Modeling and evaluation of the global sea-salt aerosol distribution: sensitivity to emission schemes and resolution effects at coastal/orographic sites

M. Spada\textsuperscript{1}, O. Jorba\textsuperscript{1}, C. Pérez García-Pando\textsuperscript{2,3}, Z. Janjic\textsuperscript{4}, and J. M. Baldasano\textsuperscript{1,5}

\textsuperscript{1}Barcelona Supercomputing Center – Centro Nacional de Supercomputación, Barcelona, Spain
\textsuperscript{2}NASA Goddard Institute for Space Studies, New York, USA
\textsuperscript{3}Department of Applied Physics and Applied Math, Columbia University, New York, USA
\textsuperscript{4}National Centers for Environmental Prediction, College Park, Maryland, USA
\textsuperscript{5}Universitat Politècnica de Catalunya, Barcelona, Spain

Correspondence to: M. Spada (michele.spada@bsc.es) and O. Jorba (oriol.jorba@bsc.es)

Received: 21 March 2013 – Published in Atmos. Chem. Phys. Discuss.: 2 May 2013
Revised: 9 October 2013 – Accepted: 2 November 2013 – Published: 4 December 2013

Abstract. One of the major sources of uncertainty in model estimates of the global sea-salt aerosol distribution is the emission parameterization. We evaluate a new sea-salt aerosol life cycle module coupled to the online multiscale chemical transport model NMMB/BSC-CTM. We compare 5 yr global simulations using five state-of-the-art sea-salt open-ocean emission schemes with monthly averaged coarse aerosol optical depth (AOD) from selected AERONET sun photometers, surface concentration measurements from the University of Miami’s Ocean Aerosol Network, and measurements from two NOAA/PMEL cruises (AEROINDOEX and ACE1). Model results are highly sensitive to the introduction of sea-surface-temperature (SST)-dependent emissions and to the accounting of spume particles production. Emission ranges from 3888 Tg yr\textsuperscript{-1} to 8114 Tg yr\textsuperscript{-1}, lifetime varies between 7.3 h and 11.3 h, and the average column mass load is between 5.0 Tg and 7.2 Tg. Coarse AOD is reproduced with an overall correlation of around 0.5 and with normalized biases ranging from +8.8 % to +38.8 %. Surface concentration is simulated with normalized biases ranging from −9.5 % to +28 % and the correlation is around 0.5. Our results indicate that SST-dependent emission schemes improve the overall model performance in reproducing surface concentrations. On the other hand, they lead to an overestimation of the coarse AOD at tropical latitudes, although it may be affected by uncertainties in the comparison due to the use of all-sky model AOD, the treatment of water uptake, deposition and optical properties in the model and/or an inaccurate size distribution at emission.

1 Introduction

Sea salt is one of the most abundant aerosol species globally. It perturbs the radiative fluxes directly by interacting with shortwave and longwave radiation, and indirectly by acting as cloud condensation nuclei (CCN) and thus altering marine cloud brightness and lifetime. It also influences heterogeneous chemistry mainly over coastal areas (Lewis and Schwartz, 2004) and is co-emitted with organic aerosols (Tsigaridis et al., 2013). The major uncertainties in the sea-salt life cycle are emission (Textor et al., 2006; de Leeuw et al., 2011), water uptake (Textor et al., 2006), and deposition (Textor et al., 2007). Lewis and Schwartz (2004) estimate the total sea-salt aerosol emission to vary from 0.3 Tg yr\textsuperscript{-1} to 30 Tg yr\textsuperscript{-1} and estimates from models involved in the AEROCOM project range from 3 Tg yr\textsuperscript{-1} to 18 Tg yr\textsuperscript{-1} for year 2000 (Textor et al., 2006). These uncertainties may lead to differences of a factor of two or more in the simulated monthly averaged concentrations among different models, and between simulated and observed concentrations (Textor et al., 2006). The lack of comprehensive measurement data sets hampers evaluation efforts and the improvement of sea-salt models and related parameterizations. For a given region...
and a given time period, only a few coincident measurements of surface concentration, aerosol optical depth (AOD), and particle-size distribution are available, and a few emission and deposition flux estimates at specific sites and temporal intervals can be found in literature. Additional difficulties arise from biases in satellite retrievals, particularly in the most important sea-salt production regions (e.g. Jaeglé et al., 2011).

Several approaches are typically used to parameterize the sea-salt emission process, from semi-empirical combinations of whitecap factorization and concentration measurements (Monahan et al., 1986; Smith et al., 1993; Smith and Harrison, 1998; Andreas, 1998; Hoppel et al., 2002; Gong, 2003; Petelski et al., 2005; Mårtensson et al., 2003; Clarke et al., 2006; Caffrey et al., 2006; Jaeglé et al., 2011; Fan and Toon, 2011), to empirical methods such as the use of concentration vertical profiles from aircraft observations (Reid et al., 2001). Parameterizations of sea-salt emission fluxes may account for different production mechanisms (e.g. bubble bursting, spume cutting), which may depend on different meteorological parameters. The most used parameter is wind speed at 10 m ($U_{10}$), but there have also been attempts to include dependencies on sea surface temperature (SST), wave height, increasing/decreasing wind, salinity and other parameters. Exhaustive reviews of these efforts and their performance can be found in Lewis and Schwartz (2004), O’Dowd and de Leeuw (2007), de Leeuw et al. (2011), and Grythe et al. (2013). The above-mentioned parameterizations are assumed for the open ocean. Production in the surf-zone represents an additional open issue (de Leeuw et al., 2000).

The high hygroscopicity of sea-salt requires water uptake schemes based on prescribed growth factors (Chin et al., 2002) or equations (Gerber, 1985; Ghan et al., 2001) or explicit calculations of the condensed aerosol water (Vignati et al., 2004). However, their performance is hard to assess and it remains an open topic for aerosol modeling (Textor et al., 2006).

In this contribution, we investigate the uncertainties associated with sea-salt, open-ocean emission schemes. We use a new sea-salt module coupled online to the multiscale NMmB/BSC Chemical Transport Model (NMmB/BSC-CTM) (Pérez et al., 2011; Haustein et al., 2012; Jorba et al., 2012), developed at the Barcelona Supercomputing Center in collaboration with NOAA/National Centers for Environmental Prediction (NCEP) and the NASA Goddard Institute for Space Studies. Its meteorological core, the Non-hydrostatic Multiscale Model (NMmB) (Janjic, 2005; Janjic and Black, 2007; Janjic et al., 2011; Janjic and Gall, 2012) allows for a bridging of the gap between global, regional, and local scales by using consistent dynamics and physics formulations.

In Sects. 2 and 3 we present the modeling system along with details of the sea-salt module development. We implement five emission parameterizations following the whitecap approach, in order to investigate this major source of uncertainty. Particular attention is given to the description of spume particles production and to the dependence of emissions upon SST. In Sect. 4 we present the observational data sets used for the evaluation of the global simulations. Results and discussion of 5 yr simulations (2002–2006) are presented in Sect. 5. Evaluation is performed against cruise data from the NOAA/PMEL Laboratory, sea-salt concentration monthly climatologies from the University of Miami Ocean Aerosol Network, and monthly averaged measurements from distributed AERONET sun photometers. Our results are also compared with other recent model studies, such as Jaeglé et al. (2011) and Tsagaridis et al. (2013).

2 Modeling background

The NMmB/BSC-CTM is a fully online chemical transport model coupling the atmospheric equations of NMmB with the gas-phase and aerosol continuity equations of BSC-CTM. At the present stage of development aerosol species included in the model are dust and sea-salt. The implementation and evaluation of other globally relevant aerosols is underway. Details on the dust aerosol module and gas-phase module can be found in Pérez et al. (2011) and Jorba et al. (2012), respectively. According to the features of its unified meteorological core, NMmB/BSC-CTM was conceived for short- and medium-range forecasting for a wide range of spatial scales as well as for climate studies (http://www.bsc.es/earth-sciences/mineral-dust/nmmmbsc-dust-forecast). Due to its fully online coupling, several feedback processes among gases, aerosol particles and radiation are taken into account by the model. In particular, the radiative effect of aerosols is considered, while cloud–aerosol interactions are neglected at present. The online coupling of aerosol optical properties and gas-phase photolysis reactions is also under development.

2.1 The NCEP non-hydrostatic multiscale model (NMmB)

The NMmB is the meteorological core of the modeling system, allowing simulations of scales ranging from global to large eddy simulations (LES) in global and regional domains. The regional NMmB has been used at NCEP as the regional North American Mesoscale (NAM) model since October 2011. The global model is formulated on the latitude-longitude grid, by applying conservative polar boundary conditions and polar filtering, slowing down the tendencies of basic dynamic variables (Janjic, 2009; Janjic and Gall, 2012). Rotated latitude-longitude grids are employed for regional simulations in order to obtain more uniform grid distances. In both cases, the horizontal discretization is performed on the Arakawa B-grid. In the vertical, the general hybrid sigma-pressure coordinate (Simmons and Burridge, 1981) is used with the Lorenz staggering. The “isotropic” horizontal finite volume differencing technique assures the conservation
of a number of dynamical and quadratic quantities (among
these, energy and enstrophy). More details about the nu-
erical schemes of the NMMB can be found in Janjic (1977,

A variety of physical schemes are implemented in the
model. A list of these parameterizations and their respec-
tive references were presented in Pérez et al. (2011) and fur-
For our purposes, we shortly recall the parameterizations in-
volved in the sea-salt aerosol cycle, i.e., surface layer, grid-
scale cloud microphysics, convective adjustment and pre-
cipitation, and radiation schemes. Boundary layer, and free
atmosphere turbulence are parameterized using the Mellor-
Yamada-Janjic (MYJ) turbulence closure scheme (Mellor and
Yamada, 1982; Janjic, 2001). In the surface layer the Monin–Obukhov similarity theory (Monin and Obukhov,
1954) is applied (Janjic, 1996) in combination with a vis-
cous sub-layer parameterization over oceans (Janjic, 1994).
The wind speed at 10 m ($U_{10}$), which is the key parameter of
sea-salt production schemes is computed consistently with
the surface layer parameterization. The friction velocity $u^*$
is computed as the square root of the surface layer vertical
momentum transport.

Grid-scale clouds are parameterized with the scheme of
Ferrier et al. (2002) including 5 prognostic cloud variables.
The relevant quantities for the coupling with aerosol pro-
cesses are the mixing ratios of both liquid and ice cloud water
and their conversion rates to precipitation. The Betts-Miller-
Janjic convective adjustment scheme (Betts, 1986; Betts
and Miller, 1986; Janjic, 1994, 2000) is used for sub-grid-
scale clouds. Using conservational constraints, the convec-
tive clouds are represented by reference humidity and tem-
perature profiles. Both water vapor mixing ratio and tem-
perature are relaxed toward reference values within a convec-
tion time step. In the case of deep convection, the reference pro-
files and the relaxation time are governed by the cloud effi-
ciency $E$ which depends on convective regime. This is a non-
dimensional parameter obtained as a combination of entropy
change, precipitation, and mean cloud temperature (Janjic,
1994, 2000). The shallow convection parameterization closure
uses the constraint that the entropy change must be non-negative Janjic (1994, 2000). The NMMB uses the op-
erational Geophysical Fluid Dynamics Laboratory (GFDL)
radiation package, which includes shortwave (Lacis and
Hansen, 1974) and longwave (Fels and Schwarzkopf, 1975)
schemes. Since the coupling with aerosols is not allowed by
the operational GFDL scheme, the Rapid Radiative Transfer
Model (RRTM) (Mlawer et al., 1997) was implemented in
the model (Pérez et al., 2011). By using RRTM, it is possible to
couple radiation (both long- and shortwave) and aerosols
by providing aerosol optical depth, asymmetry factor, and
single-scattering albedo.

### 2.2 The BSC-CTM dust module (BSC-DUST)

The development of the sea-salt module follows the im-
plementation of BSC-DUST (Pérez et al., 2011; Haustein
et al., 2012), i.e., the dust module of NMMB/BSC-CTM.
BSC-DUST includes 8 transport bins ranging from 0.1 µm to
10 µm in dry radius. Within each transport bin a log-normal
time-invariant sub-bin distribution is assumed. The processes
considered by the module are dust emission, horizontal and
vertical advection, horizontal diffusion and vertical transport
by turbulence and convection, dry deposition and sedimenta-
tion, and wet removal including in- and below- cloud scav-
enging from grid- and sub-grid scale clouds. Water uptake
was not considered. Given the strong uncertainties on the ac-
tivation properties of dust, solubility is obtained by applying
an intermediate hypothesis between pure hydrophobic and
pure hydrophilic aerosol. Both global and regional simul-
ations of dust optical depth have been exhaustively evaluated
in Pérez et al. (2011) and Haustein et al. (2012). In this con-
tribution, the model coarse AOD is calculated from the dust
and sea-salt components allowing the use of AERONET sta-
tions affected by dust to be included in the evaluation.

### 3 The sea-salt module

Sea-salt is assumed to be externally mixed with dust and the
continuity equation is solved for 8 prognostic size-sections:

$$
\partial_t q_k + (\mathbf{v} \cdot \nabla) h_k q_k = F_k^{(em)} - \sum_n F_{n,k}^{(sink)} + F_{k}^{(diff)},
$$

(1)

where $q_k$ are the sea-salt dry mass mixing-ratios, $\mathbf{v}$ is the
wind velocity, subscript $h$ stands for horizontal operator,
and $F_k^{(em)}$, $F_{n,k}^{(sink)}$, $F_k^{(diff)}$ represent sea-salt production,
sink/mixing, and turbulent diffusion terms, respectively. Ad-
vection and diffusion are analogous to those of moisture in
NMMB (Janjic, 2009). The production term is detailed in
Sect. 3.1 and sink processes are described in Sect. 3.2.

We assume a dry radius lower cutoff of 0.1 µm in the size
distribution. Upper size cutoff values depend on the pro-
duction parameterization (a detailed discussion are provided
in Sect. 3.1) and were fixed to 15 µm to comprehensively
account for all the different formation processes. Size-bins
are described in Table 1. Simulated sea-salt mass and optical
depth are strongly influenced by the number of size-bins
adopted, due to the strong dependence of dry deposition upon
particle size (Witek et al., 2011). Simulated values tend to
converge above 15 size-bins, while mass loss takes place oth-
erwise. We employ 8 size-bins which involves a mass loss of
5% (Witek et al., 2011) – a negligible quantity compared to
emission uncertainties – as a trade-off for doubled computa-
tional efficiency. A sub-bin log-normal approach is assumed
to calculate different moments of particle radius, such as dry
effective radius $r_d^{\text{eff}} = \sqrt{r_d^2} / \sqrt{r_d^2}$ and volume mean ra-
dius $r_d^{\text{vn}} = \left( \frac{r_d^2}{<r_d^2>} \right)^{1/3}$. We assume the canonical
log-normal distribution of Lewis and Schwartz (2004), characterized by a geometric radius at RH = 80 %, \( r_{80} \approx 0.3 \mu m \) and geometric standard deviation \( \sigma_g = 2.8 \).

### 3.1 Emissions

Strong uncertainties of up to one order of magnitude affect the estimates of sea-salt production fluxes. The most widely used technique to parameterize sea-salt emission is the so-called whitecap method, by which the flux is factorized as a product of sea-surface whitecap fraction and production per whitecap unit, both terms being affected by significant uncertainties. Parameterizations use wind-speed at 10 m \( (U_{10}) \), SST, atmospheric stability, sea-surface salinity, and ocean waves properties (height, age, relative direction respect to wind), for which Lewis and Schwartz (2004) and O’Dowd and de Leeuw (2007) provide useful reviews. In this study, we implement five widely used whitecap method schemes for open-ocean production (surf-zone production is neglected) with details provided in Table 2. Labels G03, M86, SM93, MA03, and J11 stand for schemes provided in Gong (2003), Monahan et al. (1986), Smith et al. (1993), Mårtensson et al. (2003), and Jaeglé et al. (2011), respectively. G03, M86, and SM93 are derived from observational data sets and only depend on \( U_{10} \); MA03 is derived from laboratory experiments and includes SST effects that are size-dependent. J11 emissions are formulated by multiplying the G03 scheme by a SST-dependent function equal for all particle sizes. The function was fitted using the GEOS-CHEM model and observations. In our work, we keep the function as it was derived by Jaeglé et al. (2011). With the exception of SM93, all the implemented schemes apply the same wind speed power law \( (U_{10})^{3.41} \) in the whitecap parameterization. Consequently, we do not focus on the model sensitivity to changes in this term. MA03 was derived for a temperature interval ranging from 271 K to 298 K, which does not strictly cover the annual variation of global SST. J11 is formulated for temperatures ranging from 273.15 K to 303.15 K.

For our comparison, we choose schemes differing in particle size and production mechanism description. Figure 1 shows that the strongest uncertainties appear for the ultrafine particles \( (r_d < 0.1 \mu m) \), which do not play a relevant role in the simulation of mass concentration and optical depth, and thus are beyond the scope of this work.

All considered schemes account for sea-salt formation from bubble bursting. Spume production is not described in M86 and MA03, while it is represented in SM93 (Fan and Toon, 2011), and its treatment in G03 is unclear (and, as a consequence, in J11). This leads to significant differences in emission fluxes of large particles (Fig. 1).

In addition, the above parameterizations were merged to obtain more comprehensive schemes, such as the combined M86/SM3 and MA03/M86/SM93 (Table 2). Hoppel et al. (2002) concluded that M86/SM93 may be considered as the best candidate to describe sea-salt emissions in the interval 0.15 \( \mu m \) to 15 \( \mu m \) in dry radius. M86/SM93 was then extended to ultrafine particles in other studies (Caffrey et al., 2006; Fan and Toon, 2011).

In this work, we also combined M86/SM93 and MA03 to account for the the SST effect upon sea-salt production. In MA03/M86/SM93, MA03 is applied within its range of validity and replaced by M86/SM93 beyond that range (i.e., for large particles with \( r_d > 1.4 \mu m \)). We find a similar attempt in the work of Tsyro et al. (2011), where MA03 is combined with M86 (but not with the spume production of SM93).

We choose an upper cutoff for the particle size around the maximum value allowed by the sea-salt production parameterizations implemented in our module. To perform a consistent comparison, we consider a range of [0.1–15] \( \mu m \) in dry radius for all the emission schemes, which implies an extension of M86, G03, and J11 schemes beyond their formulation intervals. Because some schemes work with wet radius \( r_{80} \) and others with dry radius \( r_d \), we assume \( r_{80} = 2r_d \) to obtain emission of dry particles following the water-uptake

### Table 1. Sea-salt size bins and their characteristic radii. \( r_d \), \( r_{dvm} \), and \( r_{deff} \) stand for dry radius, dry volume mean radius, and effective radius, respectively.

<table>
<thead>
<tr>
<th>bin</th>
<th>( r_d (\mu m) )</th>
<th>( r_{dvm} (\mu m) )</th>
<th>( r_{deff} (\mu m) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.10–0.18</td>
<td>0.14</td>
<td>0.14</td>
</tr>
<tr>
<td>2</td>
<td>0.18–0.30</td>
<td>0.24</td>
<td>0.24</td>
</tr>
<tr>
<td>3</td>
<td>0.30–0.60</td>
<td>0.43</td>
<td>0.45</td>
</tr>
<tr>
<td>4</td>
<td>0.60–1.00</td>
<td>0.77</td>
<td>0.79</td>
</tr>
<tr>
<td>5</td>
<td>1.00–1.80</td>
<td>1.32</td>
<td>1.36</td>
</tr>
<tr>
<td>6</td>
<td>1.80–3.00</td>
<td>2.27</td>
<td>2.32</td>
</tr>
<tr>
<td>7</td>
<td>3.00–6.00</td>
<td>3.98</td>
<td>4.13</td>
</tr>
<tr>
<td>8</td>
<td>6.00–15.00</td>
<td>7.39</td>
<td>8.64</td>
</tr>
</tbody>
</table>
Table 2. Sea-salt number emission fluxes implemented in NMMB/BSC-CTM. \(dF_N/d\ln r\) fluxes in units [m\(^{-2}\) s\(^{-1}\) \(\mu m^{-1}\)], \(dF_N/d\log(r)\) fluxes in units [m\(^{-2}\) s\(^{-1}\) \(\mu m^{-1}\)]. \(r_{80}\) and \(r_{4}\) stand for wet radius at RH = 80% and dry radius in units [\(\mu m\)], respectively. If \(r\) is used, dry or wet radius was not specified. \(U_{10}\) in m s\(^{-1}\). SST in K units. Formul. range stands for the size-range in the original formulation of each parameterization. The assumption \(r_{80} = 2r_d\) is used to merge wet and dry radius intervals. All schemes are applied in the range \(r_d \in [0.1–15] \mu m\).

<table>
<thead>
<tr>
<th>Production scheme</th>
<th>Reference</th>
<th>Mechanism</th>
<th>Formul. range</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\frac{dF_N}{dr_{80}}</td>
<td>(1.373 - U_{10}^{0.41} \cdot r_{80}^{-3.41}) \cdot (1 + 0.057 \cdot r_{80}^{0.45} \cdot 10^{-6} \cdot \exp(-C \cdot r_{80}^{2.4})) )</td>
<td>Gong (2003)</td>
<td>bubbles, spume: unclear</td>
</tr>
<tr>
<td></td>
<td>(A = 4.7(1 + \theta_{90})^{-0.157 \cdot 10^{-4 \cdot \theta_{90}}} = 0.3 \cdot C = (0.433 - \log(r_{80})) / 0.433 )</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(\frac{dF_N}{dr_{80}}</td>
<td>(1.373 - U_{10}^{0.41} \cdot r_{80}^{-3.41}) \cdot (1 + 0.057 \cdot r_{80}^{0.45} \cdot 10^{-6} \cdot \exp(-B \cdot r_{80}^{2.4})) )</td>
<td>Monahan et al. (1986)</td>
<td>bubbles</td>
</tr>
<tr>
<td></td>
<td>(B = (0.38 - \log(r_{80})) / 0.65 )</td>
<td></td>
<td></td>
</tr>
<tr>
<td>(\frac{dF_N}{dr_{80}}</td>
<td>)</td>
<td>Smith et al. (1993), Hoppel et al. (2002)</td>
<td>spume</td>
</tr>
<tr>
<td>(\frac{dF_N}{dr_{4}}</td>
<td>)</td>
<td>Mårtensson et al. (2003)</td>
<td>bubbles (SST dependent)</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>SST ([2.71–2.98])</td>
</tr>
<tr>
<td>(\frac{dF_N}{dr_{80}}</td>
<td>)</td>
<td>Combined M86/SM93</td>
<td>bubbles, spume</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>(\frac{dF_N}{dr_{80}}</td>
<td>)</td>
<td>Combined MA03/M86/SM93</td>
<td>bubbles (SST), spume</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
| \(\frac{dF_N}{dr_{80}} | \) | Jaeglé et al. (2011) | bubbles (SST), spume (SST) | \(r_{40} \in [0.07–20] \)
| | | | SST \([2.73.15 – 30.15]\) |

The assumption \(r_{80} = 2r_d\) is used to merge wet and dry radius intervals. All schemes are applied in the range \(r_d \in [0.1–15] \mu m\).

### 3.2 Water uptake

The sea-salt life cycle is strongly affected by water uptake. Hygroscopic growth may increase particles’ radii by a factor of 4 or more. Following Chin et al. (2002) we introduced prescribed RH-dependent growth factors \(\phi(RH) = r_w / r_d\), derived from the Global Aerosol Data Set of Köepke et al. (1997) and the database of d’Almeida (1991) (Table 3). \(r_w\) and \(r_d\) are the wet and the dry particle radius, respectively.

We assume the same factors for any radius-moment representation, such as effective and volume-mean radii.

Given \(\phi(RH)\), the water-uptake process is fully described by extending any dry particle parameter to its respective wet value. In particular we obtain wet particle radius and density as

\[
\begin{align*}
  r_d &\to r_w = \phi \cdot r_d \\
  \rho_d &\to \rho_w = f_d \rho_d + (1 - f_d) \rho_{\text{water}},
\end{align*}
\]

(3)

(4)

where \(\rho_{\text{water}}\) is the density of water and \(f_d\) is the volume fraction of dry aerosol (\(f_d = \phi^{-1}\)). The dry sea-salt density is assumed \(\rho_d = 2160 \text{ kg m}^{-3}\) for every size-bin. By using this simplified approach, all aerosol processes affected by

\[
\frac{\rho \cdot \frac{dF_k}{d\ln r_d}}{\rho_{\text{water}}} = \int_{\ln r_d - k}^{\ln r_d + k} F_k^{\text{emi}} \cdot 4\pi \cdot \rho_d \cdot r_d^2 \cdot dr_d.
\]

(2)

The emission mechanism is not explicitly coupled with the viscous sub-layer of the NMMB. However, the calculation of friction velocity and wind speed at 10 m depends on the viscous sub-layer scheme in the surface layer.
hygroscopic growth are easily reformulated by extending the parameterizations used in the dust module (dry aerosol) to the wet-particle case, i.e., by applying Eqs. (3) and (4). In the following we present a short review of the parameterizations used in the aerosol module of NMMB/BSC-CTM, pointing out the extension to wet particles in the sink and mixing terms. A more detailed description of each scheme can be found in Pérez et al. (2011). When not otherwise specified we refer to \( r_d \) as \( r_d^w \) for brevity.

### 3.3 Deposition and convective mixing

Sedimentation is governed by the gravitational settling velocity \( v_{g,k}(\phi) \), calculated for each size-bin \( k \) following the Stokes–Cunningham approximation. \( v_{g,k}(\phi) \) depends on the particle size and thus on the water uptake.

The dry deposition velocity \( v_{dep,k}(\phi) \), acting at the bottom layer, is parameterized following Zhang et al. (2001). The dependence on \( \phi \) is introduced in the surface resistance calculation, which accounts for particle size and density (Slinn, 1982).

Wet scavenging fluxes are parameterized both for grid-scale (stratiform) and sub-grid scale (convective) clouds. In-cloud scavenging flux is parameterized using a solubility parameter \( \epsilon_k \) that is defined as the fraction of aerosol contained in cloud which may eventually precipitate. For sea-salt particles, \( \epsilon_k \) is obtained from Zakey et al. (2006). Since the values found in Zakey et al. (2006) for dust represent an intermediate between pure hydrophobic and pure hydrophilic hypothesis, we assume \( \epsilon_{ss,k} = 2 \epsilon_{du,k} \). This calculation of \( \epsilon_{ss,k} \) is consistent with the values used in other state-of-the-art models (see for ex. the sensitivity study in Fan and Toon, 2011). Because small particles are more probable candidates to act as cloud condensation nuclei, \( \epsilon_{ss,k} \) decreases with size (see Table 4). Grid-scale below cloud scavenging is parameterized following Slinn (1984) in which capture efficiencies \( E_k(\phi) \) depend on the wet radius and density of the aerosol particles.

For sub-grid (convective) clouds, the scavenging fluxes are coupled with the Betts–Miller–Janjic scheme (BMJ) of the NMMB. The convective in-cloud scavenging parameterization employs solubility factors \( \epsilon_k(\phi) \) as well. After the in-cloud scavenging, the remaining sea-salt is assumed vertically mixed by performing a conservative relaxation towards reference profiles. The parameterization of sub-grid below-cloud scavenging is analogous to the case of grid-scale clouds. Within shallow non-precipitating convective clouds sea-salt is homogeneously mixed within the cloud.

### 3.4 AOD calculation

In order to calculate the sea-salt optical depth, extinction efficiencies \( Q_{\lambda,k}^{ext} \) are computed with the Mie-theory solving algorithm of Mishchenko et al. (2002) for each size-bin \( k \) and each RH range (Fig. 2). Spherical homogeneous particles are assumed. The refractive indices were derived from the Global Aerosol Data Set (GADS) (Köpke et al., 1997). Extinction efficiencies also depend on the sub-bin log-normal geometric parameters \( r_\phi \) and \( \sigma_\phi \) (Lewis and Schwartz, 2004). The optical depth is obtained as

\[
\tau_{\lambda,k} = \beta_{\lambda,k} \tilde{M}_{d,k},
\]

where \( \tilde{M}_{d,k} \) is the layer dry mass loading of each bin and \( \beta_{\lambda,k} \) is a mass extinction coefficient which accounts for water uptake:

\[
\beta_{\lambda,k} = \frac{3 Q_{\lambda,k}^{ext}(\phi, r_w^\phi, \sigma_w^\phi)}{4 r_{eff,w,k} \bar{f}_d(\phi) \rho_d,k}. 
\]

The algorithm of Mishchenko et al. (2002) also provides single-scattering albedo and asymmetry factor for radiative calculations. The total sea-salt optical depth is equal to the sum over all bins:

\[
\tau_{\lambda,T} = \sum_{k=1}^{8} \tau_{\lambda,k}.
\]

The coarse sea-salt optical depth is calculated with a lower cutoff value of 0.6 µm (the AERONET submicron cutoff) of the wet particle radius. In our description, this value is equivalent to a lower cutoff of the dry particle radius \( \bar{r}_d \) given by

\[
\tilde{r}_d = 0.6 \mu m / \phi(RH).
\]
At maritime atmospheric conditions \((\text{RH} = 80 \%)\), the sub-micron bins significantly contribute to the coarse AOD. Sub-bin contributions to the coarse optical depth are calculated assuming the log-normal distribution of Lewis and Schwartz (2004). Another useful parameter for model evaluation is the resulting AOD (total and coarse) from both sea-salt and dust. Because of their external mixing, we assume

\[
\tau_{\text{ss+du},\lambda} = \tau_{\text{ss},\lambda} + \tau_{\text{du},\lambda},
\]

where the subscripts ss and du respectively refer to sea-salt and dust.

### 4 Observational data

Figure 3 displays the location of measurement sites and cruise measurement trajectories used in the model evaluation. Names and coordinates of the sites are listed in Table 5. Quantities evaluated are sea-salt surface concentrations and AOD. For the station data we use monthly climatologies to compare with our simulated 5 yr period (2002–2006).

We consider AERONET Sun photometer measurements as the reference to evaluate the modeled sea-salt AOD. Even if algorithms tend to minimize biases due to cloud cover and other effects (e.g. Zhang and Reid, 2006), estimates from satellites remain highly uncertain and are not used in this contribution. Satellite overestimation can reach up to 0.07 in island stations compared to monthly AERONET-derived AOD (Jaeglé et al., 2011). At certain latitudes, the bias between satellite and ship AOD measurements may range from −0.2 to +0.2 (Smirnov et al., 2011). These biases exceed the typical sea-salt AOD value in the remote marine environment (∼0.07, see Smirnov et al., 2011). AOD measurements from the AERONET Maritime Aerosol Network (MAN) are not used in this work because of complexities in disentangling sea-salt and dust contributions from other aerosol species (such as carbonaceous and sulfate aerosols) that are currently neglected in our model.

#### 4.1 NOAA/PMEL cruises

Sea-salt cruise measurements are considered, specifically ion concentrations from two cruises of the NOAA Pacific Marine Environmental Laboratory (PMEL): the AEROSOLS99 and INDIan Ocean EXperiment (AEROINDOEX) in 1999 spanning the Atlantic and the Indian Oceans and the first Aerosol Characterization Experiment (ACE1) in 1995 crossing the Pacific Ocean. Concentrations of both \(\text{Na}^+\) and \(\text{Cl}^-\) were measured by ion chromatography (Quinn et al., 1998) at 18 m above the sea surface. The experimental aerodynamic cutoff diameter was 10 \(\mu\)m for all cruises. Instruments were kept at constant RH values during measurements. Based on these values, Jaeglé et al. (2011) assumed a dry radius cutoff of 3 \(\mu\)m for AEROINDOEX and ACE1. Hence, we use the first 6 dry model bins for the comparison. The ACE1 and

### 4.2 U-MIAMI surface concentrations

The U-MIAMI network supplied aerosol measurements from around 35 stations worldwide between the early 1980s and 1996 (Savoie and Prospero, 1977). Aerosols were collected with high-volume filter samplers and different measurement protocols were employed depending on the measurement site. We use climatologies from 15 stations (Fig. 3 and Table 5). These stations grant good data quality and are not affected by surf-zone production (J. Prospero, personal communication, 2012). The observed sea-salt mass concentrations (µg m\(^{-3}\)) were computed as 

\[\text{SS} = \text{Cl}^- + 1.47\text{Na}^+\]
Table 5. List of the observational sites used in this work, classified by network.

<table>
<thead>
<tr>
<th>Code</th>
<th>Database</th>
<th>Station</th>
<th>lat</th>
<th>lon</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>AERONET</td>
<td>Amsterdam Island</td>
<td>37.81°S</td>
<td>77.57°E</td>
</tr>
<tr>
<td>2</td>
<td>AERONET</td>
<td>Ascension Island</td>
<td>7.98°S</td>
<td>14.41°W</td>
</tr>
<tr>
<td>3</td>
<td>AERONET</td>
<td>Azores</td>
<td>38.53°N</td>
<td>28.63°W</td>
</tr>
<tr>
<td>4</td>
<td>AERONET</td>
<td>Bermuda</td>
<td>32.37°N</td>
<td>64.70°W</td>
</tr>
<tr>
<td>5</td>
<td>AERONET</td>
<td>Cape San Juan</td>
<td>18.38°N</td>
<td>65.62°W</td>
</tr>
<tr>
<td>6</td>
<td>AERONET</td>
<td>Coral-RG</td>
<td>51.60°S</td>
<td>69.32°W</td>
</tr>
<tr>
<td>7</td>
<td>AERONET</td>
<td>Coconut Island</td>
<td>21.43°N</td>
<td>157.79°W</td>
</tr>
<tr>
<td>8</td>
<td>AERONET</td>
<td>Crozet Island</td>
<td>46.43°S</td>
<td>51.85°E</td>
</tr>
<tr>
<td>9</td>
<td>AERONET</td>
<td>Dunedin</td>
<td>45.86°S</td>
<td>170.51°E</td>
</tr>
<tr>
<td>10</td>
<td>AERONET</td>
<td>Guam Island</td>
<td>13.43°N</td>
<td>144.80°E</td>
</tr>
<tr>
<td>11</td>
<td>AERONET</td>
<td>La Parguera</td>
<td>17.97°N</td>
<td>67.04°W</td>
</tr>
<tr>
<td>12</td>
<td>AERONET</td>
<td>Midway Island</td>
<td>28.21°N</td>
<td>177.38°W</td>
</tr>
<tr>
<td>13</td>
<td>AERONET</td>
<td>Nauru</td>
<td>0.52°S</td>
<td>166.92°E</td>
</tr>
<tr>
<td>14</td>
<td>AERONET</td>
<td>Reunion Island</td>
<td>20.88°S</td>
<td>55.48°E</td>
</tr>
<tr>
<td>15</td>
<td>AERONET</td>
<td>Rottnest Island</td>
<td>32.00°S</td>
<td>115.50°E</td>
</tr>
<tr>
<td>16</td>
<td>AERONET</td>
<td>Tahiti</td>
<td>17.58°S</td>
<td>149.61°W</td>
</tr>
</tbody>
</table>

SURFACE CONCENTRATIONS

- a U-MIAMI Baring Head 41.28°S 174.87°E
- b U-MIAMI Bermuda 32.27°N 64.87°W
- c U-MIAMI Cape Grim 40.68°S 144.68°E
- d U-MIAMI Cape Point 34.35°S 18.48°E
- e U-MIAMI Chatam Island 34.92°S 176.50°W
- f U-MIAMI Fanning Island 3.92°N 159.33°W
- g U-MIAMI Invercargill 46.43°S 168.35°E
- h U-MIAMI King George Island 62.18°S 58.30°W
- i U-MIAMI Marion Island 46.92°S 37.75°E
- l U-MIAMI Miami 25.75°N 80.25°W
- m U-MIAMI Midway Island 28.22°N 177.55°S
- n U-MIAMI Oahu 21.33°N 157.70°W
- o U-MIAMI Palmer 64.77°S 164.05°W
- p U-MIAMI Reunion Island 21.17°S 55.83°E
- q U-MIAMI American Samoa 14.25°S 170.58°W

following Quinn and Bates (2005) where both Cl\textsuperscript{-} and Na\textsuperscript{+} measurements were available, and as SS = 3.252Na\textsuperscript{+}, where only Na\textsuperscript{+} concentrations were supplied (J. Prospero, personal communication, 2012). Since U-MIAMI measurements are not constrained by an upper cutoff radius, we perform the comparison with the complete set of model bins.

4.3 AERONET AOD

The AEROSol RObotics NETwork (AERONET) provides automatic ground-based observations from sun photometers in a large number of stations around the globe (Holben et al., 1998; Smirnov et al., 2000). The accuracy of AERONET sun photometers is 0.01 for AOD (Holben et al., 1998; Smirnov et al., 2000). We considered a set of 16 sea-salt-dominated stations as proposed by Jaeglé et al. (2011) (Fig. 3 and Table 5). The three requirements fulfilled by the stations are sea-salt contributions to the total AOD greater than 50% as predicted by GEOS-CHEM model, availability of Level 2 quality-assured data for all the considered time ranges, and at least 3 yr of data supporting the monthly climatologies. The evaluation is performed against monthly climatologies of the AOD at 500 nm. In particular, we focus on the AOD coarse fraction, therefore limiting the influence of fine aerosol species.

5 Results and discussion

5.1 Experimental setup

We performed global simulations between 2002 and 2006 and additional simulations covering the temporal windows of the cruises. The horizontal resolution used is 1° × 1.4°. 24 vertical layers are employed and the dynamics time step is Δt = 120 s. Meteorological conditions are initialized every 24 h using the NCEP final analyses (FNL) at 1° × 1° for year > 2000 and the NCEP Global Data Assimilation System analysis (GDAS) at 2.5° × 2.5° prior to year 2000. A spinup of 1 month for sea-salt is assumed at the beginning of each simulated period. The model output is taken every 6 h to calculate monthly averages and every 1 h when comparing with cruise observations.

The five implemented emission schemes are compared with comprehensive data sets of observations dispersed over the globe. The dust AOD is indicated with the label DU. Feedback processes between aerosols and radiation are not considered in any of the simulations.

At each evaluation site we also compare the simulated wind speed with a 30 yr climatology (1981–2010) derived from the NCEP/NCAR reanalysis data set (Kalnay et al., 1996) to evaluate the representativeness of our 5 yr wind speed climatology.

5.2 Global sea-salt distribution and total budgets

Fig. 4 displays the global distribution of simulated sea-salt production, surface concentration, and AOD at 500 nm in January and August with M86/SM93. We observe a pronounced asymmetry in the summer-to-winter variation between the two hemispheres and four large regions of maximum production. The two largest monthly peaks are found in regions with enhanced westerlies, i.e., beyond the horse latitudes (lat > 30° N and lat < 30° S). Also two local maxima can be observed in correspondence with the trade winds, next to the intertropical convergence zone (around 10° N and 10° S). While sea-salt production at the southern belt only moderately changes with season, the northern belt is affected by strong variations during the year with increases in boreal winter well above +200% with respect to boreal summer. It is well known that these seasonal fluctuations are related to the asymmetric variation of the global wind speed pattern, driven by the variation of the global atmospheric angular momentum (Sandwell and Agreen, 1984).

Sea-salt production and surface concentration over the Pacific around 10° N is about half the values found at higher
Fig. 4. Seasonal regimes of sea-salt emission (left panels), AOD at 500 nm (middle panels), and surface concentration (right panels) with M86/SM93. January and August averages of a 5 yr period (2002–2006) are shown. The label emi refers to emission flux, and sconc refers to surface concentration.

Latitudes. Yet, the AOD reaches monthly mean values close to the global maximum. Because of the seasonal movement of the Intertropical Convergence Zone, the region around 10° N in the Pacific is characterized – during winter – by infrequent precipitation and low wet scavenging rate increasing particle lifetime, in contrast to the strong production belts characterized by wet extratropical cyclone activity. The RH-dependent particle size and optical properties in the model also play a relevant role in determining the AOD peaks close to the intertropical convergence zone.

Surface concentration and AOD maximum values in the Arabian Sea during the boreal summer are due to the strong southwestern winds of the monsoon circulation.

Figure 5 displays maps of annual mean sea-salt emission, surface concentration, and AOD with the five emission schemes. The two maximum production regions beyond the horse latitudes are the most sensitive to the choice of the emission scheme. G03 produces the highest concentrations with peaks above 35 µg m⁻³ in the southern belt and over 25 µg m⁻³ in the northern belt. Differences in spume production representation are clear when comparing the simple M86 with M86/SM93, for which the mean concentration is enhanced due to wind episodes exceeding the threshold $U_{10} > 9$ m s⁻¹. The relative importance of the production regions changes if SST effects are included in the emission scheme. The SST dependence in MA03/M86/SM93 produces a latitudinal modulation of the emission fluxes and surface concentration with relative enhancement in the tropics and reduction elsewhere. This effect is amplified with J11, leading to a change in maximum values of surface concentration from the high-latitude belts to the tropics. In particular, an absolute maximum value above 35 µg m⁻³ is found over the Arabian Sea.

Sea-salt AOD patterns with M86, M86/SM93, and G03 are very similar. The southern belt dominates with peaks around ~ 0.1. Peak values around ~ 0.06 are found at high latitudes and the tropical Pacific. Relevant differences are observed with MA03/M86/SM93 for which absolute maximum values of 0.1 appear next to the intertropical convergence zone. These peaks overestimate the maximum AOD from ship measurements gathered by Smirnov et al. (2011) both in the remote tropical Pacific (0.07 for total AOD at 500 nm) and the Indian Ocean (0.06, east of Madagascar). The use of the J11 scheme leads to an AOD pattern and peak values very similar to MA03/M86/SM93, with an enhancement of the SST latitudinal modulation.

Table 6 lists the annual model budgets from the different emission schemes and other recent studies. To achieve a consistent comparison, we specify values for five size intervals: all bins (ALL), fine bins up to 1 µm (F1), fine bins up to 0.5 µm (F2), coarse bins from 1 µm to 4 µm (C1), and coarse bins from 0.5 µm to 4 µm (C2).

Despite the decrease in total emissions, SST-dependent schemes lead to an enhancement of sea-salt lifetime, both in the case of MA03/M86/SM93 (with respect to M86/SM93) and J11 (with respect to G03). This effect was also observed in Jaeglé et al. (2011), where lifetime values are close to ours. However both fine (F1 and F2) and coarse (C1 and C2) lifetimes significantly increase with J11 compared to G03, in contrast to Jaeglé et al. (2011) and Tsigaridis et al. (2013). This may be related to different treatments of water uptake, deposition, and particle size distribution in the models.

With respect to AEROCOM experiments, the major difference is found in the wet deposition fraction, which is around 0.4 in our model and between 0.2 and 0.3 in AEROCOM A
and B median models. In particular, J11 produces the most compatible value with the AEROCOM inter-model variability.

Our simulated annual mean column mass load (ranging from 5.0 Tg to 7.2 Tg) is only slightly larger than the value of Jaeglé et al. (2011) and it is close to the AEROCOM Experiment A median value and about half of Experiment B (12.0 Tg) (Textor et al., 2006).

Emission is very sensitive to the upper size cutoff value and ranges from 3888 Tg yr$^{-1}$ to 8114 Tg yr$^{-1}$.

5.3 Modeled surface concentrations compared with cruise data

Cruise measurements allow a comparison with model at timescales of 2–24 h. Each measurement gathered by the vessels was averaged on space and time, thus simulated values may be affected by errors due to the adopted averaging technique. We remap the original lat/lon grid at resolution $\Delta x, \Delta y = 1^\circ, 1.4^\circ$ to a courser resolution ($\Delta x' = n \Delta x$, $\Delta y' = n \Delta y$), matching the characteristic spatial length of the cruise under consideration. The number $n$ is defined as the smallest integer satisfying the following conditions:

\[ V_{\text{cruise}} \cdot \max(T_{\text{obs}}) < n \Delta x \]  
\[ V_{\text{cruise}} \cdot \max(T_{\text{obs}}) < n \Delta y, \]  

where $V_{\text{cruise}}$ is the vessel mean speed during the cruise and $T_{\text{obs}}$ is the observation duration, which is not constant. In this way, the spatial extent of each measurement is represented by a single lower resolution grid cell. We use $n = 2$ for AEROINDOEX and $n = 4$ for ACE1. Model outputs every 1 h are then averaged over each measurement period. Cruise trajectories are displayed in Fig. 3. We recall that the values shown in this comparison refer to an upper cutoff of 3 µm in dry radius, thus we investigate the model’s ability to simulate the concentration within the first 6 bins. In this case the M86/SM93 scheme is equivalent to M86, since the larger particles produced by spume cutting are not taken into account due to the observational cutoff.

Figures 6 and 7 show a good overall correlation for AEROINDOEX and a lower correlation for ACE1. Results are similar to those obtained in Jaeglé et al. (2011)
Table 6. Model sea-salt global budgets and lifetimes compared with other recent studies. The label emi stands for total accumulated emission of sea-salt mass (Tg yr\(^{-1}\)); wetfrac = wetdep/(drydep + wetdep) where drydep accounts for both accumulated dry deposition and sedimentation (Tg yr\(^{-1}\)) and wetdep for accumulated wet deposition (Tg yr\(^{-1}\)); \(<\text{load}>\) is the annual mean column mass load (Tg yr\(^{-1}\)), and lifetime = \(<\text{load}>/(\text{drydep} + \text{wetdep})\) (h). All quantities obtained in our work are averaged over the simulation period 2002–2006 and indicated by the label o.w. The labels ALL, F1, C1, F2, and C2 refer to different dry radius intervals (µm).

<table>
<thead>
<tr>
<th>Study</th>
<th>emi</th>
<th>ALL (0.1–15)</th>
<th>F1 (0.1–1)</th>
<th>C1 (1–4)</th>
<th>F2 (0.1–0.5)</th>
<th>C2 (0.5–4)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>emi</td>
<td>(&lt;\text{load}&gt;)</td>
<td>life</td>
<td>wetfrac</td>
<td>emi</td>
<td>life</td>
</tr>
<tr>
<td>M86</td>
<td>o.w.</td>
<td>3888</td>
<td>5.0</td>
<td>11.3</td>
<td>0.486</td>
<td>499</td>
</tr>
<tr>
<td>M86/SM93</td>
<td>o.w.</td>
<td>5440</td>
<td>5.6</td>
<td>8.9</td>
<td>0.467</td>
<td>499</td>
</tr>
<tr>
<td>G03</td>
<td>o.w.</td>
<td>8114</td>
<td>6.7</td>
<td>7.3</td>
<td>0.400</td>
<td>372</td>
</tr>
<tr>
<td>MA03/M86/SM93</td>
<td>o.w.</td>
<td>5419</td>
<td>6.5</td>
<td>10.4</td>
<td>0.466</td>
<td>266</td>
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<tr>
<td>J11</td>
<td>o.w.</td>
<td>6514</td>
<td>7.2</td>
<td>9.6</td>
<td>0.368</td>
<td>298</td>
</tr>
<tr>
<td>M86(^1)</td>
<td>Tsigaridis et al. (2013)</td>
<td>471</td>
<td>32.6</td>
<td>1916</td>
<td>26.6</td>
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<tr>
<td>G03(^1)</td>
<td>Tsigaridis et al. (2013)</td>
<td>357</td>
<td>32.6</td>
<td>2327</td>
<td>26.9</td>
<td></td>
</tr>
<tr>
<td>J11(^1)</td>
<td>Tsigaridis et al. (2013)</td>
<td>310</td>
<td>31.9</td>
<td>2019</td>
<td>26.6</td>
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<tr>
<td>G03(^2)</td>
<td>Jaeglé et al. (2011)</td>
<td>5200</td>
<td>4.7</td>
<td>7.9</td>
<td>0.356</td>
<td>67</td>
</tr>
<tr>
<td>AEROCOM A median(^3)</td>
<td>Jaeglé et al. (2011)</td>
<td>4600</td>
<td>4.4</td>
<td>8.4</td>
<td>0.402</td>
<td>59</td>
</tr>
<tr>
<td>AEROCOM A mean(^3)</td>
<td>Textor et al. (2007)</td>
<td>3830</td>
<td>6.5</td>
<td>7.2</td>
<td>0.210</td>
<td></td>
</tr>
<tr>
<td>AEROCOM A stddev(^3)</td>
<td>Textor et al. (2007)</td>
<td>8200</td>
<td>7.9</td>
<td>12.0</td>
<td>0.210</td>
<td></td>
</tr>
<tr>
<td>AEROCOM B median(^3)</td>
<td>Textor et al. (2007)</td>
<td>8200</td>
<td>5.4</td>
<td>7.1</td>
<td>0.122</td>
<td></td>
</tr>
<tr>
<td>AEROCOM B mean(^3)</td>
<td>Textor et al. (2007)</td>
<td>7740</td>
<td>12.0</td>
<td>14.4</td>
<td>0.282</td>
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</tr>
<tr>
<td>AEROCOM B stddev(^3)</td>
<td>Textor et al. (2007)</td>
<td>7720</td>
<td>12.7</td>
<td>12.0</td>
<td>0.253</td>
<td></td>
</tr>
</tbody>
</table>

\(^2\) Modes: \(r_d \in [0.1–1] \)µm and \(r_d \in [1–4] \)µm. \(^3\) Modes: \(r_d \in [0.01–0.5] \)µm, \(r_d \in [0.5–4] \)µm. \(^4\) Models participating in AEROCOM experiments use different parameterizations and aerosol size representations.

Fig. 6. From top to bottom: simulated sea-salt surface concentration (sconc) with M86 (blue), G03 (green), MA03/M86/SM93 (violet), and J11 (cyan) compared to AEROINDOEX cruise measurements (black squares), simulated wind speed (red line) compared to AEROINDOEX measurements (black line) (simulated SST is also shown), and simulated precipitation (red line) compared to AEROINDOEX measurements (black line) (simulated accumulated wet deposition is also shown). M86/SM93 is not shown since it is equivalent to M86 for \(r_d < 3 \)µm. The model 1 h-output surface concentrations are plotted with solid lines and averaged over the measurement times (circles). Skill scores shown are correlation \((r)\), mean normalized bias \((\text{bias})\), and mean normalized gross error \((\text{g.err})\).
Fig. 7. From top to bottom: simulated sea-salt concentration (sconc) with M86 (blue), G03 (green), MA03/M86/SM93 (violet), and J11 (cyan) compared to ACE1 cruise measurements (black squares), simulated wind speed (red line) compared to ACE1 measurements (black line) (simulated SST is also shown), and simulated precipitation (red line) compared to ACE1 measurements (black line) (simulated accumulated wet deposition is also shown). M86/SM93 is not shown since it is equivalent to M86 for \( r_d < 3 \mu m \). The model 1 h-output surface concentrations are plotted with solid lines and averaged over the measurement times (circles). Skill scores shown are correlation \((r)\), mean normalized bias (bias), and mean normalized gross error (g.err).
NCEP/NCAR Reanalysis (Kalnay et al., 1996). We use this comparison in order to check the representativeness of our simulation temporal window (5 yr, 2002–2006) with respect to a more comprehensive model climatology. Overall, simulated sea-salt concentrations are in good agreement with observations. Significant model overestimations of a factor of two or more are found in Invercargill (g), and Marion Island (i), where all schemes are above the observed mean plus one standard deviation. In these sites, overestimation cannot be attributed to an excess of wind speed compared with the NCEP/NCAR climatology (Fig. 9). A similar overestimation is found in Jaeglé et al. (2011) and Tsigaridis et al. (2013) in Marion Island (i) and in Invercargill (g) in Tsigaridis et al. (2013). Both studies use as well global models with a horizontal resolution greater than 1 degree. Since these sites are located in regions characterized by complex topography, we hypothesize that errors may be due in part to the low model resolution used.

In the Antarctic region, schemes show opposite performances in two stations close to each other. In Palmer (o), all schemes overestimate surface concentration with the exception of J11, which reproduces well the climatology. The overestimation of the wind speed with respect to the NCEP climatology does not entirely explain the behavior of M86, M86/SM93, G03, and MA03/M86/SM93. On the other hand, J11 leads to an underestimation of the observed climatology at King George Island (h) which cannot be attributed to wind speed. Contrasting results are found in this region when comparing our work with Jaeglé et al. (2011) and Tsigaridis et al. (2013).

Significant underestimation is found in Fanning Island (f) and American Samoa (q), both located in the tropical
Jaeglé et al. (2011) and Fan and Toon (2013) includes scatterplots of observed vs. simulated surface concentration. Significant underestimation of the U-MIAMI climatological values in Fanning Island (f) and American Samoa (q) is also found in Jaeglé et al. (2011) and Tsigaridis et al. (2013). There is a significant influence of the applied emission scheme upon modeled sea-salt surface concentrations. Even the introduction of SST-dependence in MA03/M86/SM93, which only affects the smaller bins (from 0.1 µm up to 1.4 µm in dry radius), makes a relevant contribution to the simulated concentration. Fig. 10 includes scatterplots of observed and simulated values (neglecting Invercargill (g) and Marion Island (i)) and a scatterplot of simulated wind speed and NCEP climatological values. Correlation, normalized bias, normalized gross error, and a linear regression fit are provided for each scatterplot. G03 generally overestimates the climatological monthly mean concentrations, while M86 underestimates. A significant reduction in bias is obtained when using M86/SM93 instead of M86. The best agreement is obtained with the SST-dependent emission schemes MA03/M86/SM93 and J11. Overestimation with G03 may be explained by its unclear description of spume particles production, as already noted in Fan and Toon (2011). Indeed, the emission flux for particles larger than 10 µm in dry radius is nearly one order of magnitude larger than in the

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Fig. 9. Monthly mean simulated surface wind speed (2002–2006) (blue) and surface wind climatologies (1981–2010) from NCEP/NCAR Reanalysis (black) at U-MIAMI stations. Interannual standard deviations are shown for the simulation (shaded grey) and the NCEP/NCAR climatology (black bars).
other implemented schemes (Fig. 1). On the other hand, the spume production is neglected in M86, which may partly explain the underestimated concentration. The introduction of spume particles in the combined M86/SM93 improves the model results. This improvement is more evident in stations and months characterized by frequent episodes of wind speed greater than 9 m s\(^{-1}\), such as for example during January, February and March in Bermuda (b) (Fig. 8). In the tropics, where these episodes are infrequent, M86 and M86/SM93 provide similar results.

The introduction of the SST-dependence in the emission scheme (both for MA03/M86/SM93 and J11) improves the overall statistics, with a reduction in bias and gross error (Fig. 10). The wind speed scatterplot suggests that biases could not be related to a weak representativeness of the simulated 5 yr period.

5.5 Aerosol optical depth

Simulated coarse AOD are compared with monthly climatologies at 16 AERONET sites (Fig. 11). The model AOD is all-sky in contrast to AOD measurements, which are clear-sky. The differences between all-sky and clear-sky results in models are currently uncertain and are thought to be moderate for sea-salt and very low for dust (Shindell et al., 2013).

To support the analysis, Fig. 12 displays the simulated wind speed and the NCEP wind speed climatology at each site. Overall, the simulated coarse AOD is in agreement with observations. Significant discrepancies are found in Ceilap-RG (6), Dunedin (9), Reunion Island (14), and Tahiti (16) with all schemes. Overestimation in Ceilap-RG (6) affects the entire seasonal cycle mostly due to errors in dust emissions from South America. Neglecting the dust contribution, the nearly constant seasonal cycle and its mean value (~0.02) are well reproduced. Overestimation in Bermuda (4), Dunedin (9), Reunion Island (14), and Tahiti (16) takes place mainly during austral winter (JJA). At Bermuda (4), Dunedin (9), and Tahiti (16) model peaks may be partly related to an overestimated wind speed (Fig. 12).

Both at Reunion Island (14) and Dunedin (9) the significant dust contribution leads to uncertainties in the comparison. At Ascension Island (2), Bermuda (4), Cape San Juan (5), La Parguera (11), and Midway Island (12), the model’s ability to reproduce the dust cycle is decisive for a proper simulation of the coarse AOD.

Results outline a close behavior among G03, M86 and M86/SM93, in contrast to SST-dependent schemes (MA03/M86/SM93 and J11). The latter tend to overestimate the AOD over warm sea surfaces in/near the tropics (e.g. Bermuda (4), Coconut Island (7), Guam Island (10), Midway Island (12), Reunion Island (14), and Tahiti (16)). These results are mainly related to differences in the emitted size-distribution and the hygroscopic growth of sub-micron aerosols affecting the coarse AOD. Figure 1 shows close to an order of magnitude difference in the number emission flux for particles with dry radius in the range 0.15–1.4 μm. Hygroscopic growth of particles above 0.6 μm adds up to explain the higher coarse AOD when using an SST-dependent scheme. Simulated wind speeds are stronger than NCEP climatological winds in Coconut Island (7), Guam Island (10), and Tahiti (16) (Fig. 12), which may also partly explain the overestimation in these sites. On the contrary, in Bermuda (4), Midway Island (12), and Reunion Island (14) the simulated wind speed is lower than the NCEP climatology.

In the subset of stations in/near the tropics, Jaeglé et al. (2011) found that their model generally underestimates the observed climatologies and the introduction of a SST-dependent term leads to an improvement of their model performance. In our work, we also find an increase in AOD at low latitudes when applying SST-dependent...
emission schemes, although it produces an overestimation of AERONET climatologies.

Figure 13 displays scatterplots of simulated and observed coarse AOD, excluding Ceilap-RG (6) due to the significant errors in the dust component. MA03/M86/SM93 and J11 produce the largest positive biases (+38.8 % and +27.5 %, respectively) and gross errors (above 40 %). The wind speed scatterplot indicates that such overestimation cannot be explained by a wind speed overestimation.

6 Conclusions

We presented simulations of the sea-salt aerosol global distribution with the multiscale model NMMB/BSC-CTM. Since the main uncertainties in sea-salt modeling are related to the parameterization of emissions, we implemented five different sea-salt emission schemes and analyzed their performance. We compared global simulations covering the period 2002–2006 with climatologies from the U-MIAMI and
“sea-salt-dominated” stations from the AERONET sun photometer network. For the comparison with AERONET, we use the coarse fraction of the AOD.

We found a strong sensitivity of sea-salt aerosol lifetime to the emission scheme, ranging from 7.3 h to 11.3 h. The use of SST-dependent emission schemes produces an enhancement of the sea-salt lifetime, which increases from 7.3 h with G03 to 9.6 h with J11 and from 8.9 h with M86/SM93 to 10.4 h with MA03/M86/SM93.

The SST latitudinal modulation with J11 and MA03/M86/SM93 also leads to marked differences in the global patterns of surface concentration and AOD compared with M86, M86/SM93, and G03. In particular, maximum AOD values are reached at high latitudes with M86, M86/SM93, and G03, and in the tropics with J11 and MA03/M86/SM93.

SST-dependent emission schemes lead to a clear improvement of the simulated surface concentration, with a
significant reduction of bias and gross error. In particular, J11 shows the best agreement with observations. However, the simulated coarse AOD with J11 and MA03/M86/SM93 is affected by positive biases at several AERONET sites located in the tropics. Factors that may explain the AOD overestimation include the use of all-sky model AOD in the comparison and the treatment of the water uptake, deposition, and optical properties in the model. Further research may aim at investigating SST effects upon particle size distribution.

As in previous studies, the model shows a strong overestimation in sites characterized by steep topography (Invercargill (g) in New Zealand and Marion Island (i)), independently from the applied emission scheme. Our preliminary results with a high-resolution regional simulation suggest that smaller scales play a key role in these sites. A detailed investigation at these sites is underway.

The development of the sea-salt module of the NMMB/BSC-CTM is a step forward towards an aerosol model, including dust (Pérez et al., 2011), black and organic carbon, sulfate, and its online coupling with the gas-phase chemistry (Jorba et al., 2012) to obtain a unified online multiscale chemical weather forecasting system.

Acknowledgements. We would like to thank the scientists of the AERONET Program, the University of Miami Ocean Aerosol Network, the NOAA/PMEL Laboratory, and the AEROCOM Project for establishing and providing data from the stations/cruises/models used in this work. In particular, we thank J. Prospero for his personal communications and M. Schulz for providing postprocessing of the University of Miami Ocean Aerosol Network data set. We also thank F. Benincasa for technical support. BSC acknowledges the support from projects CGL2010/19652, “Supercomputación and e-ciencia” Project (CSD2007-0050) from the Consolider-Ingenio 2010 program of the Spanish Ministry of Economy and Competitiveness and the support from the grant SEV-2011-00067 of Severo Ochoa Program, awarded by the Spanish Government.

Simulations were performed in the Marenstrum Supercomputer at BSC. The two Anonymous Reviewers are gratefully acknowledged for their detailed and helpful comments.

Edited by: A. Pszenn

References

de Leeuw, G., Andreas, E. L., Angelova, M. D., Fairall, C. W., Lewis, E. R., O’Dowd, C., Schulz, M., and Schwartz, S. E.: Pro-


Kalnay, E., Kanamitsu, M., Kistler, R., Collins, W., Deaven, D., Gandin, L., Iredell, M., Saha, S., White, G., Woollen, J., Zhu,
M. Spada et al.: Modeling and evaluation of the global sea-salt aerosol distribution


Atmos. Chem. Phys., 13, 11735–11755, 2013 www.atmos-chem-phys.net/13/11735/2013/

