Supplement to "Key chemical NOx sink uncertainties and how they influence top-down emissions of nitrogen oxides"

T. Stavrakou[1], J.-F. Müller[1], K. F. Boersma[2,3], R. J. van der A[2], J. Kurokawa[4], T. Ohara[5], and Q. Zhang[6]

[1]Belgian Institute for Space Aeronomy, Avenue Circulaire 3, 1180, Brussels, Belgium
[2]Royal Netherlands Meteorological Institute (KNMI), Wilhelminalaan 10, De Bilt, The Netherlands
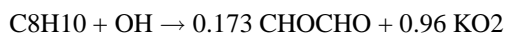[3]Eindhoven University of Technology, Fluid Dynamics Lab, Eindhoven, The Netherlands
[4]Asia Center for Air Pollution Research, Niigata, Japan
[5]National Institute for Environmental Studies, Tsukuba, Ibaraki, Japan
[6]Center for Earth System Science, Tsinghua University, Beijing, China

1 NMVOC chemistry

The model includes 22 precursor NMVOCs. The degradation mechanism for most model compounds (ethene, propene, α-pinene, acetylene, ethane, propane, formaldehyde, acetaldehyde, glyoxal, methylglyoxal, acetone, methyl ethylketone, biacetyl, acetic acid, methanol, glycolaldehyde and a lumped species accounting for other NMVOCs) is adapted from Stavrakou et al. (2009), with only minor updates. The isoprene chemical mechanism is adapted from the Mainz Isoprene Mechanism version 2 (MIM2) (Taraborrelli et al., 2009), as presented by Stavrakou et al. (2010). The chemical production and loss of formic acid are implemented as in Stavrakou et al. (2012). The chemical degradation of aromatics (Stavrakou et al., 2009) is adjusted to reproduce the yields of formaldehyde and glyoxal estimated from the Master Chemical Mechanism (MCMv3.1) (Bloss et al., 2005). More specifically, the reaction of benzene, toluene and xylenes with OH is written as

$$C6H6 + OH \rightarrow 0.333\ CHOCHO + 0.333\ KO2$$
$$C7H8 + OH \rightarrow 0.223\ CHOCHO + 0.694\ KO2$$
$$C8H10 + OH \rightarrow 0.173\ CHOCHO + 0.96\ KO2$$

where KO2 denotes the lumped peroxy radical formed from the reaction of methylethylketone with OH.

2 Aerosol model description

2.1 Organic Aerosol

The model includes four sources of organic aerosol:

- Direct emission of Primary Organic Aerosol (POA) due to anthropogenic sources (Bond et al., 2004) (9.3

TgC/year or 16.7 Tg/year) and biomass burning (van der Werf et al., 2010) (14.1 TgC/year or 25.3 Tg/year). The seasonal variation of POA emissions due to heating is based on a heating degree days approach based on the study of Cabada et al. (2002). POA has hydrophilic and hydrophobic components. Conversion of hydrophobic to hydrophilic POA due to aging is represented by an e-folding time of 1 day (Martin et al., 2003). The OA/OC ratio for POA is taken equal to 1.8.

- Reversible Secondary Organic Aerosol (SOA) formation due to the partitioning of semi-volatile organic compounds. The approach of the two-product model is adopted for SOA due to isoprene, terpenes and aromatics. The product yields and partitioning parameters for aromatics Henze et al. (2008), isoprene Henze et al. (2006) and sesquiterpenes Chang et al. (2010) are based on laboratory data. For monoterpenes, we adopt the ten-product model of Ceulemans et al. (2011), with parameters derived from simulations using a detailed oxidation and SOA formation model (BOREAM). The model accounts for a dependence of the yields on the NOx regime and on the nature of the primary oxidant (O_3, OH or NO_3). The partitioning coefficients account for water uptake (Ceulemans et al., 2011). In this study, the yields of semi-volatile compounds generated in the oxidation of monoterpenes at low-NOx were reduced by a factor of 5 in order to improve the agreement with observations over biogenically-influenced areas (i.e., the Eastern US during the summer and tropical ecosystems during the wet season).

- Irreversible SOA formation due to the production of essentially non-volatile compounds due to the uptake of glyoxal and methylglyoxal by cloud droplets and sulfate aerosols. It is included as an irreversible process (Fu et al., 2008; Stavrakou et al., 2009) and assumed to be very fast in liquid clouds. On aqueous aerosols,

Correspondence to: T. Stavrakou (jenny@aeronomie.be)

a constant reactive uptake coefficient ($2.9 \cdot 10^{-3}$) is assumed.

- An additional organic aerosol source is included, as explained in the main body of this article, taken equal to three times the POA source of (Bond et al., 2004) over the U.S., and five times this source of the rest of the world. It has no seasonal variation. The emitted OA is assumed to behave like POA, notwithstanding the fact that it might be largely constituted of SOA.

The size distribution of carbonaceous aerosols is assumed log-normal, with an effective radius (for calculating aerosol surface are) taken equal to 70 nm (Köpke et al., 1997; Martin et al., 2003). The molecular weight of POA is taken equal to 162, the value for levoglucosan, a typical component of biomass burning aerosols (Puxbaum et al., 2007). The molecular weight of SOA is model-calculated, depending on the respective abundances of condensable products. Water uptake is calculated using water activity coefficients estimated for α-pinene SOA using a detailed model (Ceulemans et al., 2011). The hygroscopic growth factor is equal to 1.17 at 90% RH (assuming MW=162), in the range of reported values, e.g. 1.12 for biomass burning aerosol and 1.21 for wet season organic aerosol over Amazonia (Rissler et al., 2006). Note that hydrophilic carbonaceous aerosols are assumed to be always liquid.

2.2 Black carbon

The model includes emissions of black carbon (BC) due to anthropogenic sources (Bond et al., 2004) (5 TgC/year) and biomass burning (van der Werf et al., 2010) (2 TgC/year). Like POA, BC has hydrophilic and hydrophobic components. Hydrophobic BC is converted to hydrophilic BC with a lifetime of 1 day.

2.3 Inorganic aerosol

The EQSAM model (Metzger et al., 2002) is used for sulfate/ammonium/nitrate/water aerosols. Composition- and temperature-dependent deliquescence relative humidities (DRH) are calculated according to ISORROPIA (Nenes et al., 1998). Below the DRH, and above the crystallisation relative humidity (CRH) (Carrico et al., 2003b), the aerosol state is dependent on its history through the hysteresis effect (Tang et al., 1995); here the aerosol is assumed to be solid only if relative humidity remained below the DRH during the last 24 hours. The aerosol physical state is highly relevant to heterogeneous chemistry: heterogeneous uptake on solid particles is very slow in the case of N_2O_5 (Davis et al., 2008) and HO_2 (Taketani et al., 2008).

Dust and sea-salt aerosols are not considered in the model. Anthropogenic SOx (59 TgS/year) and NH_3 (44 TgN/year) emissions include anthropogenic emissions obtained from EDGAR 3.2 FT2000

(http://themasites.pbl.nl/tridion/en/themasites/edgar/), overwritten by REASv1 (Ohara et al., 2007) over Asia, and EMEP over Europe (http://www.ceip.at/). Over the US, the EDGAR SO_2 emission is scaled to the National Emission Inventory (NEI, http://www.epa.gov/ttn/chief/eiinformation.html) total for each year. Biomass burning emissions (3.2 TgS/year and 3.2 $TgNH_3$–N) are obtained from van der Werf et al. (2010). The model also includes SO_2 emissions from degassing volcanoes (4.9 TgS/year) from GEIA (http://www.geiacenter.org/presentData/volcano.html) as well as oceanic emissions of NH_3 from the EDGARv2 inventory. The model accounts for the SO_2 production due to the photochemical degradation of dimetyl sulfide (DMS) emitted by the oceans (AEROCOM intercomparison, ftp://ftp-projects.zmaw.de/aerocom/emissions/) and the biosphere (Pham et al., 1995) as well as of reduced sulfur compounds, CS_2 and H_2S (Pham et al., 1995).

SO_2 is converted into particulate sulfate by either (1) gas-phase reaction with OH resulting in sulfuric acid assumed to either nucleate or condense on existing aerosols, and (2) aqueous-phase reactions of S(IV) with H_2O_2 and O_3 in liquid clouds, following e.g. Rodriguez and Dabdub (2004).

The effective radius of dry sulfate/ammonium/nitrate aerosol is taken equal to 130 nm, in the range of reported values (Köpke et al., 1997; Jones et al., 2001).

3 Comparison with observations

3.1 Ground-based measurements

The Tables 1-2 list the ground-based measurements of organic aerosol used for comparison with the model. Although the observations are obtained in different years, they are compared with model simulations conducted for year 2007. In addition, we also used the OC and SO_4 measurements in PM2.5 of the IMPROVE (Interagency Monitoring for Protection Of Visual Environments) network for the year 2007 (http://vista.cira.colostate.edu/improve/).

The results of this comparison for OC are summarized in Table 3 and in Fig. 1-2. The model results are given both with and without the *ad hoc* adjustment of OA sources described above. Thanks to this adjustment, the model bias is strongly reduced over most regions. Very large understimations (more than a factor of 5) are still found at two Portuguese sites (Aveiro and Braganza) during winter, suggesting the existence of an intense source due to biofuel use (Puxbaum et al., 2007). The model also generally underestimates OC at tropical sites impacted by intense vegetation fires (Morogoro, Ivory Coast and Central Africa) during the dry season; however it is possible that the fires were less intense in these regions in 2007 than during the years of the observations. In contrast, the model often slightly overestimates the observations at tropical sites during the wet season

(Balbina and Rondonia in South America, Ivory Coast during the wet season), confirming that the biosphere is only a moderate source of organic aerosol in absence of a strong anthropogenic influence. The seasonal variation of OC concentrations is generally well reproduced by the model over the United States (Fig. 2) and over other polluted areas (3). Over the Southeast U.S., the overestimated seasonal amplitude suggests again an overestimated role biogenic SOA in the model. Overall, the agreement is very satisfactory, even though large discrepancies remain at many individual sites.

Fig. 4-5 shows the comparison of modelled SO_4 with IMPROVE observations in 2007. Although a generally positive model bias is found (except over the Southeast U.S.), the seasonal variation of OC concentration is fairly well reproduced, and most annually averaged concentrations are reproduced to within a factor of 2.

3.2 Aircraft measurements

We use OC measurements from campaigns over the New England in the Eastern U.S. during summer (NEAQS/ITCT) and over China Sea in spring (ACE-Asia). OC was measured by Fourier Transform Infrared Spectroscopy (FTIR) filter measurements during ACE-Asia (Maria et al., 2003), whereas water-soluble organic carbon (WSOC) was measured during NEAQS/ITCT using the Particle-Into-Liquid Sampler (PILS) technique (Sullivan et al., 2006). These measurements represent sub-micron aerosol. Both campaigns were heavily impacted by anthropogenic emissions. The model reproduces fairly well both the average boundary layer OC concentrations and the vertical gradient between the PBL and the lowermost 3 kms above the surface (Fig. 6). The unadjusted model (results not shown) underestimates OC over China sea by a factor of about 3, consistent with results obtained using GEOS-Chem (Heald et al., 2011).

Table 1. In situ measurements of organic carbon (OC) concentrations at European sites expressed in ng m^{-3}. The superscript ^{f} means fine particles with diameter of less than 3 μm.

Location	Coordinates	Period	OC	Reference
Aveiro, Portugal	40.6 N 351.4 E	Oct-Mar 2003/5	7500^f	Pio et al. (2007)
Aveiro, Portugal	40.6 N 351.4 E	Apr-Sep 2003/5	3100^f	id.
Puy de Dome, France	45.8 N 3 E	Apr-Sep 2003/5	2400	id.
Moitinhos, Portugal	40.6 N 351.4 E	Jul 2004	2500^f	Duarte et al. (2008)
Anadia, Portugal	40.5 N 351.6 E	Aug 1996	3506	Castro et al. (1999)
Areao, Portugal	40.5 N 351.2 E	Nov-Aug 1993/4	2730^f	Pio et al. (1996)
Mt. Cimone, Italy	44.2 N 10.7 E	Jul-Sep 2004	1490^f	Marenco et al. (2006)
Birmingham, UK	52.5 N 358.1 E	Nov-May 2005/6	3200^f	Harrison and Yin (2008)
Melpitz, Germany	51.5 N 13 E	Jul-Sep 2004	1840^f	Spindler et al. (2005)
id.	id.	Oct-Dec 2004	2840^f	id.
id.	id.	Jan-Dec 2004/6	1970^f	id.
Schauinsland, Germany	47.9 N 7.9 E	Apr-Sep 2003/5	3400	id.
K-puszta, Hungary	46.9 N 19.5 E	Oct-Mar 2003/5	7400^f	id.
id.	id.	Apr-Sep 2003/5	4900^f	id.
Montseny, Spain	41.7 N 2.3 E	Jan-Dec 2002/7	2100^f	Querol et al. (2009)
id.	id.	Nov-Apr 2002/7	1775^f	id.
id.	id.	May-Oct 2002/7	2425^f	id.
Finokalia, Greece	35.3 N 25.7 E	Jan-Dec 2004/6	1800^f	Querol et al. (2009)
id.	id.	Nov-Apr 2004/6	1720^f	id.
id.	id.	May-Oct 2004/6	1880^f	id.
French Alps	45.5 N 7 E	Aug 2000	3400	Aymoz et al. (2004)
Jülich, Germany	50.9 N 6.4 E	Jul 2003	4600^f	Kourtchev et al. (2008)
Aspvreten, Sweden	58.8 N 17.4 E	Jun-Jul 1996	2200	Zappoli et al. (1999)
Preila, Lithuania	55.4 N 21.1 E	Jun 2006	1290	Milukaite et al. (2007)
Braganza, Portugal	41.8 N 353.8 E	Jul-Jun 2002/3	4100	Yttri et al. (2007)
id.	id.	Apr-Sep 2002/3	2740	id.
id.	id.	Oct-Mar 2002/3	5570	id.
Illmitz, Austria	47.8 N 16.8 E	Jul-Jun 2002/3	5570	id.
id.	id.	Apr-Sep 2002/3	4820	id.
id.	id.	Oct-Mar 2002/3	6280	id.
Stara Lesna, Slovakia	49.1 N 20.2 E	Jul-Jun 2002/3	4320	id.
id.	id.	Apr-Sep 2002/3	4960	id.
id.	id.	Oct-Mar 2002/3	3680	id.
Kosetice, Czech Rep.	49.6 N 15.1 E	Jul-Jun 2002/3	4580	id.
id.	id.	Apr-Sep 2002/3	4080	id.
id.	id.	Oct-Mar 2002/3	4860	id.
Langenbrugge, Germany	52.8 N 10.8 E	Jul-Jun 2002/3	4300	id.
id.	id.	Apr-Sep 2002/3	3630	id.
id.	id.	Oct-Mar 2002/3	4980	id.
Kollumerwaard, Netherlands	53.3 N 6.3 E	Jul-Jun 2002/3	2590	id.
id.	id.	Apr-Sep 2002/3	2330	id.
id.	id.	Oct-Mar 2002/3	2850	id.
Mace Head, Ireland	53.3 N 350.8 E	Jul-Jun 2002/3	1200	id.
id.	id.	Apr-Sep 2002/3	1030	id.
id.	id.	Oct-Mar 2002/3	1400	id.
Penicuik, Scotland	55.8 N 356.8 E	Jul-Jun 2002/3	1530	id.
id.	id.	Apr-Sep 2002/3	1370	id.
id.	id.	Oct-Mar 2002/3	1680	id.
Birkenes, Norway	58.3 N 8.3 E	Jul-Jun 2002/3	1570	id.
id.	id.	Apr-Sep 2002/3	1920	id.
id.	id.	Oct-Mar 2002/3	1250	id.
Aspvreten, Sweden	58.8 N 17.4 E	Jul-Jun 2002/3	2120	id.
id.	id.	Apr-Sep 2002/3	2560	id.
id.	id.	Oct-Mar 2002/3	1690	id.
Virolahti, Finland	60.5 N 27.6 E	Jul-Jun 2002/3	2080	id.
id.	id.	Apr-Sep 2002/3	2530	id.
id.	id.	Oct-Mar 2002/3	1650	id.
Hyytiala, Finland	61.8 N 24.3 E	May 2004	814^f	Saarikoski et al. (2005)
id.	id.	Jul-Aug 2005	2200^f	Kourtchev et al. (2008a)
id.	id.	Aug 2007	2700^f	Maenhaut et al. (2009)
K-puszta, Hungary	46.9 N 19.5 E	Jun 2003	4000^f	Kourtchev et al. (2009)
id.	id.	Jun 2006	3230^f	Maenhaut et al. (2009)

Table 2. Same as Table 1 for sites in America, Asia and Africa. Notes: [a]=http://data.eol.ucar.edu/codiac/dss/id=69.1530 ; [b]=http://data.eol.ucar.edu/datafile/nph-get/69.091/aerosol_OC-EC_v1Readme.txt

Location	Coordinates	Period	OC	Reference
Rondônia, Brazil	10.8 S 297.6 E	Sep 2002 (dry)	26900[f]	Decesari et al. (2006)
id.	id.	Oct 2002 (trans)	8000[f]	id.
id.	id.	Nov 2002 (wet)	1420[f]	id.
Balbina, Brazil	1.9 S 300.5 E	Jul 2001 (wet)	1190[f]	Claeys et al. (2004)
Central Africa	2-12 N 17-19 E	Nov 1996 (dry)	13021	Ruellan et al. (1999)
Ivory Coast	6.2 N 355 E	Dec-Feb 1981/2 (dry)	11500	Cachier et al. (1989)
id.	id.	Sep-Nov 1982	1200	Cachier et al. (1989)
Djougou, Benin	9.7 N 1.9 E	Jan-Dec 2006	3100[f]	Liousse et al. (2008)
Meru, Kenya	0.1 N 37.7 E	May-Jun 1999	6000[f]	Gatari and Boman (2008)
Nanyuki, Kenya	0 N, 37.1 E	May-Jun 1999	13000[f]	id.
Morogoro, Tanzania	7 S 3 7E	May-Jun 2005 (wet)	6700	Mkoma et al. (2008)
id.	id.	Jul-Sep 2005 (dry)	11000	id.
id.	id.	Mar-May 2006 (wet)	2260[f]	id.
Skukuza, S.Africa	25 S 31.5 E	Aug-Sep 2001 (dry)	6000[f]	Maenhaut and Claeys (2007)
Nylsvley, S.Africa	24.7 S 28.4 E	May 1997	8200	Puxbaum et al. (2000)
Lake Daihai, China	40.7 N 112.5 E	Jan-Dec 2005/7	11800[f]	Han et al. (2008)
Dunhuang, China	40.2 N 94.5 E	Mar-Apr 2001	500[f]	[a]
Inland China 0.5-3km	30-32 N 104-109 E	Aug-Sep 2003	24300[f]	Wang et al. (2007)
Changdao Island, China	37.9 N 120.7 E	Mar-Apr 2003	7830[f]	Feng et al. (2007)
id.	id.	Aug 2003	3530[f]	id.
id.	id.	Oct-Nov 2003	7260[f]	id.
id.	id.	Dec-Jan 2003/4	14790[f]	id.
Japan Sea Ron Brown	34-40 N 130-136 E	Apr 2001	2400[f]	[b]
Cheju Island, Korea	33.3 N 127.2 E	Dec-Mar 1994/7	3100[f]	Kim et al. (2000)
id.	id.	Jul-Aug 1994	3734[f]	id.
Chao-Chou, Taiwan	22.2 N 121 E	Mar-Oct 2005	10200[f]	Wang et al. (2008)
Lu-Lin obs., Taiwan	23.5 N 120.8 E	Dec 2003	2000[f]	Huang et al. (2005)
id.	id.	Mar 2004	6000[f]	id.
Meinung, Taiwan	22.9 N 120.5 E	Dec-Jan 2002/3	9100[f]	Tsai and Chen (2006)
Hok Tsui, Hong Kong	23.5 N 120.8 E	Jan-Feb 2001	6300[f]	Cao et al. (2003)
id.	id.	Jun-Jul 2002	3400[f]	id.
id.	22.1 N 114.2 E	Aug-Sep 2004	5600[f]	Duan et al. (2007)
id.	id.	Feb 2005	5900[f]	id.
id.	id.	Dec-Feb 2001	5500[f]	Ho et al. (2006)
id.	id.	Jun-Aug 2001	1500[f]	id.
Nagarkot, Nepal (ramp-up)	27.7 N 85.5 E	Oct-Jan 1999/00	6250[f]	Carrico et al. (2003a)
id., peak	id.	Feb-May 1999/00	14400[f]	id.
id., monsoon	id.	Jun-Sep 1999/00	2000[f]	id.
Manora Peak, India	29.4 N 79.5 E	Jan-Dec 2005/6	8700	Ram et al. (2008)
Mt. Abu, India	24.6 N 72.7 E	Jan-Dec 2005/6	3700	id.
Delhi, India	28.1 N 77 E	Jan-Feb 1998	22050	Sarkar et al. (2001)
id.	id.	Sep 1998	2590	id.
Indian Ocean	15 N 70 E	Feb-Mar 1998	9000	id.
id.	11 N 68 E	Feb-Mar 1998	7200	id.
id.	5 N 70 E	Feb-Mar 1998	3200	id.
id.	10 S 65 E	Feb-Mar 1998	1700	id.
Altzomoni, Mexico	19.1 N 261.4 E	Mar 2006	3300	Gilardoni et al. (2009)

Table 3. Averaged organic carbon concentration (μg m^{-3}) measured at ground stations, and corresponding values calculated using IMAGESv2 with either unadjusted (U) or adjusted (A) organic aerosol sources. n is the number of stations considered in the average.

|	n	obs.	model (U)	model (A)
U.S. (IMPROVE)	164	1.1	0.7	1.1
Europe (EMEP)	33	3.1	0.7	2.5
Europe (excl. EMEP)	29	2.9	0.8	2.4
China/Korea	20	6.7	1.6	5.2
India/Nepal	11	7.9	2.6	8.9
Africa and South America	15	8.0	4.5	5.3
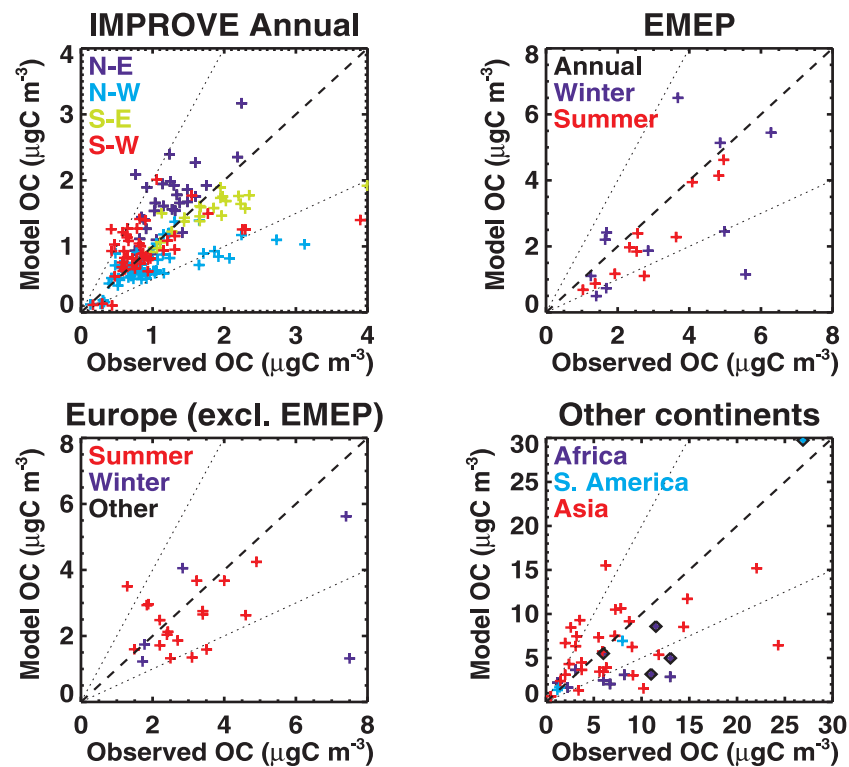
Fig. 1. Averaged organic carbon (OC) concentrations (μg m^{-3}) measured vs. model-calculated (using adjusted OA sources) over the United States, Europe (EMEP and non-EMEP stations) and over other continents. The sectors considered for the IMPROVE plot are N-E (lat>38 N, lon<90 W), N-W (lat>38 N, lon>90 W), S-E (lat>38 N, lon<100 W), S-W (lat<38 N, lon<100 W). Diamond-shaped symbols denote measurements strongly influenced by biomass burning (over Asia, Africa and South America).
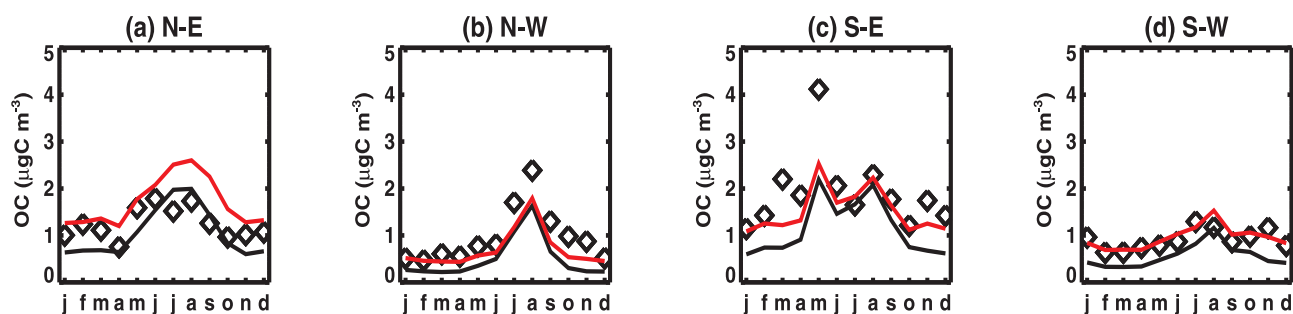
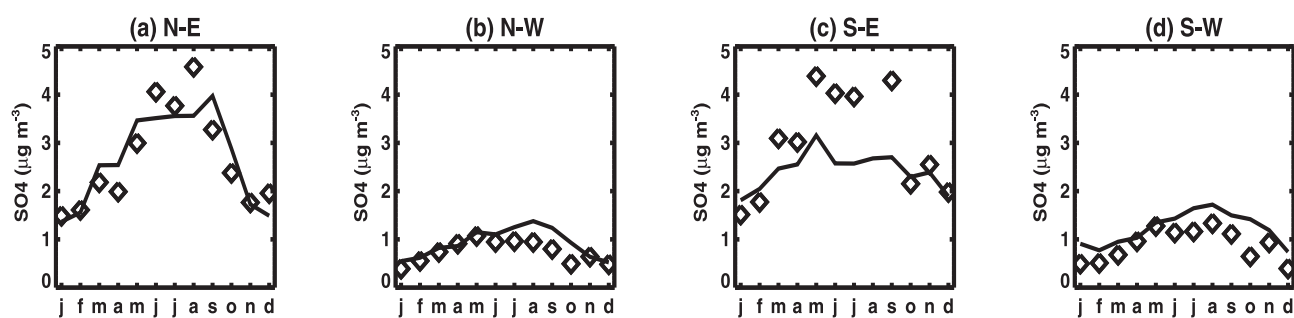**Fig. 2.** Seasonal variation of organic carbon concentrations (in μg m^{-3}) observed at IMPROVE sites in 2007, averaged over 4 sectors (defined as in Fig. 1), and comparison with values calculated using IMAGESv2 with either unadjusted (black line) or adjusted OA sources (red line).
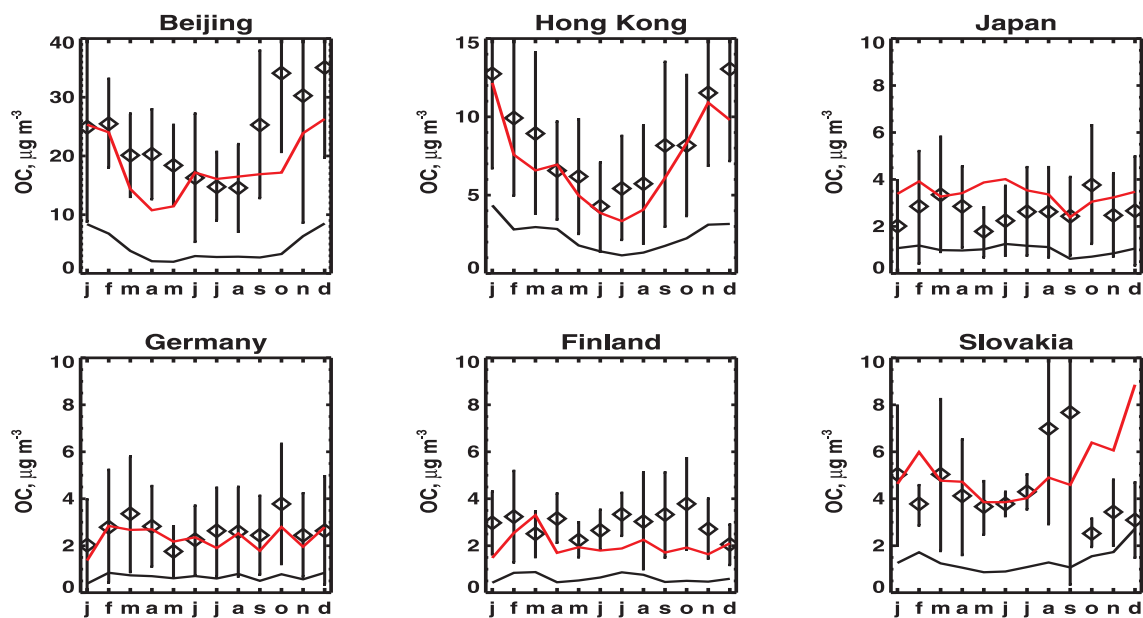

Fig. 3. Seasonal variation of organic carbon concentrations (μg m^{-3}) measured in the Beijing area, Hong Kong, Japan, Germany, Finland, and Slovenia, as reported by Bahadur et al. (2009), and comparison with values calculated using IMAGESv2 with either unadjusted (black line) or adjusted OA sources (red line).
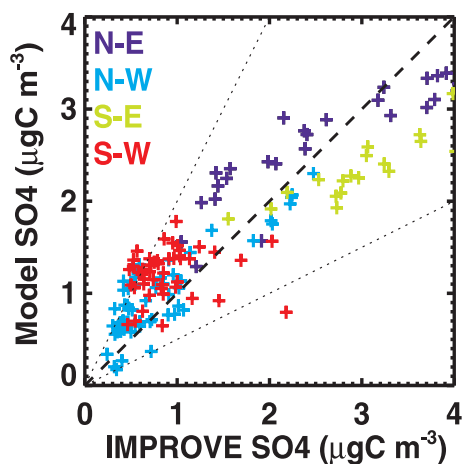
Fig. 4. Annually averaged sulfate concentrations (μg m^{-3}) in 2007 measured vs. model-calculated at IMPROVE sites. The colors define sectors defined as in Fig. 1.


Fig. 5. Seasonal variation of sulfate concentrations (μg m^{-3}) observed at IMPROVE sites in 2007, averaged over 4 sectors (defined as in Fig. 1), and comparison with modelles values.
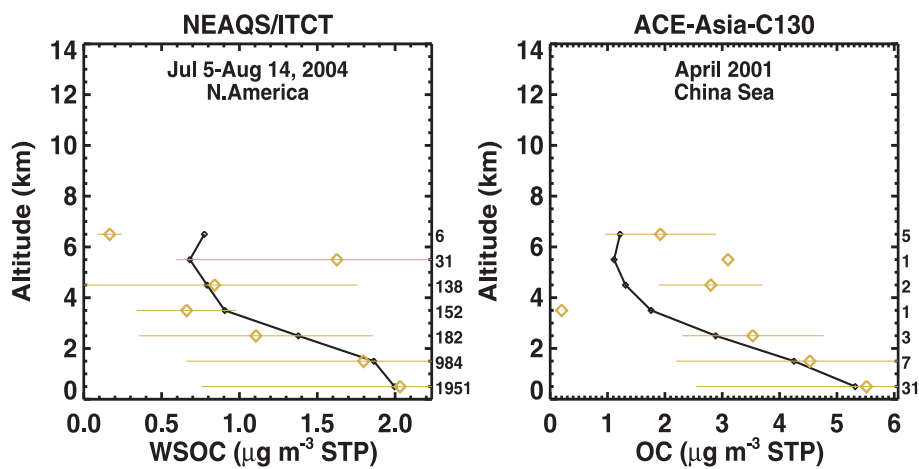
Fig. 6. Water-soluble organic carbon (WSOC) concentrations measured during the campaigns NEAQS/ITCT and ACE-Asia over China Sea (longitude < 130 E), and corresponding values calculated with IMAGESv2 using adjusted OA sources. The number of measurements in each 1-km bin is indicated on the right side of each plot. STP stands for standard temperature and pressure (i.e. 298 K and 1 atm).
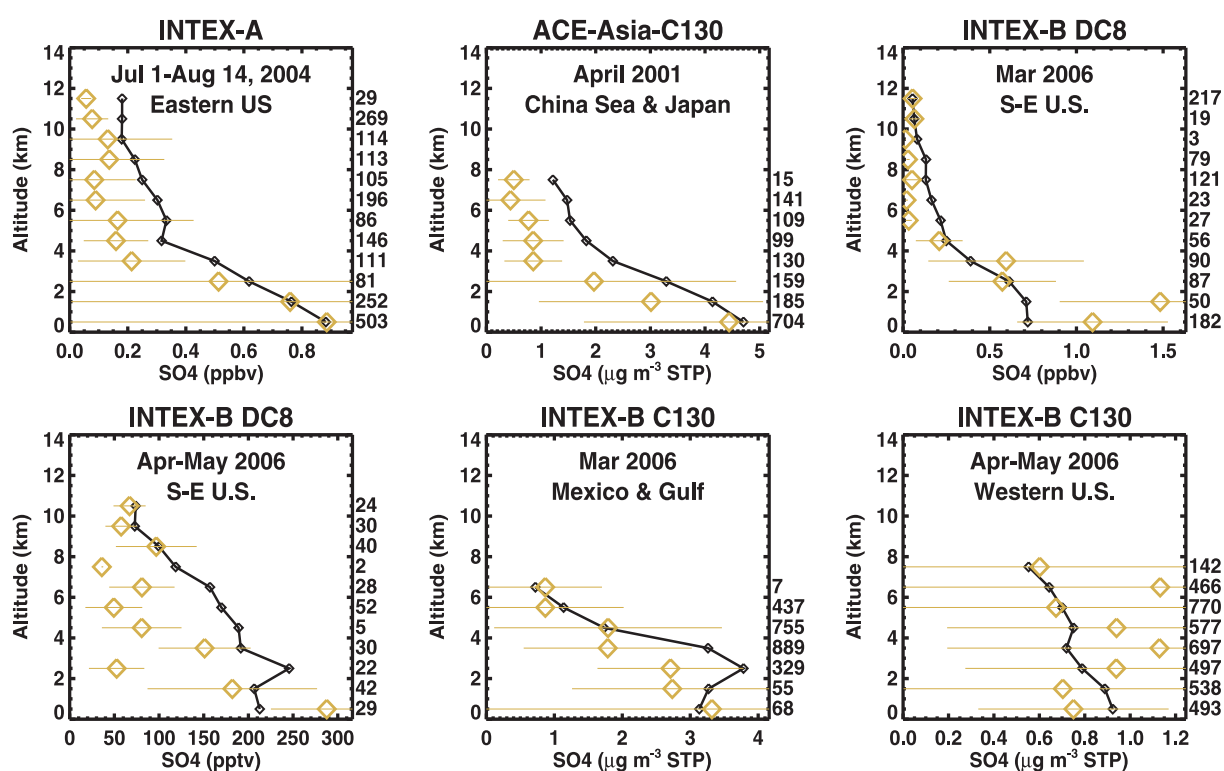
Fig. 7. Sulfate concentrations measured during the aircraft campaigns INTEX-A, ACE-Asia and INTEX-B, and corresponding values calculated with IMAGESv2. The number of measurements in each 1-km bin is indicated on the right side of each plot. STP stands for standard temperature and pressure (i.e. 298 K and 1 atm).

References

Aymoz, G., J.-L. Jaffrezo, V. Jacob, A. Colomb, C. George: Evolution of organic and inorganic components of aerosol during a Saharan dust episode observed in the French Alps, Atmos. Chem. Phys., 4, 2499–2512, 2004.

Bahadur, R., G. Habib, and L. M. Russell: Climatology of PM2.5 organic carbon concentrations from a review of ground-based atmospheric measurements by evolved gas analysis, Atmos. Environ., 43, 1591–1602, 2009.

C. Bloss, V. Wagner, M. E. Jenkin, R. Volkamer, W. J. Bloss, J. D. Lee, D. E. Heard, K. Wirtz, M. Martin-Reviejo, G. Rea, J. C. Wenger, and M. J. Pilling: Development of a detailed chemical mechanism (MCMv3.1) for the atmospheric oxidation of aromatic hydrocarbons, Atmos. Chem. Phys., 5, 641–664, 2005.

Bond, T. C., D. G. Streets, K. F. Yarber, S. M. Nelson, J.-H. Woo, and Z. Klimont : A technology-based global inventory of black and organic carbon emissions from combustion, J. Geophys. Res., 109, D14203, doi:10.1029/2003JD003697, 2004.

Cabada, J. C., S. N. Pandis, and A. L. Robinson: Sources of atmospheric carbonaceous particulate matter in Pittsburgh, Pennsylvania, J. Air Waste Manag. Assoc., 2002.

Cachier, H., M.-P. Bremond, and P. Buat-Ménard: Determination of atmospheric soot carbon with a simple thermal method, Tellus, 41B, 379–390, 1989.

Cao, J., S. Lee, K. Ho, S. Zou, X. Zhang, and J. Pan: Spatial and seasonal distributions of atmospheric carbonaceous aerosols in Pearl River Delta region, China, China Particuology, 1, 33–37, 2003 (http://www.cse.polyu.edu.hk/ cesclee/papers/O-24.pdf).

Cao, J.-J., B.-Q. Xu, J.-Q. He, X.-Q. Liu, Y.-M. Han, G. Wang, and C. Zhu: Concentrations, seasonal variations, and transport of carbonaceous aerosols at a remote Mountainous region in western China, Atmos. Environ., 43, 4444–4452, 2009.

Castro, L. M., C. A. Pio, R. M. Harrison, and D. J. T. Smith: Carbonaceous aerosol in urban and rural European atmospheres: estimation of secondary organic carbon concentrations, Atmos. Environ., 33, 2771–2781, 1999.

Carrico, C. M., M. H. Bergin, A. B. Shrestha, J. E. Dibb, L. Gomes, and J. M. Harris: The importance of carbon and mineral dust to seasonal aerosol properties in the Nepal Himalaya, Atmos. Environ., 37, 2811–2824, 2003a.

Carrico, C. M., P. Kus, M. J. Rood, P. K. Quinn, and T. S. Bates: Mixtures of pollution, dust, sea salt, and volcanic aerosol during ACE-Asia: Radiative properties as a function of relative humidity, J. Geophys. Res., 108, D23, 8650, doi:10.1029/2003JD003405, 2003b.

Ceulemans, K., J.-F. Müller, and S. Compernolle: Parameterising secondary organic aerosol from α-pinene using a detailed oxidation and aerosol formation model. Atmos. Chem. Phys. Discuss., 11, 23421-23468, 2011.

Chang, E. I. and J. F. Pankow: Organic particulate matter formation at varying relative humidity using surrogate secondary and primary organic compounds with activity corrections in the condensed phase obtained using a method based on the Wilson equation, Atmos. Chem. Phys., 10, 5475–5490, 2010.

Claeys, M., B. Graham, G. Vas, W. Wang, R. Vermeylen, V. Pashynska, J. Cafmeyer, P. Guyon, M. O. Andreae, P. Artaxo, and W. Maenhaut: Formation of Secondary Organic Aerosols Through Photooxidation of Isoprene, Science, 303, 1173–1176, 2004.

Dan, M., G. S. Zhuang, X. X. Li, H. R. Tao, and Y. H. Zhuan: The characteristics of carbonaceous species and their sources in PM2.5 in Beijing, Atmos. Environ., 38, 3443–3452, 2004.

Davis, J. M., P. V. Bhave, and K. M. Foley : Parameterization of N_2O_5 reaction probabilities on the surface of particles containing ammonium, sulfate, and nitrate, Atmos. Chem. Phys., 8, 5295–5311, 2008.

Decesari, S., S. Fuzzi, M. C. Facchini, M. Mircea, L. Emblico, F. Cavalli, W. Maenhaut, X. Chi, G. Schkolnik, A. Falkovich, Y. Rudich, M. Claeys, V. Pashynska, G. Vas, I. Kourtchev, R. Vermeylen, A. Hoffer, M. O. Andreae, E. Tagliavini, F. Moretti, and P. Artaxo: Characterization of the organic composition of aerosols from Rondônia, Brazil, during the LBA-SMOCC 2002 experiment and its representation through model compounds, Atmos. Chem. Phys., 6, 375–402, 2006.

Duan, F.K., K. B. He, Y. L. Ma, F. M. Yang, X. C. Yu, S. H. Cadle, T. Chan, P. A. Mulawa: Concentration and chemical characteristics of PM2.5 in Beijing, China: 2001–2002, Sci. Total Environ., 355, 264–275, 2006.

Duan, J., J. Tan, D. Cheng, X. Bi, W. Deng, G. Sheng, J. Fu, and M. H. Wong: Sources and characteristics of carbonaceous aerosol in two largest cities in Pearl River Delta Region, China, Atmos. Environ., 41, 2895–2903, 2007.

Duarte R. M. B. O., C. L. Mieiro, A. Penetra, C. A. Pio, and A. C. Duarte: Carbonaceous materials in size-segregated atmospheric aerosols from urban and coastal-rural areas at the Western European Coast, Atmos. Res., 90, 253–263, 2008.

Feng, J., Z. Guo, C. K. Chan, and M. Fang: Properties of organic matter in $PM_{2.5}$ at Changdao Island, China – A rural site in the transport path of the Asian continental outflow, Atmos. Environ., 41 (9), 1924–1935, 2007.

Fu, T.-M. et al.: Global budgets of atmospheric glyoxal and methylglyoxal, and implications for formation of secondary organic aerosols, J. Geophys. Res., 113, doi:10.1029/2007JD009505, 2008.

Gatari, M. J, and J. Boman, Black carbon and total carbon measurements at urban and rural sites in Kenya, East Africa, Atmos. Environ., 37, 1149–1154, 2008.,

Gilardoni, S., S. Liu, S. Takahama, L. M. Russell, J. D. Allan, R. Steinbrecher, J. L. Jimenez, P. F. De Carlo, E. J. Dunlea, and D. Baumgardner: Characterization of organic ambient aerosol during MIRAGE 2006 on three platforms, Atmos. Chem. Phys., 9, 5417–5432, 2009.

Han, Y. M., Z. W. Han, J. J. Cao, J. C. Chow, J. G. Watson, Z. S. An, S. X. Liu, and R. J. Zhang: Distribution and origin of carbonaceous aerosol over a rural high-mountain lake area, Northern China and its transport significance, Atmos. Environ., 42, 2405–2414, 2008.

Harrison, R. M., and J. Yin: Sources and processes affecting carbonaceous aerosol in central England, Atmos. Environ., 42, 1413–1423, 2008.

Heald, C. L., H. Coe, J. L. Jimenez, R. J. Weber, R. Bahreini, A. M. Middlebrook, L. M. Russell, M. Jolleys, T.-M. Fu, J. D. Allan, K. N.

Bower, G. Capes, J. Crosier, W. T. Morgan, N. H. Robinson, P. I. Williams, M. J. Cubison , P. F. DeCarlo , and E. J. Dunlea: Exploring the vertical profile of atmospheric organic aerosol: comparing 17 aircraft field campaigns with a global model, Atmos. Chem. Phys., 11, 12673–12696, 2011.

Huang, H.-E., C.-T. Lee, and H.-W. Chen: Long-range transport of biomass burning aerosols from Southeast Asia to Taiwan, Abstracts of the European Aerosol Conference 2005, W. Maenhaut, ed., Ghent, Belgium, 2005.

Henze, D. K. and J. H. Seinfeld: Global secondary organic aerosol from isoprene oxidation. Geophys. Res. Lett., 33, L09812, doi:10.1029/2006GL025976, 2006.

Henze, D. K. et al.: Global modeling of secondary organic aerosol formation from aromatic hydrocarbons: high- vs. low-yield pathways, Atmos. Chem. Phys., 8, 2405–2421, 2008.

Ho, K. F., S. C. Lee, J. J. Cao, Y. S. Li, J. C. Chow, J. G. Watson, and K. Fung: Variability of organic and elemental carbon, water soluble organic carbon, and isotopes in Hong Kong, Atmos. Chem. Phys., 6, 4569–4576, 2006.

Jones, A., D. L. Roberts, M. J. Woodage, and C. E. Johnson: Indirect sulphate aerosol forcing in a climate model with an interactive sulphur cycle, J. Geophys Res., 106, 20293–20310, 2001.

Kim, Y. P., K.-C. Moon, and J. H. Lee: Organic and elemental carbon in fine particles at Kosan, Korea, Atmos. Environ., 34, 3309–3317, 2000.

Köpke, P., M. Hess, I. Schult, and E. P. Shettle: Global Aerosol Data Set, report, Max-Planck Inst. für Meteorol., Hamburg, Germany, 1997.

Kourtchev, I., J. Warnke, W. Maenhaut, T. Hoffmann, and M. Claeys: Polar organic marker compounds in $PM_{2.5}$ aerosol from a mixed forest site in western Germany, Chemosphere, 73, 1308–1314, 2008.

Kourtchev, I., T. M. Ruuskanen, P. Keronen, L. Sogacheva, M. Dal Maso, A. Reissell, X. Chi, R. Vermeylen, M. Kulmala, W. Maenhaut, and M. Claeys: Determination of isoprene and α-/β-pinene oxidation products in boreal forest aerosols from Hyytiälä, Finland: diel variations and possible link with particle formation events, Plant Biology, 10, 138–149, 2008a.

Kourtchev, I., L. Copolovici, M. Claeys, and W. Maenhaut: Characterization of Atmospheric Aerosols at a Forested Site in Central Europe, Environ. Sci. Technol., 43 (13), 4665–4671, 2009.

Liousse, C., et al.: Aerosol modeling during AMMA-EOP with updated biomass burning emission inventories, poster at IGAC Conference, 2008.

Maenhaut, W., and M. Claeys: Characterization and sources of carbonaceous aerosols, Report D/2007/1191/521, Belgian Science Policy, 2007.

Maenhaut, W., M. Claeys, I. Janssens, and M. Kulmala: Formation mechanisms, marker compounds and source apportionment of biogenic atmospheric aerosols "BIOSOL", Final Report SD/AT/02A, Science for a Sustainable Development, Belgian Science Policy, 2009.

Marenco, F., P. Bonasoni, F. Calzolari, et al., Characterization of atmospheric aerosols at Monte Cimone, Italy, during summer 2004: Source apportionment and transport mechanisms, J. Geophys. Res., 111, D24202, doi:10.1029/2006JD007145, 2006.

Maria, S. F., L. M. Russell, B. J. Turpin, R. J. Porcja, T. L. Campos, R. J. Weber, and B. J. Huebert: Source signatures of carbon monoxide and organic functional groups in Asian Pacific Regional Aerosol Characterization Experiment (ACE-Asia) submicron aerosol types, J. Geophys. Res., 108, 8637, doi:8610.1029/2003JD003703, 2003.

Martin, R. V. et al. Global and regional decreases in tropospheric oxidants from photochemical effects of aerosols: J. Geophys. Res., 108, 4097, doi:10.1029/2002JD002622, 2003.

Metzger, S., F. Dentener, S. Pandis, and J. Lelieveld : Gas/aerosol partitioning, 1, A computationally efficient model, J. Geophys. Res., 107, 4312, doi:10.1029/2001JD001102, 2002.

Milukaite, A., K. Kvietkus, and I. Rimelytek: Organic and elemental carbon in crystal aerosol of the Baltic Sea Lithuanian Journal of Physics, 47 (2), 203–210, 2007.

Nenes, A., C. Pilinis, and C. N. Pandis: ISORROPIA: A new thermodynamic equilibrium model for multiphase multicomponent inorganic aerosols, Aquat. Geochem., 4, 132–152, 1998.

Mkoma, S. L.: Physico-Chemical Characterisation of Atmospheric Aerosols in Tanzania, with Emphasis on the Carbonaceous Aerosol Components and on Chemical Mass Closure, Ph.D. Thesis, Department of Analytical Chemistry, University of Ghent, 2008.

Ohara, T., H. Akimoto, J. Kurokawa, N. Horii, K. Yamaji, X. Yan, and T. Hayasaka : An Asian emission inventory of anthropogenic emission sources for the period 1980–2020, Atmos. Chem. Phys., 7, 4419–4444, doi:10.5194/acp-7-4419-2007, 2007.

Pio, C. A., L. M. Castro, M. A. Cerqueira, I. M. Santos, F. Belchior, and M. L. Salgueiro: Source assessment of particulate air pollutants measured at the south-west European coast, Atmos. Environ., 30, 3309–3320, 1996.

Pio C. A., M. Legrand, T. Oliveira, J. Afonso, C. Santos, A. Caseiro, P. Fialho, F. Barata, H. Puxbaum, A. Sanchez-Ochoa, A. Kasper-Giebl, A. Gelencsér, S. Preunkert, and M. Schock: Climatology of aerosol composition (organic versus inorganic) at nonurban sites on a west-east transect across Europe, J. Geophys. Res., 112, D23S02, doi:10.1029/2006JD008038, 2007.

Pham, M., J.-F. Müller, G. P. Brasseur, C. Granier, and G. Mégie: A three-dimensional study of the sulfur cycle, J. Geophys. Res., 100, D12, 26061–26092, DOI: 10.1029/95JD02095, 1995.

Puxbaum, H., J. Rendl, R. Allabashi, L. Otter, and M. C. Scholes: Mass balance of the atmospheric aerosol in a South African subtropical savanna (Nylsvley, May 1997), J. Geophys. Res., 105, 20,697–20,706, 2000.

Puxbaum, H., A. Caseiro, A. Sanchez-Ochoa, A. Kasper-Giebl, M. Claeys, A. Gelencser, M. Legrand, S. Preunkert, and C. Pio: Levoglucosan levels at background sites in Europe for assessing the impact of biomass combustion on the European aerosol background, J. Geophys. Res., 112, D23S05, doi:10.1029/2006JD008114, 2007.

Querol, X., A. Alastuey, J. Pey, M. Cusack, N. Perez, N. Mihalopoulos, C. Theodosi, E. Gerasopoulos, N. Kubilay, and M. Koçak: Variability

in regional background aerosols within the Mediterranean, Atmos. Chem. Phys., 9, 4575–4591, 2009.

Ram K., M. M. Sarin, and P. Hegde: Atmospheric abundances of primary and secondary carbonaceous species at two high-altitude sites in India : Sources and temporal variability, Atmos. Environ., 42, 6785–6796, 2008.

Rissler, J., A. Vestin, E. Swietlicki, G. Fisch, J. Zhou, P. Artaxo, and M. O. Andreae: Size distribution and hygroscopic properties of aerosol particles from dry-season biomass burning in Amazonia, Atmos. Chem. Phys., 6, 471-491, 2006.

Rodriguez, M. A., and D. Dabdub: IMAGES-SCAPE2: A modeling study of size- and chemically resolved aerosol thermodynamics in a global chemical transport model, J. Geophys., 109, D02203, doi:10.1029/2003JD003639, 2004.

Ruellan S., H. Cachier, A. Gaudichet, P. Masclet, and J.-P. Lacaux: Airborne aerosols over central Africa during the Experiment for Regional Sources and Sinks of Oxidants (EXPRESSO), J. Geophys. Res., 104, 30,673–30,690, 1999.

Saarikoski, S., T. Mäkelä, R. Hillamo, P. Aalto, V.-M. Kerminen, and M. Kulmala: Physico-chemical characterization and mass closure of size segregated atmospheric aerosols in Hyytiälä, Finland, Boreal Env. Res., 10, 385–400, 2005.

Sarkar, A. K., M. Jain, D. C. Parasharn, A. P. Mitra, U. C. Kulshrestha, and H. Cachier: Measurements of carbonaceous aerosols at urban and remote marine sites, Current Science (suppl.), 80, 176–179, 2001.

Spindler, G., E. Brüggemann, T. Gnauk, A. Grüener, H. Herrmann, K. Müller, T. M. Tuch, M. Wallasch, and A. Wiedensohler: Size-segregated physical-chemical characterization of particles depending from air mass origin at Melpitz (Germany), Abstracts of the European Aerosol Conference 2005, W. Maenhaut, ed., Ghent, Belgium, 2005.

Spindler, G., E. Brüggemann, T. Gnauk, A. Grüner, K. Müller, T. M. Tuch, M. Wallasch, A. Wehner, and H. Herrmann: Size-segregated physical and chemical long-time characterization of particles depending from air mass origin at German lowlands (Saxony, Melpitz site), EMEP Particulate Matter Assessment Report, Part B, Annex A, NILU, report EMEP/CCCReport 8/2007, ref. O-7726, 178–221, 2007.

Stavrakou, T., J.-F. Müller, I. De Smedt, M. Van Roozendael, M. Kanakidou, M. Vrekoussis, F. Wittrock, A. Richter, and J. P. Burrows: The continental source of glyoxal estimated by the synergistic use of spaceborne measurements and inverse modelling, Atmos. Chem. Phys., 8431–8446, 2009.

Stavrakou, T., J.-F. Müller, J. Peeters, A. Razavi, L. Clarisse, C. Clerbaux, P.-F. Coheur, D. Hurtmans, M. De Mazière, C. Vigouroux, N. M. Deutscher, D. W. T. Griffith, N. Jones, C. Paton-Walsh : Satellite evidence for a large source of formic acid from boreal and tropical forests, Nature Geosci., 5, 26–30, doi:10.1038/ngeo1354, 2012.

Stavrakou, T., J. Peeters, and J.-F. Müller, Improved global modelling of HOx recycling in isoprene oxidation: evaluation against the GABRIEL and INTEX-A aircraft campaign measurements, Atmos. Chem. Phys., 10, 9863–9878, 2010.

Sullivan, A. P., R. E. Peltier, C. A. Brock, J. A. de Gouw, J. S. Holloway, C. Warneke, A. G. Wollny, A and R. J. Weber: Airborne measurements of carbonaceous aerosol soluble in water over northeastern United States: Method development and and investigation into water-soluble organic carbon sources, J. Geophys. Res., 111, D23S46, doi:10.1029/2006JD007072, 2006.

Taketani, F., Kanaya, Y., and Akimoto, H. : Kinetics of heterogeneous reaction of HO_2 radical at ambient concentration levels with $(NH_4)_2SO_4$ and NaCl aerosol particles, J. Phys. Chem. A, 112, 2370–2377, doi:10.1021/jp0769936, 2008.

Tang, I. N., K. H. Fung, D. G. Imre, and H. R. Munkelwitz: Phase transformation and metastability of hygroscopic aerosols, Atmos. Environ., 27(4), 467–473, 1993.

Taraborrelli, D., M. G. Lawrence, T. M. Butler, R. Sander, and J. Lelieveld : Mainz Isoprene Mechanism 2 (MIM2): an isoprene oxidation mechanism for regional and global atmospheric modelling, Atmos. Chem. Phys., 9, 2751–2777, 2009.

Tsai, Y. I., and C.-L. Chen: Atmospheric aerosol composition and source apportionments to aerosol in southern Taiwan, Atmos. Environ., 40(25), 4751–4763, 2006.

Wang, G., K. Kawamura, S. Hatakeyama, A. Takami, H. Li, and W. Wang: Aircraft measurement of organic aerosols over China, Environ. Sci. Technol., 41 (9), 3115–3120, 2007.

Wang, W. C., K. S. Chen, S. J. Chen, C. C. Lin, J. H. Tsai, C. H. Lai, and S. K. Wang: Characteristics and Receptor Modeling of Atmospheric PM2.5 at Urban and Rural Sites in Pingtung, Taiwan, Aerosol and Air Quality Research, 8, 112–129, 2008.

van der Werf, G. R., J. T. Randerson, L. Giglio, G. J. Collatz, M. Mu, P. S. Kasibhatla, D. C. Morton, R. S. DeFries, Y. Jin, T.T. van Leeuwen : Global fire emissions and the contribution of deforestation, savanna, forest, agricultural, and peat fires (1997-2009), Atmos. Chem. Phys., 10, doi:10.5194/acp-10-11707-2010, 2010.

Yttri, K. E., W. Aas, A. Bjerke, D. Ceburnis, C. Dye, L. Emblico, M. C. Facchini, C. Forster, J. E. Hanssen, H. C. Hansson, S. G. Jennings, W. Maenhaut, J. P. Putaud, and K. Torseth: Elemental and organic carbon in PM10: a one year measurement campaign within the European Monitoring and Evaluation Programme EMEP, Atmos. Chem. Phys. 7, 5711–5725, 2007.

Zappoli, S., A. Andracchio, S. Fuzzi, M. C. Facchini, A. Gelencsér, G. Kiss, Z. Krivácsy, A. Molnár, E. Mészáros, H.-C. Hansson, K. Rosman, and Y. Zebühr: Inorganic, organic and macromolecular components of fine aerosol in different areas of Europe in relation to their water solubility, Atmos. Environ., 33, 2733–2743, 1999. ation Programme EMEP, Atmos. Chem. Phys. 7, 5711–5725, 2007.