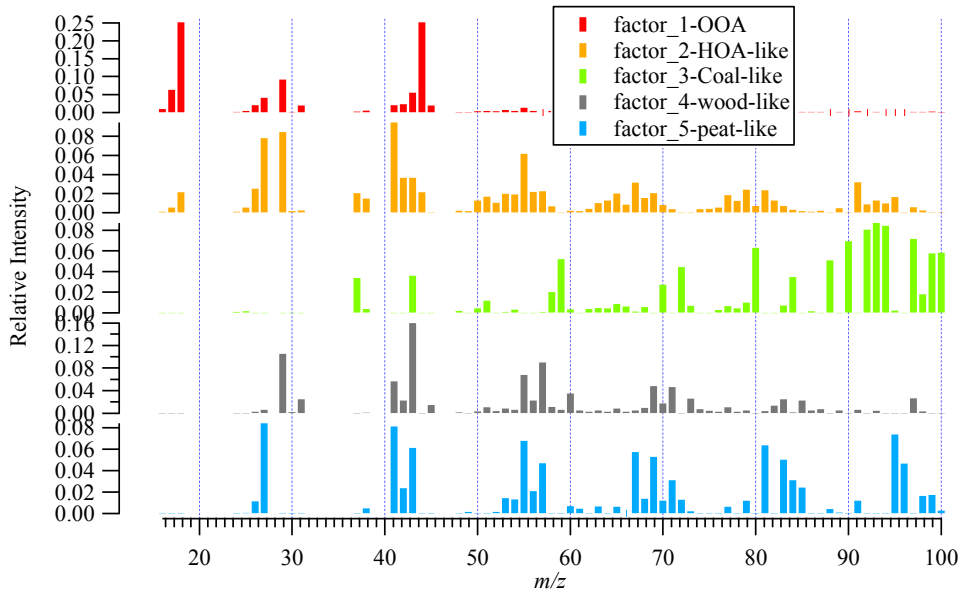


Figure S8. Profiles of 5-factor free PMF solution. Factor 1 is a typical OOA profiles with high m/z 44 signal. Factor 2 shows no signal at m/z 60 and has a higher fraction of signals at lower m/z values, and it is HOA-like. Factor 3 has a higher fraction at higher m/z values, thus it is coal-like. In contrast, both factor 4 and 5 have elevated signals at m/z 60, and the allocation of signals at m/z 29, makes factor 4 wood-like and factor 5 peat like. However, primary factors from free PMF are highly mixed due to rotational ambiguity arising from similar emission time. Thus, it is inappropriate to use this solution to estimate the contribution of different sources.

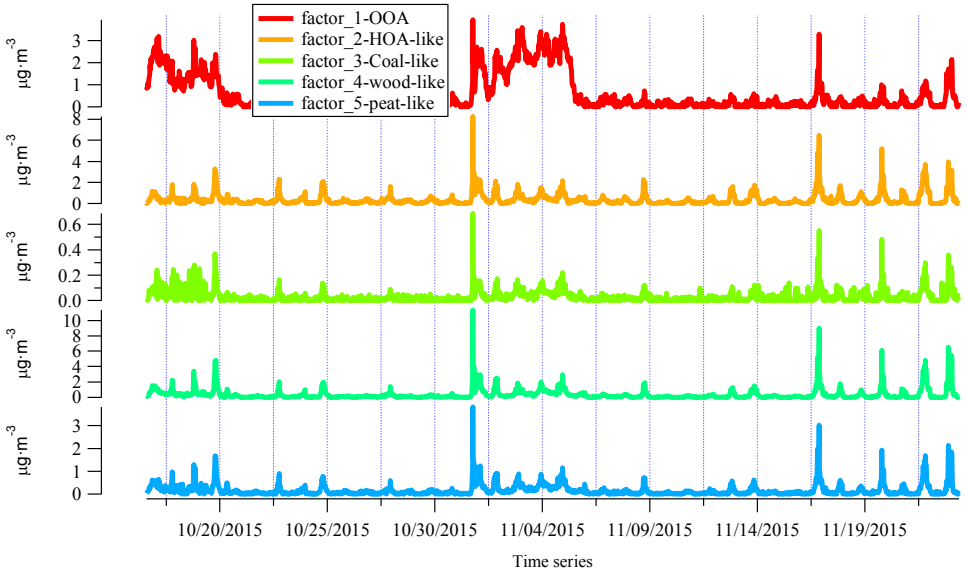


Figure S9. Time series of 5-factor free PMF solution.

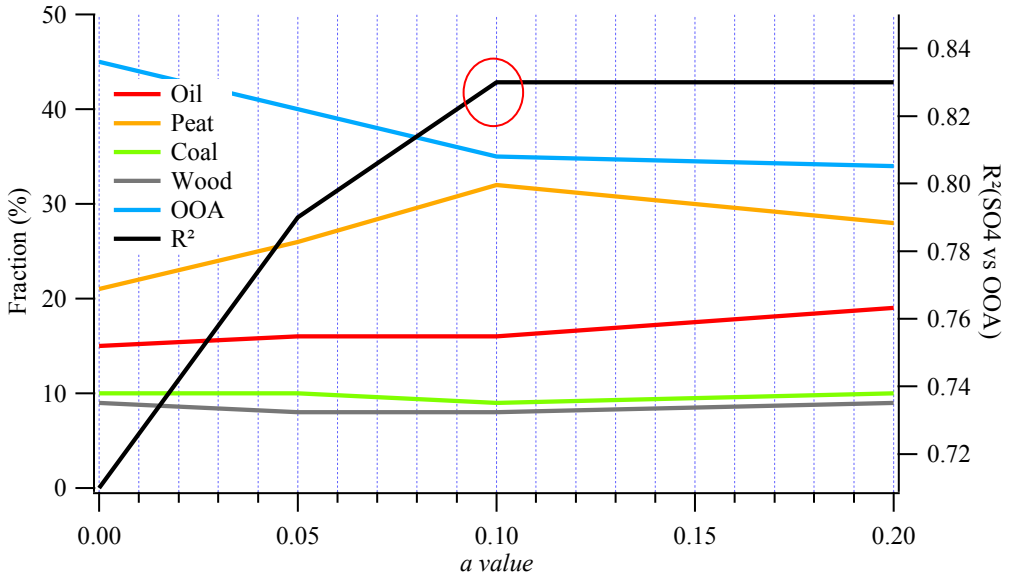


Figure S10. The relative contribution of oil, peat, coal, wood, and OOA (left axis) over the whole periods to total OA mass as well as correlation (R^2) between sulfate and OOA (right axis) as a function of *a* value. An *a* value of 0.1 was selected (red cycle) from which the R^2 starts to level off.

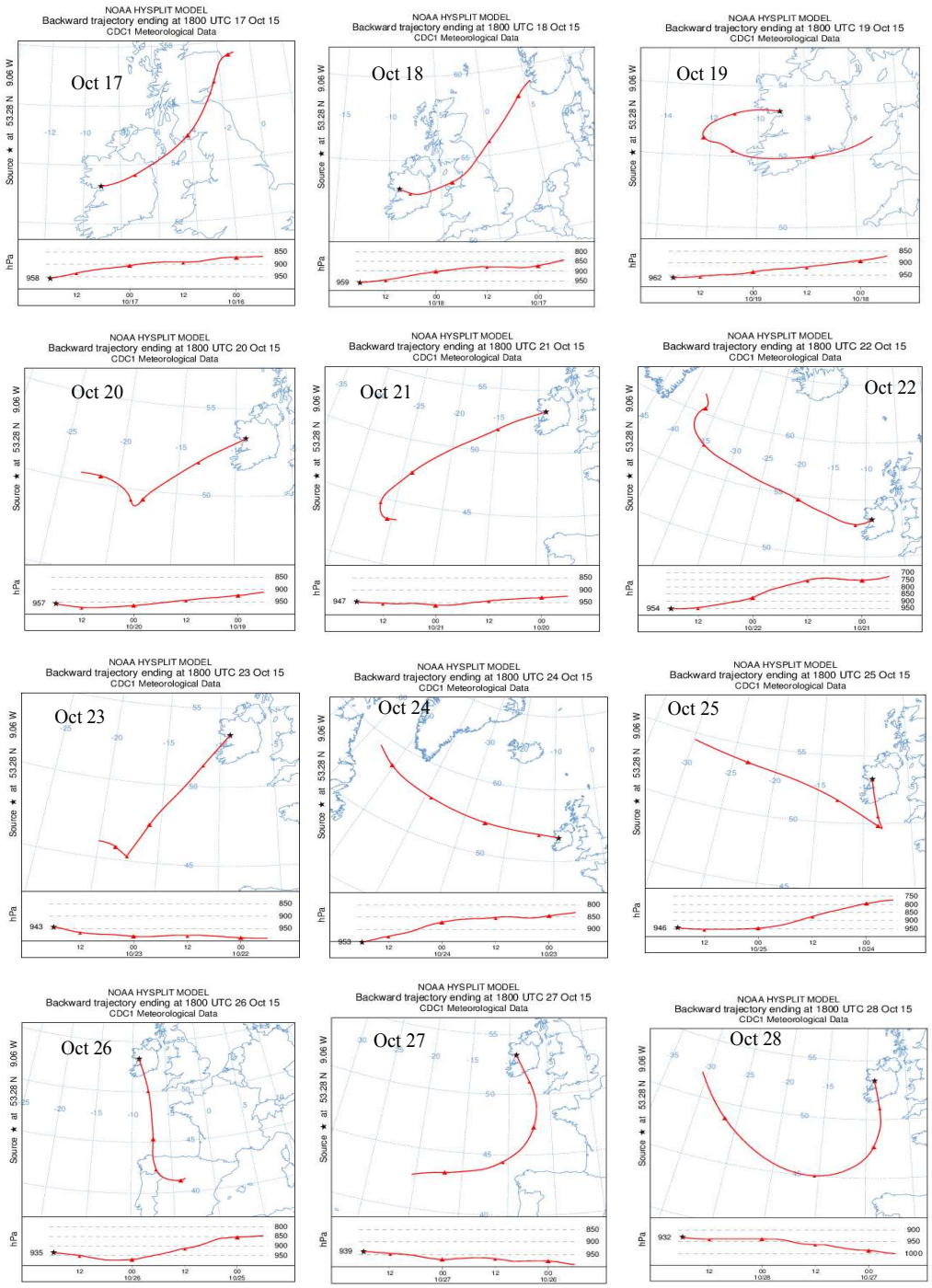


Figure S11. Backward trajectory analysis for 48 h by NOAA Hysplit4 model[7] ending at 18:00 from October 17 to November 21, 2015 in Galway, Ireland. From Oct 17 to 19 (or S1 in Figure 3) and from Nov 1 to 4 (or S2 in Figure 3), the air masses have a continental origin (from the mainland Europe, the UK, and Ireland itself). From Oct 19 to 31 (or M1 in Figure 3) and Nov 5 to 21 (M2 and M3 in Figure 3), the air masses have a marine origin with short stay in Ireland.

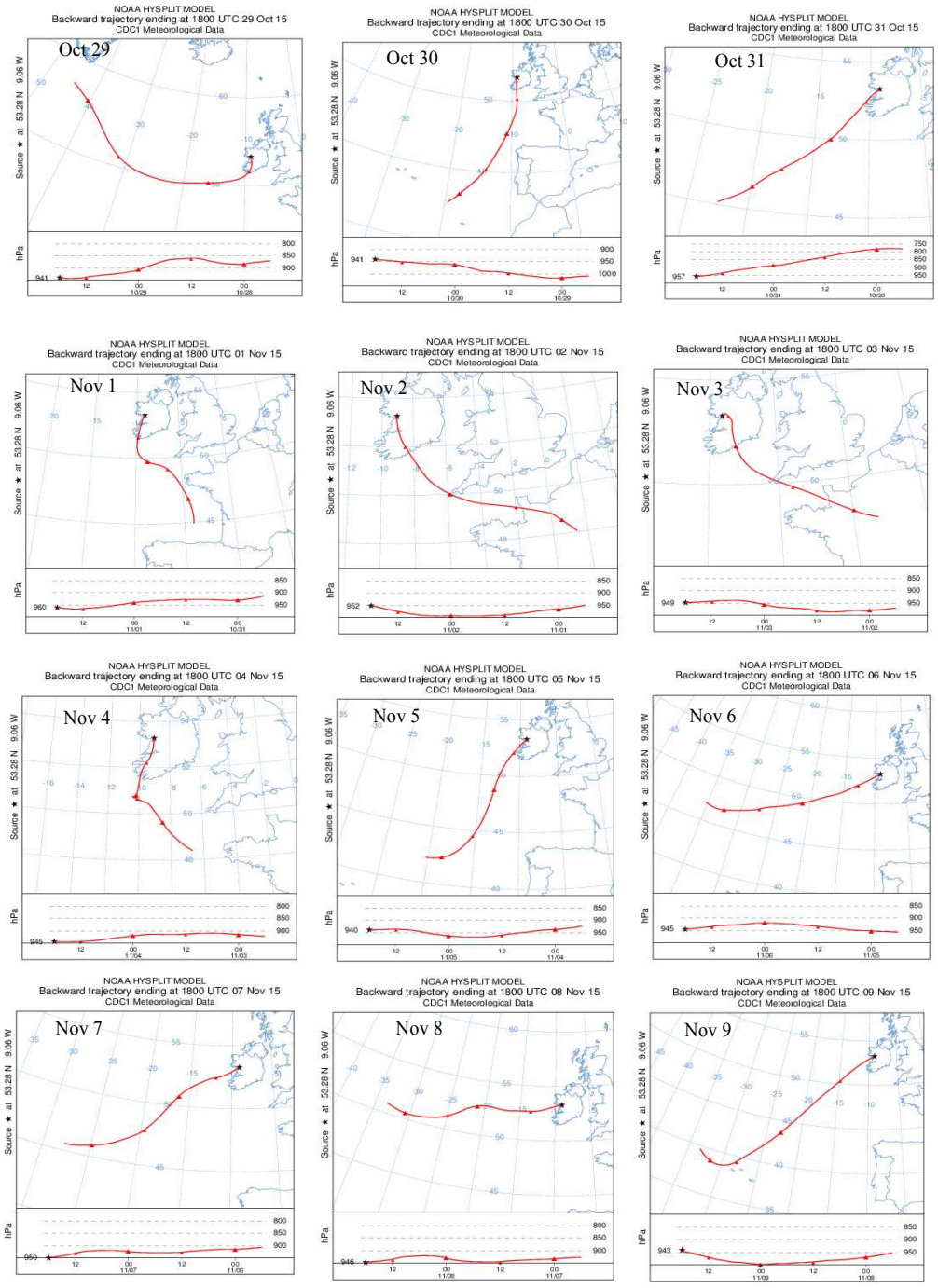


Figure S11. continued

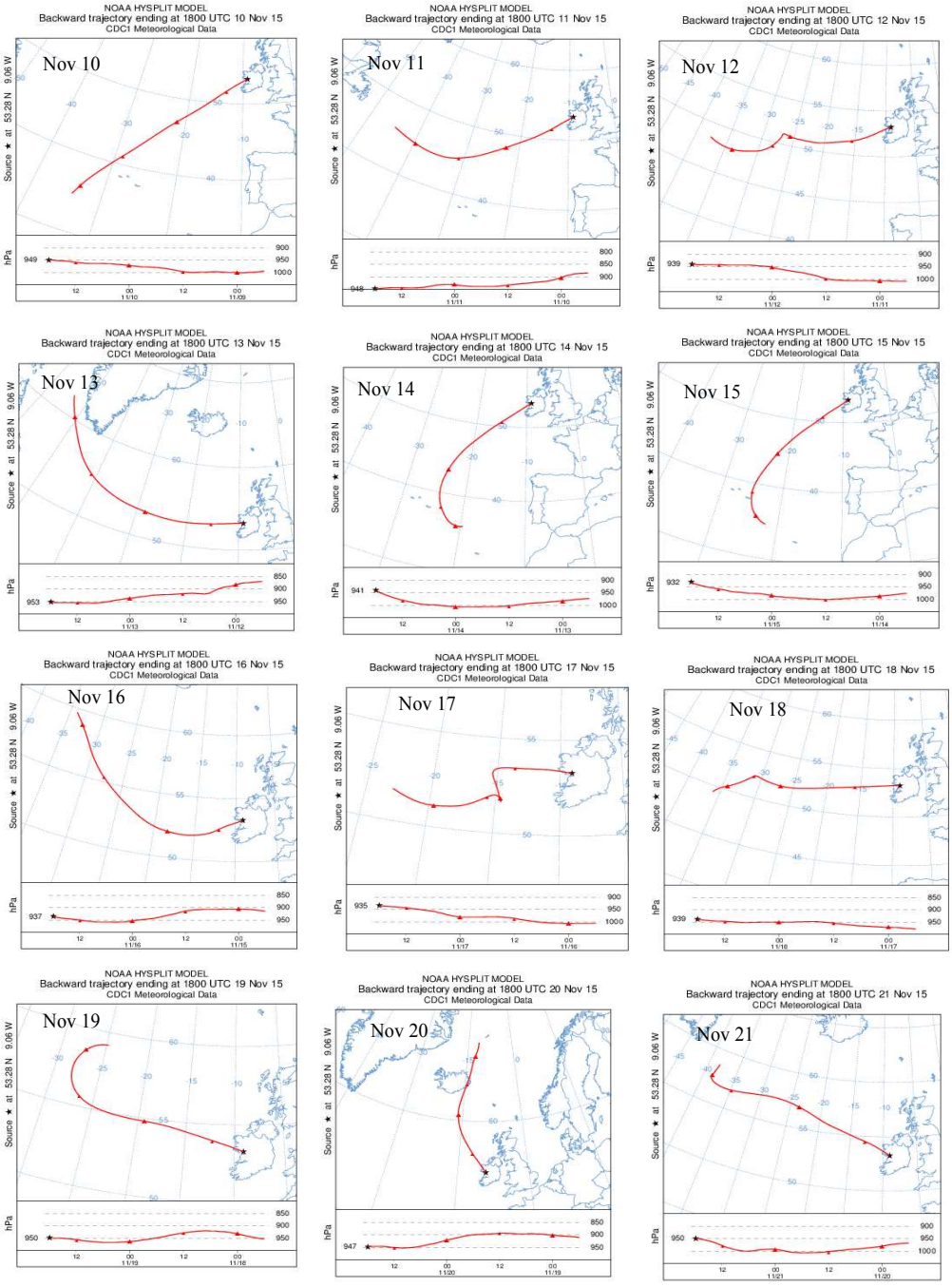


Figure S11. Continued.

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