Supporting Information

R ²	ATOFMS total	ATOFMS EC-rich	ATOFMS K-rich	PM ₁₀	PM ₁	BC	ACSM total	ACSM SO4 ²⁻	ACSM NH4+	ACSM SV- OOA	ACSM LV- OOA	PILS- IC SO4 ²⁻	PILS- IC NH4 ⁺	OPS 0.3- 0.579 μm particles	*PILS Oxalate	*WSOC	**ATOFMS fresh sea salt	** PILS SSA	**OPS 0.579-2.156 μm particles	*** PILS MSA	*** ACSM NO3 ⁻	*** PILS NO3 ⁻
ATOFMS total	1																					
ATOFMS EC-rich	0.73	1																				
ATOFMS K-rich	0.48	0.28	1																			
PM ₁₀	0.21	0.20	0.09	1																		
PM ₁	0.44	0.46	0.30	0.17	1																	
BC	0.55	0.50	0.37	0.19	0.40	1																
ACSM total	0.72	0.66	0.46	0.17	0.54	0.70	1															
ACSM SO42-	0.66	0.61	0.38	0.14	0.46	0.48	0.81	1														i
ACSM NH4 ⁺	0.65	0.62	0.37	0.12	0.44	0.47	0.81	0.93	1													
ACSM SV-OOA	0.46	0.43	0.35	0.11	0.38	0.57	0.69	0.33	0.32	1												1
ACSM LV-OOA	0.59	0.59	0.46	0.17	0.50	0.67	0.88	0.61	0.61	0.63	1											i
PILS-IC SO42-	0.58	0.52	0.25	0.12	0.41	0.41	0.61	0.81	0.75	0.21	0.42	1										[
PILS-IC NH4 ⁺	0.44	0.43	0.23	0.08	0.30	0.30	0.46	0.53	0.53	0.18	0.33	0.68	1									
OPS 0.3-0.579 μm particles	0.73	0.69	0.49	0.24	0.54	0.57	0.82	0.71	0.70	0.54	0.75	0.60	0.46	1								
*PILS Oxalate	0.35	0.41	0.25	0.14	0.36	0.37	0.50	0.28	0.29	0.41	0.54	0.27	0.23	0.42	1							
*WSOC	0.44	0.37	0.29	0.24	0.42	0.46	0.50	0.19	0.18	0.60	0.58	0.19	0.09	0.44	0.55	1						
**ATOFMS fresh sea salt	0.04	0.07	0.05	0.04	0.02	0.04	0.12	0.07	0.05	0.15	0.16	0.06	0.18	0.02	0.01	N/A	1					
**PILS SSA	0.11	0.01	0.02	0.35	0.00	0.00	0.09	0.05	0.06	0.13	0.17	0.02	0.25	0.01	0.04	N/A	0.81	1				i
**OPS 0.579-2.156 μm particles	0.42	0.03	0.02	0.24	0.03	0.01	0.01	0.00	0.00	0.03	0.07	0.01	0.08	0.03	0.00	N/A	0.81	0.88	1			
***PILS MSA	0.18	0.19	0.06	0.05	0.16	0.06	0.21	0.35	0.37	0.001	0.17	0.48	0.54	0.20	0.08	0.03	0.06	0.00	0.02	1		
***ACSM NO3-	0.58	0.53	0.34	0.25	0.54	0.41	0.75	0.71	0.75	0.35	0.66	0.75	0.55	0.65	0.35	0.28	0.05	0.02	0.04	0.46	1	
***PILS NO ₃ -	0.07	0.06	0.02	0.10	0.04	0.00	0.04	0.13	0.14	0.02	0.03	0.34	0.33	0.13	0.15	0.00	0.01	0.24	0.36	0.53	0.20	1

Table 1. Pearson's correlation coefficient (R²) between mass concentrations from ATOFMS (PM_{2.5}), TEOM (PM₁₀ & PM₁), ACSM (PM₁), PILS (PM₁₀) and MAAP (PM_{2.5}) measurements and OPS number concentrations (#/cm³) for the full sampling period, and during specific periods.

*Correlations calculated from 21st June-4th August.

******Correlations calculated from 20-27th June.

***Correlations calculated from 23rd July-5th August



Figure 1. Individual HYSPLIT 120-hour back trajectories for each cluster for Period 1 during ADRIMED and SAF-MED.



Figure 2. Daytime average aerosol optical depth at 550 nm (AOD₅₅₀) derived from MSG/SEVIRI (Thieuleux et al., 2005) from 17th to 20th June 2013 showing a dust plume transport over the western Mediterranean basin (produced by <u>http://www.icare.univ-lille1.fr</u>). Land and cloudy pixels in dark and light grey, respectively.