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Atmospheric Research xxx (2010) xxx

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Research Highlights

Atmospheric Research xxx (2010) xxx–xxx

Sea-salt aerosol forecasts compared with daily measurements at the island of Lampedusa (Central Mediterranean)

P. Kishcha [a,*], S. Nickovic [b], B. Starobinets [a], A. di Sarra [c], R. Udisti [d], S. Becagli [d], D. Sferlazzo [c], C. Bommarito [c], P. Alpert [a]

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► In order to evaluate the model performance in the open sea, the numerical simulations of sea-salt aerosol were compared with sea-salt ground-based measurements taken at the tiny Mediterranean island of Lampedusa, Italy. ► As estimated for all 380 days used in the analysis, model-vs.-measurement comparisons at Lampedusa show a relatively high correlation of 0.7 between model data and measurements; a rather low mean bias of −0.5 μg/m^3; and a mean normalized bias less than 20%. ► The model was capable of producing reasonable SSA concentrations and their day-to-day variations over the open sea.

doi:10.1016/j.atmosres.2010.12.021

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ARTICLE INFO

Article history:
Received 16 August 2010
Received in revised form 9 November 2010
Accepted 15 December 2010
Available online xxxx

Keywords:
Sea-salt aerosol
Marine aerosols
Operational regional modeling
Sea-salt aerosol forecasts

ABSTRACT

Operational regional sea-salt aerosol forecasts have been produced on a daily basis since February 2006 over the open sea in the Mediterranean, where sea-salt aerosol concentrations and their impact on the Mediterranean weather and climate could be significant under strong winds. In order to evaluate the model performance, the numerical simulations of sea-salt aerosol (SSA) were compared with sea-salt ground-based measurements taken at the tiny Mediterranean island of Lampedusa, Italy. Considerable effort was made in order to collect and analyze SSA measurements on a daily basis, during the two-year period from 2007 to 2008. In Lampedusa, the conditions of SSA measurements are considered similar to those in the open sea, given the small dimensions of the island. As estimated for all 380 days used in the analysis, model-vs.-measurement comparisons at Lampedusa show a relatively high correlation of 0.7 between model data and measurements; a rather low mean bias of −0.5 μg/m^3; and a mean normalized bias less than 20%. Therefore, the model was capable of producing reasonable SSA concentrations and their day-to-day variations over the open sea.

1. Introduction

Sea salt aerosol (SSA) produced by surface winds, is an important component of atmospheric aerosols over the ocean. The reason for the current interest in sea-salt aerosols is their influence on climate (Lewis and Schwartz, 2004, and references therein): sea-salt could affect cloud formation by acting as cloud condensation nuclei (CCN) and contributing from 5% to 90% of CCN in the marine boundary layer (Clarke et al., 2006, Rosenfeld et al., 2002). Furthermore, SSA scatters solar radiation and thereby plays an important role in the atmospheric radiation budget (Haywood et al., 1999; Satheesh and Lubin, 2003). Some investigations have dealt with the possible effects of SSA on hurricane strength and development (Emanuel, 2003).

The Mediterranean Sea, being an almost-closed area surrounded by mountain ranges, has experienced elevated aerosol loading (Lelieveld et al., 2002; Papadimas et al., 2008). This region is of special importance because it is a crossroad where natural (Saharan dust and sea-salt) aerosols and anthropogenic aerosols from Africa, Europe, and Asia are superimposed. There are publications devoted to sea-salt aerosol studies in the Mediterranean region (Astitha et al., 2008; Athanasopoulou et al., 2008; Barnaba and Gobbi, 2004; Blot et al., 2008; Levin et al., 2005; Pace et al., 2006; Querol et al., 2004; Quinn et al., 2000; Viana et al., 2005, 2007; Zakey et al., 2008). In spite of the importance of SSA effects on the Mediterranean climate and weather, there are no regular sea-salt measurements in the open sea, where sea-salt aerosols are

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doi:10.1016/j.atmosres.2010.12.021

mainly produced, and where their concentration and impact on the Mediterranean climate could be significant. In order to partly fill the gap in our understanding of the SSA processes, our model-based daily forecasts of 3-D distribution of SSA over the Mediterranean could be helpful, providing valuable information about space and time distribution of this kind of aerosol.

In order to produce operational SSA forecasts over the open sea in the Mediterranean, the regional DREAM-Salt model with horizontal resolution 0.3° has been running daily at Tel-Aviv University, since February 2006. This study was aimed at evaluating the model performance in the open sea by comparing quantitatively daily model-predicted sea-salt aerosol concentrations with daily SSA measurements taken at the very small Mediterranean island of Lampedusa, located in the Central Mediterranean.

2. Sea-salt model

Numerical simulations of the sea-salt aerosols presented in this study were conducted using a version of the DREAM dust aerosol model (Nickovic et al., 2001; Kishcha et al., 2007, 2008) with embedded SSA component (DREAM-Salt) (Nickovic et al., 2007). The SSA component was embedded into the DREAM model, in order to produce simultaneously operational forecasts of both Saharan desert dust and sea-salt aerosols over the Mediterranean. The DREAM-Salt prediction system has been producing daily forecasts of 3-D distribution of sea-salt aerosol concentration over the Mediterranean model domain 20°W–45°E, 15°N–50°N (http://wind.tau.ac.il/salt-ina/salt.html). The model has 0.3° horizontal resolution and 24 vertical levels. Forecasts are made once every day, starting from the NCEP 12:00 UTC objective analyses and providing forecasts up to 72 h ahead.

The NCEP/Eta regional atmospheric model (Janjic, 1994, and references therein) drives the aerosol. The aerosol emission scheme is based on the viscous sub-layer model (Janjic, 1994), in which energy and mass transfers above the air–sea interface critically depend on turbulent conditions. The Janjic viscous sub-layer scheme is based on the following assumptions: (a) there are two distinct layers: a thin viscous sub-layer immediately above the surface and a turbulent layer above the viscous sub-layer; (b) at the top of the viscous sub-layer all fluxes are continuous. In the viscous sub-layer, it is assumed that (1) vertical transport is determined entirely by the molecular diffusion; and (2) vertical profiles of variables are linear since the viscous diffusivity is assumed to be constant. In the turbulent layer, the vertical transport is entirely defined by turbulent fluxes. Depending on the Reynolds roughness number, $\mathrm{Re} = z_0 \cdot u^*/\nu$, the viscous sub-layer scheme is assumed to operate in three different regimes: smooth and transitional; rough; and rough with sea spray. The parameters z_0, u^*, and ν are roughness height, friction velocity and air viscosity, respectively. When Re exceeds a prescribed critical value Re, the flow ceases to be smooth and enters the rough regime. The rough regime is characterized by combined viscous and turbulent mixing. In the rough regime with sea spray, the mixing becomes fully turbulent. Here, the breaking waves provide a mass exchange, which is more effective than that of the two previous regimes. The values of u^* at which the transitions between the different regimes occur are $u^* = 0.225$ m/s and $u^* = 0.7$ m/s.

Following Janjic (1994), the sea-salt fluxes are defined by the following expressions:

$$F_{C(VSC)} = \nu \cdot \frac{C_{INT} - C_S}{z_{INT}}; \text{ and } F_{C(TRB)} = K_C \cdot \frac{C_{LM} - C_{INT}}{z_{LM} - z_{INT}}$$

in the viscous and turbulent sub-layers, respectively. Here, K_C is the surface layer Monin–Obukhov bulk turbulent mixing coefficient; C_S, C_{INT}, and C_{LM} are sea salt concentrations at the sea surface, at the top of the viscous sub-layer and at the first computational model level, respectively; and z_{INT} and z_{LM} are the heights of the top of the viscous sub-layer and the first computational model layer, respectively. The depth of the viscous sub-layer is calculated as $z_{INT} = \frac{0.35 \cdot M \cdot \sqrt[4]{\mathrm{Re}} \cdot \sqrt[2]{S_c} \cdot \nu}{u_*}$ (Janjic, 1994). Here, S_C is the Schmidt number; the constant Mhas a value of 30 in the first regime and 10 in the second regime. The viscous sub-layer depth z_{INT} decreases as the turbulence increases. The viscous sub-layer vanishes in the last rough regime with sea spray. From the requirement for continuity of the viscous and turbulent fluxes at the viscous/turbulent interface, it follows that

$$C_{INT} = \frac{C_S + \omega \cdot C_{LM}}{1 + \omega}, \text{ where } \omega = \frac{K_C \cdot z_{INT}}{\nu \cdot (z_{LM} - z_{INT})}$$

where ω plays the role of a weighting factor. Note that ω vanishes with the disappearance of the viscous sublayer in the rough regime with spray. As a consequence, it follows that $C_{INT} = C_S$ at $z = z_0$. In the Janjic scheme, the interface value C_{INT} is considered as the lower boundary condition for the surface layer turbulence scheme in the NCEP/Eta model.

In our approach, the aerosol concentration at the top of the viscous sub-layer is used as the lower boundary condition. In this approach, the effects of the viscous sub-layer model are fully taken into account. The sea-salt emission scheme defines the lower boundary condition using the source function of Erickson et al. (1986):

$$C_S^j = 10^{-9} \cdot \alpha^j \cdot \exp(0.16 \cdot U_{10} + 1.45) \quad \text{for } U_{10} \le 15\,\mathrm{m/s},\ j = 1, N$$

$$C_S^j = 10^{-9} \cdot \alpha^j \cdot \exp(0.13 \cdot U_{10} + 1.89) \quad \text{for } U_{10} > 15\,\mathrm{m/s},\ j = 1, N$$

where U_{10} is the 10 m wind speed. In our model setup, we used N = 8 particle size bins (1.0–1.5, 1.5–2.5, 2.5–3.5, 3.5–4.5, 4.5–5.5, 5.5–6.5, 6.5–7.5, and 7.5–8.5 µm). α^j is an array describing the mass going into each of the eight particle size bins, which is in percentages 0.5, 1.5, 6.0, 11.0, 16.0, 19.0, 21.0, and 25.0 of the total source, respectively. The dependence of SSA productions and size distributions on relative humidity was not included in the model. Note that the source function of Erickson et al. (1986) was also used by Tegen et al. (1997) and Reader and McFarlane (2003) for sea-salt aerosol modeling.

In addition to the sea-salt emission, DREAM-Salt incorporates parameterizations of all other major phases of atmospheric sea-salt aerosol life such as diffusion, advection, gravitational settling, and wet removal of sea-salt aerosols (Nickovic et al., 2001).

Please cite this article as: Kishcha, P., et al., Sea-salt aerosol forecasts compared with daily measurements at the island of Lampedusa (Central Mediterranean), Atmos. Res. (2010), doi:10.1016/j.atmosres.2010.12.021

3. Model-vs.-measurement comparison in Lampedusa

Lampedusa is a tiny island in the central Mediterranean which measures several kilometers, far from large islands and continental areas (Fig. 1). Lampedusa is characterized by clean air without industrial pollution. One can consider that sea-salt aerosol measurement conditions on this tiny island are similar to those in the open sea. The Lampedusa sea-salt aerosol monitoring site (35°31′N; 12°38′E) is run by the Italian National Agency for New Technologies, Energy and Economical Sustainable Development. This site is located at approximately 10 m from the northern coastline, at 50 m elevation.

Sea-salt aerosol concentrations were obtained by means of chemical composition determination of PM10 aerosol measurements. Considerable effort was made in order to collect and analyze SSA measurements, on a daily basis, over the two-year period, 2007–2008. Aerosol is sampled on Teflon filters (Pall, 47 mm diameter, 2 µm nominal porosity) with an EN 12341 sampler operating at 2.3 m^3/h with a PM10 pre-selected cut-off head. Samples were collected over 24 h, with a start time of 00:01 UTC, in order to show day by day variations. A quarter of each filter was extracted using MilliQ water (Resistivity > 18 MΩ) in ultrasonic bath for 20 min and ionic content (including sea spray markers) was determined by ion chromatography (Udisti et al., 2004).

SSA at Lampedusa was calculated as the sum of the weight of $ssNa^+$ (i.e. sea salt Na^+), Cl^-, $ssMg^{2+}$, $ssCa^{2+}$ and $ssSO_4^{2-}$. The fraction $ssNa^+$ and $ssCa^{2+}$ was calculated by the following equations:

$$ssNa = Na - nssNa = Na - nssCa * (Na/Ca)_{crust}$$

$$nssCa = Ca - ssCa = Ca - ssNa * (Ca/Na)_{seawater}$$

$$(Na/Ca)_{crust} = 0.56[w/w] \quad \text{(Bowen, 1979)}$$

$$(Ca/Na)_{seawater} = 0.038[w/w] \quad \text{(Bowen, 1979)}$$

where $(Na/Ca)_{crust}$ is the mean ratio in the Earth crust, $(Ca/Na)_{seawater}$ the mean ratio in bulk seawater, and ss and nss stand for sea-salt and non sea-salt respectively. Ratios are expressed as weight on weight [w/w]. In the Lampedusa aerosol, the $ssNa^+$ is 96% of the total Na^+, while the $ssCa^{2+}$ is only 20% of the Ca^{2+} budget. After $ssNa^+$ calculation, the $ssMg^{2+}$ and $ssSO_4^{2-}$ contributions were evaluated by their ratios in sea water (Mg/Na = 0.129 w/w and SO_4/Mg 0.253 w/w).

It is known that Cl^- in atmospheric particulate undergoes depletion processes, mainly due to exchange reactions with H_2SO_4 and HNO_3, leading to re-emission of HCl in the atmosphere. Besides, Cl^- losses from the filter surface can

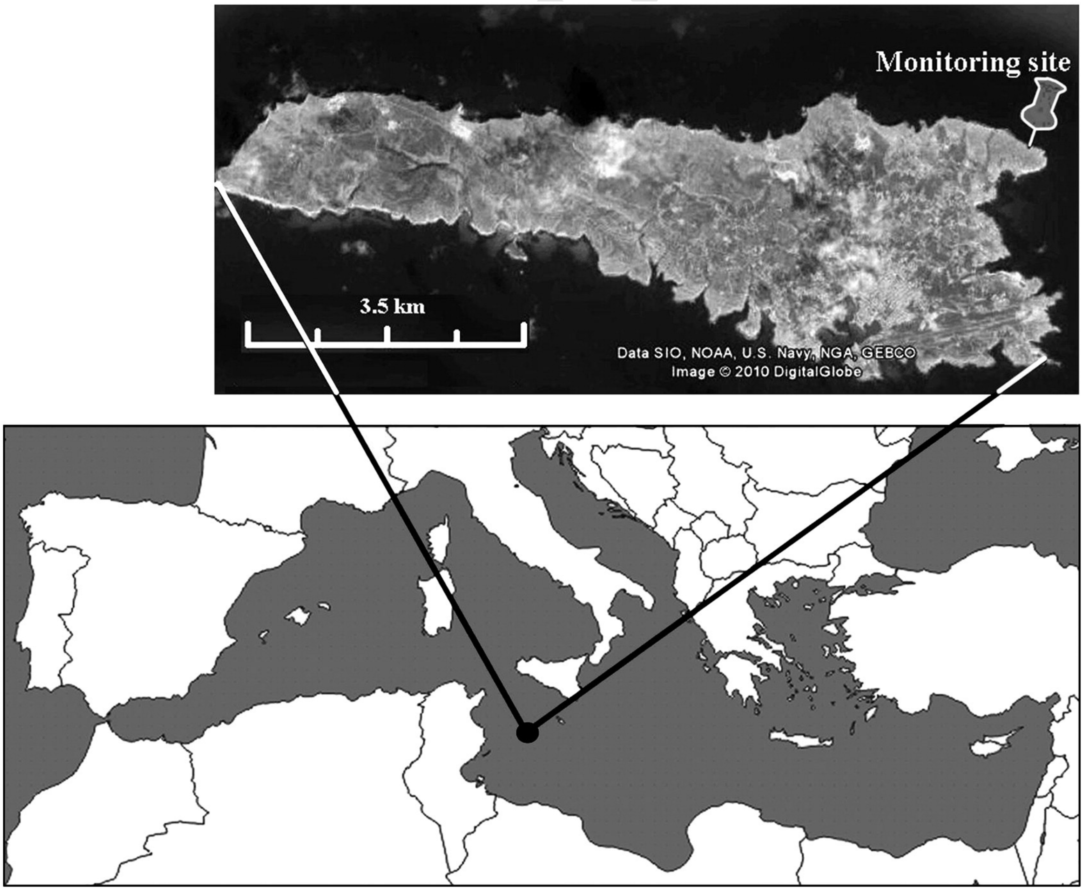

Fig. 1. A map of the Mediterranean Sea and a map of the island of Lampedusa with the monitoring site (the grey place mark).

be caused by reactions between Cl^- (from sea salt aerosol) and anthropogenic combustion products (such as NOx, O_3, HO and HO_2 radicals), producing atomic chlorine (e.g. Quinn et al., 2000; Wingenter et al., 1999, and references therein). In such aged sea spray aerosol, the Cl/Na ratio is lower than that found for fresh sea spray aerosol. At Lampedusa, the sampling site is near to the sea spray source; therefore, this effect can be considered negligible. At Lampedusa, extra sources for Cl^- are sporadic. Therefore, $ssNa^+$ and Cl^- are reliable markers for the evaluation of SSA content.

The regional DREAM-Salt model was developed for the purpose of producing operational SSA forecasts over the open sea, where SSA concentrations depend on wind speed but do not depend on wind direction. Because of coarse resolution,

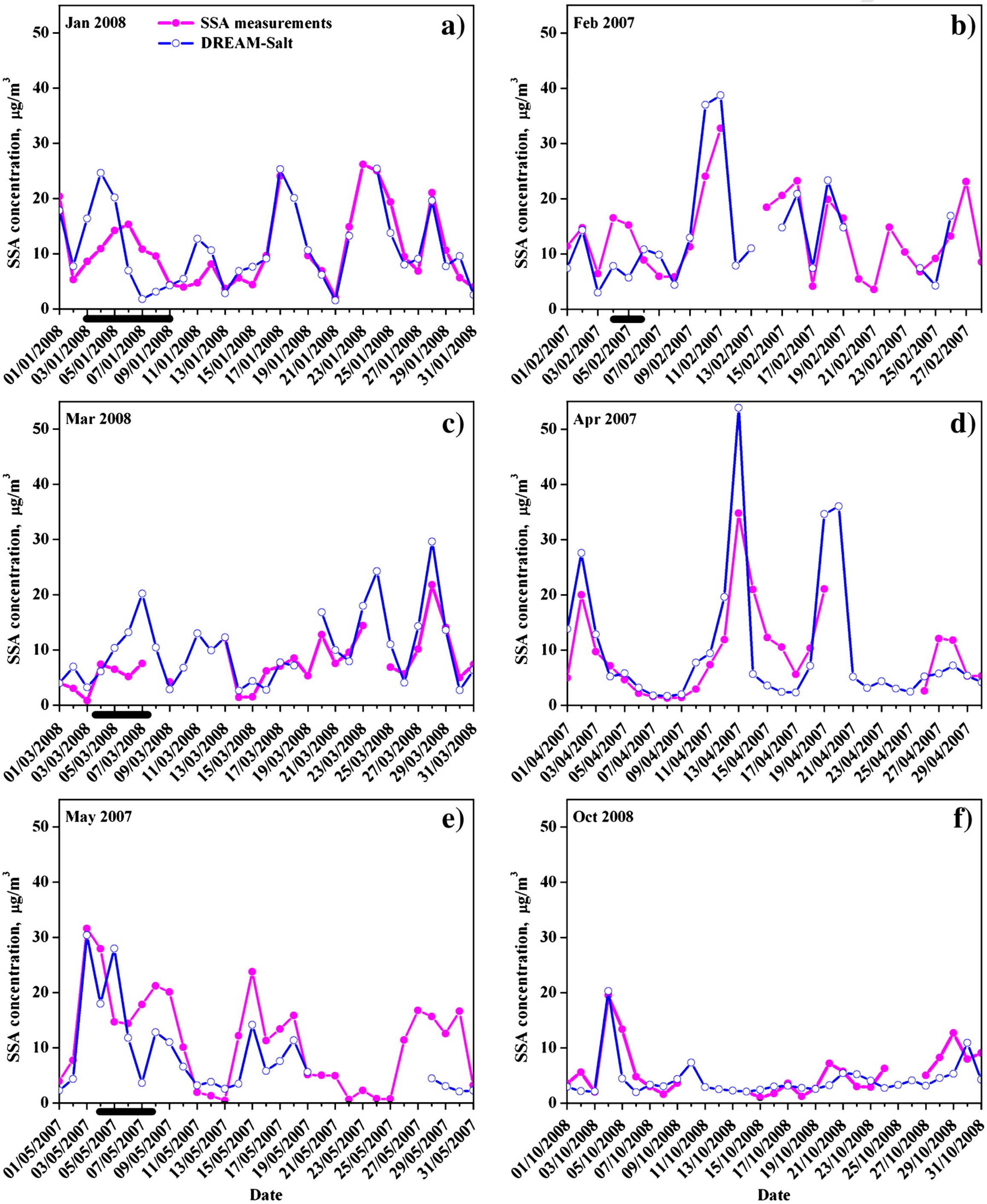

Fig. 2. Examples of the comparison between modeled and measured sea-salt aerosol concentrations at the Lampedusa site. The black underlines designate periods with discrepancies between modeled and measured SSA concentrations.

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the model cannot take into account local factors such as sea-breezes and surf zones. Despite the small size of Lampedusa, sea-breezes could exist there in the summer months. Therefore, the summer months, from June to August, were excluded from the analysis.

Fig. 2 shows examples of comparisons between modeled daily SSA-concentrations and SSA measurements taken in different months at the Lampedusa site. It is clearly seen that, in general, model data fit measurements quite well. To estimate the model performance in quantitative terms for different months, we utilized general performance metrics as shown in Table 1. As estimated, for different months during the two-year period 2007–2008, the majority of correlation coefficients between modeled SSA concentrations and measurements are mainly above 0.6, which demonstrates the ability of our model to reproduce measured SSA concentrations. The obtained correlation coefficients were found to be statistically significant at the 0.05 level. The averaged simulated aerosol concentrations ranged mainly within the same intervals as the measurements did (Table 1). The relatively high standard deviation indicates strong variability of sea-salt aerosol concentrations due to the strong variability of wind speed. Our analysis for different months shows that the mean bias could be both negative and positive. This indicates that the model could sometimes underestimate or overestimate measurements. As estimated for different months, the mean normalized bias was mainly within a range of ±25% (Table 1).

Although there is general agreement between modeled and measured SSA concentrations, for some short periods (e.g. January 3–9, 2008; February 4–6, 2007; March 3–7, 2008; and May 4–8, 2007) the discrepancies are quite large (Fig. 2). In order to understand the cause of the discrepancies between modeled and measured SSA concentrations, we analyzed the relationship between measured SSA concentrations and observed wind speed at the monitoring site (Fig. 3). One can see that during the aforementioned periods with discrepancies, there is no correlation between wind and SSA measurements. Our forecast model uses wind speed as a key parameter for sea-salt aerosol production. Therefore, the model cannot predict SSA concentrations when there is no correspondence between measured SSA concentrations and wind speed. A possible reason for the discrepancies between measured SSA concentrations and wind speed could be the fact that the measured conditions in Lampedusa only approximately correspond to those in the open sea. A comprehensive analysis of the discrepancies between measured SSA concentrations and observed wind speed at the monitoring site in Lampedusa is beyond the scope of the current study.

To demonstrate model performance under different wind directions, Table 2 represents model performance metrics for four groups of data: north winds (0°–45° and 315°–360°), south winds (135°–225°), west winds (225°–315°), and east winds (45°–135°). According to our analysis, there is no distinct dependence of the correlation between model data and measurements on wind direction: all of the correlation coefficients are above 0.6. This supports our suggestion that, in general, measurement conditions at the Lampedusa Island approximately correspond to those in the open sea. As estimated, under south and east winds, the model performance was characterized by a rather small mean bias of 0.1 μg/m³ and −0.2 μg/m³ respectively. Some noticeable negative bias (−1.7 μg/m³) was detected under weather conditions accompanied by north winds (Table 2). A plausible explanation of the negative bias under north winds is that the monitoring site is located near the northern coast and not in the middle of the island. Strong north winds create a surf zone, within which sea-waves, approaching the coastline, start breaking (De Leeuw et al., 2000). This could result in an additional source of SSA near

Table 1

Performance metrics for the DREAM-Salt model performance evaluation based on the two-year daily SSA measurements, 2007–2008, taken at the Lampedusa site (x) and corresponding model simulations (y); number of days with measurements (K); measured daily wind speed at the monitoring site (mean, minimum, and maximum values); correlation coefficient between modeled and measured SSA concentrations (R); mean SSA values (<x> and <y>); standard deviations (σ_x and σ_y); mean bias $MB = \frac{1}{K}\sum_{i=1}^{K}(y_i - x_i)$, and mean normalized bias $MNB = \frac{1}{K}\sum_{i=1}^{K}\frac{(y_i - x_i)}{x_i} \times 100\%$. Only months with K≥15 were used in the analysis. The obtained correlation coefficients were found to be statistically significant at the 0.05 significance level.

Year/Month	K	Wind m/s			R	<x> μg/m³	<y> μg/m³	σ_x μg/m³	σ_y μg/m³	MB μg/m³	MNB %
		mean	min	max							
2007											
February	19	8.1	2.9	15.1	0.85	14.0	13.8	7.6	10.2	−0.2	−1
March	26	6.1	2.4	9.3	0.54	8.4	9.0	5.3	6.1	0.6	25
April	24	5.4	1.7	11.9	0.83	9.5	10.3	8.0	12.4	0.8	18
May	23	6.9	2.2	14.0	0.70	13.2	8.6	8.4	7.9	−4.6	−1
September	25	6.2	2.4	12.4	0.51	9.1	5.3	5.2	3.4	−3.8	−30
October	15	6.2	2.3	12.1	0.75	8.5	6.1	5.6	3.0	−2.4	2
December	24	8.2	2.4	13.6	0.52	10.7	13.2	5.9	9.7	2.5	34
2008											
January	29	7.2	3.1	16.8	0.75	10.3	10.8	6.4	7.0	0.4	14
February	25	6.2	1.9	15.4	0.72	7.3	7.4	6.8	6.5	0.1	37
March	24	6.7	2.6	13.4	0.83	7.9	9.8	4.7	6.6	1.9	33
April	23	7.8	4.1	13.7	0.64	13.1	11.7	6.5	6.7	−1.4	18
May	20	5.5	2.3	10.9	0.90	9.6	10.0	8.2	10.9	0.5	7
September	26	4.9	1.9	12.9	0.58	5.8	4.4	5.9	3.5	−1.4	19
October	25	5.0	1.9	11.4	0.74	5.5	4.4	4.4	3.8	−1.1	4
November	17	5.3	2.8	7.7	0.50	5.9	4.2	2.6	1.8	−1.7	−21

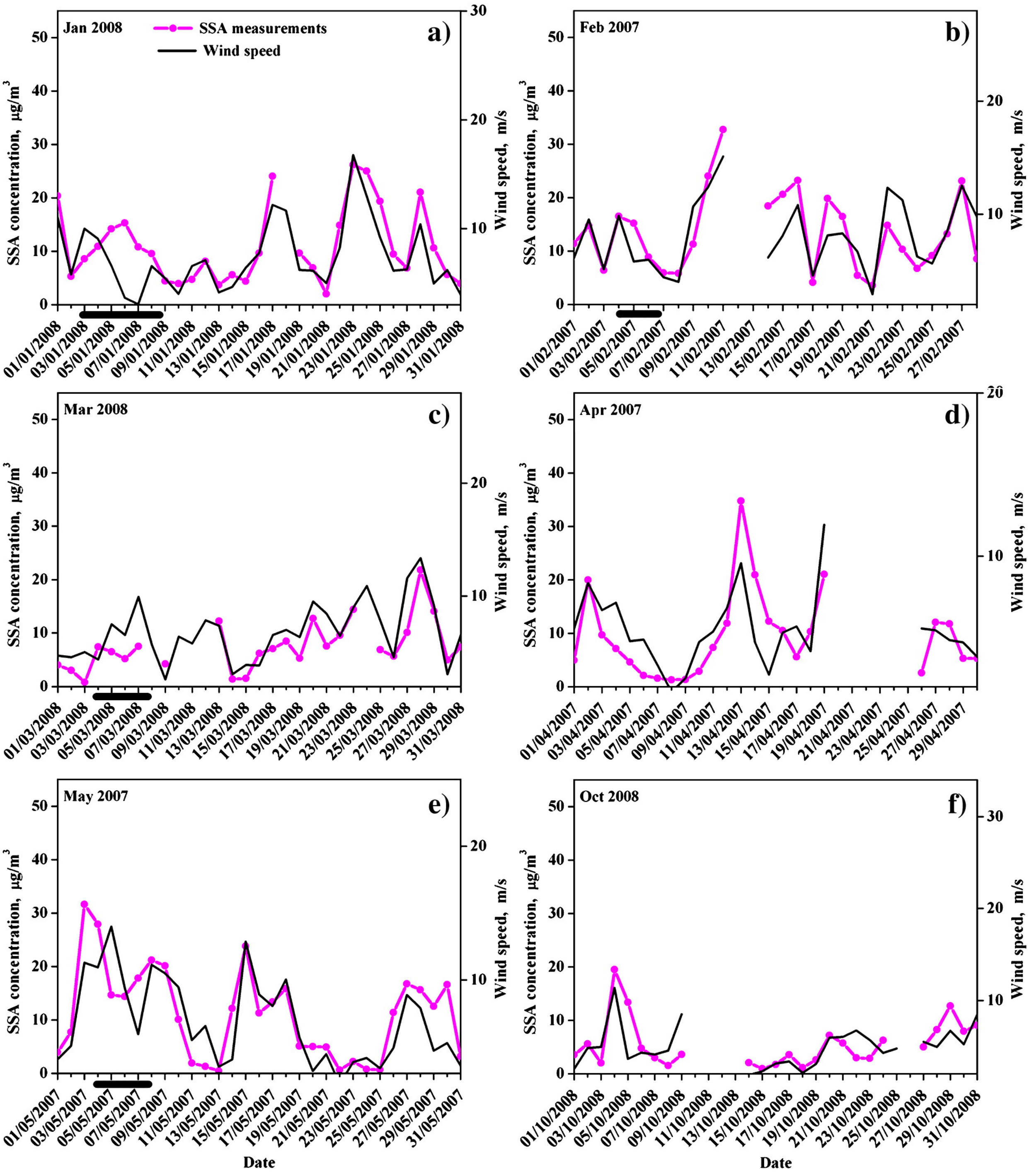

Fig. 3. Comparisons between measured sea-salt aerosol concentrations and observed wind speed at the monitoring site in Lampedusa for the examples shown in Fig. 2. The black underlines designate periods with discrepancies between measured SSA concentrations and wind speed.

the northern coast, where the monitoring site is located. Because of coarse resolution, our regional model could not take into account the effects of surf zones; this results in some model underestimation of measurements. Note that for strong winds a surf zone is created under any wind direction with respect to the island position. However, the differences, observed among the four wind directions, suggest that the production of the surf zones near the west, south and east coasts do not reach the monitoring site due to gravitational settling of sea-salt aerosols along the island.

We found that the mean normalized bias (MNB) increased from −4% under north winds to +34% under south winds (Table 2). In accordance with its definition, the mean normalized bias could have increased because of small values of measured

Table 2
DREAM-Salt performance metrics, based on daily SSA measurements taken at the Lampedusa site, for four groups of data with different wind directions: south winds (135°–225°), north winds (0°–45° and 315°–360°), east winds (45°–135°), and west winds (225°–315°). The designations are the same as in Table 1.

Groups of data	K	Wind m/s			R	<x> μg/m^3	<y> μg/m^3	σ_x μg/m^3	σ_y μg/m^3	MB μg/m^3	MNB %
		mean	min	max							
All data	380	6.4	1.7	15.4	0.70	9.2	8.7	6.8	7.7	−0.5	19
South winds	125	5.7	1.9	15.4	0.63	7.6	7.7	5.9	6.4	0.1	34
North winds	52	8.5	3.6	15.1	0.74	12.8	11.1	7.4	8.4	−1.7	−4
East winds	89	6.4	1.7	13.6	0.77	9.7	9.5	6.8	9.2	−0.2	9
West winds	114	6.3	1.9	13.3	0.61	8.9	8.2	6.4	6.9	−0.7	19

SSA concentrations: $MNB = \frac{1}{K}\sum_{i=1}^{K}\frac{(y_i - x_i)}{x_i} \times 100\%$, where x_i and y_i are measured and modeled SSA concentrations respectively, and K stands for the number of days with measurements. Mean measured SSA concentrations were estimated to be 12.8 μg/m^3, 9.7 μg/m^3, 8.9 μg/m^3, and 7.5 μg/m^3 under north, east, west, and south winds respectively (Table 2).

As estimated for all 380 days used in the analysis, the model performance was considered acceptable: it was characterized by a relatively high correlation of 0.7; a rather small mean bias of −0.5 μg/m^3; and a mean normalized bias less than 20% (Table 2). For the entire period under consideration, mean SSA concentrations were found to be 9.2 μg/m^3 and 8.7 μg/m^3, based on measured and modeled data respectively (Table 2).

4. Conclusions

The current study on sea-salt aerosol forecasts over the Mediterranean Sea uses numerical simulations of space and time distributions of sea-salt aerosols produced by the regional prediction system DREAM-Salt with an embedded SSA component. The operational regional SSA forecasts have been produced since February 2006 for the purpose of estimating daily, seasonal, and inter-annual variability of sea-salt aerosols over the open sea in the Mediterranean. There, sea-salt aerosols are mainly produced, and their impact on the Mediterranean weather and climate could be significant under strong winds.

Model performance in the open sea has been directly verified in this study by comparing quantitatively, on a daily basis, model-predicted sea-salt aerosol concentrations with available SSA-measurements, taken at the tiny Mediterranean island of Lampedusa, Italy, during the two-year period, 2007–2008. Given the small dimensions of the island, the conditions for taking SSA measurements at the Lampedusa site approximately correspond to those in the open sea. Model-vs.-measurement comparisons at Lampedusa for all 380 days with SSA measurements show that the model performance was quite acceptable: it was characterized by a relatively high correlation of 0.7; a rather small mean bias of −0.5 μg/m^3; and a mean normalized bias less than 20% (Table 2). This highlights that our model was capable of producing reasonable sea-salt concentrations and their day-to-day variations over the open sea.

The absence of correlation between measured SSA concentrations and observed wind speed at the monitoring site in Lampedusa during some short periods, as shown in Fig. 2, resulted in discrepancies between modeled sea-salt aerosol concentrations and SSA measurements (Fig. 3). An additional possible source of error is that the model did not take into consideration sea-salt aerosols produced by sea-waves in the surf zone. Future improvements of the model will resolve this issue.

Acknowledgements

This study was supported by the GLOWA-Jordan River BMBF (Germany)-MOST (Israel) project, the BMBF-MOST grant number 1946 on global change. PM10 measurements in Lampedusa were supported by the Italian Ministry for University and Research through the SNUMMAS project. Contributions by Lorenzo De Silvestri are gratefully acknowledged.

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