Supplementary Material

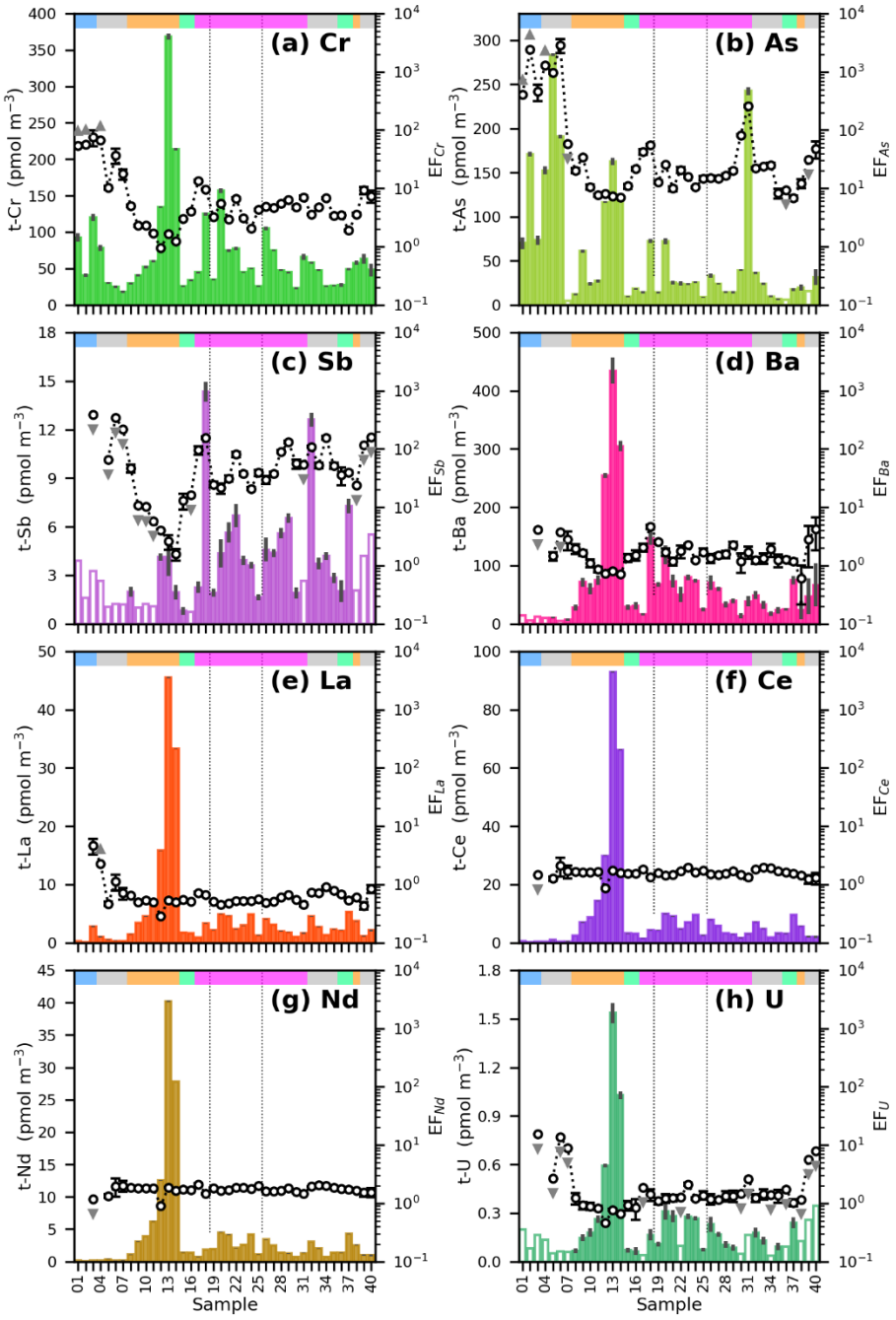

Fig. S1. Total concentrations of Cr, As, Sb, Ba, La, Ce, Nd and U in the GA04 aerosol samples, with enrichment factors relative to Al overlaid (circles). Unfilled bars indicate that analyte was below the limit of detection and bar represents 75% of the limit of detection. An EF was not determined if both Al and the element of interest were below the limit of detection. Up- / down-ward pointing grey arrows near EF markers indicate that values are minima / maxima because Al / the element were below the

limit of detection. The coloured bar at the top of each panel indicates the air mass sampled, blue = RNA, grey = WEU, orange = NAF, green = MED, pink = EEU. The dashed grey vertical lines indicate the legs of the cruise, with Legs 1-3 being left to right.

Table S1. Mean and standard deviation concentrations of elements determined in Arizona Test Dust (ATD) aliquots of ~ 11 – 14 mg, using the HNO_3-HF digestion method applied in this study (n = 8).

Element	Concentration (ppm)	Element	Concentration (ppm)
Al	113000 ± 27000	P	1520 ± 210
Ti	3730 ± 570	V	83 ± 17
Mn	634 ± 116	Ni	25.8 ± 1.8
Fe	26900 ± 4400	Cu	38.0 ± 2.7
Co	15.7 ± 3.5	Zn	104 ± 16
Th	4.6 ± 1.2	Cd	< 0.5
Pb	8.7 ± 1.8		
Cr	63.7 ± 8.0	As	22.1 ± 8.5
Sb	1.7 ± 0.2	Ba	360 ± 85
La	15.8 ± 6.0	Ce	28.8 ± 8.2
Nd	13.4 ± 3.3	U	< 0.5

Table S2. Molar elemental ratios to Al for Saharan aerosol samples from previous studies collected over the Atlantic (Shelley et al., 2015; Jickells et al., 2016) and eastern Mediterranean (Herut et al., 2016) and for all air mass types encountered during GA04.

	Shelley	Jickells	Herut	This study (GA04)				
				Air mass type				
	Saharan Aerosols			NAF	EEU	WEU	MED	RNA
P/Al		0.016	0.007	0.021	0.055	0.089	0.029	0.202
Ti/Al	0.044	0.027	0.026	0.033	0.037	0.037	0.037	0.050
V/Al	0.0015	0.010		0.007	0.010	0.016	0.009	0.016
Mn/Al	6.3×10^{-3}	5.0×10^{-3}	5.0×10^{-3}	4.5×10^{-3}	8.7×10^{-3}	9.5×10^{-3}	5.2×10^{-3}	12.9×10^{-3}
Fe/Al	0.37	0.30	0.33	0.25	0.37	0.36	0.29	0.26
Co/Al	0.16×10^{-3}	0.17×10^{-3}		0.13×10^{-3}	0.18×10^{-3}	0.25×10^{-3}	0.16×10^{-3}	1.7×10^{-3}
Ni/Al	0.00046		0.0003	0.0020	0.0036	0.0063	0.0031	0.0265
Cu/Al	0.0003	0.0005	0.0004	0.0064	0.0039	0.0125	0.0032	0.118
Zn/Al	0.0008	0.0019	0.0014	0.005	0.021	0.168	0.012	0.196
Cd/Al	4×10^{-6}	5.8×10^{-6}	1×10^{-6}	16×10^{-6}	99×10^{-6}	144×10^{-6}	31×10^{-6}	850×10^{-6}
Pb/Al	0.08×10^{-3}	0.0001	0.05×10^{-3}	0.0004	0.0012	0.0018	0.0007	0.0115
Th/Al	24×10^{-6}	18×10^{-6}		15×10^{-6}	20×10^{-6}	27×10^{-6}	18×10^{-6}	

Table S3. Representative (mean) elemental concentrations and enrichment factors (EF) for aerosols collected during shipboard sampling in the Mediterranean from previous studies and during GA04. Published data from Chester et al. (1984) (Ches84) for air mass arrivals from North Africa (Sahara) and the Mediterranean (Med.), Chester et al. (1993) (Ches93) and Moreno et al. (2010) (More10).

	Ches84 (Sahara)		Ches84 (Med.)		Ches93		More10		GA04	
	pmol m^{-3}	EF	pmol m^{-3}	EF	pmol m^{-3}	EF	pmol m^{-3}	EF	pmol m^{-3}	EF
P							660	9.5	1050	6.9
Al	74100		4150		548000		32000		47000	
Ti							970	1.3	1600	1.1
V							340	42	200	17
Mn	337	0.8	78	1.0	2900	1.6	180	2.2	230	1.5
Fe	22400	0.9	2100	1.0	181000	1.1	9500	1.8	12000	1.1
Co							4.9	4.0	6.1	1.9
Ni	53	3	22	5	240	17	95	29	68	14
Cu	42	6	31	28	190	15	92	65	63	25
Zn	128	6	184	120	400	86	280	91	640	110
Cd	3	96	4	1220	2.4	520	2.6	320	1.0	92
Pb	36	46	53	770	65	490	44	150	16	35
Th							1.0	1.9	0.7	1.2

Table S4. Representative (mean or range) elemental concentrations and enrichment factors (EF) for aerosols collected during shipboard sampling in the Black Sea from previous studies and during GA04. Published data from Hacisalihoglu et al. (1992) (Hac92) in the western (West) and eastern (East) basins and Kubilay et al. (1995) (Kub95).

	Hac92 (West)		Hac92 (East)		Kub95		GA04	
	pmol m^{-3}	EF	pmol m^{-3}	EF	pmol m^{-3}	EF	pmol m^{-3}	EF
P							1400	7.2
Al	20000		12000		13000-64000		30000	
Ti							1000	1.1
V	55	3.2	35	3.4	0.2-90	0.02-1.6	48	1.9
Mn	310	3.0	310	4.8	91-1600	1.3-4.9	230	1.5
Fe	7500	1.3	5200	1.5	4600-65000	1.2-3.6	11000	1.3
Co	4.2	1.9	2.7	2.0	0.2-9.0	0.1-1.3	4.2	1.3
Ni	84	11	51	11	36-200	7.1-8.2	25	2.2
Cu	2.2	0.5					54	6.9
Zn	700	72	400	66	46-1100	7.2-34	330	24
Cd					2.7-40	230-700	2.7	110
Pb	290	440	180	450	8.7-230	21-110	30	32
Th	0.5	1.4	0.2	1.1			0.6	1.2

Table S5. Mean, median and ranges of dry deposition fluxes for soluble Al (s-Al), Ti (s-Ti), V (s-V), Co (s-Co), Cu (s-Cu), Pb (s-Pb) and Th (s-Th) over the western and eastern Mediterranean and Black Sea basins.

	s-Al	s-Ti	s-V	s-Co	s-Cu	s-Pb	s-Th
Western Med	nmol m^{-2} d^{-1}	nmol m^{-2} d^{-1}	nmol m^{-2} d^{-1}	nmol m^{-2} d^{-1}	nmol m^{-2} d^{-1}	nmol m^{-2} d^{-1}	nmol m^{-2} d^{-1}
Mean	847	1.27	18.2	1.72	5.9	1.40	0.059
Median	688	1.11	12.1	0.90	2.1	1.26	0.037
min	116	0.18	0.1	0.03	0.5	0.03	0.006
max	2100	4.55	117	9.42	27.6	4.18	0.228
Eastern Med							
Mean	935	0.90	6.1	1.59	4.7	2.33	0.074
Median	423	0.26	2.2	0.86	1.5	1.10	0.029
min	56	0.09	0.05	0.02	0.3	0.10	0.003
max	7540	3.52	33.9	14.6	21.2	16.6	0.793
Black Sea							
Mean	1280	0.57	7.3	2.40	4.2	1.05	0.203
Median	553	0.39	8.5	1.09	4.0	0.87	0.036
min	352	0.20	0.9	0.44	1.3	0.37	0.013
max	5750	1.34	12.9	11.0	10.3	2.01	1.16

References

Chester, R., Sharples, E. J., Sanders, G. S., and Saydam, A. C.: Saharan dust incursion over the Tyrrhenian Sea, Atmospheric Environment (1967), 18, 929-935, 10.1016/0004-6981(84)90069-6, 1984.

Chester, R., Nimmo, M., Alarcon, M., Saydam, C., Murphy, K. J. T., Sanders, G. S., and Corcoran, P.: Defining the chemical character of aerosols from the atmosphere of the Mediterranean Sea and surrounding regions, Oceanol. Acta, 16, 231-246, 1993.

Hacisalihoglu, G., Eliyakut, F., Olmez, I., Balkas, T. I., and Tuncel, G.: Chemical composition of particles in the Black Sea atmosphere, Atmospheric Environment Part A-General Topics, 26, 3207-3218, 10.1016/0960-1686(92)90477-3, 1992.

Herut, B., Rahav, E., Tsagaraki, T. M., Giannakourou, A., Tsiola, A., Psarra, S., Lagaria, A., Papageorgiou, N., Mihalopoulos, N., Theodosi, C. N., Violaki, K., Stathopoulou, E., Scoullos, M., Krom, M. D., Stockdale, A., Shi, Z., Berman-Frank, I., Meador, T. B., Tanaka, T., and Paraskevi, P.: The potential impact of Saharan dust and polluted aerosols on microbial populations in the East Mediterranean Sea, an overview of a mesocosm experimental approach, Frontiers in Marine Science, 3, 226, 10.3389/fmars.2016.00226, 2016.

Jickells, T. D., Baker, A. R., and Chance, R.: Atmospheric transport of trace elements and nutrients to the oceans, Philosophical Transactions of the Royal Society of London Series A-Mathematical Physical and Engineering Sciences, 374, 20150286, 10.1098/rsta.2015.0286, 2016.

Kubilay, N., Yemenicioglu, S., and Saydam, A. C.: Airborne material collections and their chemical composition over the Black Sea, Marine Pollution Bulletin, 30, 475-483, 10.1016/0025-326x(95)00238-i, 1995.

Moreno, T., Perez, N., Querol, X., Amato, F., Alastuey, A., Bhatia, R., Spiro, B., Hanvey, M., and Gibbons, W.: Physicochemical variations in atmospheric aerosols recorded at sea onboard the Atlantic-Mediterranean 2008 Scholar Ship cruise (Part II): Natural versus anthropogenic influences revealed by PM10 trace element geochemistry, Atmospheric Environment, 44, 2563-2576, 10.1016/j.atmosenv.2010.04.027, 2010.

Shelley, R. U., Morton, P. L., and Landing, W. M.: Elemental ratios and enrichment factors in aerosols from the US-GEOTRACES North Atlantic transects, Deep Sea Research Part II: Topical Studies in Oceanography, 116, 262-272, 10.1016/j.dsr2.2014.12.005, 2015.