Supporting Information to

Chemical Characterization of Isoprene- and Monoterpene-Derived Secondary Organic Aerosol (SOA) Tracers in Remote Marine Aerosols over A Quarter Century

Tianqu Cui[†], Hilary S. Green[†‖], Paul W. Selleck[‡], Zhenfa Zhang[†], Rachel E. O'Brien[§⊥], Avram Gold[†], Melita Keywood[‡], Jesse H. Kroll[§], and Jason D. Surratt[*†]

[†] Department of Environmental Sciences and Engineering, Gillings School of Global Public Health, University of North Carolina at Chapel Hill, Chapel Hill, NC 27599, USA

[‡] CSIRO Oceans and Atmosphere, Aspendale, Victoria 3195, Australia

[§] Department of Civil and Environmental Engineering, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

[‖] Present Address: Agricultural and Environmental Chemistry Graduate Group, Department of Food Science and Technology, University of California, Davis, Davis, CA 95616, USA.

[⊥] Present Address: Department of Chemistry, College of William and Mary, Williamsburg, VA, 23185, USA

* Corresponding Author – Jason D. Surratt: surratt@unc.edu

Contents:

SUPPORTING INFORMATION

Table S1. List of analyzed samples (n=211) and field blank filters (n=26).

Filter ID	Season	# of Combined Samples (in the season)	Start (date and time)	Finish (date and time)	Collected Air Volume m^3	Extracted Filter Collection Area cm^2	Notes
345			12/18/1990 10:59	1/2/1991 10:08	2888	52.06	Total collection area was 384.3 cm2 from 1991-1999.
346			1/2/1991 10:12	1/8/1991 13:40	1013	52.06	
347			1/8/1991 13:50	1/15/1991 10:37	2134	60.02	
348	91 S	9	1/15/1991 10:42	1/22/1991 11:00	2880	50.58	
349			1/22/1991 11:07	1/29/1991 13:40	1823	60.88	
350			1/29/1991 13:48	2/5/1991 9:40	5237	58.74	
351			2/5/1991 9:45	2/12/1991 12:32	3531	60.12	
352			2/12/1991 12:36	2/19/1991 8:57	1154	68.48	
353			2/19/1991 9:03	2/26/1991 8:49	2525	68.98	
370			6/18/1991 14:26	6/25/1991 13:50	3260	83.38	
371			6/25/1991 13:56	7/2/1991 14:50	4610	91.76	
372			7/2/1991 14:58	7/9/1991 11:50	1252	99.12	
373			7/9/1991 12:00	7/17/1991 9:45	3971	84.16	
374	91 W	9	7/17/1991 9:50	7/23/1991 12:00	2640	100.84	
375			7/23/1991 12:09	7/30/1991 14:40	7049	91.04	
376			7/30/1991 14:47	8/6/1991 14:57	2137	94.48	
377			8/6/1991 15:02	8/13/1991 15:19	5039	71.34	
378			8/13/1991 15:25	8/20/1991 14:58	4008	82.16	
398			12/31/1991 11:00	1/16/1992 8:35	5375	65.06	
400			1/21/1992 11:50	1/28/1992 14:28	3248	52.52	
401	92 S	6	1/28/1992 15:00	2/4/1992 13:45	3869	81.74	
402			2/4/1992 13:50	2/11/1992 14:00	681	75.86	
403			2/11/1992 14:06	2/18/1992 13:12	1169	71.92	
404			2/18/1992 13:30	2/25/1992 14:08	887	76.02	
421			6/16/1992 13:05	6/23/1992 11:25	1245	68.48	
422			6/23/1992 12:20	6/30/1992 11:17	152	83.62	Used as a field blank
423			6/30/1992 11:59	7/7/1992 12:02	1752	62.38	
424			7/7/1992 13:00	7/14/1992 12:10	3582	54.52	
425	92 W	7	7/14/1992 13:24	7/21/1992 11:35	1464	58.52	
426			7/21/1992 12:06	7/28/1992 12:00	3894	63.08	
427			7/28/1992 13:00	8/4/1992 13:00	4764	73.28	
428			8/4/1992 13:40	8/11/1992 12:37	3018	74.60	
429			8/11/1992 13:10	8/18/1992 12:40	3271		Not extracted
448			12/22/1992 10:01	12/30/1992 13:13	2917	82.56	
449			12/30/1992 14:07	1/5/1993 13:34	1406	84.56	
450			1/5/1993 14:44	1/12/1993 15:01	5203	64.62	
451	93 S	7	1/12/1993 16:15	1/19/1993 12:00	539	61.66	
452			1/19/1993 12:20	1/26/1993 14:25	25	67.54	Used as a field blank
453			1/26/1993 14:35	2/2/1993 12:40	1187	57.24	
455			2/9/1993 13:00	2/16/1993 13:51	1142	90.22	
456			2/16/1993 14:50	2/23/1993 13:12	2309	79.76	
473			6/15/1993 14:55	6/22/1993 14:26	2590	72.24	
474			6/22/1993 15:58	6/29/1993 14:40	5068	62.56	
475			6/29/1993 15:40	7/6/1993 14:15	983	55.52	
476			7/6/1993 15:05	7/13/1993 13:30	158	63.82	Used as a field blank
477	93 W	8	7/13/1993 14:30	7/20/1993 14:22	2895	51.64	
478			7/20/1993 15:11	7/27/1993 14:20	1595	60.18	
479			7/27/1993 15:33	8/3/1993 15:10	2047	65.82	
480			8/3/1993 15:46	8/10/1993 14:05	2622	63.34	
481			8/10/1993 15:40	8/17/1993 13:50	2792	67.72	
494			12/21/1993 14:00	12/28/1993 11:50	1111	57.46	
495			12/28/1993 12:42	1/4/1994 10:32	4010	75.60	
496			1/4/1994 12:30	1/11/1994 13:50	549	61.62	
497			1/11/1994 14:45	1/18/1994 13:34	2092	57.90	
498	94 S	9	1/18/1994 14:50	1/25/1994 10:27	3438	65.06	
499			1/25/1994 15:00	2/1/1994 14:27	2281	60.18	
500			2/1/1994 15:55	2/8/1994 11:36	1151	60.56	
501			2/8/1994 15:00	2/15/1994 12:41	915	76.48	
502			2/15/1994 14:09	2/22/1994 9:25	1789	83.36	
519			6/14/1994 16:45	6/21/1994 13:37	4287	67.94	
520			6/21/1994 16:04	6/28/1994 13:30	563	70.88	
521			6/28/1994 16:09	7/5/1994 10:40	1123	70.00	
522			7/5/1994 12:39	7/12/1994 10:58	1311	72.22	
523	94 W	9	7/12/1994 13:26	7/19/1994 11:17	5368	59.40	
524			7/19/1994 12:30	7/26/1994 10:35	4731	74.16	
525			7/26/1994 12:02	8/2/1994 10:39	4441	59.84	
526			8/2/1994 11:38	8/9/1994 10:57	3898	74.38	
527			8/9/1994 11:33	8/16/1994 10:52	2683	76.46	
598			12/26/1995 9:39	1/2/1996 11:27	3701	93.68	
CGHV-599			1/2/1996 13:03	1/9/1996 12:48	360	64.88	
CGHV-600			1/9/1996 14:35	1/16/1996 11:55	2057	66.50	
CGHV-601	96 S	7	1/16/1996 15:25	1/23/1996 11:36	3613	60.46	
CGHV-602			1/23/1996 14:32	1/30/1996 12:00	1447	63.16	
CGHV-603			1/30/1996 14:05	2/6/1996 11:17	1899	61.74	
CGHV-604			2/6/1996 13:55	2/13/1996 13:04	1562	97.60	
CGHV-623			6/18/1996 12:19	6/25/1996 14:00	1763	87.11	
CGHV-624	96 W	8	6/25/1996 15:18	7/2/1996 13:20	2423	65.06	
CGHV-625			7/2/1996 14:31	7/9/1996 12:16	1508	67.54	
CGHV-626			7/9/1996 14:20	7/16/1996 13:53	651	66.54	

CGHV-627			7/16/1996 15:13	7/23/1996 12:32	2796	71.92	
CGHV-628			7/23/1996 14:13	7/30/1996 12:30	2070	72.92	
CGHV-629			7/30/1996 15:36	8/6/1996 11:42	2311	86.92	
CGHV-630			8/6/1996 13:18	8/13/1996 12:00	4538	80.56	
CGHV-651			12/31/1996 14:03	1/7/1997 11:45	2533	76.31	
CGHV-652			1/7/1997 13:55	1/14/1997 13:09	1782	64.27	
CGHV-653			1/14/1997 13:30	1/21/1997 11:13	1960	89.55	
CGHV-654	97 S	6	1/21/1997 13:00	1/28/1997 9:38	1355	75.79	
CGHV-655			1/28/1997 10:35	2/4/1997 11:15	449	91.27	Used as a field blank
CGHV-656			2/4/1997 12:17	2/11/1997 13:10	1486	81.47	
CGHV-657			2/11/1997 13:41	2/18/1997 13:34	647	72.87	
CGHV-675			6/17/1997 14:20	6/24/1997 12:42	1156	78.12	
CGHV-676			6/24/1997 14:12	7/1/1997 15:30	171	76.31	
CGHV-677			7/1/1997 16:49	7/8/1997 12:05	1364	77.34	
CGHV-678			7/8/1997 12:42	7/15/1997 14:02	4098	83.25	
CGHV-679	97 W	9	7/15/1997 15:30	7/22/1997 13:15	8112	87.16	
CGHV-680			7/22/1997 13:55	7/29/1997 13:46	4397	89.13	
CGHV-681			7/29/1997 14:36	8/5/1997 12:49	4764	86.91	
CGHV-682			8/5/1997 13:59	8/12/1997 12:53	3643	84.47	
CGHV-683			8/12/1997 14:13	8/19/1997 15:21	4741	86.16	
CGHV-703			12/30/1997 9:46	1/6/1998 11:27	3338	85.16	
CGHV-704			1/6/1998 12:57	1/13/1998 11:41	1458	77.40	
CGHV-705			1/13/1998 12:53	1/20/1998 11:02	856	72.18	
CGHV-706	98 S	8	1/20/1998 12:09	1/27/1998 11:56	162	94.76	
CGHV-707			1/27/1998 13:27	2/3/1998 12:42	2469	69.43	
CGHV-708			2/3/1998 13:52	2/10/1998 11:22	1148	95.71	
CGHV-709			2/10/1998 12:34	2/17/1998 11:33	2137	83.25	
CGHV-710			2/17/1998 11:57	2/24/1998 13:05	3043	74.87	
CGHV-728			6/23/1998 14:14	6/30/1998 12:20	1856	90.07	
CGHV-729			6/30/1998 13:10	7/7/1998 13:02	4273	83.19	
CGHV-730			7/7/1998 15:25	7/14/1998 13:41	4796	84.47	
CGHV-731	98 W	7	7/14/1998 14:33	7/21/1998 14:10	3292	70.71	
CGHV-732			7/21/1998 15:30	7/29/1998 14:14	3232	74.15	
CGHV-733			7/29/1998 15:31	8/4/1998 13:55	3849	79.06	
CGHV-734			8/4/1998 16:40	8/11/1998 13:36	1193	96.73	
CGHV-766			1/5/1999 11:41	1/12/1999 14:46	615	89.39	
CGHV-767			1/12/1999 15:30	1/19/1999 12:01	579	93.99	
CGHV-768			1/19/1999 13:26	1/25/1999 12:10	255	94.71	
CGHV-769	99 S	6	1/25/1999 13:40	2/2/1999 12:13	93		Used as a field blank
CGHV-770			2/2/1999 13:36	2/9/1999 11:36	547	91.38	
CGHV-771			2/9/1999 13:07	2/16/1999 10:21	76		Used as a field blank
CGHV-772			2/16/1999 11:42	2/23/1999 10:12	1534	81.28	
CGHV-773			2/23/1999 11:39	3/2/1999 11:54	268	88.64	
CGHV-782			6/15/1999 15:31	6/22/1999 12:43	3888	89.38	
CGHV-783			6/22/1999 13:43	6/29/1999 11:25	144		Used as a field blank
CGHV-784			6/29/1999 12:25	7/6/1999 13:12	3957	65.05	
CGHV-785			7/6/1999 14:30	7/13/1999 13:25	150		Used as a field blank
CGHV-786	99 W	8	7/13/1999 13:45	7/20/1999 15:17	965	82.04	
CGHV-787			7/20/1999 15:32	7/27/1999 11:55	5150	73.96	
CGHV-788			7/27/1999 12:10	8/3/1999 14:30	4725	73.21	
CGHV-789			8/3/1999 15:15	8/10/1999 13:51	5129	87.72	
CGHV-790			8/10/1999 14:15	8/17/1999 13:16	3974	83.76	
EA/EM 66			18/08/04 12:21	24/08/04 12:53	3092	104.05	Total collection area was 405.0 cm2 after from 2004-2015.
EA/EM 67			24/08/04 13:46	31/08/04 15:05	620	87.30	
EA/EM 68	04 W	5	31/08/04 15:49	07/09/04 13:51	4082	92.70	
EA/EM 69			07/09/04 14:28	14/09/04 14:37	5562	82.80	
EA/EM 70 BL							Used as a field blank
EA/EM 71			14/09/04 15:52	21/09/04 14:09	6529	99.00	
EB/EM 197			27/12/06 13:30	02/01/07 12:49	1178	115.25	
EB/EM 198BL							Used as a field blank
EB/EM 199			02/01/07 14:44	09/01/07 13:35	1420	109.85	
EB/EM 200	07 S	7	09/01/07 14:19	16/01/07 12:56	4135	110.95	
EB/EM 201			16/01/07 13:53	23/01/07 13:24	2785	120.45	
EB/EM 202			23/01/07 14:13	30/01/07 10:22	3614	106.25	
EB/EM 203			30/01/07 14:24	06/02/07 13:46	1857	93.65	
EB/EM 204			06/02/07 14:57	13/02/07 13:12	1516	101.55	
EB/EM 227			26/06/07 14:29	03/07/07 13:30	3074	92.75	
EB/EM 228			03/07/07 14:38	10/07/07 11:34	1173	99.05	
EB/EM 229			10/07/07 13:51	17/07/07 13:34	1530	105.95	
EB/EM 230BL	07 W	6					Used as a field blank
EB/EM 231			17/07/07 15:17	24/07/07 11:28	2111	104.45	
EB/EM 232			24/07/07 13:44	31/07/07 11:45	3286	108.05	
EB/EM 233			31/07/07 14:49	07/08/07 13:38	5776	117.85	
EB/EM 256			18/12/07 11:27	27/12/07 10:25	7421	104.40	
EB/EM 257			27/12/07 11:21	02/01/08 13:08	1768	99.90	
EB/EM 258BL						101.70	Used as a field blank
EB/EM 259			02/01/08 14:12	08/01/08 12:16	3379	104.40	
EB/EM 260	08 S	8	08/01/08 13:29	15/01/08 11:03	3759	117.00	
EB/EM 261			15/01/08 12:17	22/01/08 12:28	2121	109.80	
EB/EM 262			22/01/08 13:51	29/01/08 10:52	1455	104.40	
EB/EM 263			29/01/08 12:25	05/02/08 12:09	2014	103.50	
EB/EM 264			05/02/08 13:24	12/02/08 13:47	5223	113.40	
EB/EM 285			17/06/08 13:50	24/06/08 10:20	5851	103.45	
EB/EM 286			24/06/08 12:27	01/07/08 09:57	1709	118.80	
EB/EM 287			01/07/08 11:49	08/07/08 10:15	2399	111.60	
EB/EM 288	08 W	8	08/07/08 12:21	15/07/08 11:26	2523	109.80	
EB/EM 289			15/07/08 13:32	22/07/08 10:49	3323	109.80	
EB/EM 290			22/07/08 14:14	29/07/08 10:15	1275	108.90	
EB/EM 291			29/07/08 12:31	05/08/08 11:53	2490	108.90	
EB/EM 292			05/08/08 14:36	14/08/08 10:27	1970	113.40	
EB/EM 336	09 W	5	07/07/09 14:21	14/07/09 11:29	2226	99.00	
EB/EM 337			14/07/09 12:42	21/07/09 13:58	1419	86.80	

EB/EM 338			21/07/09 14:50	28/07/09 14:04	3341	91.70	
EB/EM 339			28/07/09 14:54	04/08/09 11:48	6078	98.05	
EB/EM 340			04/08/09 12:33	11/08/09 11:19	1838	108.85	
EB/EM 341BL							Used as a field blank
EC/EM 118			21/12/10 13:38	28/12/10 10:37	4448	90.05	
EC/EM 119			28/12/10 11:16	04/01/11 13:06	4090	98.15	
EC/EM 120BL							Used as a field blank
EC/EM 121	11 S	6	04/01/11 14:59	11/01/11 12:40	1662	100.85	
EC/EM 123			18/01/11 13:42	25/01/11 13:07	4112	97.15	
EC/EM 124			25/01/11 14:06	01/02/11 12:19	4704	93.65	
EC/EM 125BL							Used as a field blank
EC/EM 127A			08/02/11 13:01	15/02/11 13:51	3208	99.05	
EC/EM 147			11:23 21/06/2011	13:04 28/06/2011	7085	108.95	
EC/EM 148			14:20 28/06/2011	13:55 05/07/2011	296	111.65	
EC/EM 149			15:26 05/07/2011	13:55 12/07/2011	9037	102.60	
EC/EM 150			15:03 12/07/2011	13:42 19/07/2011	3376	105.85	
EC/EM 151BL							Used as a field blank
EC/EM 152	11 W	8	14:37 19/07/2011	26/017/11 14:07	254	99.05	
EC/EM 153			14:45 26/07/2011	11:22 02/08/2011	1263	106.40	
EC/EM 155			12:34 02/08/2011	11:29 09/08/2011	438	111.65	
EC/EM 156			12:32 09/08/2011	14:12 16/08/2011	132	99.05	
EC/EM 157BL							Used as a field blank
EC/EM 181			10:45 28/12/2011	12:50 03/01/2012	74	104.45	
EC/EM 182					0	99.05	Not extracted
EC/EM 183			14:23 03/01/2012	13:58 10/10/2012	4610	90.95	
EC/EM 184	12 S	6	14:42 10/01/2012	12:04 17/01/2012	4847	90.95	
EC/EM 185			12:44 17/01/2012	12:56 24/01/2012	3269	99.05	
EC/EM 186			14:02 24/01/2012	10:24 31/01/2012	1985	97.25	
EC/EM 187BL							Used as a field blank
EC/EM 188			12:05 31/01/2012	13:19 07/02/2012	1462	99.05	
EC/EM 212			14:58 19/06/2012	14:04 26/06/2012	4692	101.75	
EC/EM 213			15:33 26/06/2012	13:30 03/07/2012	1372	100.85	
EC/EM 214			14:13 03/07/2012	13:49 10/07/2012	2850	99.05	
EC/EM 215			14:41 10/07/2012	14:48 17/07/2012	1178	102.65	
EC/EM 216BL	12 W	8					Used as a field blank
EC/EM 217			15:59 14/07/2012	12:41 24/07/2012	2656	102.65	
EC/EM 218			13:33 24/07/2012	11:23 31/07/2012	0	102.65	Not extracted
EC/EM 219			12:24 31/07/2012	13:19 07/08/2012	2921	107.15	
EC/EM 220			14:17 07/08/2012	10:32 14/08/2012	2806	114.35	
ED/EM 066			14:12 17/12/2013	10:42 24/12/2013	2162	105.30	
ED/EM 067			11:55 24/12/2013	10:03 31/12/2013	4085	104.45	
ED/EM 068BL							Used as a field blank
ED/EM 069			11:19 31/12/2013	12:23 07/01/2014	4952	104.60	
ED/EM 070	14 S	8	13:42 07/01/2014	13:59 14/01/2014	2692	100.85	
ED/EM 071			14:51 14/01/2014	14:58 21/01/2014	2526	95.45	
ED/EM 072			16:16 21/01/2014	13:16 28/01/2014	4021	109.85	
ED/EM 073BL							Used as a field blank
ED/EM 074			16:08 04/02/2014	13:28 11/02/2014	130	111.65	
ED/EM 075			14:04 11/02/2014	10:00 18/02/2014	2166	112.55	
ED/EM 098			11:50 24/06/2014	13:51 01/07/2014	4562	113.45	
ED/EM 099			15:08 01/07/2014	14:01 08/07/2014	1946	99.05	
ED/EM 100			15:33 08/07/2014	13:55 15/07/2017	5679	97.25	
ED/EM 101BL	14 W	6					Used as a field blank
ED/EM 102			15:56 15/07/2014	14:59 22/07/2014	2105	95.45	
ED/EM 103			15:50 22/07/2014	13:43 29/07/2014	35	99.05	
ED/EM 104			14:25 29/07/2014	13:56 05/08/2014	2105	95.45	
ED/EM 130			10:26 30/12/2014	12:42 06/01/2015	3467	90.05	
ED/EM 131			14:05 06/01/2015	13:33 13/01/2015	4	101.70	
ED/EM 132			14:20 13/01/2015	11:59 20/01/2015	5545	100.85	
ED/EM 133	15 S	7	13:00 20/01/2015	13:16 27/01/2015	4209	97.25	
ED/EM 134BL							Used as a field blank
ED/EM 135			14:21 27/01/2015	13:20 03/02/2015	595	115.25	
ED/EM 136			10:00 05/02/2015	12:29 10/02/2015	833	93.65	
ED/EM 137			13:06 10/02/2015	13:02 17/02/2015	1236	100.85	

(End of Table S1)

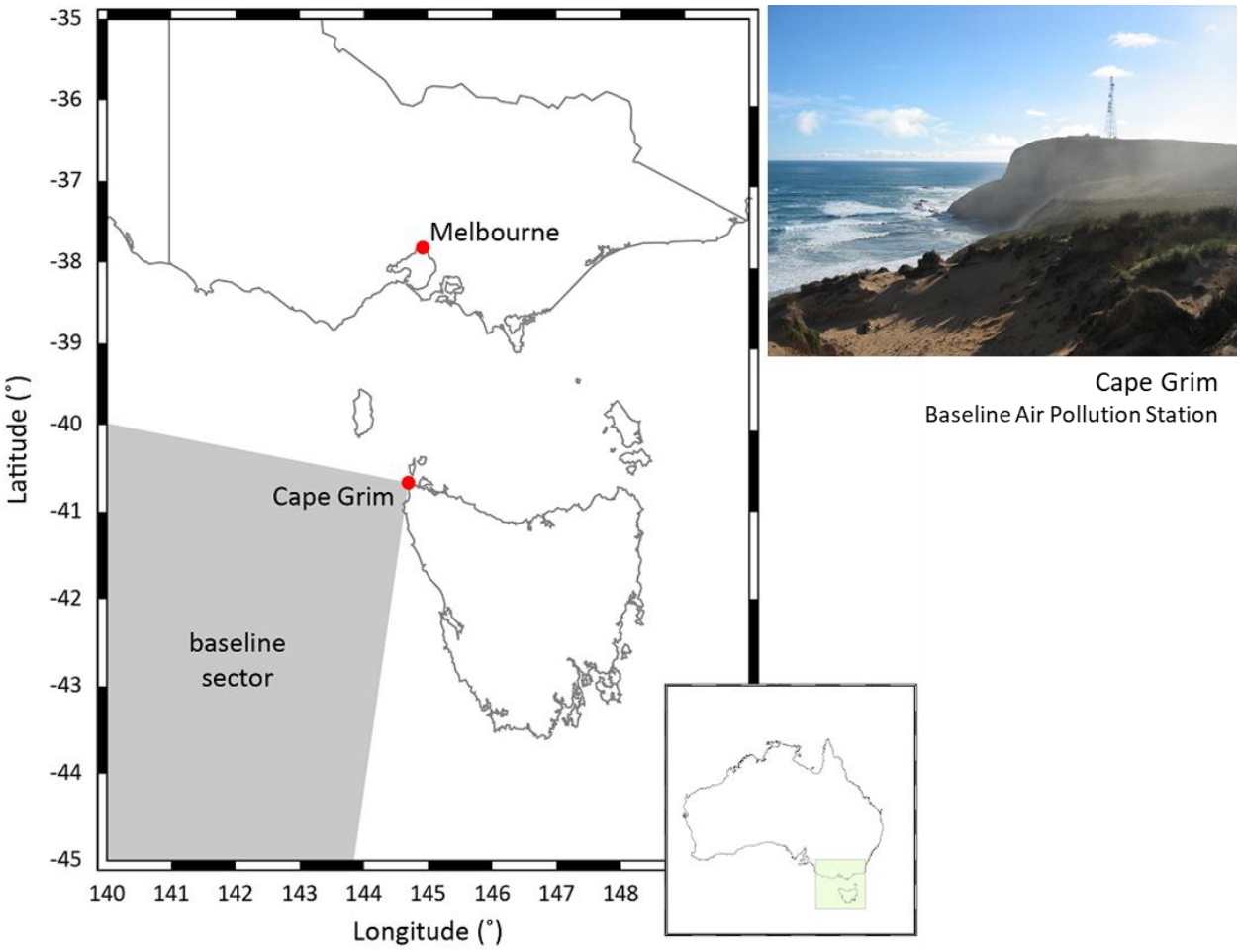

Figure S1. Location of Cape Grim site and "baseline" sector of wind direction. The Cape Grim Baseline Air Pollution Station is located at the top of 94-m cliffs on the western side of the north-west tip of Tasmania, Australia (40°40'56" S, 144°41'18" E).

1. Baseline Conditions and Supporting Data in Baseline Reports

Details of baseline conditions (i.e., Baseline Event Switch 2 and 3) can be found in Keywood, M.D (2007). Aerosol composition at Cape Grim: an evaluation of the PM_{10} sampling program and baseline event switches. Baseline Atmospheric Program Australia 2005-2006 (page 31-35). Melbourne: Australian Bureau of Meteorology and CSIRO Marine and Atmospheric Research at http://www.cmar.csiro.au/e-print/open/baseline_2005-2006.pdf.

The air mass origin maps (by U.K. Met Office) of selected events and complete baseline definitions built up from five different criteria including wind speed, wind direction, radon concentrations, condensation nuclei (CN) counts, and standard deviation of ozone, are shown by Molloy, S. B. and Galbally, E. in Analysis and identification of a suitable baseline definition for tropospheric ozone at Cape Grim, Tasmania, page 7-16 in the 2009-2010 Baseline Report at http://www.bom.gov.au/inside/cgbaps/baseline/Baseline_2009-2010.pdf.

The angular radon distribution and back trajectory data from 2009-2010 by Zahorowski et al. can be found from page 33-38 in the same Baseline Report above.

Annual or biennial Cape Grim Baseline Reports from more years since 1976 are available at http://www.bom.gov.au/inside/cgbaps/baseline.shtml.

Table S2. Concentrations of individual isoprene- and monoterpene-derived SOA tracers, levoglucosan, oxalic acid, methanesulfonic acid (MSA), and calculated chlorophyll-*a* (Chl-*a*) in aerosol samples collected from austral winter and summer seasons from 1991-2015.

Year-Season	2-MG[a]	Me.Thr.[b]	Me.Ery.[c]	Terebic Acid	Terpenylic Acid	Pinic Acid	MBTCA[d]	DTAA[e]	Levoglucosan	Oxalic Acid	MSA[f]	Cal. Chl-*a*[g]
yy(s/w)	ng/m^3	ng/m^3	ng/m^3	ng/m^3	ng/m^3	ng/m^3	ng/m^3	ng/m^3	ng/m^3	ng/m^3	ng/m^3	ng/m^3
91s	0.201	0.227	0.507	0.066	0.075	0.006	bdl[h]	0.059	0.241	n/a[i]	115.89	n/a
91w	bdl	bdl	0.090	0.023	0.043	0.004	bdl	0.038	0.176	n/a	4.49	n/a
92s	0.225	0.645	1.559	0.080	0.118	0.003	0.082	0.117	0.377	n/a	126.84	n/a
92w	0.115	0.075	0.199	0.014	0.036	0.011	0.015	0.055	0.630	n/a	5.85	n/a
93s	0.545	0.995	2.485	0.140	0.204	0.041	0.067	0.164	0.732	n/a	101.67	n/a
93w	0.099	0.096	0.226	0.011	0.049	0.035	0.005	0.157	0.524	n/a	3.51	n/a
94s	0.216	0.605	1.208	0.116	0.161	0.048	0.020	0.361	0.348	n/a	84.05	n/a
94w	0.148	0.246	0.572	0.023	0.086	0.026	0.004	0.168	0.360	n/a	4.56	n/a
96s	0.307	0.544	1.445	0.166	0.185	0.010	0.059	0.200	0.370	18.58	134.22	n/a
96w	0.108	0.155	0.306	0.025	0.111	0.014	0.001	0.070	1.367	12.52	6.73	n/a
97s	0.328	0.262	0.975	0.102	0.223	0.038	0.040	0.086	0.180	31.47	129.97	n/a
97w	0.285	0.672	1.535	0.034	0.058	0.006	0.035	0.037	1.297	8.46	2.25	n/a
98s	0.721	1.815	4.212	0.323	0.234	0.018	0.146	0.281	0.628	37.01	99.86	n/a
98w	0.237	0.258	0.629	0.045	0.091	0.013	0.070	0.080	1.183	11.44	3.18	n/a
99s	0.386	0.459	1.184	0.258	0.129	0.047	bdl	0.150	0.346	15.66	76.80	n/a
99w	0.176	0.350	0.861	0.031	0.041	0.012	0.026	0.021	0.316	7.14	3.09	n/a
04w	0.067	0.066	0.229	0.025	0.040	0.018	0.039	0.043	0.178	4.24	6.01	156.93
07s	0.229	0.720	1.792	0.181	0.137	0.007	0.086	0.357	0.276	21.20	100.44	179.59
07w	0.071	0.044	0.143	0.024	0.030	0.008	bdl	0.030	0.379	5.93	2.55	82.45
08s	0.349	1.364	3.272	0.123	0.078	0.014	0.054	0.017	0.105	15.94	65.15	160.97
08w	0.025	0.020	0.053	0.014	0.015	0.007	bdl	0.014	0.110	12.87	1.78	86.42
09w	0.057	0.042	0.103	0.007	0.007	0.005	bdl	0.008	0.563	7.10	2.94	75.33
11s	0.144	0.412	0.929	0.124	0.074	0.013	0.023	0.167	0.099	8.89	83.46	147.98
11w	0.037	0.064	0.154	0.019	0.017	0.005	0.003	0.024	0.082	1.23	0.61	99.68
12s	0.138	1.191	2.915	0.136	0.061	0.012	0.037	0.111	0.049	5.65	96.85	156.35
12w	0.021	0.012	0.050	0.022	0.011	0.006	bdl	0.010	0.116	1.21	0.84	82.13
14s	0.209	0.583	1.536	0.103	0.054	0.008	0.090	0.234	1.806	20.17	113.24	170.75
14w	0.097	0.273	0.578	0.032	0.014	0.005	0.063	0.014	0.281	12.72	3.76	99.35
15s	0.166	0.460	1.249	0.156	0.067	0.008	0.035	0.176	0.131	13.35	90.38	136.23

[a] 2-MG stands for 2-methylglyceric acid.

[b] Me.Thr. stands for 2-methylthreitol.

[c] Me.Ery. stands for 2-methylerythritol.

[d] MBTCA is 3-methyl-1,2,3-butanetricarboxylic acid.

[e] DTAA is diaterpenylic acid acetate.

[f] MSA stands for methanesulfonic acid.

[g] Calculated chlorophyll-*a* is retrieved from satellite data. See main text Section 3.4 and SI Section 7.1 for details.

[h] "bdl" indicates below detection limit.

[i] "n/a" indicates data unavailable in these years.

2. Mass of Collected PM_{10}

Gravimetric mass measurements were performed on the high-volume filters using a Sartorius Master Pro LA130S-F balance at relative humidity of approximately 50%. The resolution of the balance was 0.0001 g. Each 25 cm × 20 cm filter was weighed before and after sampling until three weights within 0.0010 g.

3. Details for Ion Chromatography (IC) Analysis

Anion and cation concentrations were determined with a Dionex ICS-3000 reagent free ion chromatograph. Anions were separated using a Dionex AS17c analytical column (2 × 250 mm), an AERS-500 suppressor and a gradient eluent of 0.75 mM to 35 mM potassium hydroxide. Cations were separated using a Dionex CS12a column (2 x 250 mm), a CERS-500 suppressor and an isocratic eluent of 20 mM methanesulfonic acid.

Seven calibration standards covering the sample concentration range are included in each analysis run for each analyte and a check standard is analyzed in each analysis run after the 7 calibration standards and then after every 20 samples.

The ion balance (IB) gives an indication of the aerosol chemistry data quality in that the total cation equivalents (positive charged ions) should equal the total anion equivalents (negative charged ions). The Global Atmospheric Watch Program (GAW) which is part of the World Meteorological Organisation (WMO) gives the IB equation and criteria for assessing valid data results in its technical report 160, “Manual for the GAW Precipitation Chemistry Programme”. Any samples that fail the IB criteria are reanalyzed.

The blank concentration is subtracted from each measurement. The blanks are also used to calculate the method detection limit (MDL). We followed the Standards Australia procedures which are those of the International Standard ISO 6879 Air quality – Performance characteristics and related concepts for air quality monitoring methods. Section 5.2.7 of the Standard states that a zero sample has a 5 % probability of causing a measured concentration above the detection limit, so that:

$$MDL = t_{0.95} \times s_{c(0)}$$

where:
$S_{c(0)}$ is the standard deviation of the blanks, and
$t_{0.95}$ is value of the 1-tailed t distribution for $P<0.05$ (i.e. the 95 % confidence limit).

4. Organic Carbon, Elemental Carbon, and Total Carbon Estimation

The organic carbon (OC), apparent elemental carbon (EC_a), and total carbon (TC) contents of the quartz filter samples were measured by the thermal-optical transmission (TOT) technique (Birch and Cary, 1996), using the procedure described in Schmid et al. (2001). This involved stepwise combustion of the aerosol carbon to form carbon dioxide (CO_2), which was then reduced to methane (CH_4) and detected by a flame ionization detector (FID). The effects of charring during pyrolysis of organic compounds in the early stages of the combustion (artifact EC_a) were corrected for by simultaneously monitoring light transmission through the filter and defining EC_a as the only carbon that evolved after the light transmittance through the sample had reached its original level. The uncertainty of the TC measurement was ±0.25 μg cm^{-2}. The average concentration of TC for the 32 valid Hi-Vol samples from 2002-2003 was 2.6 μg cm^{-2} (ranging between 0.5 and 11. 6 μg cm^{-2}). The limit of detection was 0.4 μg cm^{-2} (determined from the standard deviation of 10 field sample blanks multiplied by the appropriate t value according to ISO, 1994).

The organic matter (OM) in sample was estimated using the following equation:

$$OM = 1.8 \times OC + EC_a$$

Then the ratios of OM/PM_{10} were calculated and averaged to be 4.09% for summer (mid-December to mid-February) and 2.38% for winter (mid-June to mid-August), which were used to convert PM_{10} to OA mass when evaluating measured iSOA and mSOA as a fraction of OA from 2004-2015.

Table S3. Summed iSOA (∑iSOA), summed mSOA (∑mSOA), oxalic acid, and MSA estimated as a fraction of organic aerosol mass from 2004-2015.

	∑iSOA	**∑mSOA**	**PM_{10}**	**iSOA**	**mSOA**	**[iSOA+mSOA]**	**[iSOA+mSOA+ Oxalic acid+MSA]**
	ng/m^3	ng/m^3	µg/m^3	to OA	to OA	to OA	to OA
Summer							
2007	2.60	0.77	16.1	0.394%	0.117%	0.511%	18.955%
2008	4.73	0.29	18.9	0.612%	0.037%	0.649%	11.145%
2011	1.48	0.40	19.8	0.183%	0.049%	0.232%	11.613%
2012	4.24	0.36	15.9	0.652%	0.055%	0.707%	16.454%
2014	2.33	0.49	18.1	0.315%	0.066%	0.381%	18.416%
2015	1.87	0.44	19.6	0.233%	0.055%	0.288%	13.209%
Average	*2.88*	*0.46*	*18.1*	*0.398%*	*0.063%*	*0.462%*	14.767%
Winter							
2004	0.36	0.16	18.2	0.084%	0.038%	0.122%	2.493%
2007	0.24	0.09	15.0	0.068%	0.026%	0.094%	2.466%
2008	0.09	0.05	17.8	0.022%	0.012%	0.034%	3.492%
2009	0.20	0.03	24.3	0.035%	0.005%	0.040%	1.775%
2011	0.26	0.07	22.1	0.049%	0.013%	0.062%	0.412%
2012	0.08	0.05	19.6	0.018%	0.010%	0.028%	0.467%
2014	0.95	0.13	16.5	0.241%	0.033%	0.274%	4.461%
Average	*0.31*	*0.08*	*19.1*	*0.074%*	*0.019%*	*0.093%*	2.101%

5. Surface Ozone at Cape Grim

Ozone measurements were made at 10 m above the laboratory roof through the main station stainless steel inlet, using the absolute ozone monitor designated as TECO-2 (Model 49, Thermo Instruments, USA) for the whole period, and a newer ozone monitor (Model 49C, Thermo Instruments, USA) designated TECO-3. The absolute accuracy of the ozone monitor was checked regularly against a Thermoelectron Model 49PS ozone calibrator. Automatic calibrations were performed approximately every two weeks.

The provisional surface ozone monthly mean concentrations for all conditions for January 1990 to December 2015 are presented in the figure below.

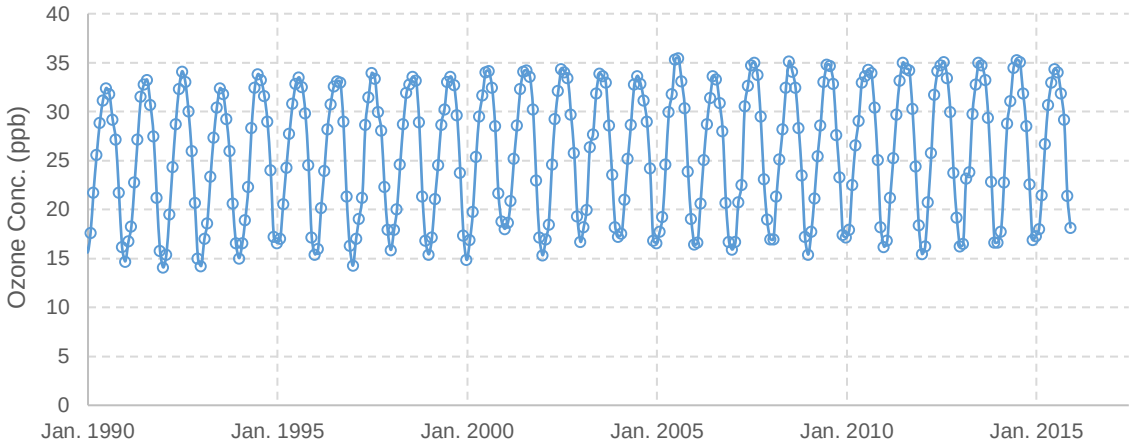

Figure S2. Monthly average surface ozone concentrations (nmol/mol, or ppb) at Cape Grim from January 1990 to December 2015.

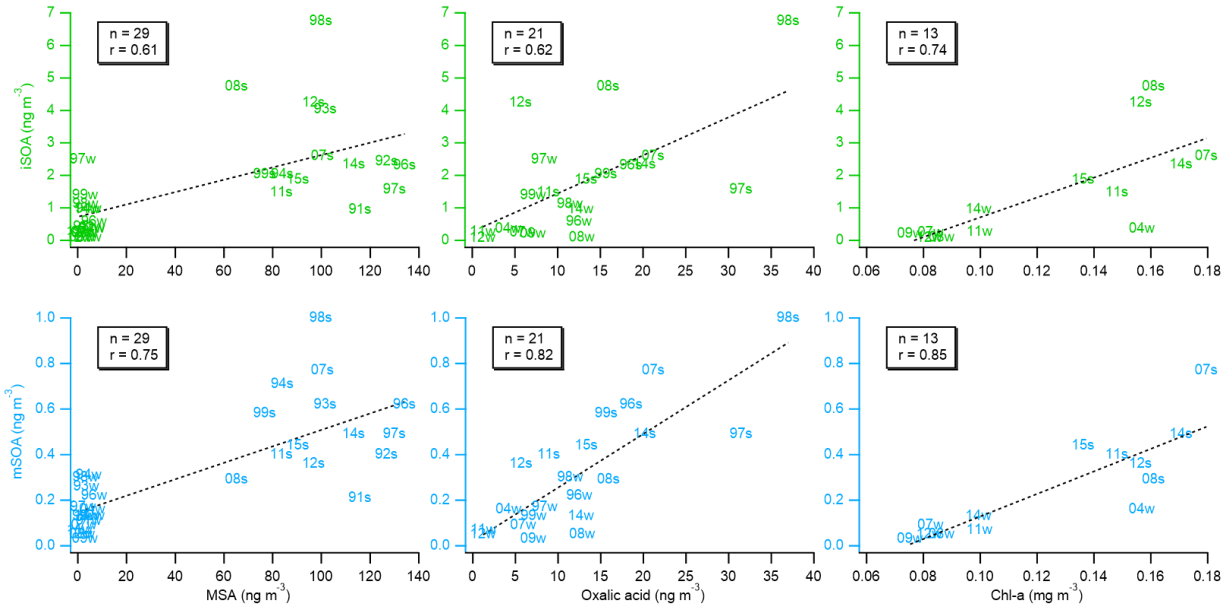

Figure S3. Correlations of the summed concentration of the three isoprene-derived SOA tracers (iSOA, green) or the five monoterpene-derived SOA tracers (mSOA, blue) with MSA/Oxalic acid/Chl-*a* (from left to right). Years and austral seasons (n=13-29) with available data are labeled in each sub-panel.

6. Chlorophyll-*a* (Chl-*a*) Data Usage and Description

6.1 Satellite measurement by remote sensing

Global Chl-*a* concentrations were derived from ocean color every day with a spatial resolution of 1-km, measured by the Moderate Resolution Imaging Spectroradiometer (MODIS) instrument aboard NASA's Terra and Aqua satellites over the entire planet. The MODIS instrument offers an estimate of the near-surface concentration of chlorophyll calculated using an empirical relationship derived from *in situ* measurements of chlorophyll and remote sensing reflectances in the blue-to-green region of the visible spectrum. Since the values retrieved from the formatted files were scaled and resampled for visualization purposes by NASA Earth Observation (NEO) and not suggested for rigorous scientific examination, we only adapted them for trend detection and simply correlation. See https://neo.sci.gsfc.nasa.gov/view.php?datasetId=MY1DMM_CHLORA for more information for the related websites, description, algorithm, and data file formats.

Total monthly Chl-*a* contents in this study were averaged and integrated from the oceanic region over [30-60°S, 90-150°E]. This region is determined based on previous back trajectory studies for suitable timescale of SOA formation and transport (approximately 4 days for correspoding air parcels to reach Cape Grim, roughly one unit of Rn's half-life = 3.84 days, reported in 2009-2010 Baseline Report, Page 33-38). On the temporal scale, the selected months also cover the same filter sampling period as precise as possible.

It is worth mentioning that the values of high-resolution Chl-*a* concentrations retrieved online in the .csv files have been scaled and resampled, and thus are basically used to examine their seasonal and temporal variation rather than accuracy.

6.2 *In situ* measurements

Chl-*a* concentration was measured in the local seawater samples obtained approximately 9 km offshore from Couta Rocks, 50 km south of Cape Grim, in the selected years from 1995-2007. One-liter ocean water samples were filtered onto 42-mm diameter glass fibre filters followed by extraction with methanol, and then determined by a published acidification method (Holm-Hansen and Riemann, 1978) using a digital fluorometer (Turner Instrument, 10AU).

When both types of Chl-*a* data were available during 2003-2006, we compare the *in situ* measurements at the offshore site made from the sea surface (depth = 0 m, data retrieved from Baseline Reports 2003-2004 (page 81-86) and 2005-2006 (page 80-84), by Cainey et al. at http://www.bom.gov.au/inside/cgbaps/baseline.shtml), and the satellite measurements at the same location and time period (Hi-Res of 0.1×0.1 degree, 8-day average), as shown in Figure S4. Although the location and time coverage between the two types of measurements were not exactly the same, the comparison reveals similar trends of Chl-*a* concentrations over the years. Therefore, it would be reliable to correlate the whole-time satellite measured Chl-*a* with the iSOA concentrations at Cape Grim.

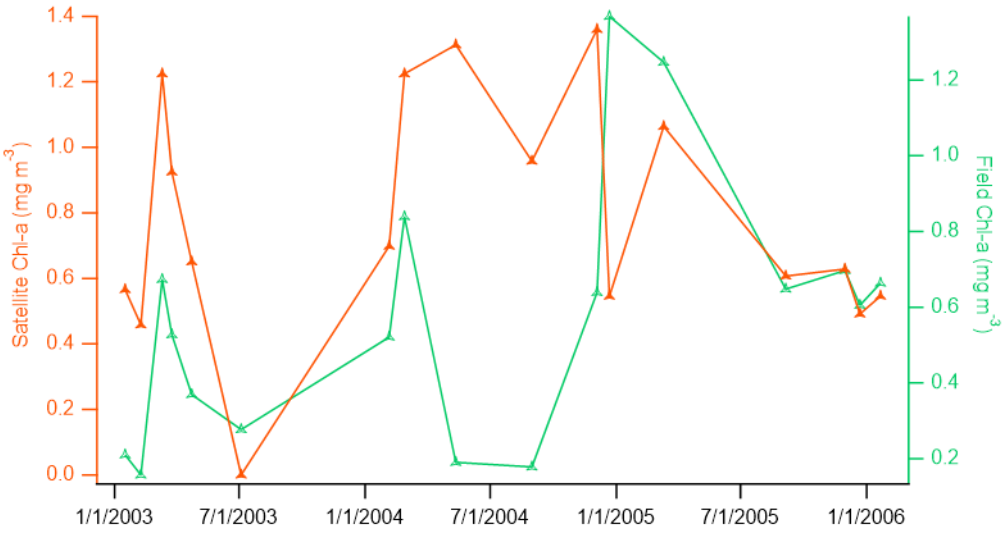

Figure S4. Comparison of satellite and *in situ* measurements of Chl-*a* during 2003-2006.

Table S4. List of molecular formulas of N-containing and other species tentatively identified by UPLC/ESI-HR-QTOFMS operated in negative (-) and positive (+) ion modes.

ESI (-)				ESI (+)
CHNO3	*CHNO4*	*Other N-Containing*	*CHO(S)*	*N-Containing*
C7H13NO3	C9H17NO4	C5H6N4O5	C4H6O4	C4H6N2
C8H15NO3	C10H19NO4	C7H5NO3S2	C5H8O4	C7H11N7O
C9H17NO3	C11H21NO4	C7H15NO5S	C6H10O4	C8H19N
C10H19NO3	C12H23NO4	C11H12N4O2	C7H10O4	C9H17NO2
C11H21NO3	C13H25NO4	C8H17NO5S	C8H6O4	C9H17NO3
C12H23NO3		C9H19NO5S	C8H12O4	C11H19N7O
C13H25NO3		C12H23NO5	C8H14O4	C12H23N7O
C14H21NO3		C10H21NO5S	C9H18O3	C13H25NO2
C14H27NO3		C11H23NO5S	C8H10O3S	C13H25NO3
C15H29NO3		C19H9ClN4O	C8H12O5	C14H16N6O2
		C17H32N2O5	C9H16O4	C18H28N6O6
		C17H30N2O3	C10H18O4	C27H41N
		C18H34N2O5	C11H20O4	
		C19H34N2O5	C12H20O4	
		C19H36N2O5	C12H20O5	
		C20H38N2O5	C9H18O6S	
		C20H36N2O3	C13H22O5	
		C16H4N2O7S2	C18H30S2	
		C21H40N2O5	C17H26O5	
		C16H28N2O10	C17H28O5	
		C12H6N4O11S	C20H14O9	
		C14H2N4O12S		
		C15H6N4O7S3		
		C12H6N4O12S2		
		C18H4N4O10S2		
		C14H4N4O15S		

Reference:

Annual or biennial Cape Grim Baseline Reports from since 1976: http://www.bom.gov.au/inside/cgbaps/baseline.shtml.

Birch, M. E. and Cary, R. A. Elemental Carbon-Based Method for Monitoring Occupational Exposures to Particulate Diesel Exhaust, *Aerosol Science and Technology*. **1996**, 25 (3), 221-241.

Schmid, H.; Laskus, L.; Abraham, H. J.; Baltensperger, U.; Lavanchy, V.; Bizjak, M.; Burba, P.; Cachier, H.; Crow, D.; Chow, J.; Even, T. G. A.; Brink, H. M.; Giesen, K-P.; Hitzenberger, R.; Hueglin, C.; Maenhaut, W.; Pio, C.; Carvalho, A.; Putaud, J-P.; Toom-Sauntry, D.; Puxbaum, H. Results of the "carbon conference" international aerosol carbon round robin test stage I, *Atmospheric Environment*, **2001**, 35 (12), 2111-2121.

Holm-Hansenm, O. and Riemann, B. Chlorophyll-*a* Determination: Improvements in Methodology, *Oikos*. **1978**, 30 (3), 438-447.