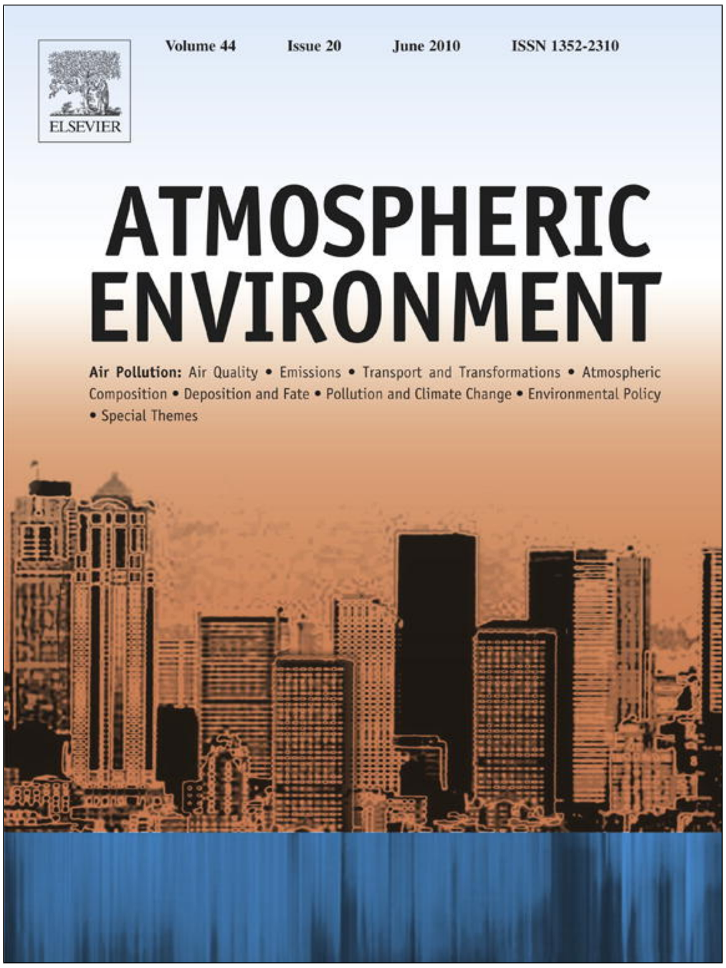
Volume 44
Issue 20
June 2010
ISSN 1352-2310
ELSEVIER
ATMOSPHERIC ENVIRONMENT
Air Pollution: Air Quality • Emissions • Transport and Transformations • Atmospheric Composition • Deposition and Fate • Pollution and Climate Change • Environmental Policy • Special Themes

Atmospheric Environment 44 (2010) 2434–2442

Contents lists available at ScienceDirect

Atmospheric Environment

journal homepage: www.elsevier.com/locate/atmosenv

Sea salt concentrations across the European continent

A.M.M. Manders [a,*], M. Schaap [a], X. Querol [b], M.F.M.A. Albert [a], J. Vercauteren [c], T.A.J. Kuhlbusch [d], R. Hoogerbrugge [e]

[a] TNO, Business unit Environment, Health and Safety, P.O. Box 80015, 3508 TA Utrecht, The Netherlands
[b] Institute of Environmental Assessment and Water Research, IDÆA, CSIC, C/Jordi Girona 11, Barcelona 08028, Spain
[c] Flemish Environment Agency (VMM), A. Van de Maelestraat 96, 9320 Erembodegem, Belgium
[d] IUTA e.V., Air Quality & Sustainable Nanotechnology Unit, D-47229 Duisburg, Germany
[e] National Institute for Public Health and the Environment (RIVM), P.O. Box 1, 3720 BA Bilthoven, The Netherlands

ARTICLE INFO

Article history:
Received 6 January 2010
Received in revised form
19 March 2010
Accepted 22 March 2010

Keywords:
Sodium
Sea salt
Regional modelling
PM10

ABSTRACT

The oceans are a major source for particles that play an important role in many atmospheric processes. In Europe sea salt may contribute significantly to particulate matter concentrations. We have compiled sodium concentration data as a tracer for sea salt for 89 sites in Europe to provide more insight in the distribution of sea salt across Europe. The annual average sea salt concentrations above land were estimated to range between 0.3 and almost 13 µg m^{-3}. Maximum concentrations are found at the Irish coast. At coastal sites along the Atlantic and North Sea coast concentrations tend to be around 5 µg m^{-3}. More inland locations up to about 300 km away from the coast tend to show concentrations between 2 and 5 µg m^{-3}, whereas sites further away from the coast are characterized by lower concentrations. An analysis of the representativity of the data with respect to a long term average showed that the long average is associated with a standard deviation of around 15%. The compilation of observations provides an improved overview of sea salt concentrations in Europe as well as an improved basis for model validation. Verification of the results of the LOTOS-EUROS model learned that the model represents well the spatial variability of the observed sea salt concentrations very well. However, the absolute concentrations are significantly overestimated due to large uncertainties in the emission and dry deposition parameterizations. Using the high explained variability in the gradients across Europe, the bias-corrected modelled distribution serves as a best estimate of the sea salt distribution across Europe for 2005.

1. Introduction

The oceans are a major source for atmospheric particles. In coastal regions sea salt may contribute tens of percents to the annual mean particulate mass, e.g. PM10 (e.g. Putaud et al., 2004). Furthermore, sea salt plays an important role in a number of physical and chemical atmospheric processes. The reaction of sea salt particles with nitric acid forms sodium nitrate, which is in contrast to ammonium nitrate not semi-volatile and needs to be accounted for in the (particulate) nitrogen budget (Tamm and Schulz, 2003). The halogens released by the reaction of acidic gases with sea salt contribute to ozone destruction (Finlayson-Pitts, 2003; Knipping and Dabdub, 2003). Sea salt contributes to the deposition of base cat-ions and its deposition flux needs to be accounted for to assess the total acid deposition to vulnerable ecosystems (Van Loon et al., 2005). Also, sea salt contributes to corrosion in coastal regions (Muster and Cole, 2005). Finally, sea salt aerosols are an important contributor to cloud condensation nuclei (CCN) in marine air masses (Yoon and Brimblecombe, 2002). Hence, sea salt aerosols are essential components of atmospheric models at urban, regional and global scales (Gong et al., 1997).

In recent years, a large number of studies have been dedicated to the modelling of atmospheric particles in general and sea salt particles in particular. Many studies of marine aerosols and their role in the climate system were performed on the global scale (e.g. Gong et al., 1997, 2003; Guelle et al., 2001; Stier et al., 2005). For air quality assessment, it is important to resolve the large gradients in sea salt levels and a number of regional models includes a description of sea salt (e.g. Foltescu et al., 2005; Bessagnet et al., 2004; Langmann et al., 2008; Schaap et al., 2008). The verification of these models is severely hampered by the number of available measurements. For example, Foltescu et al. (2005) compared their regional model results against measurements from eight monitoring stations in two

---

* Corresponding author at: TNO Environment and Geosciences, Unit Environment, Health and Safety, P.O. Box 80015, 3508 TA Utrecht, The Netherlands. Tel.: +31 (0) 88 8662260; fax: +31 (0) 88 8662044.
E-mail address: astrid.manders@tno.nl (A.M.M. Manders).

1352-2310/$ – see front matter © 2010 Published by Elsevier Ltd.
doi:10.1016/j.atmosenv.2010.03.028

countries in Europe. The model evaluations show in general a reasonable agreement between modelled and observed temporal variation but large systematic biases. Because of the limited number of validation data, an integrated picture was not available.

We present a compilation of existing data on sodium, that have (mostly) become available from PM mass closure studies, to provide more insight in the distribution of sea salt in PM10 across Europe, As such the observational basis for model verification for Europe is expanded. These data are then used verify the calculated sea salt distributions from the chemistry transport model (CTM) LOTOS-EUROS. Since 2005 had about average weather conditions and a reasonable amount of observations was available to validate the model, this year is chosen as the model year. Finally, a best estimate of the sea salt distribution based on the model to measurement comparison is provided.

2. Methodology

2.1. Compilation of observations

Sea salt aerosol consists mainly of chloride (Cl, 55.1% by weight) and sodium (Na, 30.6% by weight) (Millero, 2004), which may both be used as a tracer for sea salt. However, since reactions between chloride and acidic gases such as nitric acid may cause a chloride loss (in the form of HCl) (McInnes et al., 1994), both in the atmosphere and on a filter substrate (Slanina et al., 2001), chloride is not a conserved tracer for sea salt aerosol. Sodium, on the other hand, is a conserved tracer and has only minor non-marine sources (White, 2008). Therefore, it is a robust tracer for sea salt and we have restricted ourselves to the use of sodium data. To calculate the total sea salt concentration from the observed sodium concentration, the sodium concentration has to be multiplied by $100/30.6 = 3.26$ (Millero, 2004).

A comprehensive literature search for available data sets on atmospheric sodium in PM10 was performed. These data points were added to the data available through the EMEP network (www.emep.int). Many short-term campaign observations are available, but due to the high temporal variability of sea salt concentrations these are often not representative for a longer period. Therefore, only studies that are representative for at least a year were included, but without the restriction to daily sampling. Furthermore, we have restricted ourselves to data for which the measurement methodology was retraceable. To avoid the inclusion of stations (partly) representative for the free troposphere, mountain sites (above 700 m a.s.l.) were excluded from the analysis. For 2005, daily measurements were collected for a number of stations to verify the model results.

The comprehensive compilation of sodium observations yielded data for 89 stations throughout Europe. The sites, measurement periods and average sodium concentrations are listed in Table 1. For a number of countries data are obtained through the EMEP program, e.g. Denmark, Norway, Spain and Ireland. For other countries such as the Netherlands, Belgium and Austria the data stem from national programs or research stations. The data from national programs often cover about 60–90 measurement days spread through a year, whereas sites within the EMEP program normally have daily coverage. Since sea salt aerosol concentrations are larger near the coast, the number of sites is slightly biased towards locations not too far from the coast. In (south) eastern Europe hardly any observations are available.

2.2. The LOTOS-EUROS model

The regional air quality model LOTOS-EUROS (Schaap et al., 2008) is used to simulate the sea salt distribution across Europe. The LOTOS-EUROS model is a 3D chemistry transport model aimed to simulate air pollution in the lower troposphere. The model has been used for the assessment of particulate air pollution in a number of studies of total PM10 (Denby et al., 2008; Manders et al., 2009a), secondary inorganic components (Schaap et al., 2004a; Barbu et al., 2009) and primary carbonaceous components (Schaap et al., 2004b). The model has participated frequently in international model comparisons addressing ozone (van Loon et al., 2005; Hass et al., 1997) and particulate matter (Cuvelier et al., 2007; Hass et al., 2003; Stern et al., 2008).

The model incorporated four sea salt particle size classes: 0.14–1, 1–2.5, 2.5–5 and 5–10 μm wet diameter (at 80% relative humidity). For the generation of sub-micron particles (0.14–1 μm) the emission function by Mårtensson et al. (2003) was used. For the other modes Monahan et al. (1986) are followed. Both parameterizations use a whitecap cover as function of the wind speed, whereas Mårtensson et al. (2003) also included sea water temperature as an explanatory variable. The surf zone contributes locally to the additional generation of aerosol, depending on wind velocity and fetch (De Leeuw et al., 2000). However, since this process is very local (an area of a few hundred meters width along the coast) its contribution to the total sea salt aerosol production is likely to be small and only noticeable in the immediate vicinity of the coast. Thus, open-sea conditions are also applied to the surf zone. All sea salt aerosol is generated within the model domain, no initial or boundary conditions from other models were used.

The generation functions are generally valid for open ocean conditions with a salinity of around 35‰. But within our domain the salinity of the Baltic Sea (7‰) is much lower than that of the open ocean, leading to an overestimation of the emission flux when using open ocean conditions. Therefore, a crude approach for the Baltic Sea (between 54 and 66°N, 14–32°E) was used, in which the abovementioned emission parameterizations were applied and resulting emission strength was divided by 10. This factor is based on the experimental data presented by Mårtensson et al. (2003), as the impact of the low salinity on the emission strength is not well understood (Lewis and Schwartz, 2004).

The model applies a dry deposition scheme based on Zhang et al. (2001), with constant roughness length. Wet deposition is treated using simple scavenging coefficients.

The model was applied to a domain that ranges from 30°W to 40°E, 35°–70°N, with a normal $0.5° \times 0.25°$ longitude–latitude resolution. The domain was extended further to the west compared to its normal set-up to incorporate a reasonable fetch west of Ireland and the Iberian Peninsula. Therefore, the analysis or the results was restricted to 20°W–30°E, 35°–70°N (Fig. 1). The model is forced using 3-hourly meteorological fields from ECMWF and calculates hourly concentrations of all sea salt modes.

3. Results

3.1. Compilation of particulate sodium data

Measured average sodium concentrations (Table 1, Fig. 1) range between 0.1 and almost 4 $\mu g\ m^{-3}$. As expected, the highest concentrations are generally found near the coast, and concentrations rapidly decrease with increasing distance from the coast. At coastal sites, concentrations tend to be between 1 and 1.5 $\mu g\ m^{-3}$. Higher values are only found for stations located at the Atlantic coast of Ireland, whereas lower values are found at the Baltic coast. More inland locations up to about 300 km away from the coast tend to show concentrations between 0.5 and 1 $\mu g\ m^{-3}$, whereas remote continental sites are characterized by concentrations below 0.5 $\mu g\ m^{-3}$.

The data provided above are based on measurements for different meteorological years. As the inter-annual variability in meteorology may affect the extent to which the presented concentration data represent a long term average, an analysis on the

Table 1
Compilation of sodium observations. For each station the name, country code, location, the years during which the measurements were made, the sodium concentration (μg m^{-3}) and data source is given.

Site name	Country	Lon	Lat	Year	Na	Reference
Illmitz(AT0002R)	AUT	16.77	47.77	2004–2006	0.09	EMEP (2009)
Vienna(Auphep-1)	AUT	16.35	48.22	1999–2000	0.19	Puxbaum et al. (2004)
Vienna(Auphep-2)	AUT	15.93	48.27	1999–2000	0.13	Puxbaum et al. (2004)
Houtem	BEL	2.61	51.01	2006–2007	1.07	Vercauteren (2009)
Aarschot	BEL	4.84	50.99	2006–2007	0.74	Vercauteren (2009)
Zelzate	BEL	3.81	51.20	2006–2007	0.95	Vercauteren (2009)
Mechelen	BEL	4.48	51.03	2006–2007	0.83	Vercauteren (2009)
Borgerhout	BEL	4.43	51.21	2006–2007	0.97	Vercauteren (2009)
Hasselt	BEL	5.34	50.93	2006–2007	0.73	Vercauteren (2009)
Bern	CHE	7.43	46.95	1998–1999	0.67	Hueglin et al. (2005)
Zurich-Wiedikon	CHE	8.53	47.37	1998–1999	0.69	Hueglin et al. (2005)
Zurich-Kaserne	CHE	8.55	47.37	1998–1999	0.19	Hueglin et al. (2005)
Basel	CHE	7.58	47.53	1998–1999	0.26	Hueglin et al. (2005)
Payerne	CHE	6.93	46.82	1998–1999	0.30	Hueglin et al. (2005)
Chaumont	CHE	7.58	47.05	1998–1999	0.15	Hueglin et al. (2005)
Westerland(DE0001R)	DEU	8.31	54.93	2005–2006	1.85	EMEP (2009)
Langenbrügge(DE0002R)	DEU	10.76	52.80	2005–2006	0.39	EMEP (2009)
Schauinsland(DE0003R)	DEU	7.91	47.91	2005–2006	0.14	EMEP (2009)
Neuglobsow(DE0007R)	DEU	13.03	53.17	2005–2006	0.39	EMEP (2009)
Zingst(DE0009R)	DEU	12.73	54.43	2005–2006	0.67	EMEP (2009)
Melpitz(DE0044R)	DEU	12.93	52.53	2002–2006	0.29	EMEP (2009)
Berlin urban area	DEU	13.40	52.52	2001–2002	0.28	Beekmann et al. (2007)
PA Paulinenaue	DEU	12.72	52.68	2001–2002	0.34	Beekmann et al. (2007)
HH Hasenholz	DEU	14.02	52.57	2001–2002	0.30	Beekmann et al. (2007)
Tange(DK0003R)	DNK	9.60	56.35	2000–2006	1.03	EMEP (2009)
Keldsnor(DK0005R)	DNK	10.73	54.73	2000–2006	1.48	EMEP (2009)
Anholt(DK0008R)	DNK	11.52	56.72	1989–2006	1.59	EMEP (2009)
Ulborg(DK0031R)	DNK	8.43	56.28	2005–2006	1.40	EMEP (2009)
Campisabalos(ES0009R)	ESP	−3.14	41.28	2005–2006	0.36	EMEP (2009)
Montseny(ES0017R)	ESP	2.35	41.77	2004–2006	0.30	EMEP (2009)
Alcobendas	ESP	−3.63	40.55	2001	0.38	Querol et al. (2008)
Algeciras	ESP	−5.45	36.14	2003–2004	1.55	Querol et al. (2008)
Badajoz	ESP	−6.58	38.53	2004–2005	0.59	Querol et al. (2008)
Barcelona (IJA-CSIC)	ESP	2.12	41.38	2007	1.14	Querol et al. (2008)
Bastarrechte (Cartagena)	ESP	−0.97	37.60	2004–2005	0.81	Querol et al. (2008)
Bemantes	ESP	−8.18	43.34	2001	1.09	Querol et al. (2008)
Burgos	ESP	−3.64	42.34	2004–2005	0.47	Querol et al. (2008)
Iturrama (Pamplona)	ESP	−1.65	42.82	2003	0.37	Querol et al. (2008)
Las Palmas	ESP	−15.41	28.13	2001	3.92	Querol et al. (2008)
Madrit Escuela Aguirre	ESP	−3.68	40.43	1999–2000	0.32	Querol et al. (2008)
Montseny	ESP	2.38	41.78	2007	0.33	Querol et al. (2008)
Onda (Castellon)	ESP	−0.25	39.96	2007	0.62	Querol et al. (2008)
Palma de Mallorca	ESP	2.62	39.56	2004–2005	1.75	Querol et al. (2008)
M.Perdon (Pamplona)	ESP	−1.78	42.73	2003	0.46	Querol et al. (2008)
Ebro basin (Monagrega)	ESP	0.20	40.50	1999–2000	0.29	Rodríguez et al. (2004)
Millars valley (Onda)	ESP	0.20	39.90	1999	0.91	Rodríguez et al. (2004)
Barcelona (L'Hospitalet)	ESP	2.00	41.20	2000	0.94	Rodríguez et al. (2004)
Uto (FI0009)	FIN	21.38	59.78	2003–2006	0.54	EMEP (2009)
Virolahti (FI0017)	FIN	27.68	60.52	2003–2006	0.24	EMEP (2009)
Finland(FI0036)	FIN	24.25	68.00	2003–2006	0.15	EMEP (2009)
High Muffles(GB0014R)	GBR	−0.81	54.33	1996–2001	0.61	EMEP (2009)
East Ruston(GB0090R)	GBR	1.47	52.80	2001	1.18	EMEP (2009)
Banchory(GB0091R)	GBR	−2.53	57.08	2001	0.69	EMEP (2009)
Dundee	GBR	−2.97	56.50	1999–2000	1.16	Qin and Oduyemi (2003)
Chilton	GBR	−1.27	51.55	1957–1974	0.98	Lee et al. (1994)
Port Talbot	GBR	−3.78	51.60	1972–1973	1.05	Lee et al. (1994)
Walsall	GBR	−1.98	52.58	1976–1989	1.45	Lee et al. (1994)
Lambeth	GBR	−0.12	51.49	1976–1982	1.38	Lee et al. (1994)
Brent	GBR	−0.27	51.57	1975–1989	1.22	Lee et al. (1994)
Manchester-Rusholme	GBR	−2.22	53.45	1975–1989	1.24	Lee et al. (1994)
Trafford-Altrincham	GBR	−2.35	53.38	1978–1989	1.31	Lee et al. (1994)
Trafford-Flixton	GBR	−2.39	53.45	1975–1989	1.40	Lee et al. (1994)
Windermere	GBR	−2.94	54.36	1970–1989	0.93	Lee et al. (1994)
Finokalia (GR0002)	GR	35.30	25.70	1996–1999	1.93	EMEP (2009)
Valentia Observatory(IE0001R)	IRL	−10.24	51.94	2004–2006	1.91	EMEP (2009)
Oak Park(IE0005R)	IRL	−6.92	52.87	2005–2006	0.86	EMEP (2009)
Malin Head(IE0006R)	IRL	−7.34	55.38	2005–2006	2.44	EMEP (2009)
Carnsore Point(IE0008R)	IRL	−6.37	52.19	2005–2006	3.26	EMEP (2009)
Irafoss(IS0002R)	ISL	−21.02	64.08	2006	1.01	EMEP (2009)
Busalla-Genoa	ITA	8.95	44.57	2006–2007	0.21	Mazzei et al. (2006)
Florence	ITA	11.25	43.77	1997–1998	0.69	Lucarelli et al. (2004)
Rucava(LV0010R)	LVA	21.22	56.22	2005–2006	0.22	EMEP (2009)
Vredepeel 131	NL	5.85	51.54	2007–2008	0.70	Manders et al. (2009b)

Table 1 (*continued*)

Site name	Country	Lon	Lat	Year	Na	Reference
Breda 240	NL	4.83	51.59	2007–2008	0.85	Manders et al. (2009b)
Rotterdam 448	NL	4.46	51.92	2007–2008	1.09	Manders et al. (2009b)
Cabauw 620	NL	4.93	51.97	2007–2008	0.76	Manders et al. (2009b)
Hellendoorn 738	NL	5.71	52.11	2007–2008	0.52	Manders et al. (2009b)
Schiedam (DCMR)	NL	4.40	51.92	2007–2008	1.21	Manders et al. (2009b)
Birkenes(NO0001R)	NOR	8.25	58.38	1999–2007	0.39	EMEP (2009)
Skreådalen(NO0008R)	NOR	6.72	58.82	1999–2004	0.36	EMEP (2009)
Tustervatn(NO0015R)	NOR	13.92	65.83	1999–2007	0.28	EMEP (2009)
Kårvatn(NO0039R)	NOR	8.88	62.78	1999–2007	0.17	EMEP (2009)
Osen(NO0041R)	NOR	11.78	61.25	1999–2003	0.11	EMEP (2009)
Spitsbergen (NO0042G)	NOR	11.88	78.90	1999–2007	0.25	EMEP (2009)
Karasjok(NO0055R)	NOR	25.22	69.47	1999–2007	0.22	EMEP (2009)
Birkenes	NOR	8.20	58.32	1978–1979	0.53	EMEP (2009)
Stará Lesná(SK0004R)	SVK	20.28	49.15	2006	0.20	EMEP (2009)
Iskrba(SI0008R)	SVN	14.87	45.57	2004–2006	0.09	EMEP (2009)

inter-annual variability of sodium concentrations was performed for the sites which have at least four years of observations (Fig. 2).Yearly values may deviate from the average up to 35% depending on the number of years (N), therefore standard deviation is also provided, though based on a limited number of years. Using this approach, the spread in the variability between stations is reduced with a central value of 15%. At some locations the variability is lower.

3.2. Modelled sodium distribution for 2005

The modelled sodium distribution (Fig. 3a) shows the highest sea salt concentration over the open ocean (Atlantic, Mediterranean). Above the Atlantic, higher concentrations are reached than above the Mediterranean due to higher wind speeds. These high wind speeds are associated with westerlies in Europe, and large quantities of sea saltcan be transported inland during these conditions. Consequently, the concentrations gradually decrease from the western European coastline to inland locations. Easterly winds often do not reach such high speeds as westerly winds and are associated with considerably shorter fetches, thus causing lower sea salt emissions and loads. The slower transport results in steeper gradients along the eastern coasts.

The modelled annual mean concentrations are systematically larger than the observed concentrations. For 2005 (Fig. 4a), the spatial correlation between model results and observations is high, with an explained variability of 85%, but the model overestimated the absolute value with a factor of 2.46. Concentrations at stations tend to be more overestimated at the west than at the east coastal stations. For the full compilation set (Fig. 4b), the variability in the diagram is higher as the model and observation data are no longer paired in time. Nevertheless, the comparison confirms the findings for 2005 only data.

The model performance was tested on a daily basis for a site in Ireland (Carnsore Point), close to open sea, and a site in Germany (Langenbrugge), reflecting a more continental location. The observed seasonal variation, with higher concentrations in autumn and winter (storms) and lower concentrations in summer, is reflected in the modelled fields (Fig. 5). Furthermore, the model is able to capture the main features of the temporal variability at both the Irish and the German site, indicating that the processes of generation and transport are modelled realistically. The time series for Langenbrugge clearly illustrates the general feature that the peaks are modelled higher than observed, whereas the baseline concentrations compare better to observations.

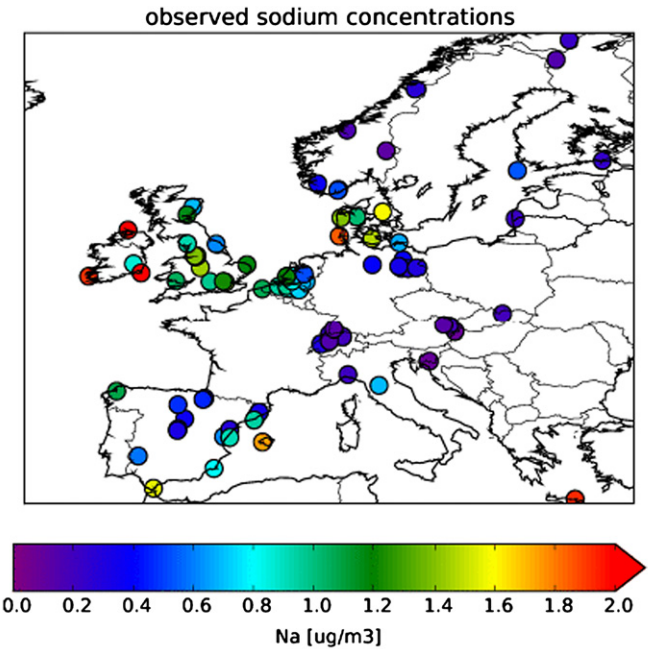

Fig. 1. Annual average sodium concentrations (µg m^{-3}) for all sites compiled in this study.

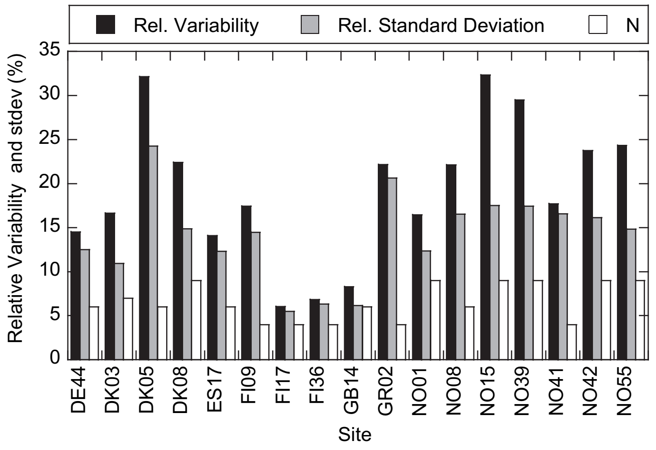

Fig. 2. Comparison of indicators for inter-annual variability at all sites with a minimum of 4 years of data. The indicators are the maximum relative difference between the concentration in a single year and the long term average (Black) and the relative standard deviation (Grey). *N* (white) is the number of years, that contribute to the calculation of the indicators is given.

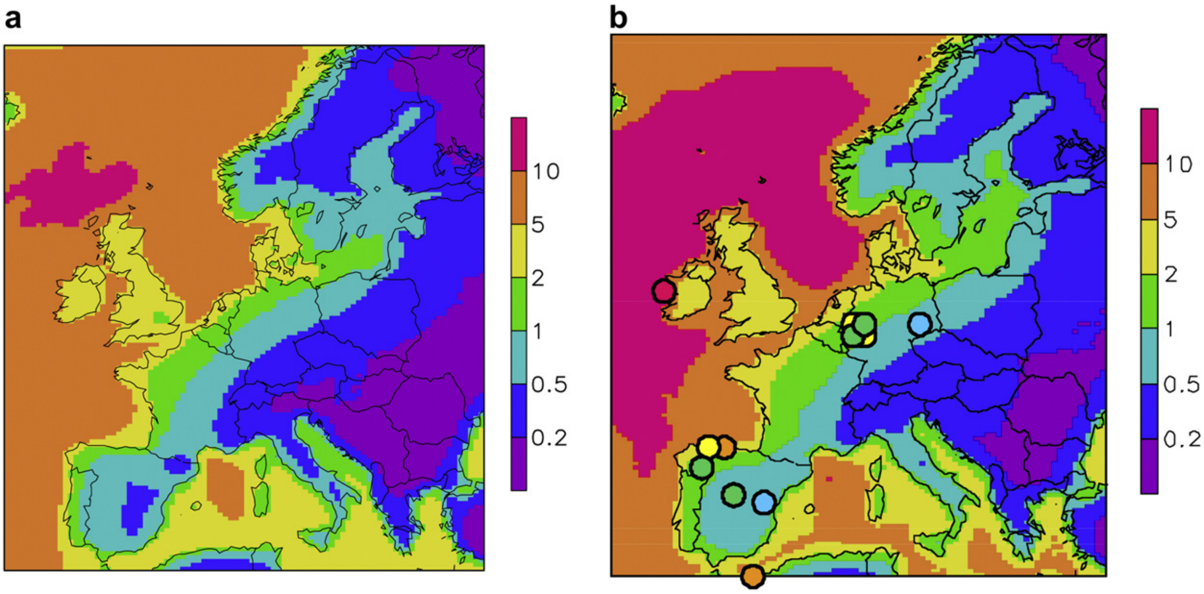

Fig. 3. (a) Modelled annual average sodium concentration (μg m^{-3}) and (b) the estimated annual average sea salt concentration, based on the scaling of sodium distribution (SS = Na × 3.26/2.46), across Europe for 2005. Filled circles indicate the concentrations at the verification locations (Table 3).

We provide paired and unpaired annual averages and standard deviations as well as the correlation between model and observations (Table 2). The differences between model results for the whole year compared to an average of values only on observation days were small, which may indicate that the annual average concentrations based on regular sampling once every few days are indeed representative for a full year. The correlation coefficients are generally moderate ($0.45 < R < 0.7$). Only for Onda the correlation is very poor. The variation around the average concentration shows a similar behaviour in both the modelled time series and the observations.

The size of the modelling domain had impact on the concentrations. Compared to simulations using the 10°W western boundary, the westward extension to 30°W led to a significant increase in modelled sea salt concentrations in the western part of the domain. This did not only apply for stations in Ireland and Spain, for which concentrations were otherwise significantly off the trend line, but, to a lesser extent, also for countries like Denmark and the Netherlands. The extension of the domain has improved the correlation for Irish and Spanish stations (not shown) to the range found for stations further into the standard domain. Hence, to model the sea salt concentrations realistically in Spain, Portugal and Ireland the standard LOTOS-EUROS model domain has to be extended westwards to include a large enough fetch or source area.

The simple adaptation of the sea salt source function for the low salinity in the Baltic Sea, based on the experiments by Mårtensson et al. (2003), appeared to be fruitful, as the observations in Latvia and Finland are also found close to the trend line in Fig. 4a. For the Black Sea, which falls mostly outside our present modelling domain, also a scaling might apply, since it has a salinity of around 17‰. However, this is above the critical value of 10‰ indicated by Mårtensson et al. (2003) and we do not have observations to verify or falsify the approach.

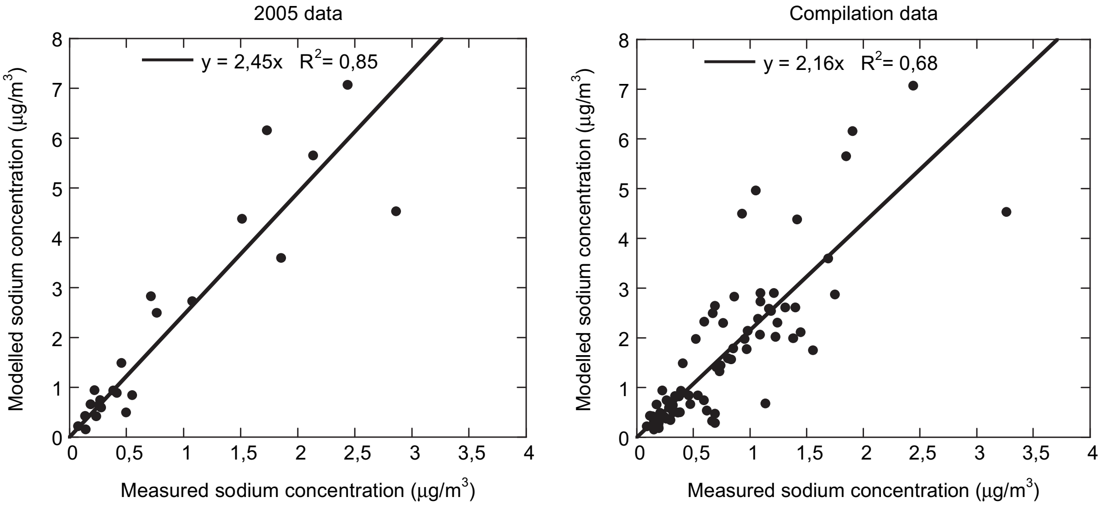

Fig. 4. Comparison of the modelled annual average concentrations to measurement data for 2005 paired in time (left) as well as for the long term averages from the entire compilation of observations (right).

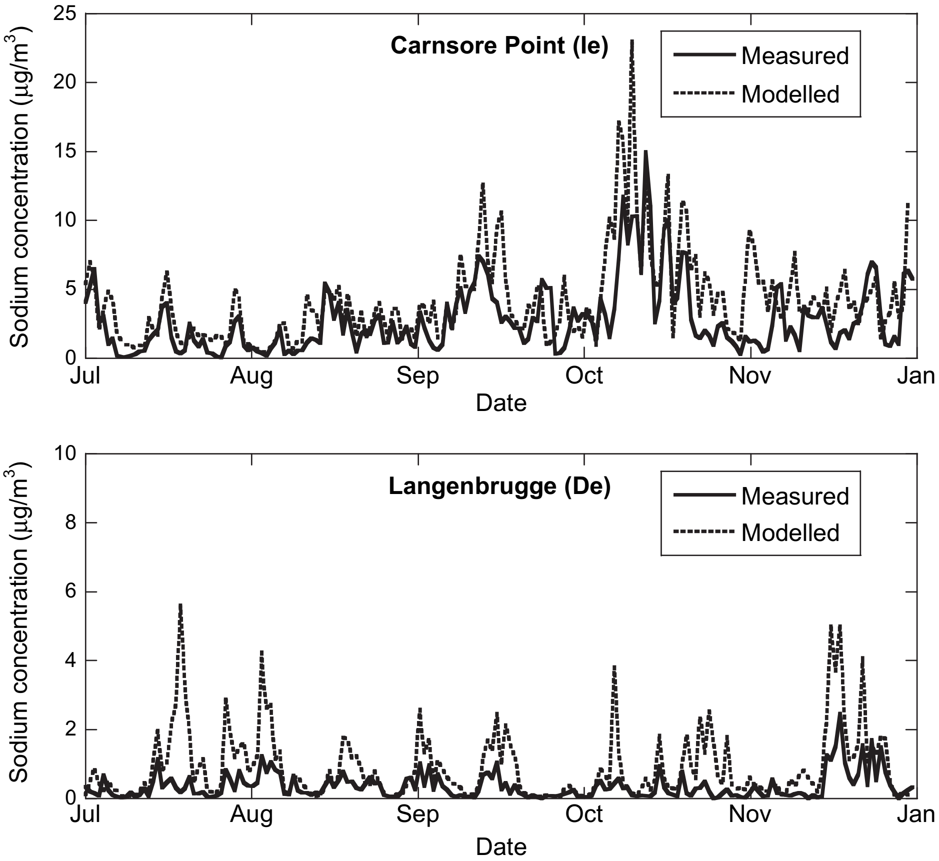

Fig. 5. Time series of observed and modelled concentrations at Langenbrugge (Germany) and Carnsore Point (Ireland) for 2005.

3.3. Annual average sea salt distribution for 2005

The comparison of the observations and modelling results showed that, although the absolute level is overestimated, the modelled spatial sodium distribution is satisfactory. Therefore, we feel that it is justified to divide the annual average modelled distribution by 2.46 to obtain a best estimate for the sea salt sodium distribution across Europe for 2005. To arrive at a best estimate for the sea salt distribution the sodium concentrations were multiplied by 3.26 to account for the other components of sea salt. The resulting distribution is shown in Fig. 3b.

The northern and western coasts of Ireland and the UK exhibit the peak annual average concentrations, reaching up to 13 µg m^{-3}. Sea salt concentrations are around 5 µg m^{-3} at the Atlantic and North Sea coasts, while at a distance of 200–300 km, when no mountains are present which prohibit transport inland, concentrations are between 2 and 5 µg m^{-3} (Ireland, the UK, the Netherlands, Denmark). Similar sea salt concentrations are predicted for the Atlantic coast of Spain and at the Mediterranean Islands. Less sea salt affected areas (above 0.5 µg m^{-3}) are mainland Spain, France, Germany, southern Italy and southern Scandinavia. Remote continental locations show the lowest sea salt concentrations (averages below 0.5 µg m^{-3}).

The estimation of the annual average sea salt distribution is evaluated against data that belong to additional measuring sites in Spain, Germany and Ireland (Table 3, Fig. 3b), which were not used for the scaling of the model results and are thus fully independent. Comparison between the annual mean sea salt concentrations

Table 2
Statistical comparison between the observed and modelled sodium concentrations for 2005. The annual average concentration (µg m^{-3}) and the standard deviation for the observations and the model are given, both for the sampling days (paired) and for the whole simulation period, as well as the correlation of the paired data. The number (N) of available data points are indicated for each station.

Station (*N*)	Observed average (µg m^{-3})	Observed stdev (µg m^{-3})	Modelled average (µg m^{-3})	Modelled stdev (µg m^{-3})	Correlation	Modelled average (µg m^{-3})	Modelled stdev (µg m^{-3})
			Paired	Paired		All year	All year
Ulborg (347)	1.52	1.33	4.39	5.14	0.71	4.43	5.13
Anholt (360)	1.85	3.28	3.60	2.96	0.45	3.57	2.92
Langenbrugge (209)	0.39	0.39	0.93	1.05	0.70	0.92	0.98
Malin Head (360)	2.42	2.15	7.07	7.58	0.56	7.11	7.62
Oak Park (361)	0.71	0.58	2.83	1.73	0.67	2.84	1.73
Carnsore Point (245)	2.85	2.44	4.53	3.32	0.67	4.10	3.12
Barcelona (80)	0.88	0.68	0.68	0.45	0.49	0.65	0.46
Montseny (99)	0.26	0.24	0.64	0.43	0.43	0.63	0.53
Onda (62)	0.48	0.31	0.54	0.43	0.06	0.49	0.36

Table 3

Additional data set for sodium used for evaluation of the derived sea salt distribution. For each station the name, country code, location, the years during which the measurements were made, the sodium concentration (μg m^{-3}) and data source is given.

Site name	Country	Lon	Lat	Year	Na	Reference
Melpitz	DEU	12.9	51.5	2004–2005	0.36	Spindler et al., (2007)
Hortenkopf	DEU	7.81	49.27	2002–2003	0.57	Kuhlbusch et al. (2003)
Mainz	DEU	8.26	50,06	2002–2003	0.53	Kuhlbusch et al. (2003)
Koblenz	DEU	7.58	50.35	2002–2003	0.59	Kuhlbusch et al. (2003)
Ludwigshafen	DEU	8.43	49.46	2002–2003	0.63	Kuhlbusch et al. (2003)
Duisburg	DEU	6.79	51.43	2002–2003	0.66	Quass et al. (2004)
Villar del Arzobispo	ESP	0.50	39.44	2004–2005	0.24	Viana et al. (2008)
Melilla	ESP	−2.94	35.29	2007	1.92	SME (2009)
Madrid	ESP	−3.68	40.43	2007	0.38	SME (2009)
Santander	ESP	−3.80	43.45	2007	2.02	SME (2009)
Torrelavega	ESP	−4.03	43.33	2007	1.08	SME (2009)
Ponferrada	ESP	−6.57	42.55	2007	0.42	SME (2009)
Mace Head	IRL	−9.9	53.33	2001–2006	3.98	Cerburnis (2009)

based on these measurements and the respective modelled and scaled PM10 values shows a satisfactory fit, even considering that the modelled year and the years when the experimental data were obtained are not coincident. Hence, this limited verification gives additional confidence in the quality of the derived distribution.

4. Discussion

The compilation of sodium observations, collected from different European studies, provides a good insight in the sea salt distribution across Europe. At coastal sites along the Atlantic and North Sea coast concentrations tend to be around 5 μg m^{-3}. More inland locations up to about 300 km away from the coast tend to show concentrations between 2 and 5 μg m^{-3}, whereas sites further away from the coast are characterized by lower concentrations. These concentration ranges are highly consistent with earlier compilations directed to the chemical speciation of PM10 (e.g. Putaud et al., 2004; Querol et al., 2004a, 2004b, 2009). This is not a surprise since many of the stations included in these earlier compilations are also used here, though we have limited ourselves to sea salt estimates based on sodium as a tracer only. Compared to the abovementioned studies we have considerably expanded the number of sites, incorporated recent data and updated the concentration data when longer time series were available. As such, the compilation of observations provides a larger data set for model validation in Europe than used before.

The compiled data set is heterogeneous with respect to measurement approach and representativity in time. The representativity in time can be addressed through the inter-annual variability, which was found to be moderately high. The inter-annual variability was also studied in the NATAIR project (Grice et al., 2008) by modelling the marine emissions around Europe for several years. They arrived at an inter-annual variability of up to 40%. The present compilation of observations supports these results. Also, using an older version of LOTOS-EUROS the period 2000–2006 has been modelled, leading to similar results (not shown). Ideally, longer time series are needed for a good representation of the inter-annual variability, but the present estimates give a consistent indication.

The modelling of the processes that impact the sea salt concentration remains a challenge. The model represented the spatial and temporal variability of the observed sea salt concentrations rather well. However, the absolute concentrations are severely overestimated. This means that the temporal and spatial variability of important parameters such as the emission strength and deposition velocities are understood in general terms. In contrast, the balance between the source and the sinks, i.e. deposition, is not. This is a general problem evidenced by many model evaluations that show reasonable temporal correlation coefficients between observed and modelled daily concentrations in combination with an under- or overestimation of the measured annual mean concentrations up to a factor three (e.g. Foltescu et al. (2005); Witek et al. (2007b); Textor et al., 2006).

The available emission parameterizations, e.g. Monahan et al. (1986), Smith et al. (1993), Gong et al., 2003; Mårtensson et al., 2003; Clarke et al. (2006), show a considerable variability. The source functions, Mårtensson et al. (2003) and Monahan et al. (1986), which are used here, are well established, but are empirical approaches. This implies that they may not be universally valid. Recent reviews give an uncertainty of a factor of 2–3 (O'Dowd and De Leeuw, 2007; Clarke et al., 2006), which is a less pessimistic estimate than the factor 7 previously estimated by Lewis and Schwartz (2004). The uncertainty in the emission strength causes a corresponding uncertainty in sea salt aerosol concentrations, which makes the model results very sensitive to the choice of the source function.

Most emission parameterizations are based on campaign data obtained on open ocean or are based on tank measurements. Generation of sea salt particles is mainly parameterised as function of wind speed and only Mårtensson includes a sea surface temperature effect. However, a large number of other factors influence whitecap cover and thereby the emission strength. These include e.g. wave state, salinity and concentration and type of surface active materials (Anguelova and Webster, 2006; Witek et al., 2007a). The impact of these parameters is neglected in current parameterizations but may be important for near coastal areas and smaller sea bodies, such as the Mediterranean Sea and the Baltic. Witek et al. (2007a) showed that the inclusion of wave related parameters such as orbital velocity in the source function, may improve the correlation of modelled sea salt concentrations with observed concentrations.

Also, Callaghan et al. (2008) showed that the whitecap cover increases less strongly with the 10 m wind than is parameterised in Monahan and Muircheartaigh (1980) above about 10 m s^{-1}. In a sensitivity analysis, we put a maximum on the source function by cutting off the effective wind speed at 12.5. This indeed reduced the annual mean concentration up to 40% on the locations with the highest wind speeds but on the continent the effect is smaller than 20% (not shown). Hence, compared to the abovementioned uncertainty this effect on continental concentrations is small. New approaches to estimate and parameterize whitecap cover through for example remote sensing (Anguelova and Webster, 2006) or to measure emission fluxes (Norris et al., 2008) may improve our understanding and emission parameterizations in the future.

Another source for uncertainty is associated with the dry deposition process. Textor et al. (2006) recognized the large differences between different global models with respect to (dry) deposition fluxes and efficiencies. LOTOS-EUROS includes besides the

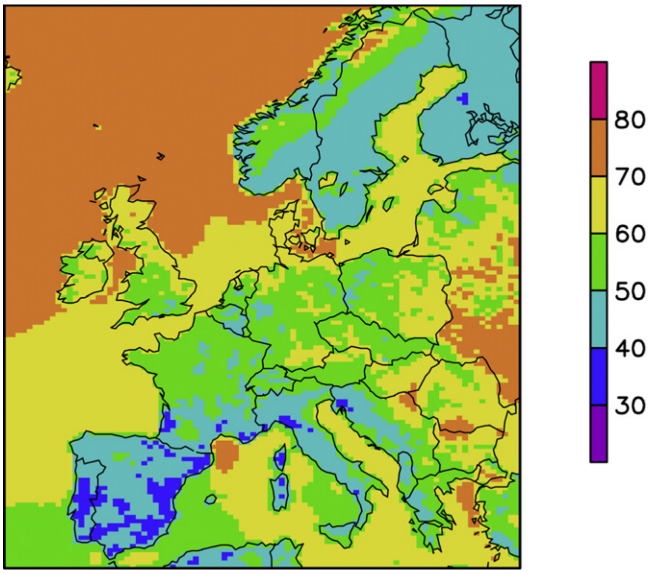

Fig. 6. Ratio (%) of the annual average sodium concentration using the DEPAC dry deposition module to that using the Zhang et al. (2001) scheme across Europe.

Zhang et al. (2001) scheme a second approach, DEPAC, for calculation of dry deposition velocities following Erisman et al. (1994). The latter is based on empirical data obtained in the Netherlands and provides systematically higher deposition velocities than in Zhang et al. (2001). A sensitivity simulation shows that sea salt concentrations are nearly a factor 2 lower when DEPAC was used (Fig. 6). In particular at sea coarse aerosol is deposited more efficiently. This is at least partly caused by the high gravitational settling velocities in DEPAC, which are based on particles of 10 μm diameter. In short, particle dry deposition is also a major area for model improvement.

The comparison of the observations and modelling results showed an overestimation of a factor of 2.46 of the model. This factor is well within the uncertainty of the parameterization of the source function and the dry deposition process, as discussed above. Also, the model explained the spatial gradients observed in ambient sodium data very well. Both arguments support the approach followed here to scale the annual mean modelled concentrations to provide a domain covering estimate of the sea salt distribution that is consistent with observations. The limited verification of the distribution supports the results for the annual mean distribution. However, for a day by day assessment of the sea salt concentrations we feel that the model needs to be improved on the above-mentioned points and that the results can only be used in a more indicative or qualitative sense.

5. Conclusions

In Europe sea salt may contribute significantly to particulate matter concentrations. The annual average sea salt concentrations above land were estimated to range between 0.3 and almost 13 μg m^{-3}. Maximum concentrations are found at the Irish coast. At coastal sites along the Atlantic and North Sea coast concentrations tend to be around 5 μg m^{-3}. More inland locations up to about 300 km away from the coast tend to show concentrations between 2 and 5 μg m^{-3}, whereas sites further away from the coast are characterized by lower concentrations. An analysis of the representativity of the data with respect to a long term average showed that annual mean sodium concentrations are associated with a standard deviation of around 15%. The compilation of observations provides an improved overview of sea salt concentrations in Europe as well as an improved basis for model validation. Verification of the results of the LOTOS-EUROS model learned that the represents the spatial variability of the observed sea salt concentrations very well. However, the absolute concentrations are significantly overestimated due to large uncertainties in the emission and dry deposition parameterizations. Using the high explained variability in the gradients across Europe, the bias-corrected modelled distribution serves as a best estimate of the sea salt distribution across Europe for 2005.

Acknowledgement

The authors are grateful for the support through the Netherlands Research Program on Particulate Matter (BOP), funded by the Netherlands Ministry of Housing, Spatial planning and the Environment (VROM). The technical assistance of M. Jozwicka (TNO) with the compilation of the data set is gratefully acknowledged.

References

Anguelova, M.D., Webster, F., 2006. Whitecap coverage from satellite measurements: a first step toward modeling the variability of oceanic whitecaps. J. Geophys. Res. C: Oceans 111 (3) art. no. C03017, doi:10.1029/2005JC003158.

Barbu, A.L., Segers, A.J., Schaap, M., Heemink, A.W., Builtjes, P.J.H., 2009. A multi-component data assimilation experiment directed to sulphur dioxide and sulphate over Europe. Atmos. Environ. 43 (9), 1622–1631.

Beekmann, M., Kerschbaumer, A., Reimer, E., Stern, R., Möller, D., 2007. PM measurement campaign HOVERT in the Greater Berlin area: model evaluation with chemically specified particulate matter observations for a one year period. Atmos. Chem. Phys. 7 (1), 55–68.

Bessagnet, B., Hodzic, A., Vautard, R., Beekmann, M., Cheinet, S., Honoré, C., Liousse, C., Rouil, L., 2004. Aerosol modeling with CHIMERE: preliminary evaluation at the continental scale. Atmos. Environ. 38, 2803–2817.

Callaghan, A., De Leeuw, G., Cohen, L., O'Dowd, C.D., 2008. Relationship of oceanic whitecap coverage to wind speed and wind history. Geophys. Res. Lett. 35 L23609.

Clarke, A., Owens, S., Zhou, J., 2006. An ultrafine sea-salt flux from breaking waves: implications for CCN in the remote marine atmosphere. J. Geophys. Res. 111 (D06202) doi:10.1029/2005JD006565.

Cerburnis, D. (2009). Personal communication.

Cuvelier, C., Thunis, P., Vautard, R., Amann, M., Bessagnet, B., Bedogni, M., Berkowicz, R., Brandt, J., Brocheton, F., Builtjes, P., Coppalle, A., Denby, B., Douros, G., Graf, A., Hellmuth, O., Honoré, C., Hodzic, A., Jonson, J., Kerschbaumer, A., de Leeuw, F., Minguzzi, E., Moussiopoulos, N., Pertot, C., Pirovano, G., Rouil, L., Schaap, M., Stern, R., Tarrason, L., Vignati, E., Volta, M., White, L., Wind, P., Zuber, A., 2007. CityDelta: a model intercomparison study to explore the impact of emission reductions in European cities in 2010. Atmos. Environ. 41, 189–207.

De Leeuw, G., Neele, F.P., Hill, M., Smith, M.H., Vignati, E., 2000. Production of sea spray aerosol in the surf zone. J. Geophys. Res. 105, 29397–29409.

Denby, B., Schaap, M., Segers, A., Builtjes, P., Horalek, J., 2008. Comparison of two data assimilation methods for assessing PM10 exceedances on the European scale. Atmos. Environ. 42, 7122–7134.

EMEP (2009). www.emep.int.

Erisman, J.W., van Pul, A., Wyers, P., 1994. Parametrization of surface-resistance for the quantification of atmospheric deposition of acidifying pollutants and ozone. Atmos. Environ. 28, 2595–2607.

Finlayson-Pitts, B.J., 2003. The tropospheric chemistry of sea salt: a molecular-level view of the chemistry of NaCl and NaBr. Chem. Rev. 103, 4801–4822.

Foltescu, V.L., Pryor, S.C., Bennet, C., 2005. Sea salt generation, dispersion and removal on the regional scale. Atmos. Environ. 39, 2123–2133.

Gong, S.L., Barrie, L.A., Blanchet, J.-P., 1997. Modeling sea salt aerosols in the atmosphere: 1. Model development. J. Geophys. Res. 102, 3805–3818.

Gong, S.L., Barrie, L.A., Blanchet, J.-P., von Salzen, K., Lohmann, U., Lesins, G., Spacek, L., Zhang, L.M., Girard, E., Lin, H., Leaitch, R., Leighton, H., Chylek, P., Huang, P., 2003. Canadian aerosol module: a size-segregated simulation of atmospheric aerosol processes for climate and air quality models. 1. Module development. J. Geophys. Res. 108 (D1) 2007.

Grice, S., Sturman, J., Dore, Ch., Woodfield, M., 2008. Modelling emissions of marine aerosol from the ocean at a European scale, publication of the NATAIR project, http://natair.ier.uni-stuttgart.de/GriceetalNatairseasaltemission.pdf.

Guelle, W., Schulz, M., Balkanski, Y., Dentener, F., 2001. Influence of the source formulation on modeling the atmospheric global distribution of sea salt aerosol. J. Geophys. Res. 106, 27509–27524.

Hass, H., Builtjes, P.J.H., Simpson, D., Stern, R., 1997. Comparison of model results obtained with several European regional air quality models. Atmos. Environ. 31, 3259–3279.

Hass, H., van Loon, M., Kessler, C., Stern, R., Matthijsen, J., Sauter, F., Zlatev, Z., Langner, J., Foltescu, V., Schaap, M., 2003. Aerosol modelling: results and intercomparison from European regional – scale modelling systems. Special Rep. EUROTRAC-2 ISS, Munchen.

Hueglin, C., Gehrig, R., Baltensperger, U., Gysel, M., Monn, C., Vonmont, H., 2005. Chemical characterisation of PM2.5, PM10 and coarse particles at urban, near-city and rural sites in Switzerland. Geophys. Res. Lett. 39, 637–651.

Knipping, E.M., Dabdub, D., 2003. Impact of chlorine emissions from sea-salt aerosol on coastal urban ozone. Environ. Sci. Technol. 37, 275–284.

Kuhlbusch, T., John, A., Romazanowa, O., Top, S., 2003. Identifizierung von PM10-Emissionsquellen im Rahmen der Maßnahmenplanung zur Reduktion der PM10-Immissionsbelastung in Rheinland-Pfalz, Abschlussbericht an das Landesamt für Umweltschutz und Gewerbeaufsicht Rheinland-Pfalz Duisburg, Germany.

Langmann, B., Varghese, S., Marmer, E., Vignati, E., Wilson, J., Stier, P., O'Dowd, C., 2008. Aerosol distribution over Europe: a model evaluation study with detailed aerosol microphysics. Atmos. Chem. Phys. 8 (6), 1591–1607.

Lee, D.S., Garland, J.A., Fox, A.A., 1994. Atmospheric concentrations of trace elements in urban areas of the United Kingdom. Atmos. Environ. 28, 2691–2713.

Lewis, E.R., Schwartz, S.E., 2004. Sea Salt Aerosol Production. AGU, Washington, DC.

Lucarelli, F., Mando, P.A., Nava, S., Prati, P., Zucchiatti, A., 2004. One-year study of the elemental composition and source apportionment of PM10 aerosols in Florence, Italy. J. Air. Waste. Manag. Assoc. 54, 1372–1382.

Manders, A.M.M., Schaap, M., Hoogerbrugge, R., 2009a. Testing the capability of the chemistry transport model LOTOS-EUROS to forecast PM10 levels in the Netherlands. Atmos. Environ. 43 (26), 4050–4059.

Manders, A.M.M., Schaap, M., Jozwicka, M., Van Arkel, F., Weijers, E.P., Matthijsen, J., (2009b). The contribution of sea salt to PM10 and PM2.5 in the Netherlands, PBL report 500099004, PBL, Bilthoven, the Netherlands, http://www.pbl.nl/en/publications.

Mårtensson, E.M., Nilsson, E.D., de Leeuw, G., Cohen, L.H., Hansson, H.-C., 2003. Laboratory simulations and parameterization of the primary marine aerosol production. J.Geophys. Res. 108.

Mazzei, F., D'Alessandro, A., Lucarelli, F., Marenco, F., Nava, S., Prati, P., Valli, G., Vecchi, R., August 2006. Elemental composition and source apportionment of particulate matter near a steel plant in Genoa (Italy). Nucl. Instrum. Methods Phys. Res. Sect. B 249 (1–2), 548–551. SPEC. ISS.

McInnes, L.M., Covert, D.S., Quinn, P.K., Germani, M.S., 1994. Measurements of chloride depletion and sulfur enrichment in individual sea-salt particles collected from the remote marine boundary layer. J. Geophys. Res. 99, 8257–8268.

Millero, F.J., 2004. Physicochemical controls on seawater. In: Holland, H.D., Turkian, K.K. (Eds.), Treatise on Geochemistry. Elsevier, Amsterdam (Chapter 6.01).

Monahan, E.C., Muircheartaigh, I.O., 12, Dec. 1980. Optimal power-law description of oceanic whitecap coverage dependence on wind speed. J. Phys. Oceanogr. 10, 2094–2099.

Monahan, E.C., Spiel, D.E., Davidson, K.L., 1986. A model of marine aerosol generation via whitecaps and wave disruption. In: Monahan, E.C., Mac Niocaill, G. (Eds.), Oceanic Whitecaps and Their Role in Air-Sea exchange.

Muster, T.H., Cole, I.S., 2005. Attachment efficiencies of salt aerosols onto infrastructure and implications for atmospheric corrosion. J. Electrochem. Soc. 152, B125–B131.

Norris, S., Brooks, I., De Leeuw, G., Smith, M.H., Moerman, M., Lingard, J., 2008. Eddy covariance measurements of sea spray particles over the Atlantic Ocean. Atmos. Chem. Phys. 8, 555–563.

O'Dowd, C.D., De Leeuw, G., 2007. Marine aerosol production: a review of the current knowledge. Philos. Trans.R. Soc. London, Ser. A 365 (1856), 1753–1774.

Putaud, J., Raes, F., Van Dingenen, R., Bruggemann, E., Facchini, M., Decesari, S., Fuzzi, S., Gehrig, R., Hueglin, C., Laj, P., Lorbeer, G., Maenhaut, W., Mihalopoulos, N., Mueller, K., Querol, X., Rodriguez, S., Schneider, J., Spindler, G., ten Brink, H., Torseth, K., Wiedensohler, A., 2004. A European aerosol phenomenology—2:chemical characteristics of particulate matter at kerbside, urban, rural and background sites in Europe. Atmos. Environ. 38, 2579–2595.

Puxbaum, H., Gomišcek, B., Kalina, M., Bauer, H., Salam, A., Stopper, S., Preining, O., Hauck, H., 2004. A dual site study of PM2.5 and PM10 aerosol chemistry in the larger region of Vienna, Austria. Atmos. Environ. 38, 3949–3958.

Qin, Y., Oduyemi, K., 2003. Chemical composition of atmospheric aerosol in Dundee, UK. Atmos. Environ. 37, 93–105.

Quass, U., Kuhlbusch, T., Fissan, H., Schmidt, K.-G., Bruckmann, P., Pfeffer, U., Gladtke, D., Zang T. 2004. Identification of Source groups for Fine Dust, final report to the Ministerium für Umwelt und Naturschutz, Landwirtschaft und Verbraucherschutz des Landes Nordrhein-Westfalen, Duisburg, Germany.

Querol, X., Alastuey, A., Ruiz, C.R., Artiñano, B., Hansson, H.C., Harrison, R.M., Buringh, E., Ten Brink, H.M., Lutz, M., Bruckmann, P., Straehl, P., Schneider, J., 2004a. Speciation and origin of PM10 and PM2.5 in selected European cities. Atmos. Environ. 38, 6547–6555.

Querol, X., Alastuey, A., Moreno, T., Viana, M.M., Castillo, S., Pey, J., Rodríguez, S., Artiñano, B., Salvador, P., Sánchez, M., Garcia Dos Santos, S., Herce Garraleta, M. D., Fernandez-Patier, R., Moreno-Grau, S., Negral, L., Minguillón, M.C., Monfort, E., Sanz, M.J., Palomo-Marín, R., Pinilla-Gil, E., Cuevas, E., de la Rosa, J., Sánchez de la Campa, A., 2004b. Spatial and temporal variations in airborne particulate matter (PM$_{10}$ and PM$_{2.5}$) across Spain 1999–2005. Atmos. Environ. 42, 3964–3979.

Querol, X., Alastuey, A., Pey, J., Cusack, M., Pérez, N., Mihalopoulos, N., Theodosi, C., Gerasopoulos, E., Kubilay, N., Kocak, M., 2009. Variability in regional background aerosols within the Mediterranean. Atmos. Chem. Phys. 9, 4575–4591.

Querol, X., Alastuey, A., Moreno, T., Viana, M.M., Castillo, S., Pey, J., Rodríguez, S., Artiñano, B., Salvador, P., Sánchez, M., Garcia Dos Santos, S., Herce Garraleta, M.D., Fernandez-Patier, R., Moreno-Grau, S. , Negral, L., Minguillón, M.C., Monfort, E., Sanz, M.J., Palomo-Marín, R., Pinilla-Gil, E., Cuevas, E., de la Rosa, J., Sánchez de la Campa, A., 2008. Spatial and temporal variations in airborne particulate matter (PM10 and PM2.5) across Spain 1999–2005. Atmos. Environ. 42, 3964–3979.

Rodríguez, S., Querol, X., Alastuey, A., Viana, M.M., Alarcón, M., Mantilla, E., Ruiz, C. R., Jul 26 2004. Comparative PM10-PM2.5 source contribution study at rural, urban and industrial sites during PM episodes in Eastern Spain. Sci Total Environ. 328 (1–3), 95–113.

Schaap, M., van Loon, M., ten Brink, H.M., Dentener, F.D., Builtjes, P.J.H., 2004a. Secondary inorganic aerosol simulations for Europe with special attention to nitrate. Atmos. Phys. Chem. 4, 857–874.

Schaap, M., Denier Van Der Gon, H.A.C., Dentener, F.J., Visschedijk, A.J.H., van Loon, M., Ten Brink, H.M., Putaud, J.-P., Guillaume, B., Liousse, C., Builtjes, P.J.H., 2004b. Anthropogenic black carbon and fine aerosol distribution over Europe. J. Geophys. Res. 109 (D18201). doi:10.1029/2003JD004330.

Schaap, M., Timmermans, R.M.A., Sauter, F.J., Roemer, M., Velders, G.J.M., C Boersen, G.A., Beck, J.P., Builtjes, P.J.H., 2008. The LOTOS-EUROS model: description, validation and latest developments. Int. J. of Environ. and Pollution 32 (No. 2), 270–290.

Slanina, J., ten Brink, H.M., Otjes, R.P., Even, A., Jongejan, P., Khlystov, A., Waijers-Ijpelaan, A., Hu, M., Lu, Y., 2001. Continuous analysis of nitrate and ammonium in aerosols by the Steam Jet Aerosol Collector (SJAC). Atmos. Environ. 35, 2319–2330.

SME 2009. Levels, composition and sources of PM10, PM2.5 and PM1 in Spain: Cantabria, Castilla, León, Madrid and Melilla. SME report, Madrid, Spain (in Spanish).

Smith, M.H., Park, P.M., Consterdine, I.E., 1993. Marine aerosol concentrations and estimated fluxes over the sea. Q. J. R. Meteorolog. Soc. 119 (512), 809–824.

Spindler, G., Brüggemann, E., Gnauk, T., Grüner, A., Müller, K., Tuch, Th.M., Wallasch, M.B., Wehner, A., Herrmann, H., August 2007. 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/CCC-Report 8/2007, ref. O-7726, 178–221.

Stern, R., Builtjes, P., Schaap, M., Timmermans, R., Vautard, R., Hodzic, A., Memmesheimer, M., Feldmann, H., Renner, E., Wolke, R., Kerschbaumer, A., 2008. A model inter-comparison study focussing on episodes with elevated PM10 concentrations. Atmos. Environ. 42, 4567–4588.

Stier, P., Feichter, J., Kinne, S., Kloster, S., Vignati, E., Wilson, J., Ganzeveld, L., Tegen, I., Werner, M., Balkanski, Y., Schulz, M., Boucher, O., Minikin, A., Petzold, A., 2005. The aerosol-climate model ECHAM5-HAM. Atmos. Chem. Phys. 5 (4), 1125–1156.

Tamm, S., Schulz, M., 2003. Open-ocean aerosol composition obtained during 15 months on a North Sea ferry. Atmos. Environ. 37 (Suppl. 1), S133–S143.

Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Feichter, H., Fillmore, D., Ghan, S., Ginoux, P., Gong, S., Grini, A., Hendricks, J., Horowitz, L., Huang, P., Isaksen, I., Iversen, T., Kloster, S., Koch, D., Kirkevåg, A., Kristjansson, J. E., Krol, M., Lauer, A., Lamarque, J.F., Liu, X., Montanaro, V., Myhre, G., Penner, J., Pitari, G., Reddy, S., Seland, Ø., Stier, P., Takemura, T., Tie, X., 2006. Analysis and quantification of the diversities of aerosol life cycles within AeroCom. Atmos. Chem. Phys. 6 (7), 1777–1813.

Van Loon, M., Tarrason, L., Posch, M., 2005. Modelling base cations in Europe, EMEP MSC-W Technical Report 2/05, EMEP, OSLO, Norway, www.emep.int.

van Loon, M., Vautard, R., Schaap, M., Bergström, R., Bessagnet, B., Brandt, J., Builtjes, P. J.H., Christensen, J.H., Cuvelier, K., Graf, A., Jonson, J.E., Krol, M., Langner, J., Roberts, P., Rouil, L., Stern, R., Tarrasón, L., Thunis, P., Vignati, E., White, L., Wind, P., 2007. Evaluation of long-term ozone simulations from seven regional air quality models and their ensemble average. Atmos. Environ. 41, 2083–2097.

Vercauteren, J., 2009. Chemkar PM10: Chemische karakterisatie van fijn stof in Vlaanderen, VVM-report D/2009/6871/015, VMM, Belgium, http://www.vmm.be/publicaties/2009/CK_PM10_TW.pdf.

Viana, M., Querol, X., Ballester, F., Llop, S., Esplugues, A., Fernández Patier, R., García Dos Santos, S., Herce, M.D., 2008. Characterising exposure to PM aerosols for an epidemiological study. Atmos. Environ. 42, 1552–1568.

White, W.H., 2008. Chemical markers for sea salt in IMPROVE aerosol data. Atmos. Environ. 42, 261–274.

Witek, M.L., Flatau, P.J., Teixeira, J.., Westphal, D.L., 2007a. Coupling an ocean wave model with a global aerosol transport model: a sea salt aerosol parameterization perspective. Geophys. Res. Let. 34.

Witek, M.L., Flatau, P.J., Quinn, P.K., Westphal, D.L., 2007b. Global sea-salt modelling: results and validation against multicampaign shipboard measurements. J. Geophys. Res. 112.

Yoon, Y.J., Brimblecombe, P., 2002. Modelling the contribution of sea salt and dimethyl sulfide derived aerosol to marine CCN. Atmos. Chem. Phys. 2, 17–30.

Zhang, L., Gong, S., Padro, J., Barrie, L., 2001. A size-segregated particle dry deposition scheme for an atmospheric aerosol module. Atmos. Environ. 35, 549–560.