Supplementary Material for:

Summertime surface PM$_1$ aerosol composition and size by source region at the Lampedusa island in the central Mediterranean Sea

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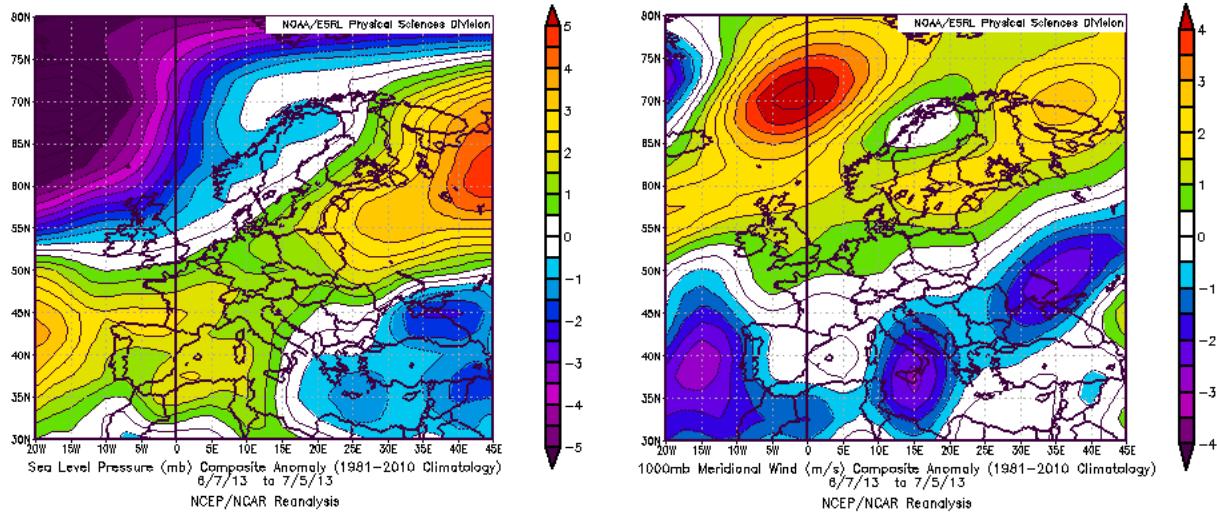
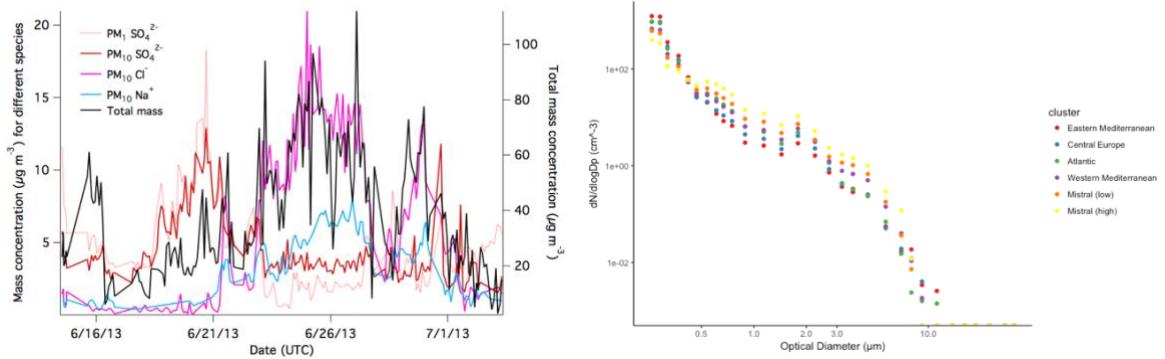
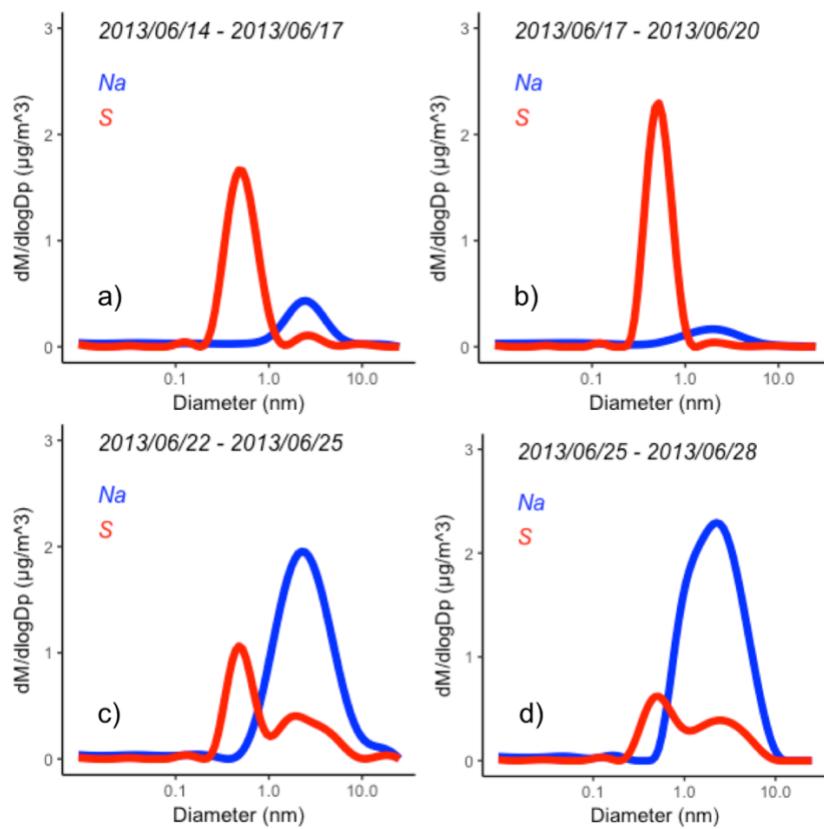
Synoptic scale meteorology

Supplementary Figure S1 Sea level pressure (left) and 1000 mb meridional wind component (right) composite anomalies of the ADRIMED campaign period with respect to the 1981-2010 climatology obtained from the NCEP/NCAR Reanalysis. Images were provided by the NOAA/ESRL Physical Sciences Division, Boulder Colorado (http://www.esrl.noaa.gov/psd/).

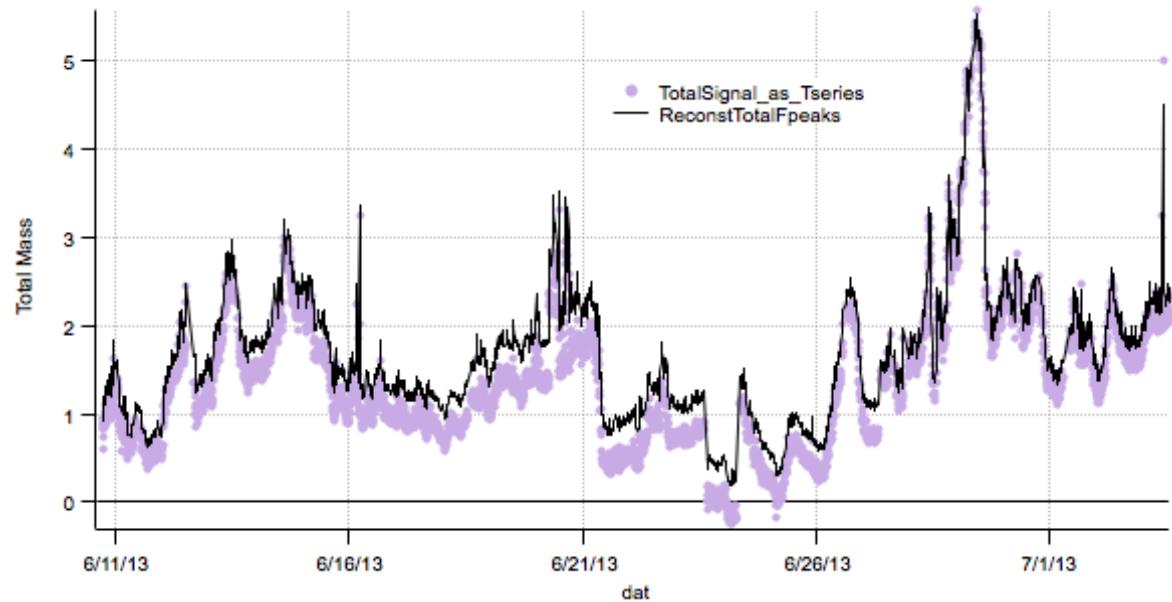
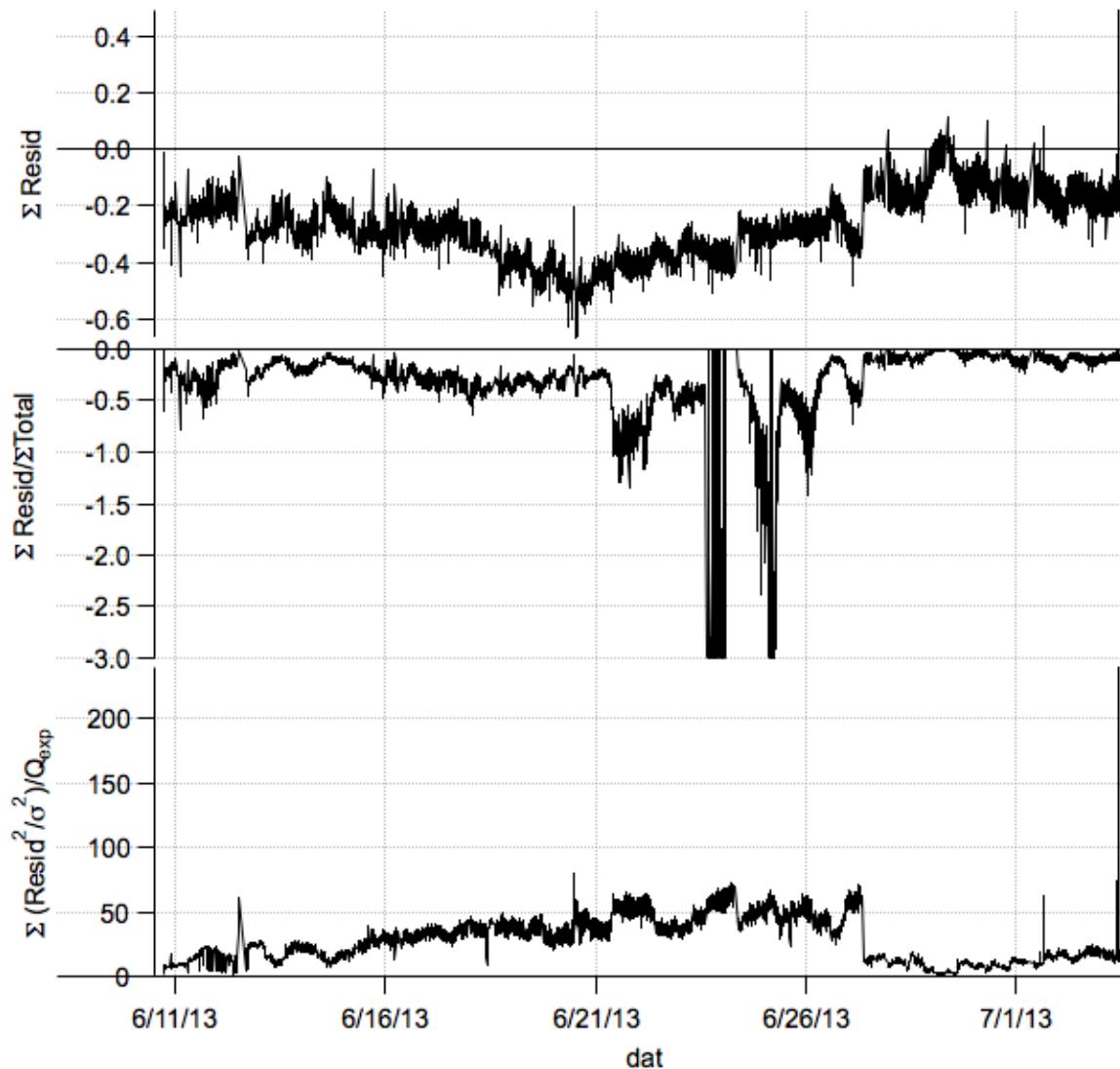
Origin of Sulphates

The total averaged aerosol mass concentration (PM_{total}) measured by the FDMS-TEOM during the campaign was 37 µg m^{-3} (maximum value up to 126 µg m^{-3}). The PM_{total} was relatively well correlated with TSP concentrations of ions associated with sea salt (Na^+, Mg^+, Cl^-) measured by the PILS (R^2 ~ 0.5, not shown). The total TSP sea salt concentration, was less than half of the PM_{total}, suggesting that a significant fraction of coarse particles was not sea salt or soluble species. There was a positive correlation (R^2=0.80) throughout the measurement period between the Na^+ and Cl^- mass concentrations and wind speed (measured at the Lampedusa site) which is indicative of sea spray aerosol. Other trace elements associated with dust were low in concentration over the entire measurement period. As shown in Section 3.2, the highest observed sea salt concentrations throughout the campaign were observed during the "Mistral" air mass origin periods. These periods were accompanied by the highest wind speeds, likely a result of the rapid descent of the air masses after passing over southern France. Despite a relatively small accumulation mode, these Mistral air masses had the highest number of coarse mode particles which is consistent with the high sea salt concentrations.

For most of the campaign, there was good correlation between the PM_1 and TSP SO_4^{2-} concentrations, with the exception of periods of high sea salt concentrations. The 3-day

samples obtained by the multistage cascade impactor show that majority of the elemental sulphur mass was within the PM_1 size range.

However, during periods of high concentrations of sea salt, up to 48% of the elemental sulphur mass was associated with particles larger than 1 µm. Some of this can be explained by the presence of seasalt-S, although up to 40% of the total nss-S was still associated with coarse mode particles. The two samples in the period 14-17 June and from the 17-20 June, characterized mostly by central European and eastern Mediterranean air masses, show relatively low sea salt ion concentrations. For those, the non-sea-salt fraction of elemental sulphur (nss-S) was nearly all under 1 µm. During the periods of higher sea salt concentrations, there was a more pronounced partitioning of the nss-S between sub- and super-micron particles.

Supplementary Figure S2 (left) the time series of TPS Cl⁻ and Na⁺, PM_{10} and PM_1 SO_4^{2-} and the total particle mass from the TEOM and (right) the size distribution above 0.26 µm, measured by the GRIMM OPC, with colours representing averages for different air mass origins.

Supplementary Figure S3 Interpolated mass size distribution of Na⁺, Cl⁻ and nss-S for four different 3-day sampling periods, collected on a 13-stage cascade impactor. Note that the vertical scales are different in each graph.

PMF mass reconstruction and residuals

Supplementary Figure S4 Total organic signal and reconstructed from 4-factor PMF solution

Supplementary Figure S5 Time series of residuals from 4-factor PMF solution

Supplementary compositional material

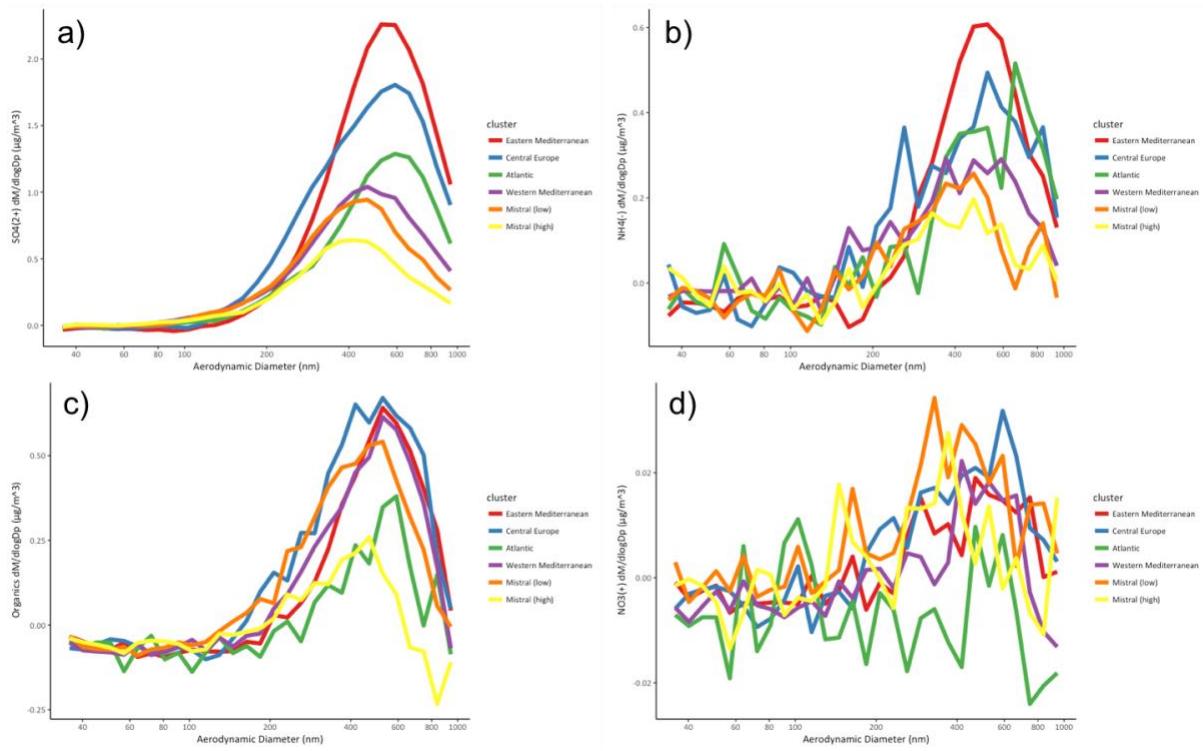
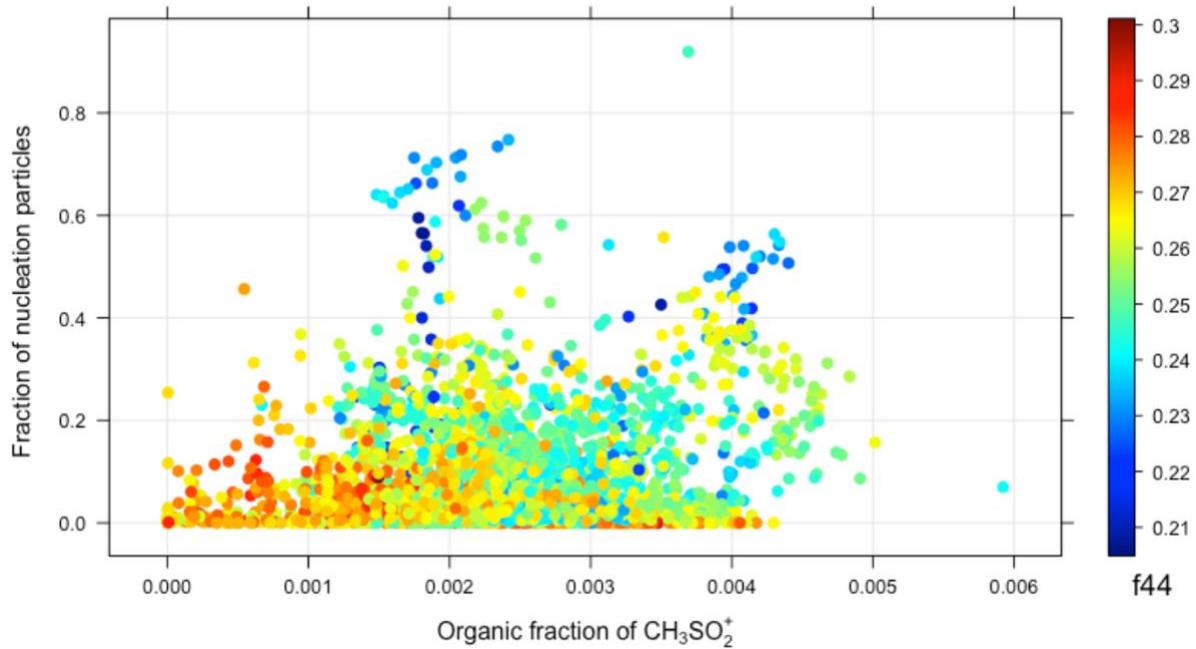
Supplementary Table S1 A summary of studies that have investigated NR-PM$_1$ composition (including PMF of OA) at coastal sites surrounding the Mediterranean basin. Only studies that have investigated PMF-based OA source apportionment are reported. MO-OOA: More oxidised oxygenated organic aerosol, LO-OOA: less oxidised oxygenated organic aerosol, HOA: Hydrocarbon-like Organic Aerosol, SV-OOA: Semi-volatile oxygenated Organic Aerosol, LV-OOA: Low-volatility oxygenated Organic Aerosol, BBOA: Biomass burning Organic Aerosol, COA: Cooking Organic Aerosol, OOA: Oxygenated Organic Aerosol, F4: "Factor -4" (unidentified PMF factor), IndOA: Industry-related Organic Aerosol, OB-OA: "Olive-branch Organic Aerosol. PMF factors in bold indicate secondary organic aerosol. After the results of this study, observations are ordered according to longitude (west to east).

AUTHORS (YEAR)	LOCATION	PERIOD	INSTRUMENT	PM$_1$ MASS AND COMPOSITION	PMF FACTORS

THIS STUDY	Lampedusa (35°31'5"N, 12°37'51"E, 45 m a.s.l.)	10 June - 5 July 2013 (summer)	cToF-AMS	10.1 µg m^{-3} (OA: 30%, SO$_4^{2-}$: 44%, NH$_4^+$: 18%, NO$_3^-$: 1%, seasalt: 1%, eBC: 5%)	HOA (8%) **MSA-OOA (12%)** **OOA-2 (28%)** **LV-OOA (53%)**
(MOHR ET AL., 2012)	Barcelona (41°23'24"N, 02°6'58.1"E, 80 m a.s.l.)	March 2009 (Spring)	HR-ToF-AMS	18.5 µg m^{-3} (OA: 43%, SO$_4^{2-}$: 16%, NH$_4^+$: 9%, NO$_3^-$: 18%, BC: 12%, Chl: 1%)	HOA (16%) BBOA (11%) COA (17%) **SV-OOA (27%)** **LV-OOA (28%)**
(MINGUILLÓN ET AL., 2016)	Barcelona (41°23'14"N, 02°06'56"E, 78 m a.s.l.)	16 August - 6 September 2013	ACSM	10 - 14 µg m^{-3} (OA: 59 - 57%, SO$_4^{2-}$: 20%, NH$_4^+$: 9 - 10%, NO$_3^-$: 4 - 5%, EC: 8%)	COA (17%) HOA (8 - 10%) **SV-OOA (38 - 40%)** **LV-OOA (36%)**
EL HADDAD ET AL., 2013	Marseille (43°18'20''N, 5°23'40"E, 64 m a.s.l.)	30 June - 14 July 2008 (Summer)	cToF-AMS	PM$_1$ concentrations not explicitly stated. SO$_4^{2-}$ concentrations where generally similar or higher than OA.	HOA (19%) F4 (6%) **SV-OOA (34%)** **LV-OOA (41%)**
(BOZZETTI ET AL., 2017)	Marseille (43°18'20''N, 5°23'40"E, 64 m a.s.l.)	August 2011 - July 2012	HR-ToF-AMS (nebulized PM2.5 filters)	10 - > 50 µg m^{-3} (OA: 37% - 51%, SO$_4^{2-}$: 12%, NH$_4^+$: 8%, NO$_3^-$: 9%, EC: 9%, Other: 9%)	Summer: HOA (17%) BBOA (5%) COA (4%) **OOA (55%)** IndOA (11%) Winter: BBOA (48%) COA (4%) **OOA (27%)** IndOA (17%)

Supplementary Table S2 The average speciated PM$_1$ concentrations during periods of different air mass origin. Units are µg m^{-3}.

Sector	Organics	SO$_4^{2-}$	NH$_4^+$	NO$_3^-$	Seasalt	eBC
Atlantic ocean	2.8±0.9	4.0±1.5	1.7±0.6	0.91±0.03	0.25±0.09	0.46±0.08

	(MO-OOA: 55%, LO-OOA: 24%, MSA-OOA: 11%, HOA: 9%)					
Western Europe	3.0±1.3 (MO-OOA: 44%, LO-OOA: 38%, MSA-OOA: 10%, HOA: 8%)	3.5±1.6	1.4±0.6	0.10±0.05	0.35±0.18	0.51±0.09
Mistral (high)	1.7±0.4 (MO-OOA: 37%, LO-OOA: 36%, MSA-OOA: 23%, HOA: 4%)	2.0±0.5	0.9±0.2	0.06±0.02	0.29±0.04	0.36±0.14
Mistral (Low)	3.2±1.1 (MO-OOA: 41%, LO-OOA: 40%, MSA-OOA: 12%, HOA: 7%)	2.9±1.4	1.3±0.5	0.11±0.06	0.41±0.17	0.50±0.10
Central Europe	3.8±0.8 (MO-OOA: 58%, LO-OOA: 23%, MSA-OOA: 11%, HOA: 8%)	6.4±1.8	2.4±0.5	0.13±0.05	0.26±0.07	0.53±0.17
Eastern Med.	3.1±0.6 (MO-OOA: 71%, LO-OOA: 14%, MSA-OOA: 6%, HOA: 8%)	6.7±3.1	2.8±1.3	0.09±0.03	0.19±0.06	0.55±0.12
Campaign average	3.0±1.1 (MO-OOA: 53%, LO-OOA: 28%, MSA-OOA: 12%, HOA: 8%)	4.5±2.7	1.9±1.0	0.10±0.05	0.30±0.15	0.50±0.21

Supplementary Figure S6 The average size distribution of sulphates, ammonium, organics and nitrates, measured by the cToF-AMS with colours representing the different air mass origins.

Supplementary Figure S7 The fraction of nucleation particles (particle number concentration between 14 and 25 nm divided by the particle number concentration between 14 and 600 nm), as a function of the organic fraction of PM_1 $CH_3SO_2^+$