



Do new sea spray aerosol source functions improve the results of a regional aerosol model?

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ABSTRACT

Sea spray aerosol particle is a dominating part of the global aerosol mass load of natural origin. Thus, it strongly influences the atmospheric radiation balance and cloud properties especially over the oceans. Uncertainties of the estimated climate impacts by this aerosol type are partly caused by the uncertainties in the particle size dependent emission fluxes of sea spray aerosol particle. We present simulations with a regional aerosol transport model system in two domains, for three months and compared the model results to measurements at four stations using various sea spray aerosol particle source functions. Despite these limitations we found the results using different source functions are within the range of most model uncertainties. Especially the model's ability to produce realistic wind speeds is crucial. Furthermore, the model results are more affected by a function correcting the emission flux for the effect of the sea surface temperature than by the use of different source functions.

1. Introduction

Sea spray aerosol particle (SSA) (de Leeuw et al., 2011) play an important role in the atmospheric aerosol load and atmospheric processes above the ocean. They affect cloud formation by acting as cloud condensation nuclei (Ayash et al., 2008) and impact on the solar radiation reaching the surface directly by scattering radiation or indirectly via their effects on clouds (Lewis and Schwartz, 2004). Since ocean areas cover 70 % of the Earth's surface these influences are important at the global scale. The importance of these effects is still uncertain. For example, Lewis and Schwartz (2004) calculated a decrease of the solar radiation reaching the surface between -6.2 and -0.08 W m^{-2} due to scattering by SSA in the cloud-free atmosphere. Furthermore, the indirect effect on the radiation through the effect of SSA on clouds is found to range between -2.9 and 0.53 W m^{-2} in different models (Ma et al., 2008; Partanen et al., 2014). The uncertainties in the effects are affected by the uncertainties in the SSA emission fluxes within the aerosol models.

The majority of the SSAs with relevance for radiation and cloud effects are emitted from the ocean surface via the bursting of air bubbles. These bubbles result from the breaking of wind-induced waves. During its collapse a bubble can produce up to several 100 film droplets. These droplets can have radii (r_{80}) from 10 nm to $1 - 2 \mu\text{m}$ at 80 % relative humidity (Gong et al., 1997; Mårtensson et al., 2003; Sellegri

et al., 2006; Veron, 2015). Small amounts of these film droplets have larger sizes (Afeti and Resch, 1990; Spiel, 1998). Due to the remaining bubble cavity at the ocean surface a water jet is formed and raised to a few millimetres above the surface. When it breaks it forms up to 6 jet droplets with a typical size range of $1 \lesssim r_{80} \lesssim 50 \mu\text{m}$ (Lewis and Schwartz, 2004; de Leeuw et al., 2011, 2014; Veron, 2015).

There are several atmospheric and oceanic parameters influencing the number-size distributions of the freshly emitted SSA, which are summarised in the literature (Lewis and Schwartz, 2004; de Leeuw et al., 2014; Salter et al., 2014). Here we only consider the influence of the surface wind speed and the sea surface temperature SST. Up to now these are the only parameters (besides wave state (Ovadnevaite et al., 2012; Partanen et al., 2014)) that are used in atmospheric modelling of SSA emission and transport. The surface wind speed is the major driver for the emission (Lewis and Schwartz, 2004; de Leeuw et al., 2011, 2014). A higher wind speed leads to higher emission fluxes for all particle sizes. This dependence is usually parameterized with an exponent of 3.41–3.74 to the wind speed at 10 m above the surface (U_{10}). The effect of the SST is less clear, but it appears that the emission rates of smaller particles ($r_{80} < 1 \mu\text{m}$) are decreased at higher temperatures whereas the emission rates of larger particles increase. The transition between the small and large particle sizes is still under discussion. The physical basis and available observations are summarised in e.g. Lewis and Schwartz (2004); Salter et al. (2014, 2015).

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Table 1

List of selected aerosol models with their best SSA source function found in comparison studies. Since some of the studies were carried out a few years ago, the actual configurations of the models may differ. The acronyms in the third column stand for: c. combination of; b. based on; m. modification of.

Model	Scale	Source function	Source
COSMO-ART	regional	c. Mårtensson et al. (2003), Monahan et al. (1986), Smith et al. (1993)	Lundgren et al. (2013)
EMEP-chem. transp. mod.	regional	c. Mårtensson et al. (2003), Monahan et al. (1986)	Tsyro et al. (2011)
LOTUS-EUROS	regional	c. Mårtensson et al. (2003), Monahan et al. (1986)	Manders et al. (2010)
WRF-chem	regional	Gong (2003)	Chen et al. (2016)
CAMS	global	c. Mårtensson et al. (2003), Monahan et al. (1986)	Meskhidze et al. (2011)
ECHAM-HAM	global	Guelle et al. (2001); c. Monahan et al. (1986), Smith and Harrison (1998)	Stier et al. (2005)
GEOS-chem	global	m. Gong (2003)	Jaeglé et al. (2011); Gantt et al. (2012)
GISS-THOMAS	global	Clarke et al. (2006)	Westervelt et al. (2012)
SILAM	global	New; b. Mårtensson et al. (2003) and Monahan et al. (1986)	Sofiev et al. (2011)

Table 2

List of acronyms for the source functions.

G03	SSA source function of Gong (2003)
G14	SSA source function of Grythe et al. (2014)
L11	SSA source function of Long et al. (2011)
MMS	Combined SSA source function of Mårtensson et al. (2003); Monahan et al. (1986) and Smith et al. (1993)
S11	Source function of Sofiev et al. (2011) without part for SST
S15	SSA source function of Salter et al. (2015)
J11T	SST dependent correction for SSA source function by Jaeglé et al. (2011)
S11T	SST dependent correction for SSA source function by Sofiev et al. (2011)

In the past decades, many studies from local to global scale compared measurements with simulation results using various SSA source functions. This resulted in different source functions producing the best results in the different models. Table 1 shows a summary for some of these functions.

Grythe et al. (2014) compared global SSA emission rates calculated with the source function of Monahan et al. (1986) within the seven different aerosol models presented by Lewis and Schwartz (2004). The results for the different models ranged from 3.3 to 11.7 Pgyr⁻¹. Gong (2003) noted that the quality of a source function strongly depends on the dynamics of the atmospheric model. This is a reason for the spread in the simulated global SSA emission fluxes in addition to the fact that different aerosol models use different source functions (Table 1). Thus, it is problematic to generalise the results of one modelling study for other models. Therefore we focus on the comparison of different SSA source functions with each other in relation to possible uncertainties in the model system and the model-measurement comparison.

A comparison of SSA source functions was carried out by Grythe et al. (2014) by analysing the performance of more than 50 source functions. They excluded several of the functions due to duplication and incomplete descriptions. Thus, 21 remained for their study. They compared the results of the trajectory model FLEXible PARTicle dispersion model FLEXPART in backward mode with 15341 individual measurements of PM₁₀ spread over the globe. Some of the source functions were incompatible with the measurements, but several parameterizations resulted in similar high correlations of model results with the observations. In consequence they suggested a new source function based on Smith and Harrison (1998). It includes a term for the SST dependence published by Jaeglé et al. (2011). This new source function resulted in an improved model performance.

Since Grythe et al. (2014) used a Lagrangian dispersion model driven by re-analysed wind fields from European Center for Medium-range Weather Forecast ECMWF their results should be close to reality. However, using an aerosol model in forecast mode instead, the results and conclusions can be different. Grythe et al. (2014) also found that the quality of the SSA source functions was different in different regions, thus regional effects may have an influence.

Here we use the regional aerosol transport model COSMO-MUSCAT in a short-time forecast mode. It is typically used in European and north African regions (Renner and Wolke, 2010; Heinold et al., 2011a; b;

2012; Wolke et al., 2012; Meier et al., 2012). Thus, we focus on these regions while studying the quality of five SSA source functions. Considering the results of Grythe et al. (2014) we selected the functions of Gong (2003); Sofiev et al. (2011) and Grythe et al. (2014). Additionally, the combined source function MMS (Mårtensson et al., 2003; Monahan et al., 1986; Smith et al., 1993) is included, because it is used in several aerosol models (Table 1). Furthermore, we include the source function of Long et al. (2011), which was excluded in the simulations of Grythe et al. (2014). Finally, the source function of Salter et al. (2015) is also part of our study.

A clear dependence of the SSA production on the SST was found (Jaeglé et al., 2011; Sofiev et al., 2011; Grythe et al., 2014). With Jaeglé et al. (2011) and Sofiev et al. (2011) two functions are available to consider this effect in the calculation of the SSA emission fluxes. Validating both Grythe et al. (2014) found the function of Jaeglé et al. (2011) performing better in the model-measurement comparison. Both correction functions were developed to be applied to existing SSA source functions, but up to now they were only used within the original published combinations. Since the quality of source functions depends on the model dynamics, we compare different combinations of SST correction functions and SSA source functions. The SSA source function by Salter et al. (2015) includes also a term for the SST dependence.

In Section 2 we describe the different functions for SSA emission and the SST effects, before in Section 3 the model, its setup and the used measurements are explained. Section 4 describes the model results and possible uncertainties in the model studies. In Section 5 we discuss the effects of the different SSA source functions in relation to the influence by the SST and put them in the context of the uncertainties.

2. Description of the SSA source functions

In several studies it was found that the source function of Monahan et al. (1986) provided the best results in the size range $0.2 \leq r_{80} \leq 1 \mu\text{m}$ (ex.: Andreas et al., 1995; Guelle et al., 2001; Gong, 2003). Thus, it is often used but modified to perform better outside of that size range. The source function of G03 is one of these modifications. It was developed to reproduce the measurements of O'Dowd et al. (1997) with the model CAM and is valid in the size range of $0.07 < r_{80} < 20 \mu\text{m}$. Another source function based on Monahan et al. (1986) is that of S11. The authors modified the original function to be able to reproduce the laboratory measurements by Mårtensson et al. (2003). This function has additional terms to account for the dependence on the ocean salinity and the SST. The term for the salinity is not considered here. The measurements by Mårtensson et al. (2003) were carried out for particles with a dry diameter of $D_{p_{dry}} < 2.8 \mu\text{m}$. However, the derived SSA source function was extrapolated beyond that size range to $0.01 \leq D_{p_{dry}} \leq 10 \mu\text{m}$. Another approach to combine the outcome of Monahan et al. (1986) and Mårtensson et al. (2003) is used by several authors (ex.: Manders et al., 2010; Tsyro et al., 2011; Lundgren et al., 2013). They take the two source functions in predefined size ranges. We include the definitions of Lundgren et al. (2013) with their addition of the source function of

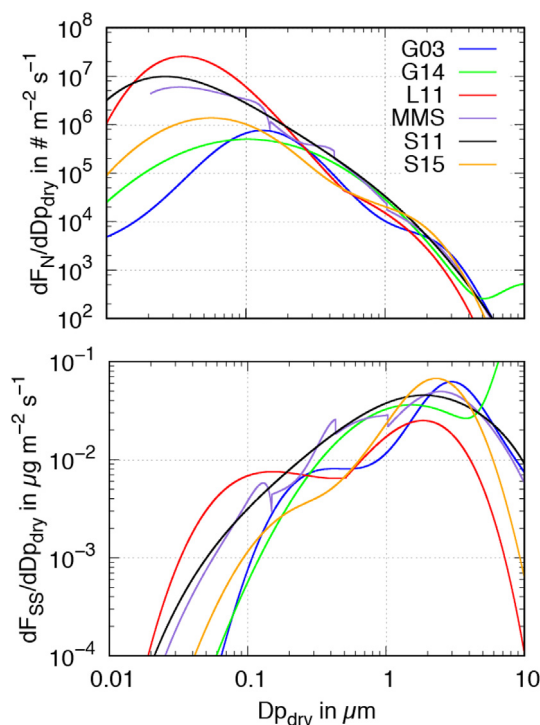


Fig. 1. Effective number (top) and mass (bottom) emission fluxes for all 6 sea spray source functions at 10 m above the surface. The wind speed at 10 m is 9 m s^{-1} and the SST was set to 25°C for MMS and S15. The acronyms are explained in Table 2.

Smith et al. (1993) for the large particles (MMS) to our study. G14 modified the source function of Smith and Harrison (1998) based on backward trajectory analyses. The source function of L11 is based on results of tank experiments by Keene et al. (2007) and is valid in the size range $0.044 < D_{p80} < 24 \mu\text{m}$. Finally the source function described by S15 is a result of their measurements in a sea spray chamber. It is valid in the size range $0.01 \lesssim D_{p_{dry}} \lesssim 10 \mu\text{m}$.

The equations of all SSA source functions used in this work are given in Appendix A and the particle size dependencies of the emission fluxes are presented in Fig. 1. The figure shows that the number and mass emission fluxes differ up to three orders of magnitude between the different parameterizations for particles with $D_{p_{dry}} \lesssim 0.1 \mu\text{m}$. These discrepancies likely result from measurement uncertainties of small particles (Blot et al., 2013, and citations therein) and the extrapolation of the source functions. The emission fluxes of all source functions agree within one order of magnitude in the middle size range ($0.1 \lesssim D_{p_{dry}} \lesssim 5 \mu\text{m}$), while they differ again for the large particles. This may be due to the different measurement techniques that the source functions are based on (see de Leeuw et al., 2011, for a detailed comparison) or the extrapolation of the SSA source functions.

The first source function incorporating a SST dependence term was published by Mårtensson et al. (2003), but it is only valid for particles with $D_{p_{dry}} < 2.8 \mu\text{m}$. Sofiev et al. (2011) used their measurements to develop a function to correct the original wind speed dependent emission fluxes for the effects of the SST (S11T). Jaeglé et al. (2011) found an overestimation of the modelled SS mass concentration in high latitudes with $T_W < 10^\circ\text{C}$ and an underestimation in low latitudes with $25^\circ\text{C} < T_W$ by GEOS-chem. They developed a function for the SST dependent correction of the source function to fix that (J11T). In contrast to S11 this function is independent of the particle sizes. The SST correction function S15 is an internal part of source function by Salter et al. (2015).

The three SST correction functions can be found in appendix A. Additionally they are plotted in Fig. 2. The comparison of Salter et al.

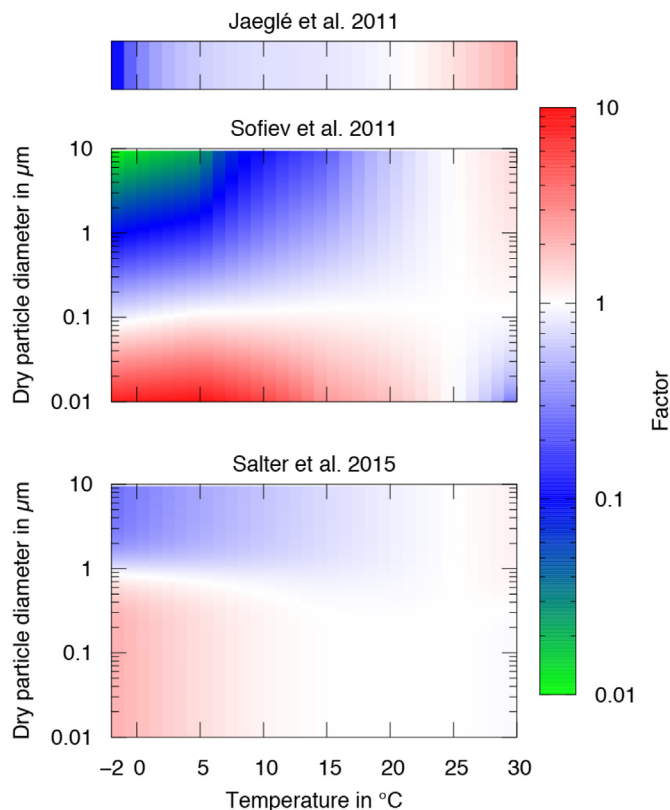


Fig. 2. Correction factors of the sea spray source functions based on SST. Jaeglé et al. (2011) does not have a size dependence. The very low values by Sofiev et al. (2011) may be due to extrapolation.

(2015) to the other functions is done in the way that the emission fluxes ($U_{10} = 9 \text{ m s}^{-1}$) were calculated for the given temperatures and then normalized by division by the emission fluxes at $T_W = 25^\circ\text{C}$. This value was used by Sofiev et al. (2011) as the temperature where the original source function is valid without correction. Fig. 2 shows that all correction functions agree in the fact that the emission flux of larger particles (J11T is mass dependent) increase with increasing temperature, while differing in other aspects like the strength of the increase. Furthermore, S11T and S15 show a decrease of the emission flux with increasing temperature for small particles but again with different magnitudes.

3. Model setup

3.1. Description of the model system COSMO-MUSCAT

The model simulations were carried out with the regional model COSMO-MUSCAT (Wolke et al., 2012). This is an online coupled model system consisting of the weather model of the German weather service COSMO (Schättler et al., 2009) and the aerosol chemical transport model MUSCAT (Wolke et al., 2012). The model system was developed for mesoscale air quality (Schrödner et al., 2014) and regional dust transport studies (Heinold et al., 2011b). It treats aerosol and gas phase transport processes including advection, turbulent diffusion, sedimentation and size dependent wet and dry deposition. Additionally, chemical and microphysical transformations can be simulated by the model, but are not considered in this work (Wolke et al., 2012).

The SSA size distribution is described with a mass based spectral approach using 15 logarithmically spaced size bins. They range from 10 nm to $10 \mu\text{m}$. Only the aerosol species SS and OM are regarded within the simulations. The size of the wet aerosols is calculated from the hygroscopicity of SS using the approach of Lewis and Schwartz

(2006) and assuming that OM has no effects on the water uptake. This wet particle size is also used to calculate the AOD based on the Mie theory (Mishchenko et al., 2002).

The emission fluxes and their dependence on plSST are calculated using the equations described in Section 2. The different combinations included in the comparison are explained in Section 4. The salinity dependence of the emission of dry SS was accounted for through the calculation of the particle size at formation using the actual salinity from the model. Knowing this value, a corresponding particle size (dry or at 80 % relative humidity) is calculated for a salinity of 35 g kg^{-1} . For that particle size the emission flux is calculated and assigned to the current particle size in the model. The amount of OM emitted within the SSA is calculated with the approach of Long et al. (2011), but are not focus of this work.

3.2. Observational data

The validation of the modelled SS mass concentration is done by calculation the corresponding sodium mass concentration using a conversion factor of 0.3061 (Seinfeld and Pandis, 2006). Sodium is the best tracer for SS, because it has only few other sources than the ocean, which are negligible (Tsyro et al., 2011). The model simulations were compared to the measured $\text{PM}_{2.5}$ - and PM_{10} -mass concentrations from the EMEP-stations (European Monitoring and Evaluation Programme) Auchencorth Moss, Cabauw and Virolahti II (Fig. 3). Some of these stations were already used in a model-measurement-comparison for the validation of modelled SS (Tsyro et al., 2011). These three stations cover several different geographical situations, like low salinity (Virolahti II) or long wind fetches (Auchencorth Moss) in the mid latitudes. The measurements are described in detail in Aas et al. (2012). Since the measured sodium mass in PM_{10} includes that in $\text{PM}_{2.5}$, we consider the larger size fraction by the difference $\text{PM}_{10}-\text{PM}_{2.5}$. Melpitz (Middle Germany) is an additional station where we had access to reliable measurement data. A comparison to those data is shown in the supplement. It is not useful for the validation of sea spray source functions due to its long distance to the coast.

Additionally, observations from the CVAO are used to include a station from the subtropics with higher SST (Fig. 3). Aerosol measurements from a five-staged Berner-impactor are available from an intensive campaign in December 2007 (Müller et al., 2010).

3.3. Description of case study and model setup

The model simulations were carried out using two domains and

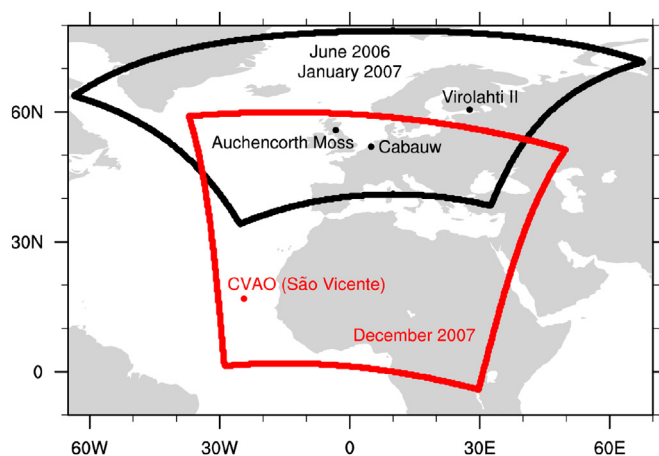


Fig. 3. The two model domains (“European” in black and “North-African” in red) used for the study together with the measurement stations. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

Table 3
Model configuration.

Description	Parameter	Value
Horizontal grid resolution	$dx; dy$	0.25° ($\approx 28 \text{ km}$)
Number of vertical grid cells; maximum altitude of grid	$N(z); H(z_{max})$	$30 \pm 10211 \text{ m}$ (MUSCAT) $40 \pm 23588.5 \text{ m}$ (COSMO)
Height of lowest grid cell	$dz(0)$	20 m
Temporal resolution	dt	80 s (MUSCAT) 300 s (COSMO)
Initialization time	dt_{init}	5 d
Lateral boundary conditions	$dc/dx; dc/dy$	0

three different time periods (June 2006; January 2007 and December 2007) shown in Fig. 3. These domains are named “European” domain, which covers the EMEP-stations and “North-African” domain to focus on the Cape Verde region. Table 3 shows the horizontal, vertical and temporal resolution for all three domains. Furthermore, the simulations were carried out with a spin-up period of five days and gradient zero for the boundary conditions of the aerosol. This means that the values in all grid cells at the boundaries of the model domain are mirrored outside of the model boundary resulting in a gradient of zero at all boundaries.

The input data for COSMO at the boundaries of the model domain are provided by reanalysis data of GME. The data of the SST and the sea ice extent are also provided via GME. The sea surface salinity was taken from the “World Ocean Atlas 2001” (described in Boyer et al., 2005, and available at “www.nodc.noaa.gov/OC5/WOA01/qd_ts01.html”; accessed: 4th of May 2011) using the yearly averaged maps with 0.25° horizontal resolution.

4. Model results

We conducted 11 simulations with different setups per domain and time periods. Every individual setup consisted of a SSA source function (G03, G14, L11, MMS, S11, S15) which was multiplied by a SST-correction function (J11T, S11T) or NO for model simulations without SST correction. The latter represents the temperature point where the correction functions reach the conserving value of 1. This is circa 21°C for J11T and 25°C for S11T. The acronyms are described in Table 2. The nomenclature used in the following sections is written in the way that it starts with the source function followed by the correction function. As an example: G03 + S11T means SSA source function from Gong (2003) in combination with the SST correction function by Sofiev et al. (2011).

4.1. Comparison of the emission parametrizations

The model results focussing on the effect of different source functions are shown in Fig. 4 for $\text{PM}_{2.5}$ (left column), $\text{PM}_{10}-\text{PM}_{2.5}$ (middle column) and AOD (right column). The top panel (Fig. 4a–c) shows the averaged model results using L11 + S11T in the European domain for January 2007. This function produces the lowest results. When dividing the other simulations (G03 + S11T, G14 + S11T, MMS + S11T, S11 + S11T) by these results the ratio is larger than 1, thus facilitating the comparison (Fig. 4d–o).

The simulated surface concentrations of sodium in $\text{PM}_{2.5}$ (Fig. 4a) range up to values of $0.95 \mu\text{g m}^{-3}$ with a regional average of $0.18 \mu\text{g m}^{-3}$. $\text{PM}_{10}-\text{PM}_{2.5}$ (Fig. 4b) has maximum values around $2.7 \mu\text{g m}^{-3}$ with a regional average of $0.45 \mu\text{g m}^{-3}$. The AOD (Fig. 4c) ranges up to 0.08 with a regional average of 0.016. The model results are low compared to the literature. For example, Tsyro et al. (2011) (Fig. 2) calculated annual mean concentrations of SS in PM_{10} ranging from 1 to $25 \mu\text{g m}^{-3}$ in the same region for the year 2006. Jaeglé et al. (2011) used AOD measurements of typical ocean sites for model validation. These values ranged from 0.05 to 0.4 in a monthly multi-year (2005–2008) average.

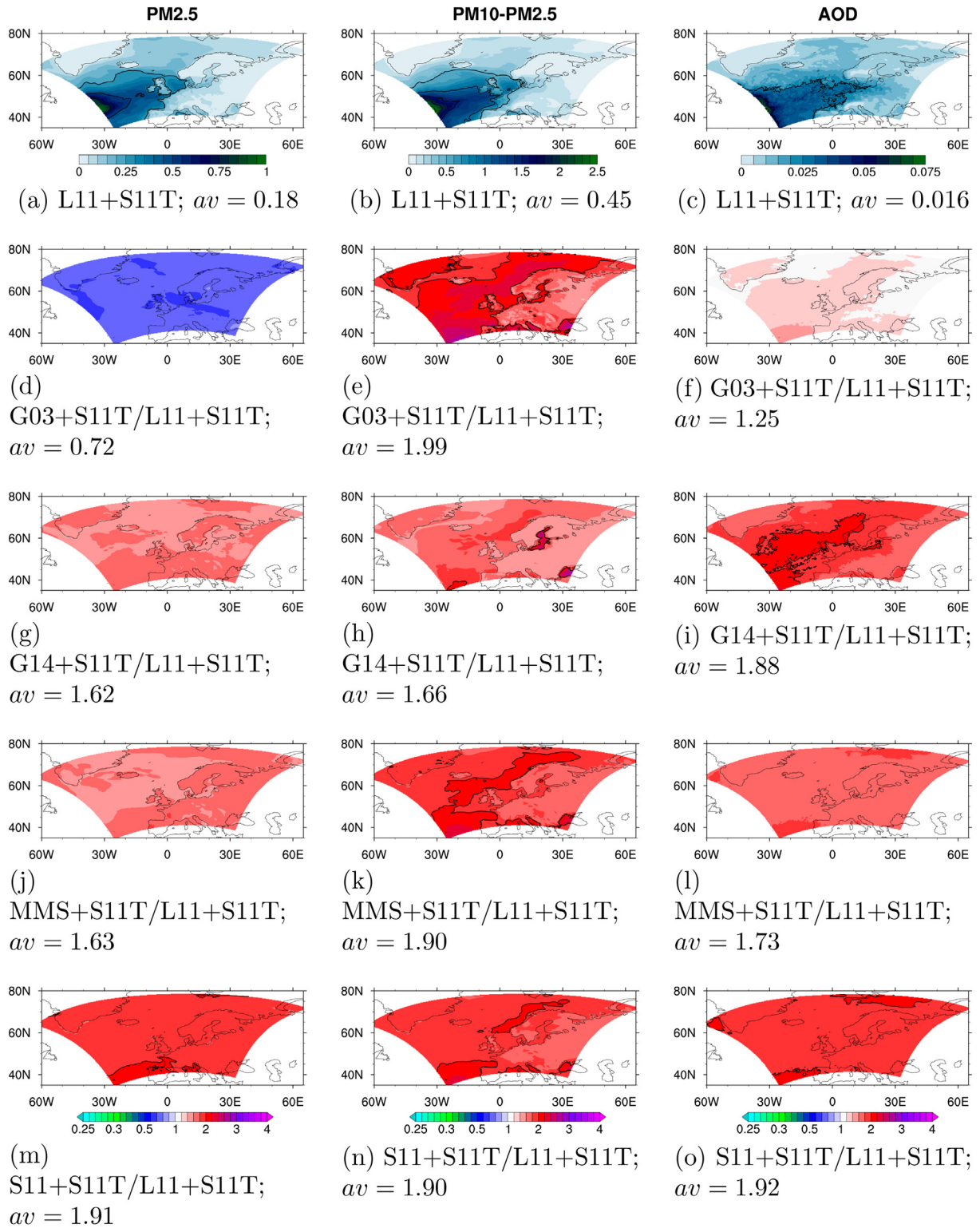


Fig. 4. Simulated surface concentrations of sodium (in $\mu\text{g m}^{-3}$) in $\text{PM}_{2.5}$ (left column), in $\text{PM}_{10-\text{PM}_{2.5}}$ (middle column) and AOD (right column) averaged for January 2007. The top panel (a,b,c) show the absolute values from the model simulation with L11 + S11T, while the figures in the lower panels show the ratio of the simulation to the results with L11 + S11T. “av” stands for the field averaged value.

Of course, these results are not directly comparable due to different time periods and regions. In contrast to Jaeglé et al. (2011) here only SSA is considered in AOD. But they indicate that the results from the L11 + S11T simulation are at the lower end of published simulation results.

The comparison of the results obtained with different SSA source

functions to L11 shows that G03 calculates slightly lower sodium concentrations in $\text{PM}_{2.5}$ (Fig. 4d). The other three source functions produce higher concentrations with G14 (Fig. 4g) and MMS (Fig. 4j) having comparable ratios (field average of 1.62, 1.63). S11 produces the highest concentrations with an averaged ratio of 1.91 (Fig. 4m). The ratios are higher for $\text{PM}_{10-\text{PM}_{2.5}}$ (Fig. 4e, h, k, n), but still ranging

around a ratio of 2 in the regional average. The ratios for the AOD are again smaller than those at PM_{10} - $PM_{2.5}$. Except for G03 they reach ratios around 2 in the regional average. For G03 the ratio is only a little increased to 1.25. Overall, the ratios of the source functions to L11 are ranging around 2 (maximum up to 4) for each regarded parameter. Furthermore, the ratio of G14, MMS and S11 to each other are very close to 1.

All simulations that are shown in (Fig. 4) were conducted in combination with the SST correction function S11T and for a winter period, where the SST is lowest. Further results are shown in the Supplement. For June 2006 the ratios are a little higher than in January. As an example, the field averaged ratio for $PM_{2.5}$ and S11 + S11T to L11 + S11T has a value of 2.14 in June and 1.91 in January. Also in the Supplement the ratios of G03 to L11 without the effects of the SST are shown. The comparison indicates that the SST-function has an effect on the ratio especially for PM_{10} - $PM_{2.5}$ and the AOD by increasing these parameters, but this increase is small.

4.2. Comparison of the SST correction functions

The analysis with the focus on the effect of the SST correction functions includes simulations with J11T, S11T and without SST correction NO). We used G03 as SSA source function. The choice of the source function affects the results only weakly (comparison in Supplement). The monthly averaged results for January 2007 are shown in Fig. 5. The top panel presents the total values of the simulations without accounting for the SST and the second and third panels show the ratios of the results with to without SST correction. The left column of the figure shows the sodium concentration in $PM_{2.5}$, the middle column PM_{10} - $PM_{2.5}$ and the right column the AOD.

The surface concentrations of sodium in $PM_{2.5}$ (Fig. 5a; regional average of $0.41 \mu g m^{-3}$) is clearly lower than that in the size range PM_{10} - $PM_{2.5}$ (Fig. 5b; regional average of $5.3 \mu g m^{-3}$). This indicates that the

bulk of the particle mass is present in the larger particles. Thus, changes in the concentration of smaller particles ($PM_{2.5}$) have only weak effects on the total aerosol mass in the model. All parameters are far higher than L11 + S11T (Fig. 4a–c), due to the effects of the different source function and the SST correction. The difference can reach a ratio of 10 for PM_{10} - $PM_{2.5}$.

Fig. 5d–f shows the effect of J11T. This function decreases the surface concentration of sodium in all sizes and the AOD by a field averaged factor of 0.6. The correction function produces the strongest decrease in the cold regions of the Arctic Sea. In Fig. 5g–i the influence of S11T on the surface concentration and the AOD is shown. It decreases the modelled sodium concentration in $PM_{2.5}$ by a factor of 0.3 (field average) and has the strongest effect on PM_{10} - $PM_{2.5}$ (0.18), but also decreases the AOD by a factor of 0.23. In very cold regions the decrease can reach down to factors smaller than 0.1. The differences between the parameters are due to the dependence of the SST correction function on particle size (Fig. 2).

The results of the simulations with the source function S15 for the surface concentrations of sodium in $PM_{2.5}$ and PM_{10} - $PM_{2.5}$ and the AOD are given in Fig. 6a–c averaged for January 2007. The values are close to the G03 + NO results (Fig. 5a–c). While the concentrations for $PM_{2.5}$ are higher than G03 + NO, they are smaller for PM_{10} - $PM_{2.5}$ and the AOD is nearly equal. This is reflected by the ratio of both simulations (Fig. 6d–f). Fig. 6d shows nearly no latitudinal dependence of the ratio for $PM_{2.5}$. This means that the effects due to the SST dependence in S15 cancel each other out in that size range. For the large particles (PM_{10} - $PM_{2.5}$) there is a clear SST dependence in the ratio (Fig. 5e). It decreases from north to south. The same is the case for the AOD, but less obvious. Fig. 6g and h and 6j–l present the ratio of S15 to the results with the two SST correction functions J11T and S11T, which were combined with the source function G03. The ratios of G03 + J11T and G03 + S11T to S15 (Fig. 6d–f) increase in all parameters from north to south. This indicates that J11T and S11T has stronger effects on the emission fluxes

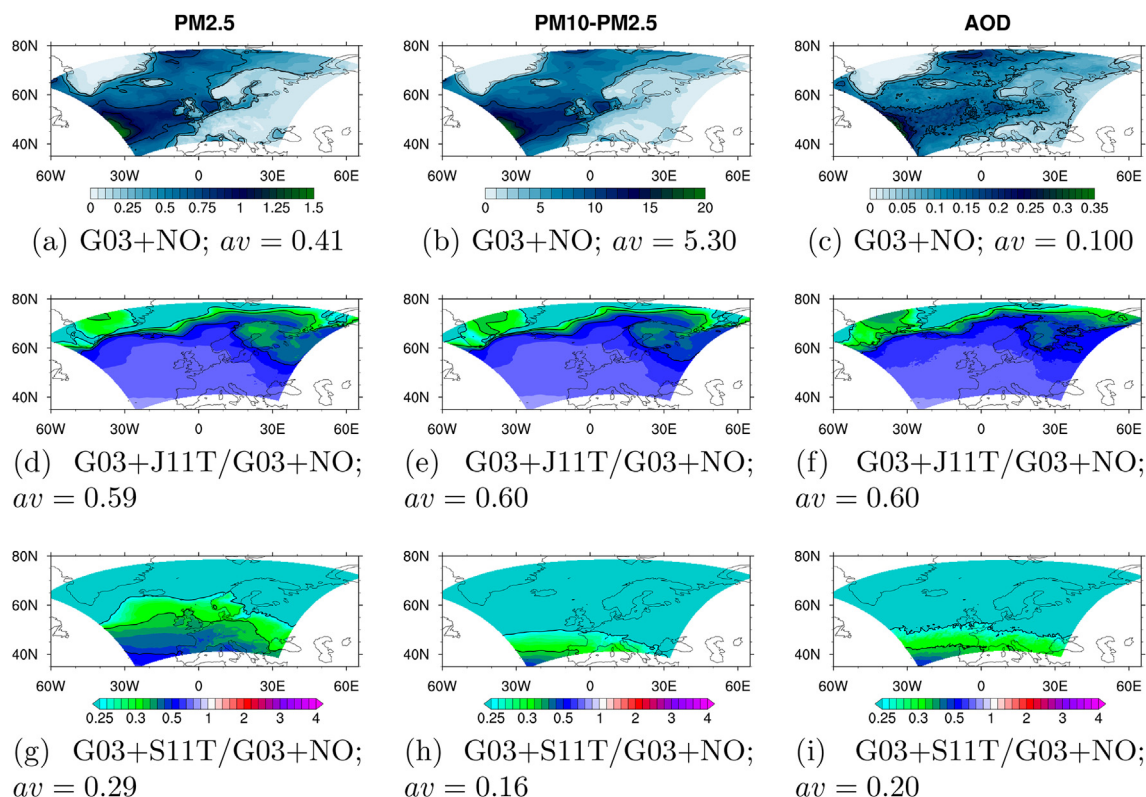


Fig. 5. Surface concentration of sodium (in $\mu g m^{-3}$) in $PM_{2.5}$ (left column), in PM_{10} - $PM_{2.5}$ (middle column) and AOD (right column) averaged for January 2007. The top panel (a,b,c) show the absolute values from the model simulation with G03 + NO, while the lower panels show the ratio of the simulation to the results with G03 + NO. “av” stands for the field averaged value.

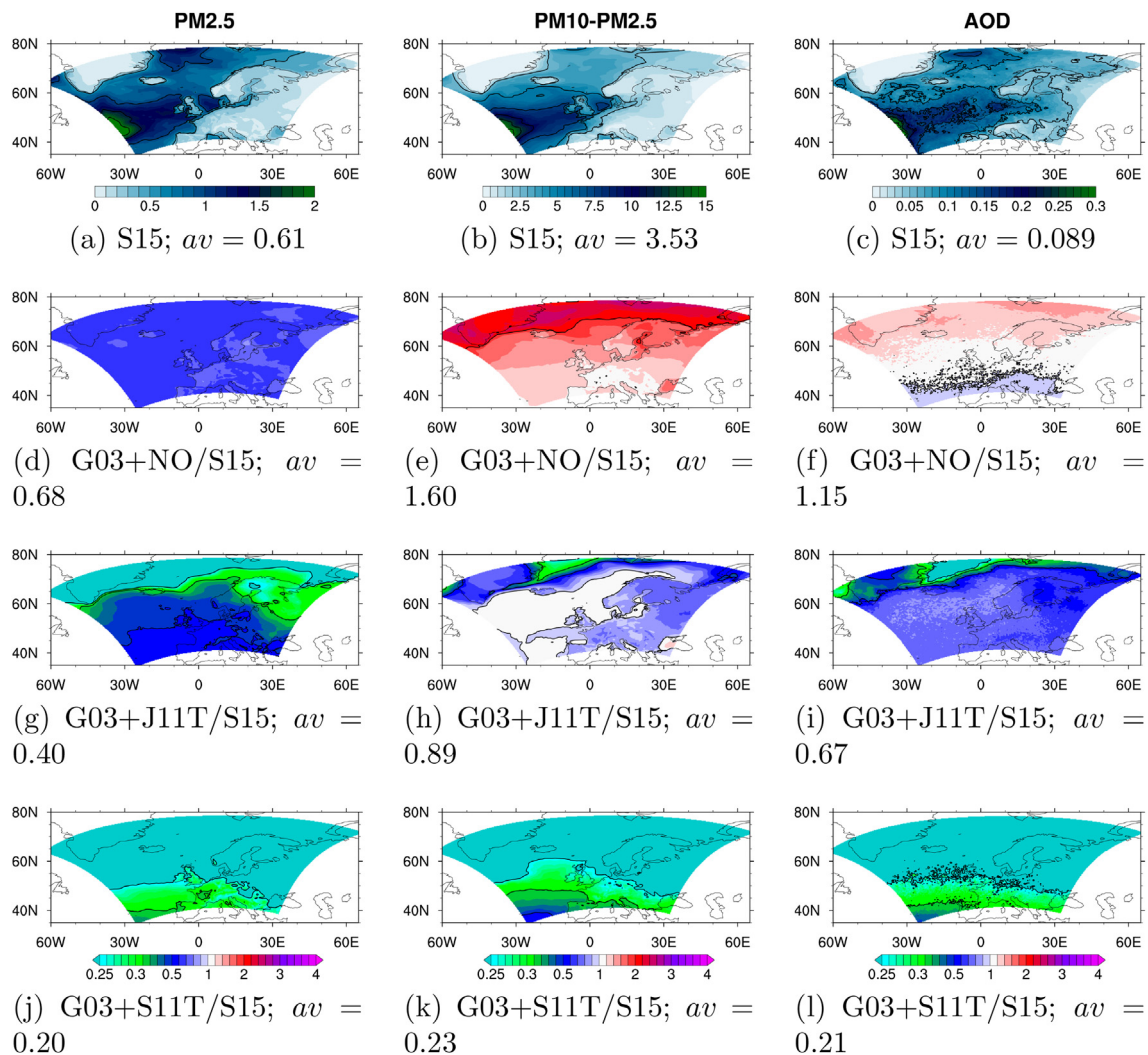


Fig. 6. Surface concentration of sodium (in $\mu\text{g m}^{-3}$) for $\text{PM}_{2.5}$ (left column), $\text{PM}_{10-\text{PM}_{2.5}}$ (middle column) and AOD (right column) averaged for January 2007. The top panel (a,b,c) show the absolute values from the model simulation with S15, while the lower panels show the ratios of the simulation to the results with S15. “av” stands for the field averaged value.

due to the SST than S15. This is also evident in the average values.

The strongest decrease of the sodium surface concentration in $\text{PM}_{2.5}$ and $\text{PM}_{10-\text{PM}_{2.5}}$ and the AOD occurs in the simulations with the SST correction function S11T. Their weakest decrease is generated by S15. These results are supported by further results for June 2006 that are shown in the Supplement together with results for the combination with L11 as SSA source function.

4.3. Comparison of model results with measurements

Model results are validated with the concentration data described in Section 3.2 in terms of monthly averages, monthly percentiles, slopes, mean relative error, correlation. They are provided in the Supplement together with the time series and scatter plots. Comparisons of monthly sodium concentrations are given for the EMEP stations in Tables 4 and 5 for $\text{PM}_{2.5}$ and $\text{PM}_{10-\text{PM}_{2.5}}$. The data in the tables include only days for which back trajectory analyses showed that the air mass stayed over land less than 6 h. The colour of the numbers in the table indicated the under or overestimations of the modelled concentrations compared to the observations by factors 2, 3, 4 or more.

The observed sodium concentrations in $\text{PM}_{2.5}$ are higher in January than in June (Table 4). All simulations show the same effect at Auchencorth Moss and Cabauw. The effect is weaker in the simulations than observed at Auchencorth Moss, while it is nearly reproduced at

Cabauw. This is reflected by the ratio between simulation and measurement (brackets). At Cabauw it is nearly the same in June and January, while at Auchencorth Moss the factor is lower in January. At Virolahti II the ratio between model results and measurements is lower in January than in June. This is mainly caused by the inability of the model to reproduce the higher concentrations in January.

The model simulations produce more often lower sodium concentrations than measured for $\text{PM}_{2.5}$ (illustrated by the domination of blue, green and turquoise colors in the table). Higher concentrations than measured are produced by the model only for Virolahti II and Auchencorth Moss in June and only for some of the setups. The overestimation has a maximum ratio of 2.29 (G14 + J11T), while the largest underestimation was produced by G03 + S11T (0.39). The majority of the simulation results agree within a factor of 2 with the observations. At Cabauw in June as well as for all stations in January all modelled sodium concentrations underestimate the measurements with a minimum of 0.13 for G03 + S11T at Cabauw. The simulation G14 + J11T produces highest sodium concentrations and is closest to the observations. This setup performs best in 3 of 6 cases. Including further statistics (see Supplement) the G14 + J11T function retrieves the best results for $\text{PM}_{2.5}$.

The observed sodium concentrations in the coarse fraction ($\text{PM}_{10-\text{PM}_{2.5}}$) are higher at Auchencorth Moss and Cabauw in January than in June, while it is the reverse at Virolahti II. The majority of the

Table 4

Sodium concentration in $\text{PM}_{2.5}$ (in $\mu\text{g m}^{-3}$) at the EMEP stations. The value in the brackets stand for the standard deviation of the observations and for the ratio of the model simulation to the observation. The colors represent the ratios of model concentrations to the measurement: turquoise: $\approx 1/4$ and less, green: $\approx 1/3$, blue: $\approx 1/2$, black: ≈ 1 , orange: ≈ 2 , red: ≈ 3 , violet: ≈ 4 and more.

Station	Obs.	G03+ J11T	G03+ NO	G03+ S11T	G14+ J11T	G14+ S11T	L11+ J11T	L11+ NO	L11+ S11T	MMS+ S11T	S11+ S11T	S15
June 2006												
Virolahti II	0.13 (0.06)	0.14 (1.11)	0.18 (1.42)	0.08 (0.62)	0.30 (2.29)	0.14 (1.11)	0.15 (1.17)	0.19 (1.50)	0.09 (0.66)	0.14 (1.10)	0.17 (1.32)	0.26 (1.98)
Auch. Moss	0.15 (0.26)	0.11 (0.75)	0.14 (0.99)	0.06 (0.39)	0.32 (2.18)	0.13 (0.88)	0.13 (0.92)	0.18 (1.21)	0.07 (0.49)	0.13 (0.90)	0.15 (1.06)	0.20 (1.34)
Cabauw	0.74 (0.33)	0.18 (0.25)	0.24 (0.32)	0.09 (0.12)	0.45 (0.61)	0.20 (0.27)	0.22 (0.30)	0.29 (0.39)	0.11 (0.15)	0.19 (0.26)	0.23 (0.32)	0.34 (0.45)
January 2007												
Virolahti II	0.15 (0.14)	0.07 (0.46)	0.12 (0.79)	0.03 (0.20)	0.14 (0.96)	0.06 (0.41)	0.09 (0.57)	0.14 (0.94)	0.04 (0.27)	0.06 (0.43)	0.07 (0.49)	0.15 (0.99)
Auch. Moss	0.61 (0.62)	0.17 (0.29)	0.23 (0.38)	0.09 (0.15)	0.42 (0.70)	0.21 (0.34)	0.24 (0.40)	0.33 (0.54)	0.13 (0.21)	0.22 (0.36)	0.24 (0.40)	0.33 (0.54)
Cabauw	1.06 (0.50)	0.29 (0.27)	0.40 (0.37)	0.14 (0.13)	0.69 (0.65)	0.32 (0.30)	0.40 (0.38)	0.55 (0.52)	0.20 (0.19)	0.33 (0.31)	0.38 (0.36)	0.56 (0.53)

simulations captured that difference. The results shown in Table 5 indicate that the majority of the model results overestimate the observations. While the highest overestimation (maximum 9.72 for G03 + NO at Auchencorth Moss) occurs when no SST correction function is used, the best fit for all stations occurs with the SST correction function S11T. The combination with the source function L11 is in 3 of 6 cases closest to the measurements, but overall tends to underestimation. When also including the statistics shown in the Supplement, the picture is less clear. While the L11 + S11T performs best at some points, results computed with other source functions also fit better at other points. Therefore, there is no clear best source function.

Measurements of aerosol particles from a five-stage Berner-impactor operated at the CVAO (Fig. 3) were included in the model evaluation (Table 6). We summed the sodium concentrations in the first and the second stages spanning the size range from $0.05 < Dp_{amb} < 0.42 \mu\text{m}$, because of the low concentration and the high uncertainty of the source functions for very small particles (see Sec. 2). In this size range all model simulations underestimate the observed concentrations by a factor of 2–3. Thus the highest model concentration by L11 + NO fits best to the observations. For the third stage ($0.42 < Dp_{amb} < 1.2 \mu\text{m}$) all model results agree within a factor of 2 with the measurements. The best agreement for the model setup L11 + J11T. Similar results are found for the concentrations in the fourth impactor stage ($1.2 < Dp_{amb} < 3.5 \mu\text{m}$). All simulations are close to the observations with a slight advantage of G03 + S11T. For the large particles (fifth impactor stage; $3.5 < Dp_{amb} < 10 \mu\text{m}$) the majority of the source

functions clearly overestimate the measurements by a factor of up to 5 (G03 + J11T). Here the simulation with the lowest concentration (L11 + S11T) agrees best with the measurements. Including further statistics (see Supplement) the best agreement of the different model setups remains unclear for the first four impactor stages. At impactor stage five the model results with the source function L11 + S11T clearly agrees best with the observations.

4.4. Uncertainties in the comparison of model results and measurements

There are several sources of uncertainties in such a comparison of modelled SSA concentrations to measurements. First they occur within the model itself, resulting from the uncertainties of the implemented parametrizations, the grid resolution or the numerical integration scheme. Only some of these uncertainties can be quantified.

The meteorology of the model has a strong influence on the uncertainties of the modelled SSA concentration via the wind speed and direction. Wind is not only responsible for the transport, but also strongly affects the emission of SSA itself. Typical source functions have a wind speed dependence of $U_{10}^{3.41}$ or $U_{10}^{3.74}$ (see Appendix A). The atmospheric model we used in these simulations was found to tend to underestimate the wind speed at land stations in Northern Africa (Heinold et al., 2007, 2009), but also tend to overestimate the wind speed especially at stable conditions (Vautard et al., 2012). The latter conditions are typical above ocean. Heinold et al. (2007) mentioned a maximum underestimation of 4 m s^{-1} at moderate wind speeds. With

Table 5

Sodium concentration in $\text{PM}_{10}\text{-PM}_{2.5}$ (in $\mu\text{g m}^{-3}$) at the EMEP stations. The value in the brackets stand for the standard deviation of the observations and for the ratio of the model simulation to the observation. The colors represent the ratios to the measurement: turquoise: $\approx 1/4$ and less, green: $\approx 1/3$, blue: $\approx 1/2$, black: ≈ 1 , orange: ≈ 2 , red: ≈ 3 , violet: ≈ 4 and more.

Station	Obs.	G03+ J11T	G03+ NO	G03+ S11T	G14+ J11T	G14+ S11T	L11+ J11T	L11+ NO	L11+ S11T	MMS+ S11T	S11+ S11T	S15
June 2006												
Virolahti II	0.25 (0.15)	1.00 (4.04)	1.28 (5.17)	0.38 (1.53)	1.68 (6.77)	0.51 (2.05)	0.36 (1.46)	0.47 (1.88)	0.14 (0.57)	0.33 (1.34)	0.31 (1.27)	0.96 (3.87)
Auch. Moss	0.21 (0.27)	1.56 (7.40)	2.05 (9.72)	0.51 (2.43)	1.47 (6.96)	0.41 (1.93)	0.68 (3.25)	0.90 (4.29)	0.24 (1.16)	0.49 (2.31)	0.49 (2.32)	1.63 (7.75)
Cabauw	0.59 (0.43)	1.78 (3.04)	2.33 (3.96)	0.59 (1.00)	1.64 (2.79)	0.45 (0.77)	0.64 (1.09)	0.84 (1.43)	0.23 (0.39)	0.52 (0.88)	0.50 (0.85)	1.64 (2.79)
January 2007												
Virolahti II	0.17 (0.13)	0.75 (4.28)	1.18 (6.77)	0.17 (0.96)	1.10 (6.28)	0.18 (1.02)	0.38 (2.16)	0.57 (3.28)	0.10 (0.59)	0.17 (1.00)	0.18 (1.04)	0.82 (4.68)
Auch. Moss	0.42 (0.42)	2.92 (7.00)	3.94 (9.44)	0.82 (1.98)	2.38 (5.71)	0.72 (1.73)	1.53 (3.67)	2.06 (4.94)	0.49 (1.16)	0.84 (2.02)	0.87 (2.08)	3.26 (7.82)
Cabauw	0.78 (0.40)	4.45 (5.73)	6.08 (7.82)	1.19 (1.53)	3.58 (4.61)	1.02 (1.31)	2.16 (2.78)	2.95 (3.79)	0.66 (0.85)	1.18 (1.52)	1.21 (1.55)	4.73 (6.08)

Table 6

Sodium concentration (in $\mu\text{g m}^{-3}$) for the Berner impactor operated at the Cape Verdes averaged for December 2007. The value in the brackets stand for the standard deviation of the observation and for the ratio of the model simulations to the observation. The colors represent the ratios to the measurement: turquoise: $\approx 1/4$ and less, green: $\approx 1/3$, blue: $\approx 1/2$, black: ≈ 1 , orange: ≈ 2 , red: ≈ 3 , violet: ≈ 4 and more.4

Size	Obs.	G03+ J11T	G03+ NO	G03+ S11T	G14+ J11T	G14+ S11T	L11+ J11T	L11+ NO	L11+ S11T	MMS+ S11T	S11+ S11T	S15
$0.05 < Dp_{amb} < 0.42 \mu\text{m}$	0.03 (0.02)	0.01 (0.46)	0.01 (0.43)	0.01 (0.43)	0.01 (0.41)	0.01 (0.35)	0.03 (0.85)	0.03 (0.88)	0.02 (0.73)	0.02 (0.70)	0.03 (0.79)	0.02 (0.70)
$0.42 < Dp_{amb} < 1.2 \mu\text{m}$	0.13 (0.06)	0.12 (0.91)	0.11 (0.85)	0.11 (0.79)	0.27 (1.98)	0.22 (1.60)	0.13 (0.99)	0.14 (1.06)	0.10 (0.77)	0.27 (2.01)	0.28 (2.08)	0.27 (2.01)
$1.2 < Dp_{amb} < 3.5 \mu\text{m}$	1.09 (0.38)	1.35 (1.23)	1.25 (1.15)	1.01 (0.93)	1.89 (1.73)	1.44 (1.32)	1.24 (1.14)	1.30 (1.20)	0.88 (0.81)	1.48 (1.36)	1.76 (1.62)	1.48 (1.36)
$3.5 < Dp_{amb} < 10 \mu\text{m}$	1.24 (0.45)	6.25 (5.04)	5.73 (4.62)	4.30 (3.47)	4.74 (3.82)	3.47 (2.80)	1.94 (1.56)	1.86 (1.50)	1.35 (1.09)	3.54 (2.86)	3.32 (2.67)	3.54 (2.86)

this value we roughly estimate of the resulting error in the emission fluxes. Assuming this error of 4 m s^{-1} between 5 m s^{-1} and 9 m s^{-1} produce an error of factor 9 in the calculated SSA emission fluxes. Furthermore, all source functions assume a steady state between ocean waves and the surface wind, which is rarely reached in the atmosphere (Gemmrich et al., 2008).

The modelled SSA concentrations are also affected by the uncertainty of the modelled relative humidity, due to its strong influence on the size of SS particles. This leads to uncertainties in the calculated sedimentation velocity of the particles, thus in effect affecting the modelled particle concentration. Since we cannot compare these values with measurements, we use the values of Vautard et al. (2012, Fig. 14 left) to roughly estimate the resulting error. There the modelled relative humidity is usually within an uncertainty of 20 %. Since the relative humidity above the ocean is usually around 80 %, we assume an uncertainty range between 70 and 90 %. Using the function of Lewis and Schwartz (2006) for the size calculation the diameter of the wet particles differ by a factor of 2/3. This results in a different sedimentation velocity by a factor of 2.

A further source of uncertainties in our simulations is the neglect of microphysical processes and other aerosol types. These processes would lead to larger aerosol particles in the model due to gas uptake and coagulation resulting from the collision with other aerosols. The size distribution is shifted to larger particles. The inclusion of the microphysical processes would lead to higher deposition rates compared to our simulations. The resulting uncertainty cannot be quantified at this point.

Besides the model errors, uncertainties result from the comparison of model results averaged in a grid box to point measurements. The comparison can be done by assuming the station is representative for a larger area or by an interpolation from the model grid. In particular, the first approach may lead to RMS spatial sampling errors that may be as high as 3.4 for a grid box of $210 \times 210 \text{ km}$ (Schutgens et al., 2016). At higher grid resolutions this error becomes smaller (see later). Furthermore, both approaches assume homogeneity of the aerosol field within the grid cell and/or in its surrounding. This is not necessarily the case in the atmosphere, especially in regions with inhomogeneous surfaces like coastlines. Thus, the change of the air mass from above the oceanic source region of SSA to land in a coastal grid cell may lead to uncertainties. SSA concentrations were found to decrease exponentially downstream from the coast when the air mass flows inland, with the strongest decrease close to the coast (Gustafsson and Franzén, 2000; Vignati et al., 2001). A discrete aerosol model is not able to reproduce such strong gradients (Wolke et al., 2012). This problem is illustrated schematically in Fig. 7. The model results (purple crosses) in a grid cell, which is partly covered with land surface, are affected by the deposition above this surface, while the measurements at a coastal station (A) are not affected by this deposition. Thus, the model results need to be smaller than the observations, even if we assume a perfect model. Using the data observed by Gustafsson and Franzén (2000) and considering our grid resolution of 28 km we roughly estimate the error that is

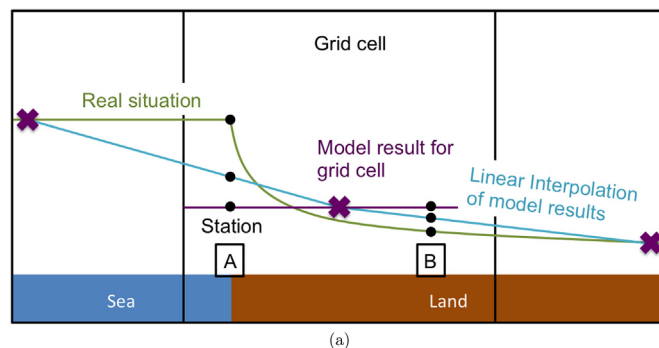


Fig. 7. This figure illustrates a problem occurring in comparisons of model results and measurements at a coastal station (A) and a few kilometres inland (B). The modelled concentrations are cell values represented by the purple crosses in the cell middles. Obviously, even “exact” measurements will be reproduced poorly. A possible alternative consists in the use of 2D-interpolated modelled concentrations for the model-measurement comparison. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

explained in detail in Appendix B. We got an underestimation by a factor of up to 2, when using the modelled grid cell averaged value or ≈ 1.4 when doing a linear interpolation between the cell neighbours. This uncertainty can be further reduced by using a higher grid resolution at the coast line or by comparing the model results to open ocean measurements. Nevertheless, only land stations provide long-term monitoring measurements, so coastal stations stay the best to use for model validation. But the described problem needs to be considered as uncertainty.

One solution to the coastal problem may be the increase of the grid resolution of the model. This will lead to a smaller error due to the comparison at a specific location, but increase the computational costs. Furthermore, aerosol models are usually tweaked to produce the best results at a particular horizontal resolution (Pope and Stratton, 2002; Schutgens et al., 2016). Thus, reducing the grid size may not necessarily produce better results due to a possible worse meteorology (Wolke et al., 2012).

The effects of the coastline on the model results are less important for column parameters such as AOD. For AOD evaluation with measurements only clean marine situations with mainly SSA present in the atmospheric column should be considered. However, even in clean marine cases the air mass can carry secondary aerosols that also affect the AOD (de Leeuw et al., 2014). Therefore, it is necessary to include further aerosol species to reproduce the AOD, which has been neglected in these simulations. But even in that situation additional uncertainties result from the modelling of these species. Another parameter affecting the modelled AOD is the relative humidity, since the AOD of SSA is strongly affected by the water vapour uptake. While the AOD is a further parameter for the model validation, it also carries uncertainties, which need to be considered.

The surface concentration measurements also contain uncertainties from the aerosol collection, the preparation of the samples and the analytic procedure. The uncertainties for $PM_{2.5}$ and PM_{10} are around $\pm 20\%$ (EMEP, 2001). Since PM_{10} - $PM_{2.5}$ includes both measurements, uncertainties from both measurements must be considered. Here we assume an error of $\pm 20\%$ for the measurement of the Berner-impactor at the CVAO.

Finally we estimate a total uncertainty using error propagation and including only the quantified uncertainties. With this we obtain an uncertainty factor of 12. These are the extreme cases which did not last a longer time within the simulations. The overall uncertainty should be smaller, but can not be quantified more accurately at this point.

5. Discussion

We compare our results with the findings by Grythe et al. (2014). They compared globally and yearly averaged production rates of many source functions based on multi-year averages of wind fields from ECMWF. They found the production rate of $G03 + NO$ (5.95 Pg yr^{-1}) having a ratio of 1.3 to $G03 + J11T$ (4.59 Pg yr^{-1}) and 2.3 to $S11 + S11T$ (2.59 Pg yr^{-1}). Furthermore, with a ratio of 0.67 it is lower than $G14 + J11T$ (8.91 Pg yr^{-1}). We calculated comparable results for the total concentration (see also Supplement). The function $G03 + NO$ related to $G03 + J11$ has a field averaged ratio of 1.5 (2.0) for the European domain in June 2006 (January 2007). In relation to $S11 + S11T$ the ratio is 4.7 (5.0) and to $G14 + J11T$ it is 1.3 (1.8). Our results are not directly comparable to Grythe et al. (2014) due to different regions, time periods and size ranges. However, the relations of the ratios are similar for the most of the functions.

The ratios between the model results using the different SSA source functions are usually smaller than 2 for $PM_{2.5}$ and AOD. The ratios for PM_{10} - $PM_{2.5}$ can reach a value of 4. Furthermore, the SST correction functions decrease the modelled concentrations by a factor of 1.7 (J11T) and around 5 (S11T) with the strongest effect on PM_{10} - $PM_{2.5}$. Thus the use of a SST correction function can lead to stronger model discrepancies than using different SSA source functions. The SST correction by S11T may decrease the emission rates by a factor of more than 10 for large particles (see Fig. 2). This decrease is due to the interpolation of the source function from its verified range below $Dp_{dry} \lesssim 2.8 \mu\text{m}$ to $10 \mu\text{m}$ and may be too strong (Sofiev et al., 2011). Therefore, it leads to underestimated sodium concentrations in PM_{10} which were already reported by Sofiev et al. (2011). Nevertheless, including a SST correction function may improve results. Chen et al. (2016) used WRF-chem with its implemented SSA source function G03 without SST correction to simulate SS concentrations across Europe. They found the model overestimates the measured sodium concentration in PM_{10} by a factor around 10 at 3 stations in the Netherlands. Table 5 shows the modelled sodium concentration at Cabauw can be decreased by a factor of up to 5 when using the SST correction by S11T in comparison to simulations without SST correction. Tsyro et al. (2011) found a discrepancy in the modelled sodium concentration in PM_{10} across Europe between summer and winter. In relation to measurements the modelled concentrations were higher in winter than in summer. This discrepancy can be fixed by a SST correction function due to their decrease of the concentration in winter. Our results together with the results shown in (Jaeglé et al., 2011; Sofiev et al., 2011; Grythe et al., 2014) emphasize the need of a SST-correction function when calculating SSA emission fluxes in aerosol models.

The SSA source function S15 produces model results in the range of other source functions, differing only in parts of the particle spectrum. While it produces sodium concentrations in $PM_{2.5}$ that are a little higher than $G03 + NO$ the AOD is comparable. For PM_{10} - $PM_{2.5}$ the modelled sodium concentration is clearly lower than $G03 + NO$ due to the SST correction. The SST correction by S15 is weaker than that of J11T and S11T, but comparable to the findings of Grythe et al. (2014). They found the measured total SSA concentration in the wind speed range

$5 - 10 \text{ m s}^{-1}$ increases with temperature according to the relationship $0.031 \cdot T_W + 0.39$. Using equation A.6 and summing the emitted mass fluxes for all particles up to $Dp_{dry} = 5 \mu\text{m}$ ($\approx PM_{10}$ at 80 % relative humidity) results in the relationship for concentration increase with temperature to be $0.0263 \cdot T_W + 0.4537$ ($R^2 = 0.9965$), which is independent of wind speed.

There are several sources of uncertainties in the comparison of model results to measurements but also within a model itself, which affects the choice of a possible best source function. All regarded parametrizations differ from each other at most by a ratio of 2 or in combination with the SST in maximum of a ratio of 10. The majority of the model results are still within the range of the uncertainties, which can reach values up to a ratio of 10. Keeping this maximum error in mind we assume all simulations within a ratio of 2 to the measurements agree well. So we conclude that nearly all considered source functions are suitable for use within COSMO-MUSCAT. Results using the parametrizations by $G14 + J11T$ and $L11 + S11T$ fit best to the measurements at some parts of the particle spectrum. The use of a SST correction is essential. In these experiments S11T performs best. The combinations $G14 + S11T$, $MMS + S11T$ and $S11 + S11T$ perform equally well at all particle sizes. They have only slight differences between each other in some regions or parts of the size range.

The best source function may differ in the use with a different model system, due to different model dynamic. Especially the ability to simulate realistic wind speeds is crucial. Due to such strong dependencies on the quality of internal model parameters, new source functions may not necessarily improve the model results for the atmospheric SSA concentrations. But including further parameters like the SST can improve the model results in specific regions or size ranges, for example for PM_{10} - $PM_{2.5}$ in Europe. Furthermore, the future developments can be expected to improve the model performances considering the meteorology. So it is desirable to develop source functions that produce most realistic SSA emission fluxes under all atmospheric and oceanographic conditions. On the other hand, worse model results of a source function do not necessarily mean that it would not give realistic results under atmospheric conditions.

6. Summary

Using the regional chemistry transport model COSMO-MUSCAT we carried out several model simulations for two months in an European domain and one month in a north-western African domain with the focus on SSA. We compared the monthly averaged model results using different SSA source functions with each other and the sodium measurements at four stations. In addition, two SST correction functions were included in our simulations. It was found that the use of such a SST correction generally improves the model results. This correction has a stronger impact on the results for different regions or time periods than using different SSA source functions. Our results are discussed in the context of possible uncertainties in the model system and in the model-measurement comparison. We found that the differences caused by using various source functions are in the range of those uncertainties. The choice of the best SSA emission parametrization may thus be model-dependent and strongly depends on the models performance for the surface wind speed. Considering the uncertainties three different SSA source functions ($G14 + S11T$, $MMS + S11T$ and $S11 + S11T$) perform equally well in the COSMO-MUSCAT model system.

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Appendix D. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.atmosenv.2018.10.016>.

Appendix A. Sea spray aerosol source functions and correction for SST

Appendix A.1. G03 - Gong (2003).

Gong (2003) calculated SSA emission fluxes with a modified version of the parameterization by Monahan et al. (1986). They improved the source function for particle sizes smaller than $r_{80} < 0.2 \mu\text{m}$ by including the dynamical parameter Θ , which was set to $\Theta = 30$:

$$\begin{aligned} \frac{dF_N}{dr_{80}} &= 1.373 \cdot U_{10}^{3.41} \cdot r_{80}^{-A} \cdot (1 + 0.057 \cdot r_{80}^{3.45}) \cdot 10^{1.607 \cdot \exp(-B^2)} \\ A &= 4.7 \cdot (1 + \Theta \cdot r_{80})^{-0.017 \cdot r_{80}^{-1.44}} \\ B &= \frac{0.433 - \lg r_{80}}{0.433} \end{aligned} \quad (\text{A.1})$$

This function is valid for $0.07 \lesssim r_{80} \approx Dp_{dry} \lesssim 20 \mu\text{m}$. We extrapolated it to be valid down to $Dp_{dry} = 0.01 \mu\text{m}$.

Appendix A.2. G14 - Grythe et al. (2014)

In a large comparison study Grythe et al. (2014) found best results for the source function by Smith and Harrison (1998). They included a term for small particles to extend its validity range to $0.01 \lesssim Dp_{dry} \lesssim 10 \mu\text{m}$.

$$\frac{dF_N}{dDp_{dry}} = 235 \cdot U_{10}^{3.5} \cdot \exp\left(-0.55 \cdot \left[\ln\left\{\frac{Dp_{dry}}{0.1}\right\}\right]^2\right) + 0.2 \cdot U_{10}^{3.5} \cdot \exp\left(-1.5 \cdot \left[\ln\left\{\frac{Dp_{dry}}{3}\right\}\right]^2\right) + 6.8 \cdot U_{10}^3 \cdot \exp\left(-1 \cdot \left[\ln\left\{\frac{Dp_{dry}}{30}\right\}\right]^2\right) \quad (\text{A.2})$$

Appendix A.3. L11 - Long et al. (2011)

Long et al. (2011) developed a source function by a two-modal fitting to the results from a bubble tank experiment by Keene et al. (2007). Thus this source function is valid for $0.044 < Dp_{80} = 2 \cdot Dp_{dry} < 24 \mu\text{m}$. We extrapolated this function down to $Dp_{dry} = 0.01 \mu\text{m}$ and neglected the influences of OM on the number emission flux:

$$\begin{aligned} \frac{dF_N}{d \lg Dp_{80}} &= 2 \times 10^{-8} \cdot U_{10}^{3.74} \cdot 10^{P_N} \\ \text{For } Dp_{80} < 1 \mu\text{m}: \\ P_1 &= 1.46 \cdot (\lg Dp_{80})^3 + 1.33 \cdot (\lg Dp_{80})^2 - \\ &1.82 \cdot \lg Dp_{80} + 8.83 \\ \text{For } Dp_{80} > 1 \mu\text{m}: \\ P_2 &= -1.53 \cdot (\lg Dp_{80})^3 - 0.081 \cdot (\lg Dp_{80})^2 - \\ &0.426 \cdot \lg Dp_{80} + 8.84 \end{aligned} \quad (\text{A.3})$$

Appendix A.4. MMS - Lundgren et al. (2013)

The source functions of Mårtensson et al. (2003); Monahan et al. (1986) and Smith et al. (1993) were combined in different ways by several authors. Here we use the definitions of Lundgren et al. (2013):

For $0.02 < Dp_{dry} \lesssim 1 \mu\text{m}$:

$$\begin{aligned} \frac{dF_N}{d \lg Dp_{dry}} &= 3.84 \times 10^{-6} \cdot U_{10}^{3.41} \cdot 298.15 \cdot A + B \\ A &= c_4 \cdot Dp_{dry}^4 + c_3 \cdot Dp_{dry}^3 + c_2 \cdot Dp_{dry}^2 + c_1 \cdot Dp_{dry} + c_0 \\ B &= d_4 \cdot Dp_{dry}^4 + d_3 \cdot Dp_{dry}^3 + d_2 \cdot Dp_{dry}^2 + d_1 \cdot Dp_{dry} + d_0 \end{aligned}$$

For $1 < Dp_{dry} \lesssim 9 \mu\text{m}$:

$$\begin{aligned} \frac{dF_N}{dr_{80}} &= 1.373 \cdot U_{10}^{3.41} \cdot r_{80}^{-3} \cdot (1 + 0.057 \cdot r_{80}^{1.05}) \cdot 10^{1.19 \cdot \exp(-B^2)} \\ B &= \frac{0.380 - \lg r_{80}}{0.650} \end{aligned}$$

For $9 \mu\text{m} < Dp_{dry}$:

$$\begin{aligned} \frac{dF_N}{dr_{80}} &= (0.676 \cdot U_{10} + 2.43) \cdot \exp\left(-3.1 \cdot \ln\left[\frac{r_{80}}{2.1}\right]\right) + \\ &(0.959 \cdot U_{10}^{0.5} - 1.475) \cdot \exp\left(-3.3 \cdot \ln\left[\frac{r_{80}}{9.2}\right]\right) \end{aligned} \quad (\text{A.4})$$

There is no extrapolation for particles with $Dp_{dry} < 0.02 \mu\text{m}$. For the parameter $c_{0...4}$ and $d_{0...4}$ see Mårtensson et al. (2003).

Appendix A.5. S11 - Sofiev et al. (2011)

The source function of Sofiev et al. (2011) includes not only the influences of wind speed, but also of SST (below) and salinity (neglected). Here we present only the wind speed part being valid for $0.01 \lesssim Dp_{dry} \lesssim 10 \mu\text{m}$:

$$\frac{dF_N}{dDp_{dry}} = 3.84 \cdot U_{10}^{3.41} \cdot \frac{\exp\left(-\frac{0.09}{Dp_{dry} + 0.003}\right)}{2 + \exp\left(-\frac{5}{Dp_{dry}}\right)} \cdot \frac{1 + 0.05 \cdot Dp_{dry}^{1.05}}{Dp_{dry}^3} \cdot 10^{1.05 \cdot \exp\left(-\left(\frac{0.27 - \lg Dp_{dry}}{1.1}\right)^2\right)} \quad (\text{A.5})$$

Appendix A.6. S15 - Salter et al. (2015)

The source function of Salter et al. (2015) is based on experiments in a sea spray chamber and obtained through a three-fold lognormal fit to the measurements. It includes the influences of the wind speed and the SST, which cannot be treated independently like Sofiev et al. (2011). It is valid for particles with $0.01 \lesssim Dp_{dry} \lesssim 10 \mu\text{m}$:

$$\frac{dF_N}{dDp_{dry}} = \sum_{j=1}^3 \frac{F_{ent} \cdot c_{S15,j}}{\sqrt{2 \cdot \pi} \cdot \lg \sigma_j} \cdot \exp\left(-\frac{1}{2} \cdot \frac{\left(\lg Dp_{dry} - \lg \overline{Dp}_{mod,j}\right)^2}{(\lg \sigma_j)^2}\right)$$

$$F_{ent} = 2 \times 10^{-8} \cdot U_{10}^{3.74}$$

$$c_{S15,j} = A_j \cdot T_W^3 + B_j \cdot T_W^2 + C_j \cdot T_W + D_j$$

$$A_j = [-5.2168 \times 10^{-5}, 0, 0]$$

$$B_j = [3.31725 \times 10^7, 7.374 \times 10^5, 1.421 \times 10^4]$$

$$C_j = [-6.95275 \times 10^8, -2.4803 \times 10^7, 1.4662 \times 10^7]$$

$$D_j = [1.0684 \times 10^{10}, 7.7373 \times 10^8, 1.7075 \times 10^8]$$

$$\overline{Dp}_{mod,j} = [9.5 \times 10^{-2}, 0.6, 1.5]$$

$$\sigma_j = [2.1, 1.72, 1.6] \quad (\text{A.6})$$

Appendix A.7. J11T - Jaeglé et al. (2011)

Jaeglé et al. (2011) developed a model based correction function for the SSA emission flux by the influence of the SST:

$$c_{J11}(T_W) = 0.3 + 0.1 \cdot T_W - 0.0076 \cdot T_W^2 + 0.00021 \cdot T_W^3 \quad (\text{A.7})$$

Appendix A.8. S11T - Sofiev et al. (2011)

The correction function for the SSA emission flux by the influence of the SST by Sofiev et al. (2011) was obtained through the extraction of the SST dependent term from the original source function. It is calculated only for the given plSST (T_{Wj}). For the temperatures in between the factor has to be gained via interpolation. We further interpolated it to temperatures up to $T_W = 30^\circ\text{C}$:

$$c_{S11}(T_{Wj}, Dp_{dry}) = a_j \cdot Dp_{dry}^{b_j}$$

$$T_{Wj} = [-2, 5, 15, 25]$$

$$a_j = [0.092, 0.15, 0.48, 1]$$

$$b_j = [-0.96, -0.88, -0.36, 0] \quad (\text{A.8})$$

Appendix B. Uncertainty of the model comparison to coastal stations

We discuss the uncertainty of the model to measurement comparison for a coastal station based on the results of Gustafsson and Franzén (2000). In Fig. 3a they mentioned a decrease of the sea salt concentration (c in $\mu\text{g m}^{-3}$) with distance to the coast (x in km). It follows the function:

$$c = 21.24 \cdot x^{-0.45} \quad (\text{B.1})$$

Furthermore, they wrote in the second point of their conclusions that the sea salt concentration decreased to 20 % at 50 km inland compared to the coast. Using equation B.1 this gives a concentration of $3.65 \mu\text{g m}^{-3}$ at 50 km and $18.26 \mu\text{g m}^{-3}$ at the coast. Due to the grid resolution it is not possible to reproduce these values. Here we attempt an estimation of the maximum error resulting from the grid based on these concentrations. To quantify this maximum error we assume a coastal station measuring the concentration $18.26 \mu\text{g m}^{-3}$, but located near the border of the grid. The coastline is assumed to be located near the edge of the grid cell (see fig. B.8). The grid cell is mainly covered by land, but the considered coastal station lays marginally inside. Since models produce cell averaged concentrations, we can calculate this value for the grid cell by integrating equation B.9 within it and dividing it by the resolution (z):

$$F(c) = \frac{1}{z} \cdot \int_0^z 21.24 \cdot x^{-0.45} dx = \frac{21.24}{0.55 \cdot z} \cdot z^{0.55} \quad (\text{B.2})$$

We used a horizontal resolution of $z = 28$ km. This results in an grid averaged concentration of $8.62 \mu\text{g m}^{-3}$. Thus the ratio of the grid average value been produced by a perfect model to the observed concentration at our coastal station will be 2.12. This can be improved by an linear interpolation to the neighbour cell. Therefore we assume that the neighbour cell has the cell averaged value of $18.26 \mu\text{g m}^{-3}$, as it is measured at the coastal station. With this linear interpolation we calculate a concentration of $13.44 \mu\text{g m}^{-3}$ at the coastal station, which is right at the boundary of the grid cells, 14 km away from the center. Finally this results in a ratio of 1.36 to the observation.

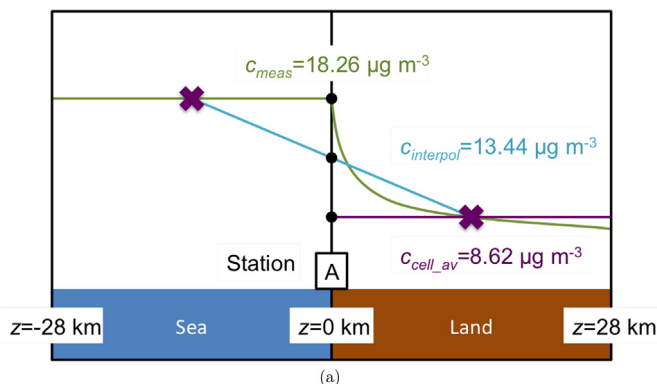


Figure B.8. The figure show a rough estimation of the error resulting from the comparison of point measurements at a coastal station (A) with results within a grid cell, like they where computed by models. The estimation is based on Gustafsson and Franzén (2000). Using our horizontal resolution of 28 km we get a cell averaged value of $c_{\text{cell_av}} = 8.62 \mu\text{g m}^{-3}$. That is clearly below the measurement at the station ($c_{\text{meas}} = 18.26 \mu\text{g m}^{-3}$).

Appendix C. List of symbols

c	Concentration of a substance or aerosol particles (in m^{-3} , $\mu\text{m}^3 \text{m}^{-3}$ or $\mu\text{g m}^{-3}$ depending on basis number, volume or mass).
c_{T11}	Function to correct the emission flux of SSA for the effect of the SST using the function of Jaeglé et al. (2011).
c_{S11}	Function to correct the emission flux of SSA for the effect of the SST using the function of Sofiev et al. (2011).
DP_{80}	Diameter of an aerosol particle (in μm) at 80% relative humidity.
DP_{amb}	Diameter of an aerosol particle (in μm) at the relative humidity of the ambient air.
DP_{dry}	Diameter of an aerosol particle (in μm) in dry air.
F_N	Number flux of aerosol particle (in $\text{m}^{-2}\text{s}^{-1}$).
r_{80}	Radius of an aerosol particle (in μm) at 80% relative humidity.
T_W	Sea surface temperature or water temperature (in $^{\circ}\text{C}$ or K).
U_{10}	Wind speed (in m s^{-1}) at 10 m above the surface.
z	Altitude (in m).

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