MODELLING EMISSIONS OF MARINE AEROSOL FROM THE OCEAN AT A EUROPEAN SCALE

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ABSTRACT

An empirical modelling approach to estimate spatially disaggregated emissions of marine aerosol (as PM_{10} and $PM_{2.5}$) has been applied across a sea/ocean coverage surrounding Europe. Datasets have been generated at a 10 km x 10 km resolution by using a GIS. Total emissions of PM_{10} in 2003 from the Caspian, Black, Mediterranean and Baltic Seas were found to be 0.52, 2.61, 21.54 and 4.75 Tg respectively. These agree well with current global emissions, although estimates from all sources are considered to be high in uncertainty. The PM_{10} emission estimates were size resolved to allow emission estimates of $PM_{2.5}$ to also be made. The extent of the study included some of the Atlantic Ocean, which was shown to be the largest source on a unit area basis. Spatial patterns of emission indicated higher emissions at locations further from land, the impact of sheltering by land being clearly observable. Seasonal variations were investigated, the emissions demonstrating their dependence on wind speed. A number of recommendations are made regarding model development.

I INTRODUCTION

Objectives

The objective of this work is to improve and apply methods for estimating emissions of marine aerosol on a European scale. This is being done as part of the NatAir project, which aims to improve methodologies for quantifying natural and biogenic emissions to the atmosphere for Europe. Currently there is no specific SNAP or NFR code for marine aerosol emissions and there is a deficit of spatially disaggregated emissions estimates in the literature. The GIS-based modelling approach developed here to meet these objectives has been specifically applied for PM_{10} and $PM_{2.5}$ in this work. However, it could potentially be developed to apply to other particle size ranges. Comments are also included on the different metrics commonly in use.

Background

Marine aerosol in the atmosphere are important for a number of reasons. They are the dominant aerosol (by mass) in remote marine surface air and can occasionally have a significant impact on continental air masses.

Marine aerosol plays a major role in a variety of processes in the atmosphere. For example, they influence radiative transfer directly by scattering solar radiation and indirectly by altering cloud droplet size distribution and concentration. This influences the albedo of marine boundary layer (MBL) clouds. Marine aerosol can also change cloud properties and lifetime

and provide media for chemical reactions (Foltescu, et al, 2004; Murphy et al., 1998). For example, the marine aerosol flux from the sea surface is an important factor in the formation of cloud condensation nuclei (CCN) in the MBL. Marine aerosol can directly supply more than 80% of the total MBL CCN when wind speeds are moderate and high, exceeding 12 m s^{-1} (Yoon and Brimblecombe, 2002). The effect of marine aerosol on climate is dependent on the marine aerosol size distribution (Gong et al., 1997).

Marine aerosol particles are chemical carriers of species containing Cl, Br, I and S and therefore play a role in the atmospheric cycles of these important elements. The halogens Br and Cl, once mobilized by heterogeneous reactions from sea-salt inorganic forms to reactive gaseous forms (e.g. Br_2 and Cl_2), can play a role in atmospheric ozone depletion and destruction of light hydrocarbons (Gong et al., 1997). Marine aerosol emissions and ambient concentrations also impact on sea surface-atmosphere exchange (Andreas et al., 1995; Pryor and Barthelmie, 2000) and heterogeneous chemistry, including the oxidation of SO_2 and NO_2 in the MBL, which is largely controlled by sea spray availability (Pryor et al., 2001). In addition, marine aerosol plays a role in plant stress and contribute to corrosion in coastal regions (Cole et al., 1999).

In terms of land-based PM concentrations, marine aerosol can make a significant contribution, especially when wind speeds are high. For example, in the UK between one and five PM_{10} episodes a year are thought to be caused by marine aerosol, which can result in peak hourly concentrations in excess of 40 μg m^{-3} in coastal locations (AQEG, 2005). Similar events are observed in coastal areas of other European countries (e.g. Ireland, Netherlands, Cyprus).

The availability of reliable spatially disaggregated marine aerosol emission estimates across a European extent provides a useful starting point for understanding and better representing many of these issues.

Mechanisms to generate marine aerosol emissions to the atmosphere

In order to produce a model and modelled estimates of marine aerosol emissions, it is important to understand the processes which generate a flux of aerosol from the sea to the air. Marine aerosol emissions, and therefore concentrations in the air, strongly reflect the state of the sea surface, which is in turn determined by meteorological conditions- especially surface wind speed (Gong et al., 1997). The rate at which the sea surface produces spray droplets is roughly estimated as the third power of the 10-meter wind speed, U_{10} (Wu, 1986).

The generation of marine aerosol can be attributed to various physical processes. The major mechanism is believed to be the entrained air bubbles bursting during whitecap formations due to the surface wind, which ejects jet and film droplets from the sea surface (Monahan et al., 1986). Most of the bubbles are formed as a result of air entrapment, primarily associated with breaking waves. The energy dissipated in wave breaking is derived from wind action. The amount of wave breaking is therefore very sensitive to wind speed (Foltescu et al. 2005). Sea spray generation by splash or bubbles produced from precipitation are of lesser magnitude than whitecapping and only occur intermittently and at a local scale (Gong et al., 1997).

It is therefore the case that generation/emission functions for marine aerosol tend to be based primarily on the relationship between whitecap coverage and wind speed. However, other variables such as sea surface temperature and salinity may also influence the flux (Martensson et al., 2003). Marine aerosol source functions have been formulated using laboratory and field experiments, arguments based on bubble-mediated production, and models describing the balance between the production and removal of the aerosol from the MBL (Gong et al., 2003).

Marine aerosol particles cover a wide size range (about 0.05 to 10 μm diameter) and have a correspondingly wide range of atmospheric lifetimes. Therefore, it is necessary to analyse their emissions and atmospheric distribution in a size-resolved model (IPCC, 2001).

There are a range of different definitions for airborne aerosol and particles. This study uses the term "marine aerosol" to refer to emissions in general from the sea. However, the empirical model (detailed in Section II) generates emissions of PM_{10} specifically. The standard reference method for measuring PM_{10} (BS EN12341) uses a gravimetric technique, and specifies the conditioning of the samples. This conditioning does not include drying of the sample, and hence the PM_{10} measurement refers to a "wet" mass of particulates. Clearly the resulting marine emissions by mass are dominated by water, and these results are therefore inappropriate for use in studies on human health.

By using the salinity of the different seas, it is possible to express the emissions as a "dry" mass, and these results are also included in Section III. It is useful to include the mass expressed in this way because many PM measurements are made using dried samples.

II APPROACH/ METHOD

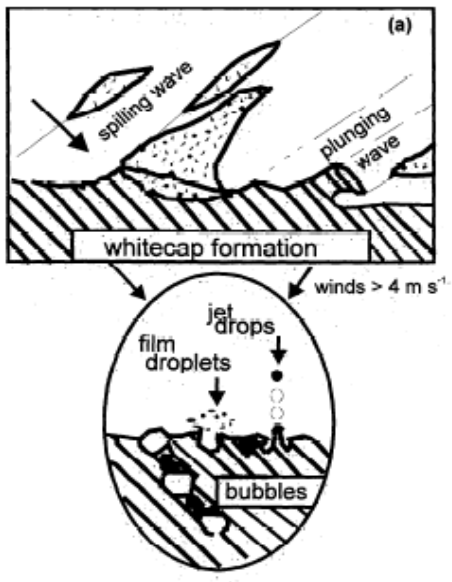
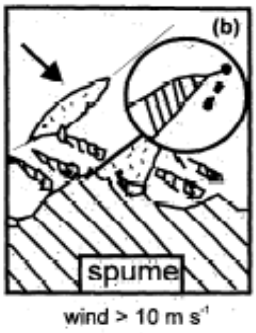
The model equations

The method for modelling marine aerosol emissions adopted here follows an approach developed by Monahan et al. (1986) and improved and applied by Gong et al. (1997) as part of a wider study to model marine aerosol emissions and concentrations for a point in the North Atlantic between Ireland and Iceland. The model presented in Gong et al. (1997) includes the following components:

- Marine aerosol (more specifically size fractionated PM) generation by surface wind;
- Vertical transport of particulates;
- Dry deposition and gravitational settling; and
- Wet removal processes

They incorporated these components into a one-dimensional version of the Canadian general climate model and ran the model over a 90 day period (January – March). Gong (2003) then proposed a refinement to the model equations so as to better represent emissions of particles with a radius of less than 0.2 μm. The study presented here only considers quantifying emission estimates, and not modelling marine aerosol particle behaviour and concentrations once emitted to the atmosphere. Hence we have only considered the first of the model components used in Gong et al. (1997). This takes the form of a semi-empirical formulation, which relates size-segregated surface emission rates to wind speed. This is expressed as density function dF/dr (particles emitted m^{-2} s^{-1} μm^{-1}). Two mechanisms of marine aerosol generation are represented in this formulation (see Figure 1).

Figure 1: Mechanisms of marine aerosol generation used in the modelling (Diagrams from Gong et al., 1997)

a) Indirect emissions through bubbles	b) Direct emissions through spumes
	
$\frac{dF_0}{dr} = 1.373U_{10}^{3.41} r^{-3} (1 + 0.057 r^{1.05}) \times 10^{1.19 e^{-B^2}}$ (Equation 1a) where $B = (0.380 - \log r)/0.650$. r is the particle radius at RH = 80% and U_{10} is the 10-m wind speed. The p	$\frac{dF_1}{dr} = \begin{cases} 0 & r < 10\mu m \\ 8.6 \times 10^{-6} e^{2.08 U_{10}} r^{-2} & 10\mu m \le r \ge 75\mu m \\ 4.83 \times 10^{-2} e^{2.08 U_{10}} r^{-4} & 75\mu m \le r \ge 100\mu m \\ 8.60 \times 10^{6} e^{2.08 U_{10}} r^{-8} & r \ge 100\mu m \end{cases}$ (Equation 1b)

The total generation rate of sea-salt aerosols (dF/dr) is then calculated as the sum of the rate of indirect generation through bubbles (dF_0/dr) and direct generation through spumes (dF_1/dr):

$$\frac{dF}{dr} = \frac{dF_1}{dr} + \frac{dF_0}{dr} \qquad \text{(Equation 2)}$$

It should be noted that for particles with a radius of less than 10 μm, there are no modelled emissions from spumes (see equation 1b). Therefore, for our modelling of PM_{10} and $PM_{2.5}$ emissions, all emissions modelled are caused by indirect emissions through bubbles as calculated in Equation 1a. Gong et al. (1997) recommend that for particles of a radius of greater than 10 μm equation 1b should be used. However they also observed that this generates too many large particles at higher wind speeds compared to observations.

These equations were scripted in a GIS to enable emission calculations to be made using spatially disaggregated input data. Details of the particle size bins for which the modelling has been carried out are given below and a discussion of the advantages and disadvantages of using a size bin approach is presented in the discussion. Similarly, details of 10m wind speed data that feed into the model equations are given below.

Particle size ranges

Gong et al. (1997) defined eight discrete particle size bins for their modelling. The total flux of marine aerosol particles emitted for each size bin was calculated by integrating the equation across the size bin.

For our work, eight consecutive size bins have also been defined, starting from 0 μm and each covering a range of 1.25 μm in diameter (see Table 1). Model runs have been carried out using the mid-point of each bin and assuming that this modelled emission applies across the whole bin. Calculated emissions from each bin were then added together as indicated in Table 1 to give total a $PM_{2.5}$ and a total PM_{10} emission.

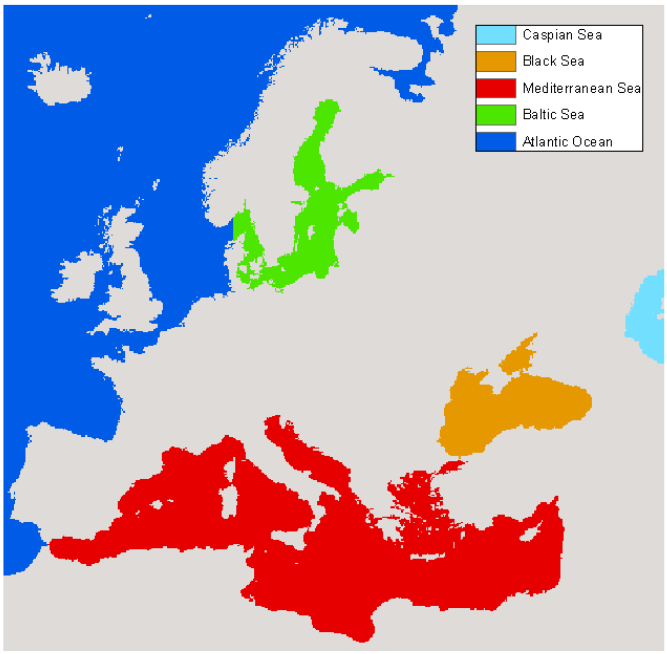
Table 1: Size bin widths (diameter) used to calculate emissions of PM_{10} and $PM_{2.5}$ from marine sources.

Minimum bin width (μm)	0	1.25	2.5	3.75	5	6.25	7.5	8.75
Maximum bin width (μm)	1.25	2.5	3.75	5	6.25	7.5	8.75	10
Calculated mass included in $PM_{2.5}$	✓	✓						
Calculated mass included in PM_{10}	✓	✓	✓	✓	✓	✓	✓	✓

Applying the model to a European scale

Gong et al. (1997) applied the model equations set out above to a point in the North Atlantic between Ireland and Iceland. The model extent defined in the NatAir project is significantly larger than this (see Figure 2). The model was run at a spatial resolution of 10 x 10 km to allow practical data handling.

Figure 2: Marine area covered by the NatAir extent



The main input data necessary to run the model is wind speed data at a 10m altitude. This was provided by the University of Stuttgart (Heiko Pfeiffer pers. comm., 2006). These data were extracted from the MM5 meteorological data set (at a resolution of 24 x 24 km), and converted to 10 x 10 km using an area weighted averaging approach. The model was run using daily average wind speeds. Although finer spatial and temporal resolutions were preferable, it was not possible to run the model for a four year period at this temporal resolution in the available timescales.

The model output therefore represented flux of PM for each size bin expressed as number of particles emitted per 10 x 10 km gridsquare per day. To calculate the mass of PM_{10} and $PM_{2.5}$ generated for each grid square, further assumptions regarding the characteristics of the particles emitted were made, to convert from particle numbers to mass:

- particles were assumed to be a perfect sphere (volume $4/3\pi r^3$)
- the density of particles was assumed to be equivalent to the density of water at 15 °C and 3.5% salinity.
- The particles are assumed to be water containing ions at a salinity representative of the relevant sea/ocean.

III RESULTS

Estimates of total annual PM_{10} and $PM_{2.5}$ for 1997, 2000, 2001 and 2003 are presented in Table 2a. This shows that for PM_{10} that total emissions across the whole NatAir extent were greatest in 2003 (128 Tg/year). Emissions in 1997 and 2000 are then the next greatest with lowest emissions in 2001 (86% of 2003 total emissions). Total emissions of $PM_{2.5}$ follow a very similar pattern with the greatest total emission modelled in 2003.

Table 2a: Annual PM_{10} and $PM_{2.5}$ Emissions- Wet (Tg/year)

PM_{10}						
Year	NatAir grid Total	Caspian Sea	Black Sea	Mediterranean Sea	Baltic Sea	Atlantic Ocean
1997	120.67	0.67	2.28	14.73	4.48	98.48
2000	120.11	0.41	2.13	14.01	4.54	98.97
2001	110.46	0.48	2.51	15.47	4.60	87.36
2003	127.76	0.52	2.61	21.54	4.75	98.27
$PM_{2.5}$						
Year	NatAir grid Total	Caspian Sea	Black Sea	Mediterranean Sea	Baltic Sea	Atlantic Ocean
1997	10.05	0.06	0.19	1.23	0.37	8.20
2000	10.01	0.03	0.18	1.17	0.38	8.24
2001	9.20	0.04	0.21	1.29	0.38	7.28
2003	10.64	0.04	0.22	1.79	0.40	8.19

As explained in Section 1, these refer to a ”wet“ mass, and a “dry” mass can be determined by using the salinity of the different seas. These are given in Table 2b below (salinity data being taken from Anthoni 2006 and Aladin & Plotnikov 2004).

Table 2b: Annual PM_{10} and $PM_{2.5}$ Emissions- Dry (Tg/year)

PM_{10}						
Year	NatAir grid Total	Caspian Sea	Black Sea	Mediterranean Sea	Baltic Sea	Atlantic Ocean
1997	4.22	0.008	0.087	0.560	0.022	3.545
2000	4.20	0.005	0.081	0.532	0.023	3.563
2001	3.86	0.006	0.095	0.588	0.023	3.145
2003	4.49	0.006	0.099	0.819	0.024	3.538
$PM_{2.5}$						
Year	NatAir grid Total	Caspian Sea	Black Sea	Mediterranean Sea	Baltic Sea	Atlantic Ocean
1997	0.35	0.0007	0.0072	0.0467	0.0019	0.2952
2000	0.35	0.0004	0.0068	0.0445	0.0019	0.2966
2001	0.32	0.0005	0.0080	0.0490	0.0019	0.2621
2003	0.37	0.0005	0.0084	0.0680	0.0020	0.2948

Figures 3a and 3b present maps of annual total emissions of PM_{10} and $PM_{2.5}$ for 1997, 2000, 2001 and 2003. This shows that the grid cells with the highest total emissions across the year are found in the North Atlantic Ocean especially in the area north of 50° latitude. The pattern of lower emissions to the east of Iceland and the UK and in the Baltic Sea probably occurs because the land masses shelter these area of sea from the prevailing wind direction, which in the North Atlantic is from the west. In terms of inter-year variability in the North Atlantic, Table 2a shows that emission totals in 1997, 2000 and 2003 are all very similar, while total emissions in 2001 are less.

In the Mediterranean, Figures 3a and 3b show that a zone of low emissions follows the coastline, with particularly high emissions in the Gulf of Lions on the French south coast and in the Aegean Sea. In 2003, total emissions of both PM_{10} and $PM_{2.5}$ in the Mediterranean are significantly higher than in other modelled years.

Figure 4 presents time series of emissions throughout the year for all four years modelled. This shows that generally emissions are highest in the winter, with a summer low and transition periods in between in the spring and autumn. The distributions all show high variability across relatively short time scales. This might reflect the non-linearity between 10m wind speeds and emissions in the generation function used. This means that on days with high wind speeds, emissions will be significantly higher than on days with lower wind speeds. It would therefore be instructive to look at the wind speed distributions throughout the year to further understand what is causing these peaks.

In terms of variability between different seas/oceans, Figure 4 show that the greatest overall seasonal variation in emissions occur in the Atlantic. However, the other seas also do exhibit some seasonal variability, which is particularly evident in the Mediterranean in 2003.

Table 3 presents average emission per grid square for different sea areas within the NatAir extent. This is useful because it shows that as well as the Atlantic having the greatest total emissions of PM, it also generates the most PM per grid square. The Baltic sea has the next most prolific PM emission per grid square, followed by the Mediterranean.

Table 3: Average emission (wet) per grid square for different sea areas within the NatAir extent (Tonnes/year)

PM_{10}						
Year	NatAir grid Total	Caspian Sea	Black Sea	Mediterranean Sea	Baltic Sea	Atlantic Ocean
1997	1451.4	523.2	496.2	582.8	1003.7	2091.8
2000	1444.7	319.4	465.2	554.3	1019.2	2102.2
2001	1328.6	373.6	547.4	612.0	1030.7	1855.6
2003	1536.7	406.4	569.8	852.1	1064.4	2087.3
$PM_{2.5}$						
Year	NatAir grid Total	Caspian Sea	Black Sea	Mediterranean Sea	Baltic Sea	Atlantic Ocean
1997	120.9	43.6	41.3	48.5	83.6	174.3
2000	120.4	26.6	38.8	46.2	84.9	175.1
2001	110.7	31.1	45.6	51.0	85.9	154.6
2003	128.0	33.9	47.5	71.0	88.7	173.9

The proportion of PM_{10} emitted as $PM_{2.5}$ shows very little variability either spatially or temporally. Typically approximately 8% of PM_{10} is emitted as $PM_{2.5}$. This is broadly in line with expectations when compared to measured concentration data. For example, APEG (1999) found that concentrations of fine marine aerosol particles at five sites in the UK made up between 20 and 30% of total coarse particle concentration. This is reasonable since fine particles are likely to have a longer residence time in the atmosphere than more coarse particles, so by the time a given parcel of sea air reaches the land-based monitoring sites, the ratio of fine to coarse particle will have increased.

For the present day climate, the total global marine aerosol flux from the ocean to the atmosphere is estimated to be 3,300 Tg/yr (IPCC, 2001) (Table 2), although this is high in uncertainty (Seinfeld and Pandis' estimate is lower at 1,300 Tg/yr).

Table 4. Marine aerosol emissions for the year 2000 (Tg/yr)- IPCC, 2001.

Diameter	Northern Hemisphere	Southern Hemisphere	Global
< 1 μm	23	31	54
1 - 16 μm	1,420	1,870	3,290
Total	1,440	1,900	3,340

A simple comparison between emissions estimated in this paper, and those by IPCC show the NatAir emissions to be 6% of the global estimate, with the NatAir extent representing 5% of the global sea coverage. The uncertainties of both emission estimates are high (and it has been assumed that the global PM_{16} emission can be linearly scaled to a PM_{10} fraction).

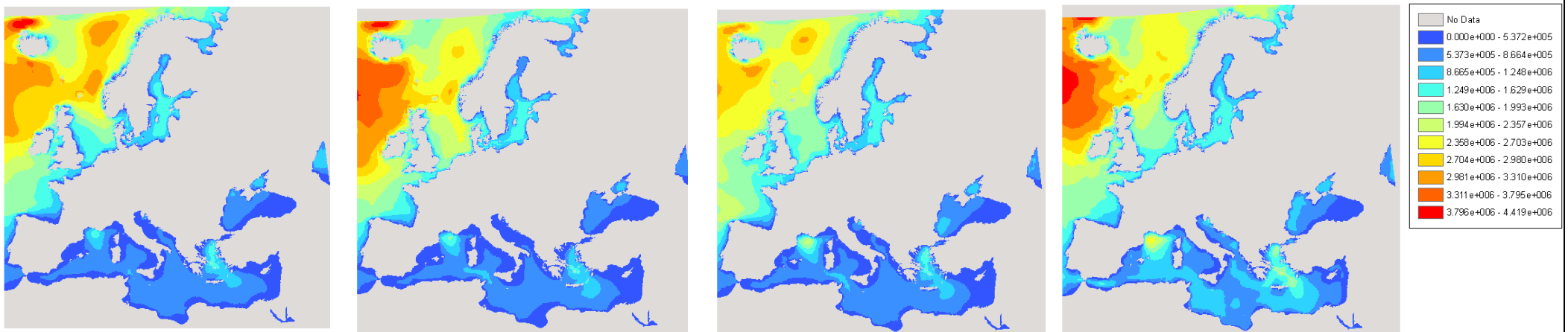
The Impact of Climate Change on Emissions

Climate change scenarios predict a range of impacts on future meteorological conditions. For marine aerosol emissions the most important parameter is wind speed, although other parameters have the potential to influence the emissions- salinity, and it has been proposed that temperature, surfactants and organic content all may impact on the level of emission to differing extents.

Within the NatAir programme it has been suggested that 2003 be used as an indicator for typical 2050 meteorological conditions, although this is more relevant for temperature. However, it is possible to make some qualitative comments on the likely future emissions of marine aerosol.

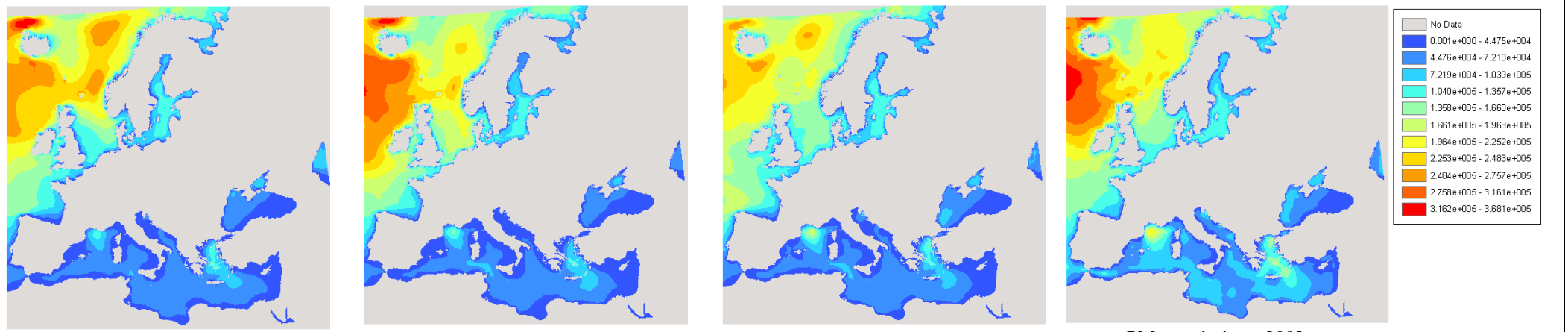
Wind speeds are expected to be more variable, and generally higher. Given that the relationship between wind speed and emission is non-linear, it is the peaks of wind speed rather than the overall average which may have the most significant impact on future emissions of marine aerosol. Given the associated uncertainties with both the current estimates, and the changes to wind speed in general, it is not possible to quantify the expected increase in emissions of marine aerosol.

Figure 3a: Total annual emissions maps of PM_{10} for 1997, 200, 2001 and 2003 (kg/gridsquare/year)



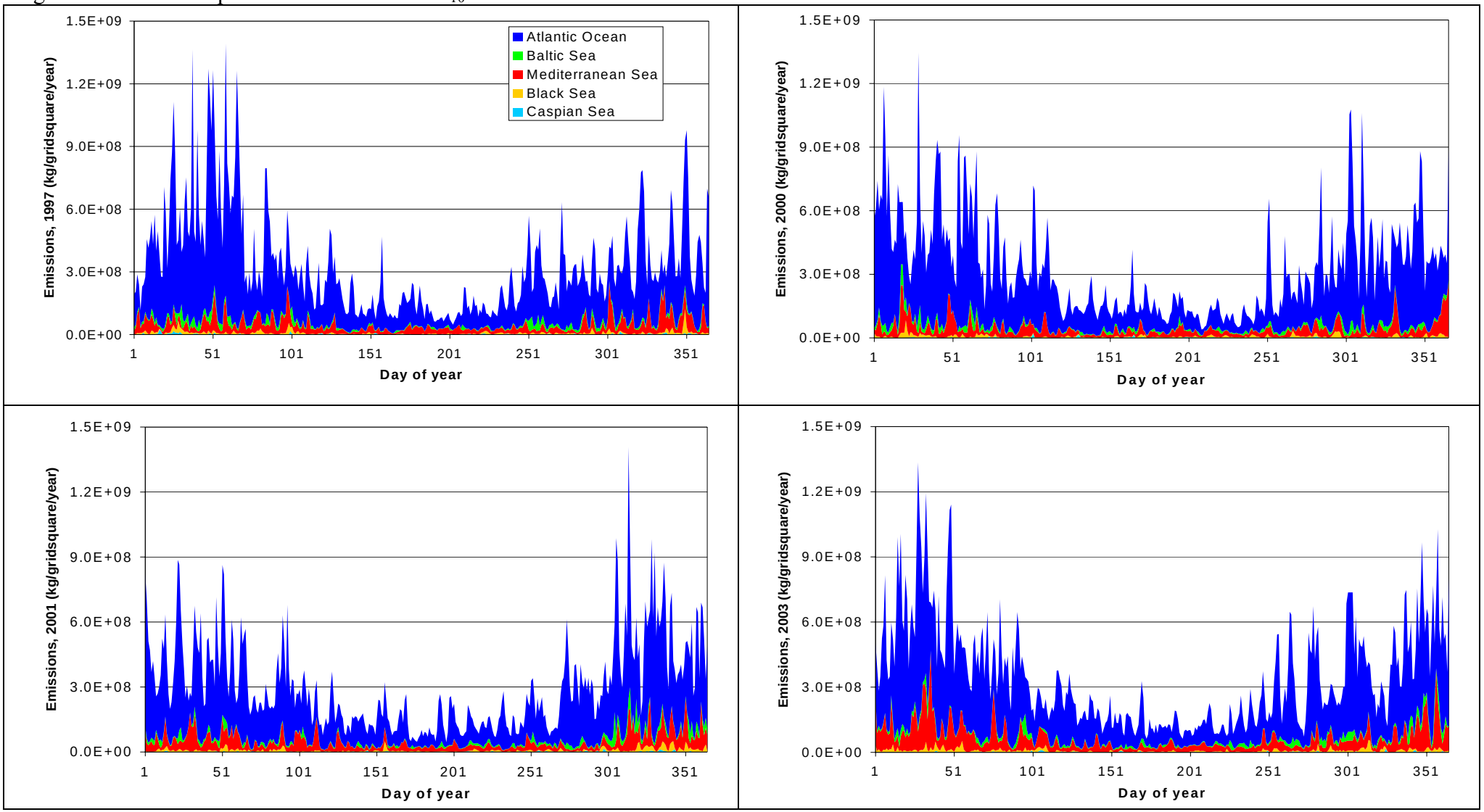
PM_{10} emissions, 1997 | PM_{10} emissions, 2000 | PM_{10} emissions, 2001 | PM_{10} emissions, 2003

Figure 3b: Total annual emissions maps of $PM_{2.5}$ for 1997, 200, 2001 and 2003 (kg/gridsquare/year)



$PM_{2.5}$ emissions, 1997 | $PM_{2.5}$ emissions, 2000 | $PM_{2.5}$ emissions, 2001 | $PM_{2.5}$ emissions, 2003

Figure 4: Time series plots of emissions of PM_{10}



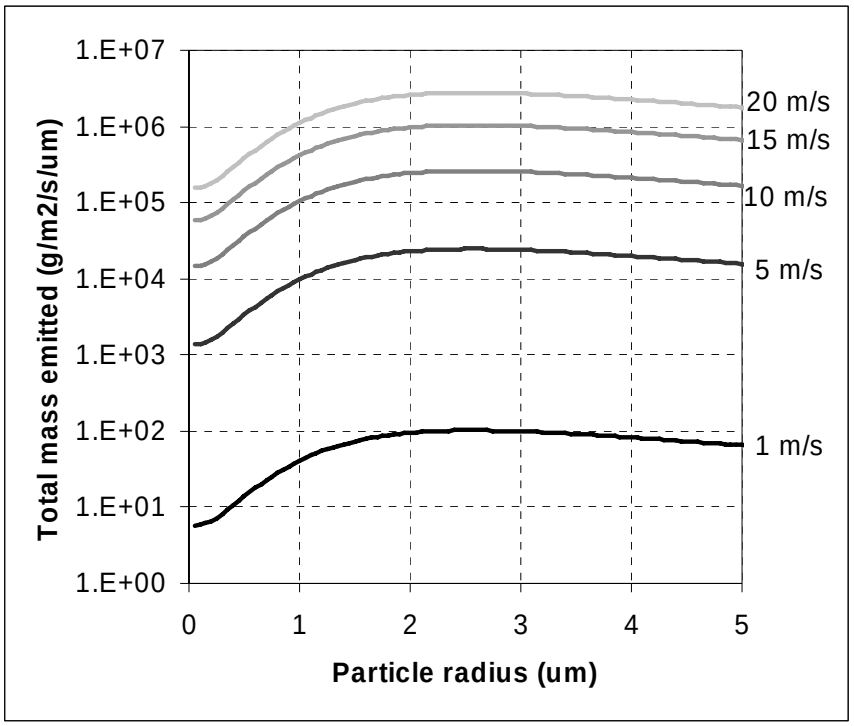
IV UNCERTAINTIES

Model equations

The model equation calculates marine aerosol emissions as a function of wind speed. This is a non-linear relationship where higher wind speeds cause disproportionately high emissions. This is shown by simple sensitivity tests on the model equations as shown in Figure 5. This shows that increasing the wind speed from 1 to 20 m s^{-1} causes the total mass of sea-salt aerosol emitted to increase by approximately 4.5 orders of magnitude for all particle sizes within the range considered in this study.

This graph also shows for a given wind speed within the range considered, the greatest mass of particles emitted are of approximately 2 μm in radius. For particles of a greater radius than this, fewer particles are release so the total mass is less. For particles smaller than this, more are released. However, they are sufficiently light not to reach the same total mass as for bigger particle sizes. Across the PM_{10} size range, this graph shows there is a variability in total mass emitted of 1.2 orders of magnitude for any given wind speed.

Figure 5: Model sensitivity to variations in wind speed for a range of particle sizes.



Other uncertainty in the model equations is introduced by omission of a model term to represent generation of sea-salt aerosol by wet precipitation (i.e. the current model equations fail to represent all processes occurring). This occurs through the impact of the raindrops which produces:

- bubbles produced in the surface water; and
- bubbles generated by secondary drops caused by to splash from the initial rain drop impact (Blanchard and Woodcock, 1957).

The Gong et al. (1997) model does not incorporate this mechanism due to the lack of information on the subject. However, compared to the wind generation mechanism, marine aerosol produced through precipitation is likely to be minor, especially since there will be significant periods of time with no precipitation occurring in a given grid square.

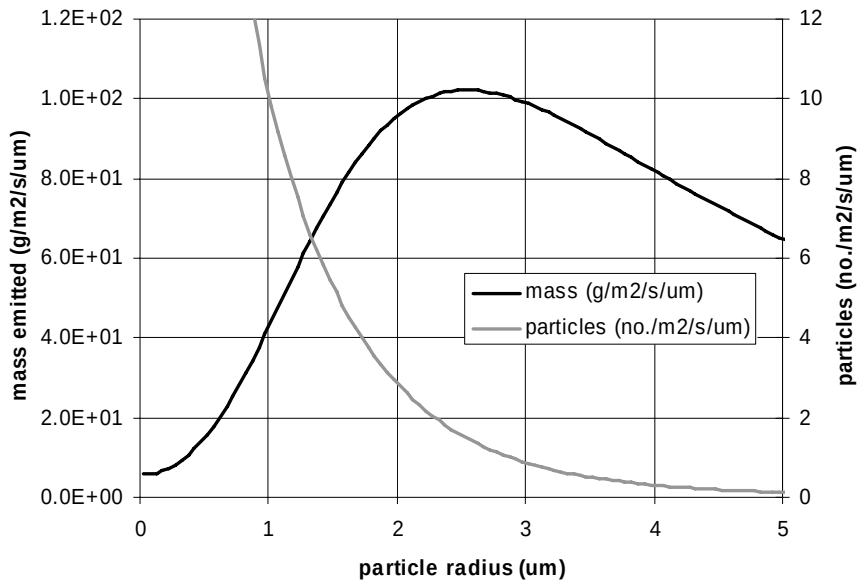
Model input data
The model has been run using daily average wind speed. However, wind speed tends to vary on much faster time scale than this. Therefore, it is likely that using the daily average wind speed will under-predict emissions as the peak wind speeds, which would cause the most significant emissions, will be smoothed out by the averaging of the meteorological data.

The spatial resolution of the modelling also introduces uncertainties into the method. This is because particularly in coastal regions, there may be intra-grid square variability in terms of wind speeds and therefore emissions. Additionally, at the land-sea interface, where there are grid squares which contain both land and sea, these have not been modelled. Omission of these grid squares is unlikely to have a significant impact upon the total emission estimate for the entire grid extent. However, for users of the spatially disaggregated map data, it may have a more significant impact. This is particularly the case for concentration modelling over land as these are the most relevant grid squares.

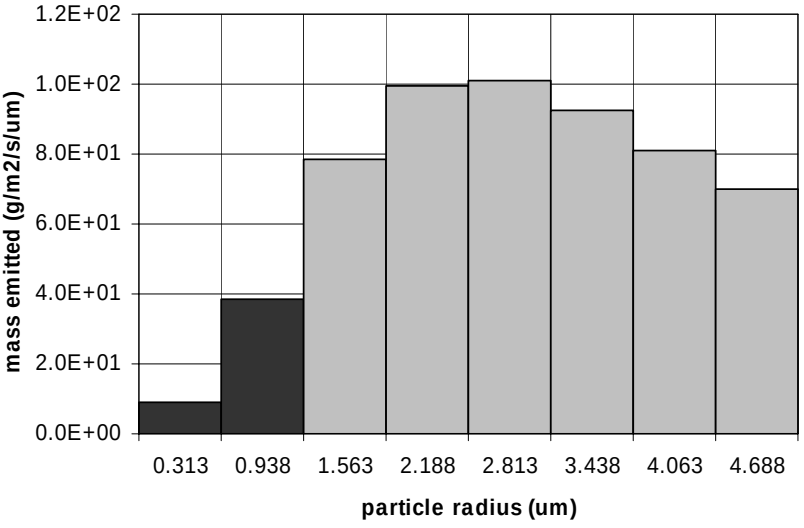
Modelling for different particle sizes
For the purposes of this modelling exercise, a series of particle size bins have been selected. The model has been run for the mid point of each bin and the total emission calculated as the width of the bin multiple by the calculated emission for the mid point value. Figure 6 compares the total emissions using the size bins selected with the actual curve generated using the model equation. This shows that whilst some error is introduced by using size bins, there is a good fit to the model equation line. Therefore, although using a size bins approach does introduce some uncertainty to the model results, it is much preferable compared with not representing particle size differences at all.

Figure 6: Total emissions (g m^{-2} s^{-1}) for a wind speed of 1 m s^{-1} using the size bins compared with the model equation line

a) Model equation lines (total emission calculated as the area under the curve)



b) Model approximation using bin widths of 0.625 μm (PM2.5 emissions =area covered by dark grey bars, PM_{10} emissions = area covered by light and dark grey bars)



A further issue associated with particle size relates to the representation of emissions of very small (sub 0.1 μm) marine aerosol particles. Gong et al., (1997) and Andreas (1998) both observed that modelled emissions for these particles significantly over estimated the number of particles generated. Gong (2003) therefore proposed revising Monahan's formula as follows:

$$\frac{dF_0}{dr} = 1.373U_{10}^{3.41} r^{-A}(1 + 0.057r^{3.45}) \times 10^{1.607e^{-B^2}} \qquad \text{(Equation 3)}$$

Where $A = 4.7(1 + \Theta r)^{-0.017r^{-1.44}}$ and B = (0.433 – log r)/0.433 and Θ in A is an adjustable parameter that controls the shape of the sub-micron size distributions.

The modified formula maintains the original form of the Monahan equation but adds a size-dependent exponential term (A) for radius r and modifies other parameters. The new source function derived by Gong (2003) should be suitable for simulating both number and mass size distributions down to about 0.03 μm in dry marine aerosol radius. This modified approach has

not been used here. However, because particles of less than 0.1 μm make up a very small proportion of PM_{10} emission by mass for any given wind speed, the impact of this over estimation on total modelled emissions is likely to be relatively small.

Conversion from number of particles emitted to total mass

The assumption made in converting number of particles emitted for each size bin to a total mass emitted for each size bin will obviously not hold true in all cases. For example, not all marine aerosol particles (if any) will be perfect spheres and they will have a range of densities. However, using this approach does enable us to estimate mass emitted, which otherwise would not be possible.

V CONCLUSIONS AND OUTLOOK

An empirical modelling approach to estimate spatially disaggregated emissions of marine aerosol (as PM_{10} and $PM_{2.5}$) has been applied across a sea/ocean coverage surrounding Europe. Datasets have been generated at a 10 km x 10 km resolution by using a GIS to allow these data to be of use for input into atmospheric transport and chemistry modelling studies. Spatial patterns of emission indicated higher emissions at locations further from land, the impact of sheltering by land being clearly observable.

Emissions have been estimated for four calendar years (1997, 2000, 2001 and 2003) to investigate the inter year variability, and allow qualitative comment on the dependence of the emissions on the input parameters and possible trends for future years.

Although emission estimates are considered to be relatively uncertain, the results for the NatAir extent were found to agree well with other estimates of marine aerosol emissions. A number of recommendations have been made for further work to improve the robustness of the emission estimates.

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