

1 **Supporting Information**
2

3 Manuscript Title: Investigating size-segregated sources of elemental composition of
4 particulate matter in the South China Sea during the 2011 Vasco Cruise
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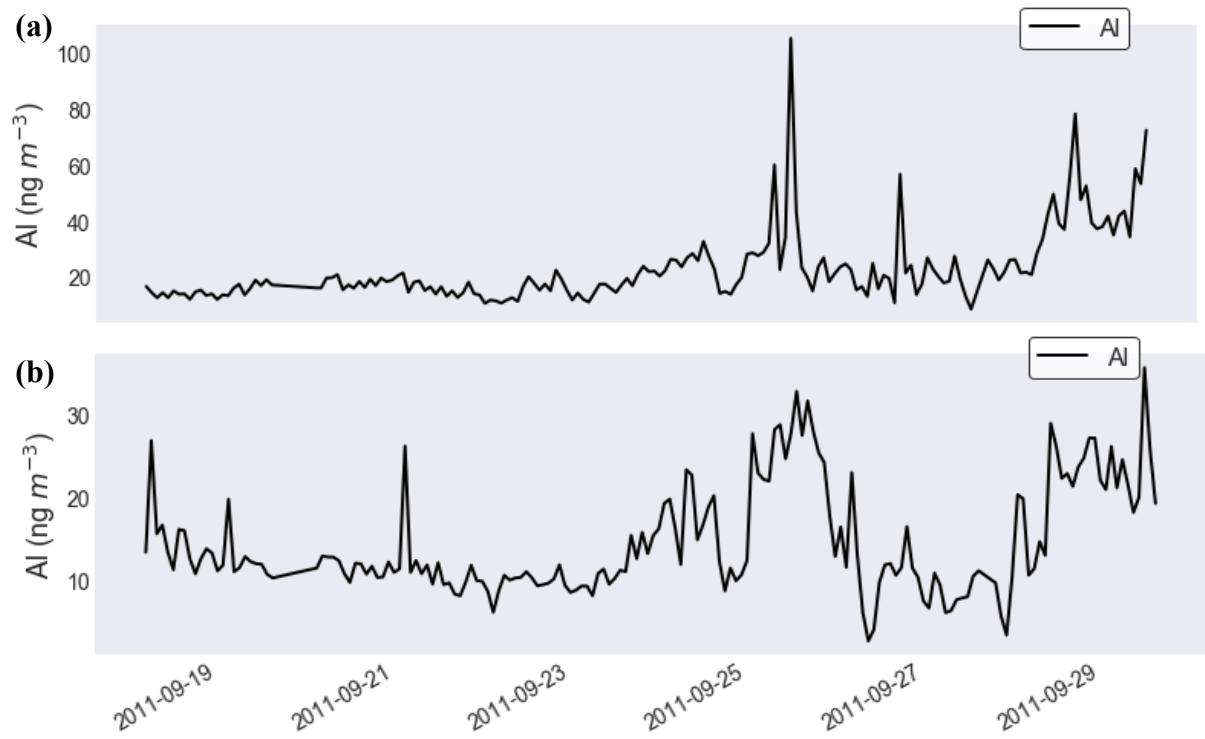
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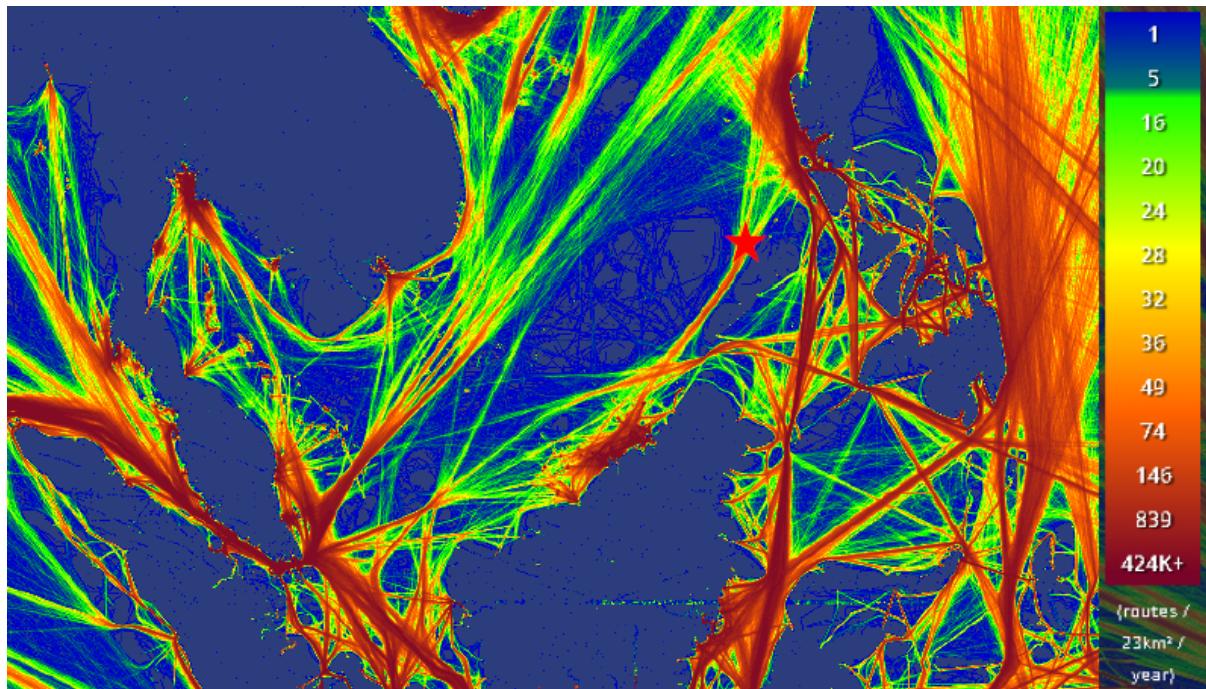
17 Supporting Information Summary: 13 Pages including Cover Page, 4 Tables, 7 Figures.

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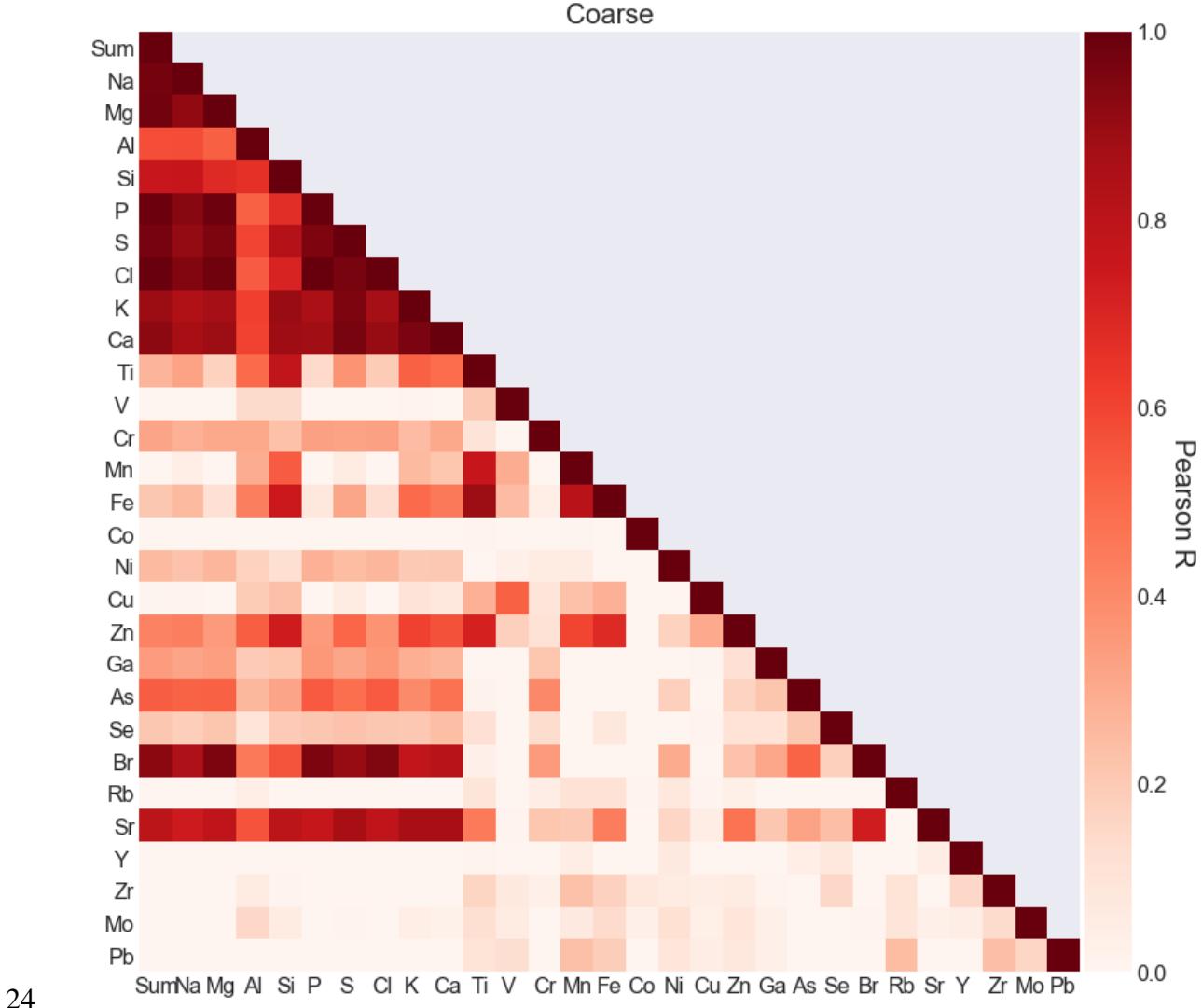
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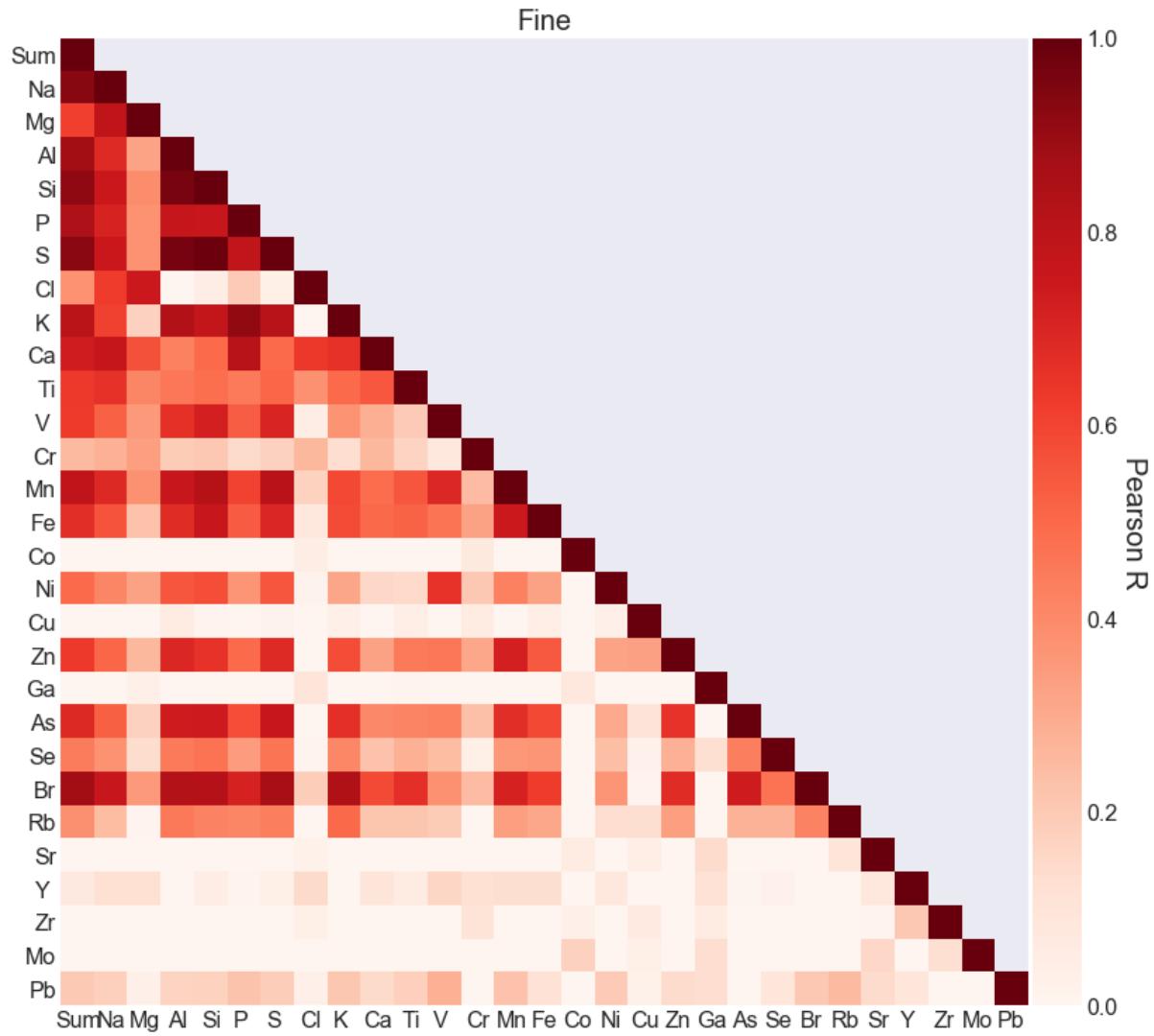
20 **Figure S1.** Time series of Al in the (a) fine and (b) ultrafine modes.



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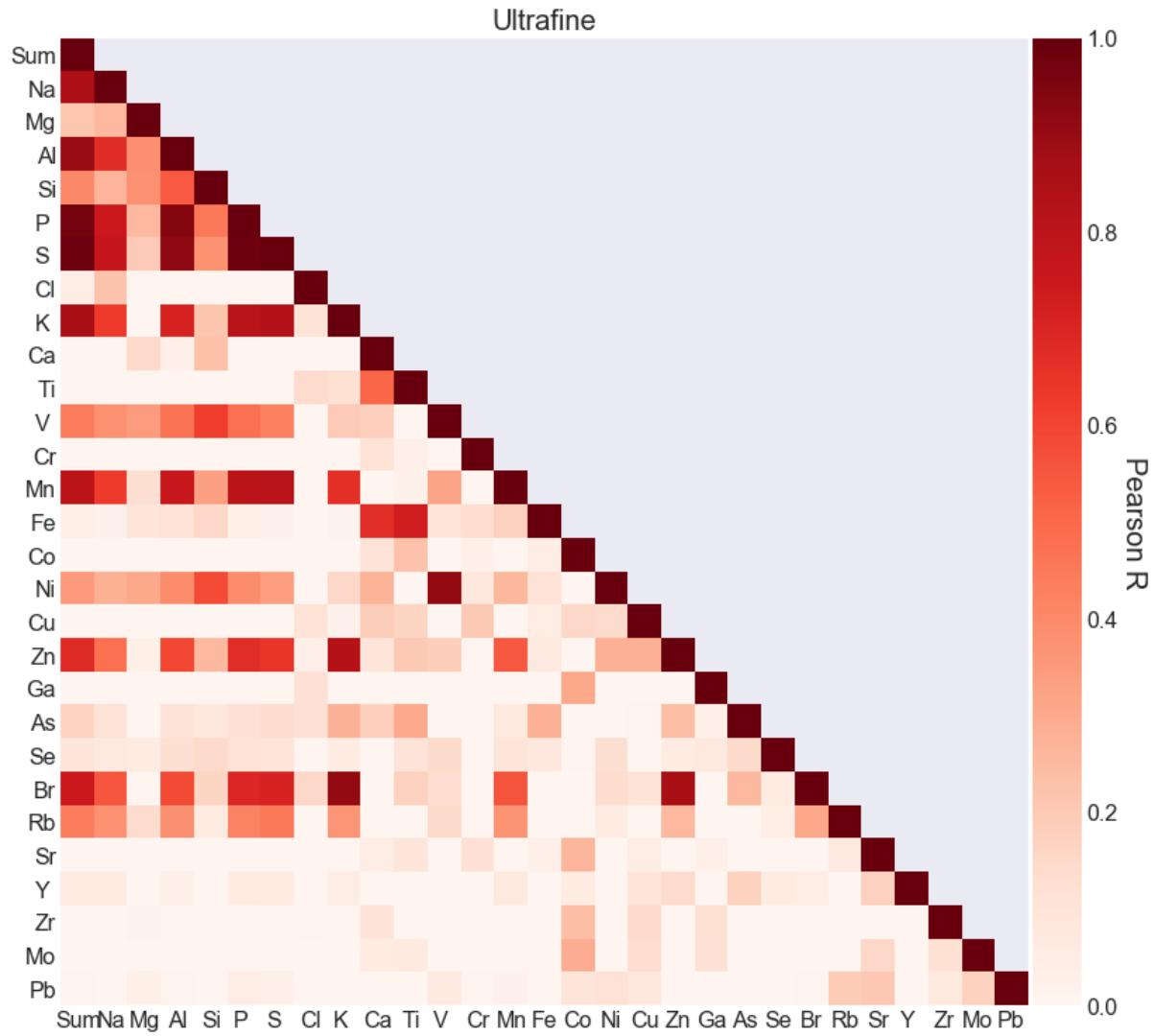
22 **Figure S2. Ship tracks near the Palawan sampling site for 2015-2016. Red star indicates**
23 **the location of the M/Y Vasco. Source: MarineTraffic.**





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27 **Figure S4. Correlation heatmap for the fine mode (0.34 – 1.15 μm).**



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29 **Figure S5. Correlation heatmap for the ultrafine mode (0.07-0.34 μm).**

Table S1. Pearson correlation matrix for the coarse mode (1.15 - 10 µm). Correlation coefficients above 0.7 are marked in bold.

	Sum	Na	Mg	Al	Si	P	S	Cl	K	Ca	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	As	Se	Br	Rb	Sr	Y	Zr	Mo	Pb
Sum	1.00																												
Na	0.97	1.00																											
Mg	0.97	0.91	1.00																										
Al	0.57	0.58	0.53	1.00																									
Si	0.76	0.76	0.68	0.66	1.00																								
P	0.99	0.93	0.98	0.52	0.67	1.00																							
S	0.97	0.91	0.95	0.60	0.82	0.95	1.00																						
Cl	0.99	0.94	0.98	0.54	0.70	1.00	0.96	1.00																					
K	0.89	0.84	0.87	0.61	0.90	0.85	0.95	0.87	1.00																				
Ca	0.92	0.87	0.89	0.60	0.88	0.88	0.96	0.90	0.96	1.00																			
Ti	0.27	0.32	0.17	0.50	0.78	0.15	0.37	0.19	0.52	0.49	1.00																		
V	-0.11	-0.10	-0.13	0.14	0.14	-0.15	-0.05	-0.13	0.01	0.00	0.20	1.00																	
Cr	0.32	0.28	0.31	0.31	0.23	0.33	0.32	0.33	0.25	0.31	0.10	-0.06	1.00																
Mn	-0.03	0.04	-0.13	0.29	0.54	-0.15	0.06	-0.11	0.25	0.21	0.76	0.29	-0.08	1.00															
Fe	0.21	0.25	0.11	0.43	0.75	0.08	0.31	0.13	0.50	0.45	0.89	0.25	0.04	0.81	1.00														
Co	-0.07	-0.04	-0.09	-0.06	-0.06	-0.09	-0.09	-0.08	-0.10	-0.09	0.01	-0.04	-0.08	0.00	-0.02	1.00													
Ni	0.25	0.22	0.26	0.17	0.12	0.28	0.25	0.26	0.20	0.21	-0.02	0.03	0.06	0.06	-0.05	-0.03	1.00												
Cu	0.01	0.01	-0.02	0.19	0.23	-0.02	0.05	-0.01	0.10	0.08	0.28	0.52	0.09	0.23	0.28	-0.02	-0.02	1.00											
Zn	0.42	0.44	0.35	0.53	0.74	0.35	0.51	0.37	0.61	0.56	0.72	0.18	0.11	0.60	0.68	-0.03	0.17	0.30	1.00										
Ga	0.35	0.32	0.34	0.20	0.21	0.36	0.31	0.36	0.29	0.26	0.00	-0.04	0.21	-0.06	0.00	-0.05	-0.06	0.00	0.12	1.00									
As	0.53	0.52	0.52	0.26	0.32	0.54	0.48	0.54	0.40	0.47	0.02	-0.13	0.40	-0.09	-0.02	-0.05	0.18	-0.02	0.16	0.22	1.00								
Se	0.21	0.19	0.22	0.09	0.20	0.21	0.22	0.21	0.21	0.24	0.12	-0.07	0.14	-0.01	0.08	-0.07	-0.10	0.01	0.11	0.11	0.21	1.00							
Br	0.92	0.84	0.95	0.45	0.56	0.95	0.90	0.95	0.78	0.82	0.03	-0.18	0.35	-0.26	-0.02	-0.09	0.30	-0.06	0.23	0.31	0.51	0.18	1.00						
Rb	-0.17	-0.15	-0.21	0.04	-0.03	-0.18	-0.15	-0.18	-0.11	-0.13	0.10	-0.02	0.05	0.11	0.11	0.01	0.08	0.01	0.04	-0.09	-0.11	-0.02	-0.19	1.00					
Sr	0.80	0.74	0.79	0.56	0.80	0.77	0.87	0.79	0.86	0.86	0.45	0.01	0.21	0.20	0.44	-0.09	0.16	0.05	0.47	0.21	0.32	0.24	0.73	-0.11	1.00				
Y	-0.09	-0.09	-0.08	-0.07	-0.05	-0.08	-0.05	-0.09	-0.06	-0.04	0.01	-0.16	-0.02	0.04	-0.01	-0.09	0.07	-0.10	-0.05	-0.08	0.04	0.08	-0.07	-0.14	0.05	1.00			
Zr	-0.15	-0.10	-0.20	0.05	0.01	-0.18	-0.17	-0.17	-0.11	-0.13	0.16	0.07	0.03	0.23	0.17	0.08	0.05	0.05	0.05	0.01	-0.10	0.16	-0.18	0.11	-0.13	0.15	1.00		
Mo	-0.02	-0.03	-0.01	0.15	0.06	-0.03	0.01	-0.03	0.04	0.03	0.12	0.06	-0.04	0.07	0.14	0.03	0.12	0.04	0.09	0.03	-0.11	-0.03	0.01	0.10	0.03	0.04	0.14	1.00	
Pb	-0.33	-0.32	-0.35	-0.01	-0.13	-0.34	-0.30	-0.34	-0.23	-0.28	0.10	0.13	-0.08	0.23	0.19	-0.09	0.09	0.05	0.08	0.03	-0.48	-0.26	-0.34	0.24	-0.13	-0.02	0.24	0.16	1.00

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Table S2. Pearson correlation matrix for the fine mode (0.34 – 1.15 µm). Correlation coefficients above 0.7 are marked in bold.

	Sum	Na	Mg	Al	Si	P	S	Cl	K	Ca	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	As	Se	Br	Rb	Sr	Y	Zr	Mo	Pb
Sum	1.00																												
Na	0.93	1.00																											
Mg	0.61	0.79	1.00																										
Al	0.88	0.68	0.32	1.00																									
Si	0.92	0.75	0.39	0.96	1.00																								
P	0.84	0.71	0.37	0.77	0.76	1.00																							
S	0.93	0.75	0.37	0.97	0.98	0.78	1.00																						
Cl	0.37	0.62	0.75	-0.08	0.05	0.20	0.04	1.00																					
K	0.80	0.61	0.17	0.83	0.78	0.91	0.81	-0.02	1.00																				
Ca	0.73	0.77	0.57	0.43	0.49	0.81	0.49	0.63	0.66	1.00																			
Ti	0.63	0.66	0.41	0.46	0.48	0.45	0.51	0.38	0.50	0.55	1.00																		
V	0.63	0.52	0.35	0.66	0.72	0.53	0.70	0.05	0.37	0.29	0.20	1.00																	
Cr	0.25	0.28	0.33	0.19	0.21	0.14	0.18	0.26	0.13	0.26	0.17	0.07	1.00																
Mn	0.79	0.69	0.38	0.76	0.82	0.60	0.81	0.17	0.59	0.49	0.55	0.69	0.25	1.00															
Fe	0.67	0.56	0.22	0.68	0.76	0.54	0.69	0.08	0.58	0.50	0.52	0.47	0.32	0.75	1.00														
Co	-0.43	-0.34	-0.12	-0.44	-0.47	-0.42	-0.47	0.04	-0.40	-0.29	-0.20	-0.42	0.07	-0.43	-0.45	1.00													
Ni	0.49	0.41	0.33	0.55	0.57	0.37	0.55	0.02	0.32	0.15	0.15	0.65	0.20	0.43	0.33	-0.20	1.00												
Cu	-0.04	-0.09	-0.13	0.06	0.01	-0.02	0.02	-0.19	0.03	-0.10	0.04	-0.01	0.06	-0.07	0.04	0.00	0.03	1.00											
Zn	0.63	0.50	0.26	0.69	0.66	0.49	0.68	0.00	0.58	0.33	0.45	0.46	0.31	0.72	0.55	-0.36	0.32	0.33	1.00										
Ga	-0.11	-0.05	0.03	-0.15	-0.17	-0.13	-0.15	0.09	-0.15	-0.10	0.02	-0.09	-0.07	-0.12	-0.18	0.08	-0.01	-0.06	-0.04	1.00									
As	0.69	0.53	0.17	0.73	0.74	0.57	0.76	-0.03	0.67	0.40	0.42	0.43	0.23	0.67	0.59	-0.33	0.30	0.10	0.66	-0.23	1.00								
Se	0.44	0.37	0.13	0.45	0.47	0.35	0.47	0.01	0.41	0.22	0.28	0.24	0.04	0.36	0.36	-0.17	0.23	0.02	0.28	0.12	0.43	1.00							
Br	0.88	0.76	0.35	0.83	0.82	0.71	0.86	0.19	0.84	0.58	0.67	0.38	0.25	0.71	0.62	-0.32	0.36	0.01	0.68	-0.10	0.74	0.47	1.00						
Rb	0.38	0.24	0.01	0.45	0.42	0.41	0.44	-0.13	0.50	0.22	0.22	0.19	-0.01	0.33	0.31	-0.15	0.13	0.13	0.34	-0.13	0.28	0.28	0.42	1.00					
Sr	-0.18	-0.14	-0.08	-0.17	-0.19	-0.21	-0.19	0.03	-0.21	-0.14	-0.06	-0.05	-0.10	-0.10	-0.11	0.06	-0.03	0.04	-0.04	0.14	-0.21	-0.01	-0.16	0.09	1.00				
Y	0.07	0.12	0.12	-0.01	0.04	0.01	0.04	0.14	-0.07	0.09	0.06	0.16	0.12	0.13	0.13	-0.10	0.08	-0.14	-0.04	0.11	0.01	0.02	-0.02	-0.22	0.08	1.00			
Zr	-0.13	-0.12	-0.01	-0.14	-0.13	-0.08	-0.15	0.04	-0.14	-0.03	-0.04	-0.05	0.10	-0.08	-0.01	0.03	-0.02	0.06	-0.06	0.06	-0.13	-0.02	-0.18	-0.11	0.01	0.21	1.00		
Mo	-0.27	-0.27	-0.21	-0.17	-0.23	-0.20	-0.22	-0.21	-0.19	-0.27	-0.11	-0.13	-0.22	-0.19	-0.23	0.17	-0.03	0.04	-0.16	0.13	-0.21	-0.16	-0.23	-0.07	0.15	-0.02	0.12	1.00	
Pb	0.20	0.18	0.03	0.17	0.17	0.22	0.19	0.03	0.21	0.14	0.18	0.28	-0.03	0.23	0.11	-0.14	0.20	0.03	0.14	0.13	-0.07	0.09	0.20	0.25	0.14	0.09	-0.11	-0.01	1.00

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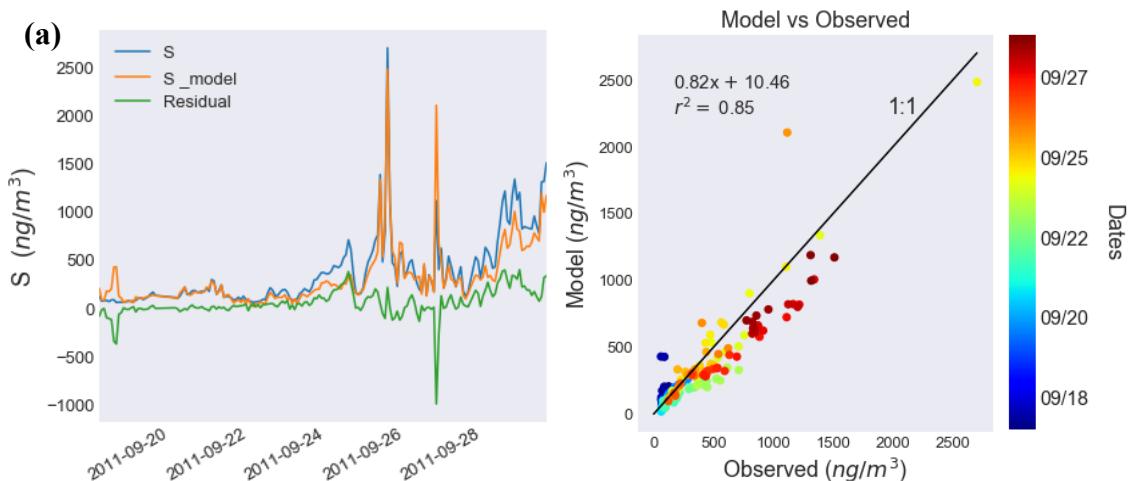
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36 **Table S3. Pearson correlation matrix for the ultrafine mode (0.07-0.34 µm). Correlation coefficients above 0.7 are marked in bold.**

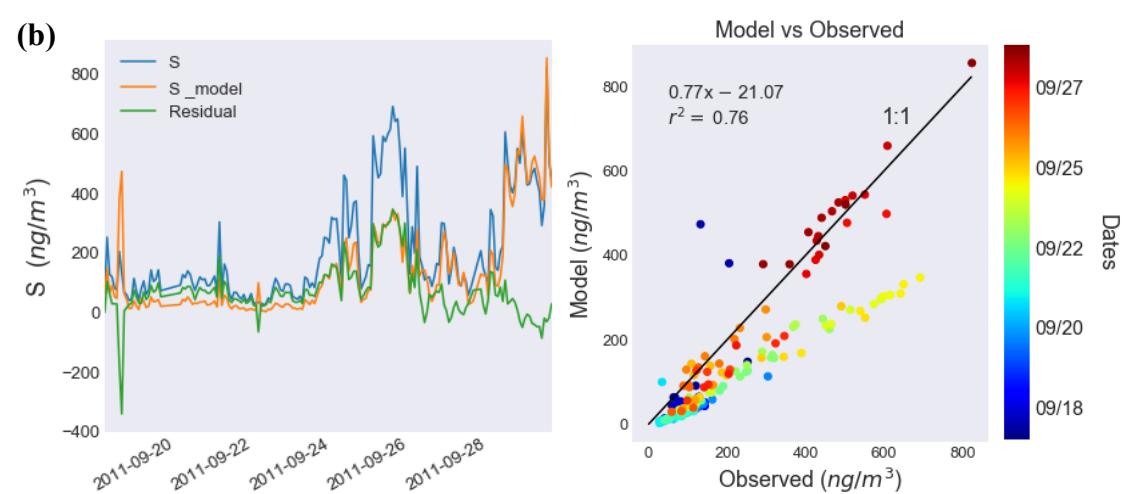
	Sum	Na	Mg	Al	Si	P	S	Cl	K	Ca	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	As	Se	Br	Rb	Sr	Y	Zr	Mo	Pb						
Sum	1																																		
Na	0.85	1																																	
Mg	0.21	0.25	1																																
Al	0.9	0.68	0.38	1																															
Si	0.4	0.27	0.38	0.54	1																														
P	0.97	0.75	0.26	0.94	0.45	1																													
S	0.98	0.77	0.2	0.91	0.37	0.98	1																												
Cl	0.05	0.22	-0.29	-0.3	-0.15	-0.1	-0.06	1																											
K	0.86	0.63	-0.01	0.71	0.22	0.81	0.83	0.1	1																										
Ca	-0.04	-0.04	0.15	0.03	0.23	-0.01	-0.07	-0.02	-0.05	1																									
Ti	-0.02	-0.04	-0.1	-0.04	0	-0.04	-0.06	0.14	0.12	0.51	1																								
V	0.44	0.38	0.34	0.47	0.62	0.48	0.43	-0.07	0.2	0.18	-0.09	1																							
Cr	-0.11	-0.11	-0.03	-0.09	-0.06	-0.12	-0.1	-0.05	-0.11	0.11	0.03	-0.06	1																						
Mn	0.8	0.63	0.12	0.76	0.33	0.8	0.81	-0.04	0.66	-0.02	0.03	0.32	-0.03	1																					
Fe	0.04	0.02	0.09	0.11	0.16	0.04	0.02	-0.02	0.02	0.67	0.73	0.09	0.13	0.17	1																				
Co	-0.62	-0.53	-0.12	-0.55	-0.21	-0.59	-0.62	-0.06	-0.51	0.1	0.22	-0.34	0.03	-0.51	0.04	1																			
Ni	0.35	0.28	0.31	0.39	0.58	0.39	0.33	-0.07	0.15	0.27	-0.04	0.91	0.08	0.26	0.11	-0.25	1																		
Cu	-0.09	-0.09	-0.19	-0.14	-0.16	-0.11	-0.11	0.11	0.02	0.19	0.16	-0.16	0.2	-0.02	0.05	0.15	0.14	1																	
Zn	0.68	0.48	0.04	0.59	0.25	0.67	0.64	0.03	0.82	0.09	0.2	0.19	-0.01	0.54	0.07	-0.32	0.28	0.28	1																
Ga	-0.23	-0.15	-0.08	-0.29	-0.2	-0.25	-0.25	0.11	-0.16	-0.19	-0.06	-0.12	-0.08	-0.24	-0.23	0.31	-0.13	-0.04	-0.17	1															
As	0.17	0.11	-0.04	0.1	0.08	0.12	0.13	0.12	0.28	0.18	0.3	-0.01	-0.03	0.07	0.28	-0.05	-0.02	0.01	0.23	0.03	1														
Se	0.09	0.07	0.06	0.12	0.14	0.11	0.09	-0.06	0.05	-0.01	0.1	0.14	-0.04	0.1	0.08	0.01	0.13	-0.11	0.05	0.07	0.14	1													
Br	0.75	0.55	-0.1	0.58	0.16	0.69	0.71	0.16	0.91	-0.03	0.17	0.13	-0.07	0.56	-0.01	-0.38	0.13	0.1	0.86	-0.08	0.26	0.05	1												
Rb	0.44	0.37	0.14	0.38	0.05	0.42	0.45	-0.09	0.36	-0.16	-0.1	0.14	-0.03	0.37	-0.13	-0.24	0.06	-0.08	0.26	-0.04	-0.09	0.04	0.31	1											
Sr	-0.21	-0.14	-0.04	-0.19	-0.12	-0.2	-0.21	0	-0.22	0.05	0.09	-0.16	0.11	-0.2	0.03	0.26	-0.14	0.05	-0.16	0.03	-0.11	-0.07	-0.17	0.07	1										
Y	0.06	0.06	-0.12	0.03	0	0.05	0.06	0	0.05	-0.13	-0.07	-0.09	-0.07	0.07	-0.08	0.06	-0.07	0.09	0.14	-0.06	0.17	0.05	0.05	-0.18	0.17	1									

Zr	-0.33	-0.28	0.02	-0.29	-0.14	-0.32	-0.33	0.01	-0.26	0.1	-0.01	-0.15	0	-0.33	-0.12	0.24	-0.06	0.15	-0.16	0.12	-0.12	0	-0.22	-0.19	0	-0.02	1		
Mo	-0.27	-0.26	-0.08	-0.25	-0.07	-0.26	-0.28	-0.01	-0.2	0.06	0.07	-0.14	-0.02	-0.25	-0.01	0.29	-0.08	0.13	-0.04	0.12	-0.04	-0.13	-0.08	-0.04	0.16	-0.07	0.12	1	
Pb	0.01	-0.04	0.04	0.01	-0.01	0.05	0.03	-0.03	-0.05	-0.06	-0.14	0.06	0.01	0.02	-0.18	0.09	0.11	0.08	-0.01	0	-0.53	-0.08	0.01	0.19	0.21	-0.1	0.07	0.17	1

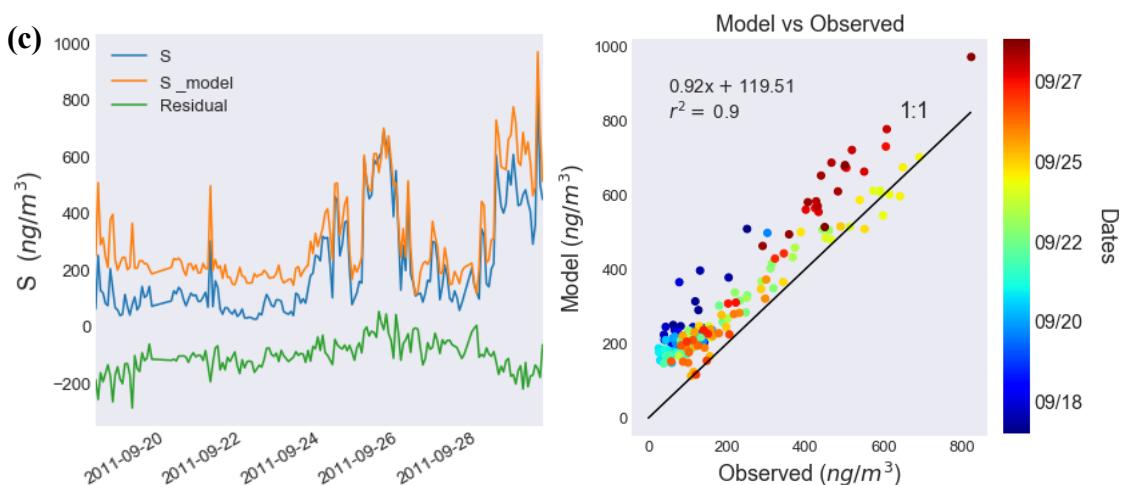
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41 **Figure S6. Multiple linear regression time series plots predicting S using tracers K and V**
 42 **in the (a) fine mode, (b) ultrafine mode, and (c) with the addition of Al in the model for**
 43 **the ultrafine mode. The residual is plotted in green, defined as the difference between the**
 44 **actual and modelled concentrations of S. Color bar indicates the time of sampling.**

45 *Multiple linear regression analysis description*

46 Having origins in both oil combustion (Balasubramanian et al., 2003; Han et al., 2006;
47 Santoso et al., 2010) and biomass burning (Artaxo et al., 1998; Han et al., 2006; Reid et al.,
48 2015), S serves as a general indicator combustion (Atwood et al., 2012). Multiple linear
49 regression was used with S as the dependent variable to determine contributions from biomass
50 burning and oil combustion in the fine and ultrafine modes. One key assumption is that
51 variations in S can be attributed to biomass burning and oil combustion, represented by their
52 tracers K and V, respectively.

53 Strong correlations, slopes near unity, and low y-intercepts in the linear regression plots
54 of actual and modelled S indicate that K and V are good predictors of S, particularly in the fine
55 mode (Fig. S6a). However, the model underestimates the concentration of ultrafine S between
56 24 to 26 September (Fig. S6b). The underestimation may be due to an additional source of
57 ultrafine S that is independent of biomass burning and oil combustion.

58 To further investigate additional elements that may explain the model's underestimation,
59 residual values between the modelled and actual concentrations of S were used to identify
60 elements that correlated strongly with the residual. In the ultrafine mode, Al was found to
61 correlate strongly with the residual. Upon adding Al to the model, the model was observed to
62 better capture variance in S, specifically between 24 and 26 September (Fig. S6c) which was
63 significantly underestimated when only K and V were included in the model (Fig. S6b). This
64 addition creates a positive discrepancy of the model over the actual concentration of S which
65 indicates source of Al distinct from S in the ultrafine mode. Although the addition of Al
66 improves the model's prediction between 24 and 26 September, we consider K and V generally
67 sufficient predictors of S.

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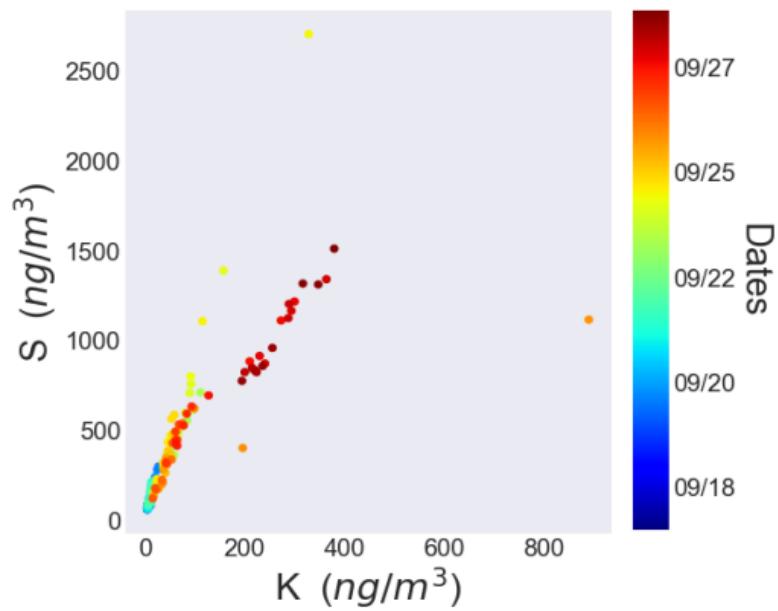
70 **Table S4. Comparison of ratio-slopes and r² correlations of various elements with stage**71 **7-8 Si.**

	<i>18-19 Sept</i>			<i>19-30 Sept</i>		
	Ratio-slope	R ²	Intercept	Ratio-slope	R ²	Intercept
P	0.06	0.76	16.36	0.83	0.96	10.45
S	1.73	0.73	11.67	22.26	0.94	-84.65
Al	0.11	0.61	9.4	0.81	0.92	3.47

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75 **Figure S7.** Linear regression of S and K in the fine mode ($0.34 - 1.15 \mu\text{m}$), colored by date.