

## Supplementary material

Table S1. Observed clean marine organic aerosol concentrations ( $\mu\text{g m}^{-3}$ )\*

Location	Lat	Lon	Collection date	Conc.	Note	Refs
Bermuda	32.25	-64.87	16-17 May 1973	0.42 $\pm$ 0.1	(a)	(1)
			17-18 May	0.36 $\pm$ 0.08		
			18-19 May	0.32 $\pm$ 0.1		
			21-22 May	0.36 $\pm$ 0.08		
			23-24 May	0.66 $\pm$ 0.15		
			24-26 May	0.21 $\pm$ 0.04		
			28-29 May	0.38 $\pm$ 0.08		
			30-31 May	0.50 $\pm$ 0.18		
Eastern Tropical Pacific	5.49	-83.72	11-12 Feb 1974	0.82 $\pm$ 0.08	(a),(b),(c)	(2)
	3.49	-87.56	12-13 Feb	0.24 $\pm$ 0.08		
	1.74	-89.42	13-14 Feb	0.57 $\pm$ 0.08		
	1.28	-89.94	14-15 Feb	0.59 $\pm$ 0.08		
	1.94	-92.49	15-16 Feb	0.57 $\pm$ 0.08		
	1.53	-93.98	16-17 Feb	0.16 $\pm$ 0.08		
	0.48	-92.47	17-18 Feb	0.08 $\pm$ 0.08		
	0.23	-89.92	18-19 Feb	0.50 $\pm$ 0.08		
	1.32	-87.35	19-20 Feb	0.16 $\pm$ 0.08		
	2.43	-84.84	20-21 Feb	0.24 $\pm$ 0.08		
	1.83	-82.87	21-22 Feb	0.41 $\pm$ 0.08		
	1.52	-81.75	22-23 Feb	0.51 $\pm$ 0.08		

Bermuda	32.25	-64.87	Jun 1975	0.52	(a), (d)	(3)
			Jun	0.41		
			Jun	0.46		
			Jun	0.29		
			Jun	0.21		
			Jun	0.22		
Hawaii	21.37	-147.70	Jul-Oct 1975	0.60	(a)	(3)
			Jul-Oct	0.50		
			Jul-Oct	0.52		
			Jul-Oct	0.56		
			Jul-Oct	0.55		
			Jul-Oct	0.55		
			Jul-Oct	0.50		
American Samoa	-14.30	-170.57	Jun-Aug 1976	0.57	(e)	(3)
			Jun-Aug	0.39		
			Jun-Aug	0.29		
			Jun-Aug	0.36		
			Jun-Aug	0.36		
			Jun-Aug	0.22		
			Jun-Aug	0.20		
			Jun-Aug	0.18		
			Jun-Aug	0.22		
Loop Head, Ireland	52.6	-9.9	Jun-Jul 1977	0.80	(f)	(4)
Tasmania	-40.68	144.68	Feb 1978	0.74	(f)	(5)

Tasmania	-40.68	144.68	Sep 1979	0.45		(6)
			Sep	0.32		
			Sep	0.36		
			Sep	0.20		
Northeast Atlantic	40	-10	Oct 1980	0.42	(c)	(7)
Western Tropical Atlantic	-20	-40	Nov 1980	0.43	(c)	(7)
Tropical Pacific	0	-110	Jul 1982	0.35	(b),(g),(h),(i)	(8)
	0	-115	Jul	0.24		
	0	-122	Jul	0.21		
	-2	-140	Jul	0.11		
	0	-140	Jul	0.17		
	10	-140	Aug	0.39		
	18	-140	Aug	0.39		
Bermuda	32.25	-64.87	Aug 1982	1.08	(j),(k),(h)	(9)
			Jan-Feb 1983	0.80		
Eastern Tropical Pacific	-15.0	-80.0	14 Mar 1981	0.27 $\pm$ 0.03	(l),(b)	(10)
			16 Mar	0.08 $\pm$ 0.01		
			20 Mar	0.21 $\pm$ 0.02		
			22 Mar	0.21 $\pm$ 0.02		
			25 Mar	0.38 $\pm$ 0.03		
			29 Mar	0.15 $\pm$ 0.02		
			1 Apr	0.08 $\pm$ 0.01		
			5 Apr	0.35 $\pm$ 0.04		
American Samoa	-14.30	-170.57	7 Jul 1981	0.18 $\pm$ 0.02	(l)	(10)

				17 Jul	0.15 $\pm$ 0.02		
				28 Jul	0.15 $\pm$ 0.02		
				5 Aug	0.10 $\pm$ 0.01		
				26 Jan	0.13 $\pm$ 0.01		
				17 Feb	0.20 $\pm$ 0.02		
Amsterdam Island	-37.8	77.6	10 Feb 1982	0.21 $\pm$ 0.02	(l)	(10)	
New Zealand coast	-34.4	172.7	20 Jun 1983	0.21 $\pm$ 0.02		(10)	
				10 Jul	0.17 $\pm$ 0.02		
				28 Jul	0.17 $\pm$ 0.02		
				19 Aug	0.18 $\pm$ 0.02		
Northeast Atlantic	35.7	-22.0	30 Apr 1985	0.80 $\pm$ 0.08	(l),(b)	(10)	
				6 May	0.42 $\pm$ 0.04		
				9 May	0.31 $\pm$ 0.03		
				13 May	0.43 $\pm$ 0.04		
Puerto Rico	18.32	-65.75	2-3 Apr 1992	0.556 $\pm$ 0.14	(m)	(11)	
				4-5 Apr	0.896 $\pm$ 0.15		
				8-9 Apr	0.568 $\pm$ 0.13		
				10-11 Apr	0.628 $\pm$ 0.14		
Puerto Rico	18.32	-65.75	12-13 Feb 1995	0.496	(n),(o)	(12)	
				13-14 Feb	0.830		
				14-15 Feb	0.588		
				15-16 Feb	0.501		
				16-17 Feb	0.267		

			13-14 Mar	0.487		
			14-15 Mar	0.329		
			15-16 Mar	0.592		
			16-17 Mar	0.491		
			30 May	0.626		
			31 May	0.678		
			1 Jun	0.578		
			2 Jun	0.676		
Tenerife	28.3	-16.5	Jun-Jul 1997	0.29	(o),(p)	(13)
Bermuda	32.25	-64.87	Apr-Jun 1998	0.53	(q),(r),(s)	(14)
Trinidad Head, California	41.05	-124.15	Apr-May 2002	0.38	(t)	(15)
Northeast Pacific	30	-160	Mar 2001	0.28 $\pm$ 0.05	(u),(v),(w)	(16)
Mace Head, Ireland	53.3	-9.9	Apr-Oct 2002	0.91	(h),(p),(x)	(17)
North Pacific	50.0	-145.0	Jul 2002	0.3 $\pm$ 0.1	(p),(y)	(18)
Mace Head, Ireland	53.3	-9.9	Winter 2002-2004	0.07	(h),(p),(x)	(19)
			Spring	0.72		
			Summer	0.68		
			Fall	0.50		
Southwest Atlantic	-44	-20	Jan 2007	0.03	(b),(z)	(20)
	-44	-60	31 Jan-2 Feb	0.32		
	-60	-40	Mar	0.02		
Amsterdam Island	-37.80	77.57	Jan 2003-2007	0.381	(aa),(ab)	(21)
			Feb	0.253		
			Mar	0.203		

			Apr	0.172		
			May	0.171		
			Jun	0.136		
			Jul	0.144		
			Aug	0.150		
			Sep	0.164		
			Oct	0.130		
			Nov	0.151		
			Dec	0.272		
Eastern Tropical Atlantic	29	-15	May 2007	0.2	(h),(p),(y)	(22)
	23	-21	May	0.8		
	21	-22	May	0.6		
	19	-25	May	0.4		
Northeast Atlantic	46.49	-44.67	Mar 2008	0.73	(h),(p)	(23)
	63.25	18.42	Apr	0.35		
	70.95	-7.80	Apr	0.27		
	70.24	-13.40	Apr	0.28		
Mace Head, Ireland	53.3	-9.9	11-18 Jan 2006	0.04	(h),(p)	(24)
			18-22 Jan	0.13		
			10-20 Feb	0.17		
			29 Mar-4 Apr	0.08		
			12-26 Apr	0.18		
			12-19 Jun	0.31		
			19-28 Jun	0.24		

			28 Jun-5 Jul	0.38		
			3-8 Sep	0.10		
			5-11 Oct	0.10		
			15-22 Nov	0.07		
			4-11 Dec	0.07		
Northwest Pacific	41	155	Jul-Aug 2008	0.88	(b),(ac),(ad)	(25)
	45	167	Aug	0.55	(b),(ac),(ae)	
Northwest Pacific	30	150	Sep 2008	1.06	(b),(ac)	(26)
	43	155	Aug	1.69	(b),(ac)	
North Pacific	31	175	Nov 1989	0.22	(b),(ac),(af)	(27)
	30	-160	Nov	0.08		
	32	-135	Nov	0.11		
North Atlantic	30	-60	Dec 1989	0.32	(b),(ac),(af)	(27)
	30	-35	Dec	0.25		
Mace Head, Ireland	53.3	-9.9	14 Aug 2009	0.16	(h),(z),(p),(ag)	(28)
			15 Aug	0.57		
			16 Aug	2.49		
			17 Aug	0.74		
			18 Aug	0.35		
			19 Aug	0.31		
Northeast Pacific	45	-130	Apr 2006	0.15	(p),(z)	(29)
Eastern Tropical Pacific	-20	-85	Oct-Nov 2008	0.10	(p),(z)	(29)
Eastern Tropical Pacific	10	-140	Sep 2009	0.02	(p),(z)	(29)

\* When applicable, concentrations are converted from  $\mu\text{g C m}^{-3}$  to  $\mu\text{g m}^{-3}$  using an organic matter/organic carbon ratio of 1.4. When reported, uncertainty ranges are depicted by  $\pm$  values.

(1) Hoffman and Duce (1974); (2) Barger and Garrett (1976); (3) Hoffman and Duce (1977); (4) Eichmann et al. (1979); (5) Eichmann et al. (1980); (6) Andreae (1982); (7) Andreae (1983); (8) Andreae et al. (1984); (9) Wolff et al. (1986); (10) Cachier et al. (1986); (11) Novakov and Penner (1993); (12) Novakov et al. (1997); (13) Putaud et al. (2000); (14) Turekian et al. (2003); (15) Allan et al. (2004); (16) Quinn et al. (2004); (17) Cavalli et al. (2004); (18) Phinney et al. (2006); (19) Yoon et al. (2007); (20) Zorn et al. (2008); (21) Sciare et al. (2009); (22) Lee et al. (2010); (23) Russell et al. (2010); (24) Rinaldi et al. (2010); (25) Miyazaki et al. (2010); (26) Miyazaki et al. (2011); (27) Fu et al. (2011); (28) Ovadnevaite et al. (2011a); (29) Shank et al. (2012)

- (a) Atmospheric samples were collected over a period of 24 to 48 hours on precombusted glass-fiber filters and extracted with chloroform
- (b) Ship-borne measurements with the average coordinates reported between the start and end of a sampling period
- (c) We have included samples having BC concentrations  $< 0.05 \mu\text{g m}^{-3}$  and a BC/total carbon ratio  $< 0.1$ .
- (d) We have omitted samples with high dust loading
- (e) Atmospheric samples were collected over a period of 7 days on precombusted glass fiber filters and extracted with chloroform
- (f) Atmospheric samples with no continental contact during 48 hr back trajectory were collected over a period of 12 to 48 hours on precombusted glass fiber filters and analyzed using a combustion method
- (g) In the aerosol fraction  $\leq 1.7 \mu\text{m}$  diameter
- (h) Samples having BC concentrations  $< 0.05 \mu\text{g m}^{-3}$
- (i) Excess potassium  $< 5 \text{ ng m}^{-3}$
- (j) Atmospheric samples were collected over a period of 24 hours on Microquartz filters and analyzed using a combustion method
- (k) In the aerosol fraction  $\leq 2.5 \mu\text{m}$  diameter
- (l) Atmospheric samples were collected over a period of 2 days to 2 weeks on precombusted glass-fiber filters and extracted with different kinds of solvent
- (m) Atmospheric samples were collected over a period of 24 hours on aluminum foil substrates and analyzed using thermal-optical reflectance method
- (n) Atmospheric samples were collected over a period of 12 to 24 hours on precombusted quartz-fiber filters and analyzed by thermal Evolved Gas Analysis
- (o) In the aerosol fraction  $\leq 0.6 \mu\text{m}$  diameter
- (p) In the aerosol fraction  $\leq 1 \mu\text{m}$  diameter
- (q) Atmospheric samples were collected over a period of 20 to 48 hours on precombusted glass-fiber filters and analyzed with Carlo-Erba NA 1500 NCS elemental analyzer
- (r) Geometric mean diameter  $< 1.2 \mu\text{m}$
- (s) Numbers in parentheses represent upper and lower estimates
- (t) Atmospheric samples were collected over a period of 20 to 48 hours on precombusted glass-fiber filters and analyzed with Carlo-Erba NA 1500 NCS elemental analyzer

- (u) Atmospheric samples were collected over a period of 2.5 to 12 hours on quartz filter and analyzed with a Sunset Labs thermal/optical analyzer
- (v) In the aerosol fraction  $\leq 1.1 \mu\text{m}$  diameter
- (w) The mass of particulate organic matter (POM) was determined by multiplying the measured OC concentration in  $\mu\text{g m}^{-3}$  by a factor of 2.1 in the marine region
- (x) Atmospheric samples were collected over a period of 7 days on tedlar foil (for water soluble organic carbon) and quartz filters (for total carbon) and analyzed with a Total Organic Carbon liquid analyzer and thermal Evolved Gas Analysis, respectively
- (y) The real-time quantitative measurements of atmospheric samples were conducted using Aerodyne Aerosol Mass Spectrometer (AMS)
- (z) The real-time quantitative measurements of atmospheric samples were conducted using Aerodyne High-Resolution-Time-of-Flight AMS (HR-ToF-AMS)
- (aa) Atmospheric samples were collected over a period of 8 to 10 days on pre-fired Whatman QMA quartz filters and analyzed with a thermo-optical transmission carbon analyzer system and total organic carbon analyzer
- (ab) Non-methanesulfonate organic carbon (nms-OC)
- (ac) Atmospheric samples were collected on precombusted quartz filters and analyzed using a Sunset lab EC/OC analyzer
- (ad) Highly biologically influenced aerosols
- (ae) Less biologically influenced aerosols
- (af) Samples having EC concentrations below detection limit (not defined) are used
- (ag) Daily average