EMISSION AND DISPERSION MODELLING OF AEROSOLS AND HUMAN EXPOSURE TO PARTICULATE MATTER

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Academic dissertation in Physics

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Finnish Meteorological Institute

Helsinki, 2016
Emission and dispersion modelling of aerosols and human exposure to particulate matter

Abstract

Atmospheric aerosols are subject to extensive research, due to their effect on air quality, human health and ecosystems, and hold a pivotal role in the Earth’s climate. The first focus of this study is to improve the modelling of aerosol emissions and its dispersion in the atmosphere, in different spatial and temporal scales, and secondly, to integrate the dispersion modelling with population activity data to estimate exposure metrics. The mathematical models used in this study are fully or partially developed by the Finnish Meteorological Institute: a regional-to-global scale chemical transport model SILAM, a local-scale point/line-source dispersion model, UDM/CAR-FMI, and a human exposure and intake fraction assessment model, EXPAND.

One of the outcomes of this work was the refinement of the emission modelling for the mesoscale dispersion model. A new parameterisation for bubble-mediated sea salt flux has been developed, taking into account the effects of wind speed and seawater salinity and temperature. The parameterization is valid for low-to-moderate wind speed, seawater salinity ranging between 0 and 33 ‰, seawater temperature ranging between -2 and 25 ºC, and can be applicable to particles with dry diameters ranging between 0.01 and 10 µm. The near-real time fire estimation system, IS4FIRES, based on Fire Radiative Power (FRP) measured by the remote sensing instrument MODIS, was refined to reduce the overestimation of particulate matter (PM) emissions by including more vegetation types, improving the diurnal variation, removing misattributed fires from the FRP data, and recalibrating the emission factors. Applying dynamic emission modelling brought more insight to the spatial distribution of these emissions, their contribution to the atmospheric budget, and possible impact on air quality and climate. The modelling shows that sea salt can be transported far over land and contribute up to 6 µg m\(^{-3}\) to PM\(_{10}\) (at an annual level). It also indicates that the Mediterranean Sea has sharp gradients of concentration, becoming an interesting area to analyse regarding the feedbacks to the regional climate. According to the predictions, upward scattering by SSA, at TOA, can be up to 0.5 W m\(^{-2}\), and there will be an overall cooling in the future for the North of Europe and warming for the South, due to SSA. The simulations for wildland fires show how the system improves after calibration and the importance vegetation type for the intensity of the emissions. By including misattributed fires, there will be up to 80% overestimation in aerosol optical depth, close to the misattributed sources.

The emissions for Helsinki Metropolitan Area (HMA) were revised to bring up-to-date the emissions for traffic and energy sectors, for urban-scale applications. The EXPAND model was revised to combine concentrations and activity data in order to compute parameters such as population exposure or intake fraction. EXPAND includes improvements of the associated urban emission and dispersion modelling system, time use of population, and infiltration coefficients from outdoor to indoor air. This refinement showed that PM\(_{2.5}\) in HMA is mainly originated from long-range transport, with the largest local contributors being vehicular and shipping (at harbours and its vicinity) emissions. At annual level, the population is mostly exposed to PM\(_{2.5}\) indoors (home and work), but the population is acutely exposed while commuting.

Emissions modelling, atmospheric dispersion modelling, exposure modelling, climate change, sea salt, wildland fire, particulate matter
To the women of my family, for their strength, wisdom and courage

Às mulheres da minha família, pela vossa força, sabedoria e coragem
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Håvard, jeg er klar. Du kan nu forsøde min kaffe med en formiddag kys for en million år.

Helsinki, April 2016,
Joana Soares
### Abbreviations and terminology

<table>
<thead>
<tr>
<th>Abbreviation</th>
<th>Description</th>
</tr>
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<tbody>
<tr>
<td>AQUA</td>
<td>Multi-national NASA scientific research satellite in orbit around the Earth</td>
</tr>
<tr>
<td>AIS</td>
<td>Automatic Identification System</td>
</tr>
<tr>
<td>AOD</td>
<td>Aerosol Optical Depth</td>
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<tr>
<td>BEM</td>
<td>Biogenic Emission Model</td>
</tr>
<tr>
<td>CAR</td>
<td>Road Network Dispersion Model</td>
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<tr>
<td>CTM</td>
<td>Chemical Transport Model</td>
</tr>
<tr>
<td>DEHM</td>
<td>Danish Eulerian Hemispheric Model</td>
</tr>
<tr>
<td>DMS</td>
<td>Dimethylsulfide</td>
</tr>
<tr>
<td>(D_p)</td>
<td>Particle diameter</td>
</tr>
<tr>
<td>DRE</td>
<td>Direct radiative effect</td>
</tr>
<tr>
<td>ECHAM5-MPIOM</td>
<td>Atmospheric general circulation model/ocean model developed by the Max Planck Institute</td>
</tr>
<tr>
<td>EMEP</td>
<td>Co-operative Programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe</td>
</tr>
<tr>
<td>EMME/2</td>
<td>Traffic planning model with interactive transportation planning</td>
</tr>
<tr>
<td>ESCAPE</td>
<td>European Study of Cohorts for Air Pollution Effects</td>
</tr>
<tr>
<td>EXPAND</td>
<td>EXposure model for Particulate matter And Nitrogen oxides</td>
</tr>
<tr>
<td>FMI</td>
<td>Finnish Meteorological Institute</td>
</tr>
<tr>
<td>FRP</td>
<td>Fire radiative power</td>
</tr>
<tr>
<td>FORE</td>
<td>Road Suspension Emissions Model</td>
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<tr>
<td>GCM</td>
<td>General circulation models</td>
</tr>
<tr>
<td>GLCC</td>
<td>Global Land Cover Characterization</td>
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<tr>
<td>GIS</td>
<td>Geographic Information System</td>
</tr>
<tr>
<td>HMA</td>
<td>Helsinki Metropolitan Area</td>
</tr>
<tr>
<td>IS4FIRES</td>
<td>Integrated Monitoring and Modelling System for wildland Fires</td>
</tr>
<tr>
<td>M03</td>
<td>Mårtensson et al. (2003)</td>
</tr>
</tbody>
</table>
M86 Monahan et al. (1986)
MARGA Monitor for Aerosol and Gases
MATCH Multi-scale Atmospheric Transport and Chemistry
MODIS Moderate Resolution Imaging Spectroradiometer
NASA National Aeronautics and Space Administration
NEAT Northeast Atlantic measurement campaign
PM Particulate matter
SD Standard deviation
SILAM System for Integrated modelling of Atmospheric composition
STEAM Ship Traffic Emissions Assessment Model
SRES A1B Emission Scenarios with the assumption that similar improvement rates apply to all energy supply and end-use technologies
SSA Sea salt aerosol
TERRA Multi-national NASA scientific research satellite in a Sun-synchronous orbit around the Earth
TOA top of the atmosphere
Trace gases Gaseous constituents that comprise less than 1% of the atmosphere
UDM Urban Dispersion Modelling System
RCA3 Rossby Centre Regional Climate model, version 3
RMSE Root mean square error
R Correlation coefficient.
VOC Volatile Organic Compounds
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Review of the papers and Author’s contribution

**Paper I** presents a new analytical formulation for bubble-mediated sea salt spray production for particles with $D_p$ ranging between 0.01 and 10 µm. In this formulation, the production of sea salt aerosol is a function of wind speed and seawater temperature and salinity. This source function was implemented in the dispersion model SILAM and applied to compute the distribution of sea salt aerosol over the North Atlantic and Western Europe, as well as globally. The source function was evaluated by comparing SILAM’s predictions with ground-based and remote sensing observations. The author contributed to the validation of the source function, by performing most of the simulations and analysing the results. The author contributed to the writing of the article.

**Paper II** presents a multi-model comparison of four dispersion models (DEHM, MATCH, EMEP and SILAM) to assess the uncertainty and robustness of the sea spray predictions over Europe, particularly over the European seas. The models were driven by the same global projection climate scenario SRES A1B for past (1990-2009) and future (2040-2059) climate but included different formulations for sea salt aerosol production. In this paper, the impact of climate change on the production and fate of sea salt aerosol was assessed, alongside with the direct radiative effect of this natural aerosol on the regional radiative budget. The author was responsible for the simulations with the SILAM and the radiative transfer (LibRadTran) models, for the collection and standardization of the sea salt aerosol simulations, and for the analysis of the results. The author of this thesis was the leading author of the article.

**Paper III** discusses the main uncertainties of wildland fires emission estimates used in the AQMEII-II case study, by quantifying the uncertainties of the wildland fire particulate matter emission estimations by IS4FIRES. IS4FIRES converts fire information data from the remote sensing instrument MODIS, to emission fluxes of atmospheric pollutants. It is used for near-real-time and historical evaluation of emissions from wildland fires. The estimation of uncertainties is done by reviewing and refining the fire emission model IS4FIRESv1 to the current version, IS4FIRESv2. The refinement and evaluation was done by comparing SILAM’s predictions with remote sensing observations. The author was involved in the refinement and validation of IS4FIRESv2, and was responsible for quantifying the uncertainties for previous and current versions of the fire emission model. The author of this thesis was the leading author of the article.
**Paper IV** presents an improved version of the exposure model (EXPAND), that estimates the human exposure metrics, in an urban area: population exposure and intake fraction. The paper describes the improvements to emission modelling and exposure estimation. Population exposure to fine particulate matter in Helsinki Metropolitan Area and in Helsinki for 2008 and 2009, respectively, were estimated with EXPAND. These estimations indicate where the population is most affected and which sources are most relevant for the exposure. The author was involved in the gathering and analysis of the results, and was the leading author of the article.

**Paper V** presents different methods to evaluate intake fractions from benzene emitted by traffic sources in Helsinki Metropolitan Area: a modified version of the exposure model framework EXPAND, for computations in a street canyon; a box-model; and a statistical model relating concentration and time-activity data. The author was responsible for the computations with EXPAND and analysis of the results, and participated in the writing of the article.
Other publications not included in this thesis


1 Introduction

An atmospheric aerosol is a mixture of liquid and solid particles suspended in air; this mixture can be of different sizes and composition (Warneck, 1988). Atmospheric aerosols can be originated from primary emissions (natural or anthropogenic sources) or formed within the atmosphere through gas-to-particle conversion (Seinfeld and Pandis, 2006). On a planetary scale, the bulk of the total atmospheric burden mainly consist of natural primary aerosol; however, the proportions can be very different if considering urban or industrial areas, where traffic and industrial are the main sources of atmospheric aerosol (Textor et al., 2006). At the source areas, the chemical composition of the aerosol is linked to the prevailing emission sources but throughout the atmospheric life-cycle, concentration and properties of the aerosol change continuously through a series of physical and chemical processes (Kulmala et al., 2004): new particle formation, condensation, evaporation, water uptake, heterogeneous chemistry, dispersion and removal.

Dispersion modelling is a widely used tool to estimate the contribution of aerosols to the atmospheric composition, and infer about its potential impact on human health, ecosystems, and climate. An atmospheric dispersion model is a mathematical simplification of the atmosphere, applied to different space and time scales, depending on the physical and chemical processes that atmospheric species undergo. The process of modelling can be divided in four stages: data input, dispersion calculations, deriving the quantities desired, and analysis. The accuracy and uncertainty of each stage should be known and evaluated to ensure a reliable assessment.

Studies, such as Im et al. (2014) and Prank et al. (2016), show that the misrepresentation of the sources and processes commonly brings under or overestimation of particulate matter (PM) levels by dispersion models. The characterization of PM has many gaps: chemical speciation of the emissions, spatial and temporal distribution of the aerosol emission profile, and misrepresentation of chemical and physical transformation if long periods and large spatial scales are required (Rudich et al., 2007).

$\text{PM}_x$ is the term used in air quality and health assessment to define the mass concentration of aerosol with maximum aerodynamic diameter of $x$ µm. PM has been identified as the largest environmental public health risk, being associated with excess morbidity and mortality (Hänninen et al., 2014). Many of the studies that assess the health impact of PM use air quality data directly from models (Amann et al., 2005) or a combination of models and monitoring (Fiala et al 2009) for the estimation of long term population exposure. The assessment of exposure usually requires the application of an integrated model chain starting from estimation of emissions, atmospheric dispersion and transformation of air pollutants (Ashmore and Dimitripoulou, 2009). Finally, the exposure model combines ambient air concentrations of pollutants and population activity data to calculate human exposure. If coupled with time-microenvironment-activity models, exposure will be weighted by the time spent by the population in a particular microenvironment, reflecting the movement of the population and the exposure to existing sources where the population is located.
The overall goal of this thesis is to assess the impact of several major non-anthropogenic aerosol sources on atmospheric composition and air quality, with a downstream application to human health. The work was concentrated on the refinement of mathematical models fully or partially developed by the Finnish Meteorological Institute (FMI): a meso-to-global scale chemical transport model SILAM, a local-scale point/line-source dispersion model UDM/CAR-FMI, and a human exposure and intake fraction assessment model EXPAND, especially regarding the emission modelling of aerosols. The specific objectives considered the development and application of these models to assess the atmospheric composition and human exposure:

- to improve the insight on the contribution of sea salt aerosol (SSA) to the atmospheric composition by developing an unified parameterisation for SSA flux, as a function of wind speed and seawater properties;
- to estimate the climate impact on SSA production and fate over Europe and European Seas and evaluate the feedback to the regional radiative balance due to the presence of SSA;
- to improve the understanding of the impact of PM emissions from wildland fires on atmospheric composition by refining a method for wildland fire emission estimation, IS4FIRES;
- to estimate the contribution from various PM$_{2.5}$ sources to the atmospheric composition and their impact on human exposure in Helsinki and in the Helsinki Metropolitan Area (HMA).

2 **Atmospheric dispersion modelling**

A dispersion model is a mathematical representation of the transport and diffusion processes that occur in the atmosphere under a set of conditions. The dispersion model intends to represent the complex physical and chemical processes that take place during transport and dispersion of trace gases and particles emitted into the environment, by a set of equations (ordinary, partial differential, parameterised and empirical). The fundamental equations solved by atmospheric dispersion models are the advection-diffusion-reaction equations, describing the evolution of tracers in space and time, considering atmospheric transport, chemical transformation, sources and sinks (Morton, 1996).

Atmospheric problems can be simulated over a variety of spatial scales, depending on the physical and chemical processes to be considered. This thesis addresses dispersion problems taking place at different spatial scales. Papers I, II and III are focused on mesoscale (20 – 100 km) processes, with the resolved time scale ranging from (tens of) minutes up to hours, and Papers IV and V are focused in microscale (< 2 km) processes with a times scale of tens of seconds. Mesoscale modelling tries to capture the diurnal cycle of the circulations, which results in the development of flow gradients and vertical stratification, and the effects of topography; both affect the air quality of a region. These models allow understanding the impact of emissions far away from the sources. Microscale should be able to allow capturing transport and mixing due to local thermal circulations and landscape characteristics. From now on, microscale modelling will be referred as urban scale modelling, since in
this thesis all the computations presented are for an urban area, involving diffusion of individual sources (e.g. power plants) and line-sources (traffic).

2.1 Mesoscale modelling: SILAM

Papers I, II and III have as their main modelling tool the SILAM model, documented in Sofiev et al. (2006, 2008, 2014 and 2015), which includes Eulerian and Lagrangian atmospheric transport descriptions. The system contains a meteorological pre-processor for evaluating the basic features of the boundary layer and the free troposphere, using the meteorological fields provided by numerical meteorological models (Sofiev et al., 2010). The model has several chemical transformation modules, including gas-phase chemistry in the troposphere and the stratosphere (Carbon Bond Mechanism (Gery et al, 1989) with updated coefficients), secondary inorganic aerosol formation (Sofiev, 2000), linearized sulphur oxides chemistry, radioactive nuclides decay, and aerosol dynamics (condensation and coagulation) computed from thermodynamic equilibrium or as dynamic. Aerosol size spectrum is described with sectional approach with user-defined bin distribution. Table 2.1 describes the different aerosols types assumed for the runs in Papers I, II and III, including the numbers of bins and respective size ranges. Depending on the particle size, mechanisms of dry deposition vary from primarily turbulent diffusion driven removal of fine aerosols to primarily gravitational settling of coarse particles (Kouznetsov and Sofiev, 2012). Wet deposition distinguishes between sub- and in-cloud scavenging by both rain and snow (Horn et al., 1987; Smith and Clark, 1989; Jylhä, 1991; Sofiev et al., 2006). The optical properties of aerosols and a selection of trace gases are calculated following Prank (2008). The particle hygroscopic growth is taken into account when deriving gravitational settling, dry deposition and optical properties of the aerosols. Emission models for sea salt (Paper I), wild-land fires (Sofiev et al., 2009, 2012a, Paper III), desert dust, allergenic pollen (Sofiev et al., 2012b, Siljamo et al., 2012, Prank et al., 2013) and volatile organic compounds (VOC) are embedded in SILAM. The bio-VOC can be estimated by MEGAN (Guenther et al., 1995) or BEM (Popkou et al., 2010). The model also includes data assimilation tools for both 3D- and 4D-VAR (Vira & Sofiev, 2010) and ensemble Kalman filter.

SILAM model has been extensively evaluated against air quality observations over Europe and the globe (daily at http://www.gmes-atmosphere.eu; Solazzo et al., 2012; Huijnen et al., 2010).

Table 2.1 Aerosol types and number of bins and size ranges assumed for the runs in Paper I, II and III.

<table>
<thead>
<tr>
<th>aerosol type</th>
<th>number of bins</th>
<th>size ranges (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>anthropogenic/fire</td>
<td>2</td>
<td>[0 2.5] [2.5 10]</td>
</tr>
<tr>
<td>dust</td>
<td>4</td>
<td>[0.01–1] [2.5] [2.5 10] [10 30]</td>
</tr>
<tr>
<td>sea salt</td>
<td>5</td>
<td>[0.01–0.1] [0.1 1.5] [1.5–6] [6 15] [15 30]</td>
</tr>
</tbody>
</table>
2.2 Urban area modelling: UDM-FMI and CAR-FMI

In Paper IV and V, the modelling system applied for evaluating emissions and atmospheric dispersion of pollution in an urban area is a combined application of UDM-FMI and CAR-FMI models. Both models are multiple source Gaussian urban dispersion models and have been addressed in detail by Karppinen et al. (2000a) and Kukkonen et al. (2001). The UDM and CAR-FMI take into account stationary and traffic (vehicular and shipping) sources, respectively.

The dispersion parameters are modelled as a function of Monin-Obukhov length, friction velocity, and boundary layer height, which are computed by the meteorological pre-processing model MPP-FMI (Karppinen et al., 2001). This model has been used with input data from the three nearest synoptic weather stations and the nearest sounding station, to evaluate an hourly meteorological time series for the dispersion modelling computations. Both dispersion models consider inert particles and gaseous compounds where simple chemical transformations, such as conversion between NO, NO\textsubscript{2} and ozone, are included. PM is treated as inert, i.e. no chemical reactions or aerosol processes are included in the calculations. The models include treatment of dry and wet deposition. The system computes hourly time series of concentrations and statistical parameters in each receptor point, which can be directly compared with air quality guidelines. The receptor points are adjustable with intervals ranging from approximately 20 m in the vicinity of the sources to 500 m further from the sources. The regional background concentrations of gaseous compounds are taken from a single measurement or interpolated from several measurements of the monitoring network, or estimated by a regional chemical transport model (CTM).

To include the shipping contribution to PM\textsubscript{2.5} surface concentrations in Paper IV, CAR-FMI was refined to include emissions from shipping. All shipping emissions were treated as line sources with an injection height of 30m above the sea level.

The CAR-FMI model has been previously evaluated against the measured data of urban measurement networks in HMA and in London, for gaseous pollutants (Karppinen et al., 2000b; Kousa et al., 2001; Hellén et al., 2005) and for PM\textsubscript{2.5} (Kauhaniemi et al., 2008; Sokhi et al., 2008; Singh et al., 2014). The performance of the CAR-FMI model has also been evaluated against the results of a field measurement campaign and other roadside dispersion models (Kukkonen et al., 2001; Öttl et al., 2001; Levitin et al., 2005). The UDM-FMI model has been evaluated against the measured data of urban measurement networks in HMA (Karppinen et al., 2000b; Kousa et al., 2001) and the tracer experiments of Kincaid, Copenhagen and Lilleström.

3 Exposure modelling

The relationship between a source and the subsequent exposure of a population depends on the ability of the released pollutant to reach individuals. The exposure efficiency is the fraction of material released that penetrates into the human body, depending on the characteristics of the source, pollutant,
environment where the pollutant is released, as well as the receptor (Harrison et al., 1986; Evans et al., 2000).

Since the urban population spends typically 80-95% of their time indoors (Hänninen et al., 2005; Schweizer et al., 2007), the exposure to pollutants is dominated by indoor exposure. Epidemiological studies based on concentrations (measured at fixed air quality monitoring sites or predicted by land-use regression models) ignore indoor sources, activity patterns of individuals, and fine-scale spatial variability of concentrations. These problems can be overcome by combining concentration of pollutants in air with time-microenvironment-activity models and indoor to outdoor concentration ratios. Microenvironment is defined as a location containing a relatively uniform concentration where the human exposure takes place. The average personal or population exposure is then estimated as a linear combination of concentrations in different microenvironments, weighted by the time spent in each of them. Therefore, exposure can be described as:

\[ E_i = \sum_{j=1}^{m} T_{ij} \cdot C_{ij} \]  

(3.1)

where \( E_i \) is the total exposure of individual \( i \) in various microenvironments \( m \) (µg m\(^{-3}\) s), \( T_{ij} \) is the time spent in microenvironment \( j \) by individual \( i \) (s) and \( C_{ij} \) is the air pollutant concentration that individual \( i \) experiences in microenvironment \( j \) (µg m\(^{-3}\)). This formulation can also be interpreted as a weighted sum of concentrations, in which the weights are equal to the time spent in each environment.

Another concept applied in exposure assessments is intake fraction. This concept has been used in life cycle assessments (Humbert et al. 2011; Apte et al., 2012), for ranking sources (Tainio et al., 2009), and to compare different control policies (Stevens et al., 2005). The intake fraction is defined as a fraction of the pollutant emission that is taken by humans via relevant exposure pathways, e.g. a portion of a source’s emissions that is inhaled by an exposed population over a defined period of time. This exposure metric is defined as it follows:

\[ iF = \frac{\text{Population Intake}}{\text{Total Emissions}} = \frac{\int_{T_1}^{T_2} \left( \sum_{i=1}^{P} C_i(t) \cdot Q_i(t) \right) dt}{\int_{T_1}^{T_2} E(t) \ dt} \]  

(3.2)

where \( iF \) is the intake fraction for the exposed population, \( T_1 \) and \( T_2 \) (s) are the starting and ending times of the emission, \( P \) is the number of people in the exposed population, \( Q_i \) (m\(^3\) s\(^{-1}\)) is the breathing rate for individual \( i \) at time \( t \), \( C_i \) (µg m\(^{-3}\)) is the concentration attributable to a specific source at time \( t \) in the breathing zone of individual \( i \), and \( E \) (g s\(^{-1}\)) is the source's emissions at time \( t \). In practice, the integral of the numerator is evaluated until the incremental concentration attributable to the source of interest is negligibly small. For instance, an intake fraction of one in a million (10\(^{-6}\)) means that for every tonne of a pollutant emitted, 1 g is inhaled by the exposed population. This allows quantifying
emission-to-intake relations, independently of the emission, location or exposure pathway for non-reactive compounds.

3.1 EXPAND

EXPAND has been used to evaluate the spatial and temporal variation of the average exposure of a urban population to air pollution, in different microenvironments (Kousa et al., 2002). This system combines information on the concentration in ambient air and the population activity and location, utilizing the geographical information system (GIS) MapInfo (Figure 3.1). Papers IV and V are an example of how this modelling framework can be used for health risk assessment. The first paper revises this model to include several improvements related to urban emission estimations, the dispersion module, treatment of the time-use of population, and infiltration coefficients from outdoor to indoor air. The revised model version can also be used for estimating intake fraction; which is then discussed in Paper V. A comparison between the original (Kousa et al. 2002) and the current version (Paper V) is shown in Table 3.1.

Table 3.1 A summary of the refinements of the EXPAND model.

<table>
<thead>
<tr>
<th></th>
<th>Original (Kousa et al., 2002)</th>
<th>Current (Paper IV, V)</th>
</tr>
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<tbody>
<tr>
<td>Emissions</td>
<td>Vehicular (exhaust), stationary</td>
<td>Vehicular (exhaust and suspension), shipping, stationary</td>
</tr>
<tr>
<td>Pollutants</td>
<td>NOx, NO2, ozone</td>
<td>NOx, NO2, ozone, PM2.5</td>
</tr>
<tr>
<td>Time activity data</td>
<td>Working age population, year 2000</td>
<td>All population, year 2010 wider range of activities</td>
</tr>
<tr>
<td>Model results</td>
<td>Population exposure, microenvironment-</td>
<td>Population exposure and intake fractions, microenvironment-, source-population group-specific</td>
</tr>
<tr>
<td></td>
<td>and source-specific</td>
<td></td>
</tr>
<tr>
<td>Coordinate systems</td>
<td>Finnish</td>
<td>international and national</td>
</tr>
</tbody>
</table>

The concentration in ambient air for vehicular traffic and shipping are obtained using CAR-FMI, and in case of stationary sources using UDM-FMI. Both models are described in Section 2.2. The emissions serving as input for the models are described in Section 4, and Papers IV and V.

The population activity data is available for four microenvironments: home, workplace, traffic and other activities (shops, restaurants, etc.). The dataset provides the total number, age distribution and geographic information on people living in a particular building, working at a particular workplace, located at a particular road, and spending time at other activities. The methodology for compiling the activity data is described in Papers IV and V.
All the data are interpolated into a rectangular grid defined by the user, then exposure or intake fraction are calculated for the same grid. MapInfo is subsequently utilized in the post-processing and visualization of the results.

4 Emission modelling

Estimating emissions is a key element for assessing air quality. Emissions are typically estimated based on models ranging from simple look-up tables to sophisticated and complex systems. In general, emission models apply methods that combine emissions factors with activity data to calculate emissions; both demanding a variety of parameters which have an impact on the resulting emissions. Emissions can be compiled considering emission totals, derived from aggregated data, then apportioned to smaller areas using proxy data (top-down approach), or using information on emission and activity of single sources to estimate total emissions (bottom-up approach).

Atmospheric models can be driven by a combination of emission inventories and dynamic emission modelling, to estimate the emission of gases and aerosols into the atmosphere. Emission inventories contain the emissions estimates for types of sources (point, area or line), together with complementary data such as location, emission height and sector. These inventories are typically defined for a given time period and with poor time-resolution. Dynamic emission modelling, on the other hand, can be applied to obtain a geographical distribution of individual events and its emissions, reflecting the temporal and spatial variation of the emissions more accurately.

In the sections below, the emission modelling systems for sea spray and wildland fire applied for mesoscale modelling (Paper I, II and II) are presented. Both sources depend on weather conditions and local factors. The dynamic modelling becomes inevitable for accurate assessment of aerosol load, air quality and health impact. Paper III also included the dynamic modelling of wind-blown desert dust and the MACCity emission dataset (Granier et al., 2011) for anthropogenic gases and aerosols. For the urban-scale modelling, Paper IV describes the emission databases compiled for PM$_{2.5}$ originated from traffic and energy production in Helsinki and in HMA. A short description of the methodology to generate those databases is presented in Section 4.3.

4.1 Sea salt aerosol for mesoscale applications

SSA originates from sea spray droplets resulting from waves breaking on the seawater surface, forming whitecaps and causing the entrainment of air into the water. The two main mechanisms responsible for sea spray formation are air bubble bursting during whitecap formation and decay, and direct tearing of droplets from the top of the breaking waves. Therefore, the formation of primary SSA is mainly dependent on wind speed: the emission of SSA is generally considered to be proportional to surface winds cubed (Monahan et al., 1986), suggesting that small changes in surface winds can have a substantial impact on the emission flux. Studies on marine aerosol size distribution (e.g. Covert et
al., 1998; Russell and Heintzenberg, 2001; Bates et al., 2002; Huebert et al., 2003) suggest that for high wind speeds, the production of very coarse SSA ($D_p > 20$ mm) increases. Other parameters influencing the formation of primary SSA have been identified. Laboratory studies by Mårtensson et al. (2003), hereafter referred as M03, and in situ measurements by Nilsson et al. (2007) show that for nano-sized particles, the aerosol number emission decrease with increasing seawater temperature, and for particles with $D_p > 100$ nm, the number SSA increase with increasing seawater temperature. These reflect different sea spray formation processes. Seawater salinity also affects the droplet formation, where formation of particles with $D_p < 0.2$ µm are not affected by salinity, but for larger $D_p$, salinity impact is substantial: higher salinity contributes to higher production (Mårtensson et al., 2003).

The majority of the CTMs have SSA source based on M03 parameterisation for formation of sub-micron aerosols and white-cap-area parameterisations of Monahan et al. (1986), hereafter referred as M86, for super-micron particles. The idea behind the new parameterisation described in Paper I, was to obtain a unified function that would cover such size ranges and would take into account the temperature and the salinity of seawater. Besides, the numerical implementation of M03 function showed a high sensitivity to the $6^{th}$ polynomial fit: $D_p$ should range between 0.02 to 2.8 µm of dry diameter, which results in the SSA flux shown in Figure 4.1 (left). If the size ranges are defined differently from the ones tabulated in that study, it can result in a very different outcome.

![Figure 4.1 SSA number flux density. Left: computed with M03 parameterisation for different seawater temperatures. Right: computed with Paper I parameterisation (red) and with M86 parameterisation (solid blue line) and extrapolated (dashed blue lines), M03 data (green dots) and its uncertainty (light-green dashed line) for $T_w=25^\circ$C, $S_w=33\%$, and SEAS data (brown).](image)

The M03 observations for a seawater surface temperature of 25 °C and a seawater salinity of 33 %, and the SEAS campaign (Clarke et al., 2006) were used to extrapolate the M86 function to particle sizes down to 20 nm. The M86 function was chosen to be extrapolated because it is widely used, which simplifies the comparison with other models. The function is monotonous and requires few
adjustments to fit into the M03 and SEAS data. All the data and functions used to obtain the SSA flux unified function are show in Figure 4.1 (right). The only significant correction refers to sub-0.1 µm particles, for which an exponential term had to be introduced to reduce the production under 0.03 µm. The resulting curve was then extrapolated from 8 to 10 µm to cover the whole target range of $D_p$ from 0.01 to 10 µm. The resulting sea salt flux function $(dF_p/dD_p)$ for particles with $D_p$ ranging from 0.01 to 10 µm, for seawater temperature of 25 °C and salinity of 33 ‰ is described below and is depicted in red in Figure 4.1 (right):

$$
\left(\frac{dF_p}{dD_p}\right)_{33‰,25°C} = 1 \cdot 10^6 \cdot \frac{\exp\left(-\frac{0.09}{D_p + 3 \cdot 10^{-3}}\right)}{2 + \exp\left(-\frac{5}{D_p}\right)} \cdot \frac{1 + 0.05 D_p^{0.05}}{D_p^{1.05}} \cdot 10 \cdot \frac{0.27 \cdot \lg D_p}{1.1}.
$$

(4.1)

To calculate the SSA production for other seawater temperatures and salinities, correction factors were derived from the M03 experimental data. The correction functions were derived by dividing the M03 observed fluxes for seawater temperatures at 15 °C, 5 °C and -2 °C by the fluxes observed at 25 °C. Figure 4.2 (left) shows that the ratio between the M03 production flux, at these three temperatures and the flux at 25 °C, are smooth and monotonically decreasing with increasing particle size. The correction function for the seawater temperature ($F_{TW}$) and salinities ($F_{SW}$) are described below, for $D_p$ in µm.

$$
F_{TW=15°C}(D_p) = 0.48 \cdot D_p^{-0.36}
$$

$$
F_{TW=5°C}(D_p) = 0.15 \cdot D_p^{-0.88}
$$

$$
F_{TW=-2°C}(D_p) = 0.092 \cdot D_p^{-0.96}
$$

$$
F_{SW=0.0092} = 0.12 \cdot D_p^{-0.71}
$$

$$
F_{SW=0} = 5.85 \cdot 10^{-5} \cdot D_p^{-1.7}
$$

(4.2)

(4.3)

For $D_p \sim 0.1-0.2$ µm and $D_p \sim 2$ µm a few outliers appear in two of the correction functions (shown in green and in light blue in Figure 4.2, left). These are related to non-monotonicity of the particle size distributions measured by M03 at different temperatures. Nevertheless, the correction factors for other temperatures seem to be easily obtained by linear interpolation, since water temperature dependency seems to be monotonic.

The effect of salinity is evaluated following the same procedure (Figure 4.2, right). The salinity of 33 ‰ is taken as the reference and for other salinities the correction functions are derived using the ratios of the M03 fluxes for 9.2 ‰ (e.g., Baltic Sea) and 0 ‰ (“fresh” water) to that at 33 ‰ (Atlantic).
Testing the formulation above has resulted in adjustments to the temperature correction function. The seawater temperature reference for the unified shape function is currently 20 °C, instead of 25 °C as referred in Paper I. The changes to the main function are presented in Paper III, section 2.3.4.

4.2 Wildland fires for mesoscale applications

Wildland fires also have strong regional and local contribution, quite often becoming the dominant source and strongly contributing to exceedances of the daily limit value established by air quality directives (Saarikoski et al, 2007). The impact of fire emission on the atmospheric composition depends on the dynamics of the fire and meteorology. The amount of emitted tracer is typically assumed to be proportional to the area affected by the fires (burnt area) and the empirical coefficients characterising the combustion process (Crutzen et al., 1979):

$$E_i = EF_i \times BA \times BD \times CF$$  \hspace{1cm} (4.4)

where $E_i$ (kg) is the total emission of the emitted specie $i$, $EF_i$ is the emission factor for the emitted specie $i$ (g kg$^{-1}$) dry matter burned, $BA$ is the size of the burned area (km$^2$), $BD$ is the biomass density (g kg$^{-1}$ km$^2$), and $CF$ is the combustion completeness factor reflecting combustion efficiency of the fires.

For large scale applications, emission factors are usually extrapolated from laboratory experiments or field campaigns. Apart from extrapolation errors, variables in Eq. (4.4) also inherit uncertainties, such as the spatial extent and duration of the fires; amount and distribution of available biomass or fuels; and fraction of biomass or fuel consumed from the different carbon reservoirs (French et al., 2004). Kaiser et al. (2012) shows that bottom-up approaches such as these, tend to underestimate PM.
emissions. The same study suggests that top-down approaches, based on active-fire remote-sensing observations, could be a better choice. Based on Kaufman et al. (1998) and Ichoku and Kaufman (2005) it is possible to relate the energy of the fire with the rate of biomass consumption and derive a relationship similar to Eq. (4.4), by relating the physical quantities of the biomass burned \((BA^*BD^*CF)\) with radiant component of the energy release of the fire. This energy release is the so-called fire radiative power.

\[ E_i = C_{ia} \cdot FRP \]  

(4.5)

where \(E_i\) (kg) is the total emission of the emitted specie \(i\), \(C_{ia}\) is the emission coefficient \((kg \, MJ^{-1})\) for specie \(i\) and vegetation type \(a\), and \(FRP\) is the fire radiative power \((MJ)\).

IS4FIRES is an operational, near-real-time assessment system for wildland fires based on the active-fire observation products of MODIS (http://modis.gsfc.nasa.gov, Justice et al., 2002; Kaufman et al., 1998) and SEVIRI (Kaiser et al., 2009; Roberts and Wooster, 2008). The system provides PM fire emission compiled from individual-fire FRPs registered daily by MODIS, via Equation 4.5. Temporal evolution of the fire intensity is derived from FRP observation from SEVIRI, for different vegetation classes (Sofiev et al., 2013). The development of the IS4FIRES system for wildland fire emission has been ongoing since 2006. To-date, IS4FIRES has two releases: v1 (Sofiev et al., 2009) and v2 (Paper II). The difference between the versions of IS4FIRES is described in the table 4.2.

For the FRP scaling, the emissions coefficients are obtained offline from the top-down calibration, which is performed once and involves the solution of the inverse dispersion problem for the fire smoke plumes. During the calibration step, SILAM (described in Section 2.1) is used to calculate the atmospheric dispersion of the emitted masses, thus producing both near-surface PM concentrations and aerosol optical density (AOD). The obtained plumes are attributed to the vegetation type prevailing at the location where the fire occurred, based on the Global Land Cover Characterization (GLCC) inventory (Loveland et al., 2000). The emission factor for each land-use type is obtained via fitting the modelled PM concentrations (only IS4FIRESv1) and AOD into the observed ones. The global distribution of AOD is provided by MODIS instruments on-board NASA satellites Aqua and Terra: Level-2 data from Collection 5 (before 2009) and 5.1 (Kaufman et al., 2002; Remer et al., 2008). IS4FIRESv1 calibration is described in Sofiev et al. (2009), Section 4. For IS4FIRESv2, the calibration is based on a long-term comparison (2002-2013) of remote sensing measurements and SILAM results. Both predicted and observed AOD data were projected to a global 1° x 1° grid, on an hourly basis. The MODIS-AOD pixels falling into the same grid cell were averaged; a minimum of 25 pixels per grid-cell were required to avoid biased AOD values. These two steps ensured the maximum possible co-location of the observations and model results, both in space and in time. The calibration used only the fire-dominated cells as predicted by SILAM: daily mean fire-induced AOD was requested to be bigger than combined AOD from all non-fire sources (sea-salt, wind-blown dust, primary anthropogenic and secondary inorganic aerosol). The observed AOD is then corrected by subtracting the non-fire SILAM-AOD. This correction is made under the assumption that fire-induced
AOD over fire-dominated pixels is the most-uncertain part of the total AOD predicted. The final step of the optimisation was to run an unconstrained minimisation of the root mean square error (RMSE) between the SILAM and MODIS AOD by adjusting the C_{ia} for each vegetation type. The optimisation is run independently for each year, maintaining the initial scaling factors as a starting point for the optimisation. A single scaling factor for each vegetation type is attained by averaging the values obtained from every optimisation run.

Upon obtaining the emission factors, the emission bottom-up estimation is performed, resulting in daily biomass-burning emission maps. The daily emissions are then scaled with diurnal variation profiles. Finally, the fire plume is given a prescribed height that can be static, prescribing always the same value or prescribing climatological fields (Sofiev et al., 2013), or dynamic, changing every-time step of the model (Sofiev et al., 2012a).

Elvidge et al. (2013) indicated that some sources, such as gas flares and large industrial installations, could be misinterpreted by MODIS as fires. An effort to mask-out these sources has been undertaken in IS4FIRESv2 by calculating the frequency of fires occurring in each 3*3 km² pixel over the globe, based on the MODIS-FRP. Grid-cells burning over 50 days per year, for at least 4 years over the 12-year period, would be flagged as possible highly energetic sources and removed from the FRP database.

Table 4.2 Comparison between previous (v1) and current (v2) versions of IS4FIRES.

<table>
<thead>
<tr>
<th></th>
<th>v1 (Sofiev et al., 2009)</th>
<th>v2 (Paper III)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Akagi et al (2011)</td>
</tr>
<tr>
<td>vegetation classes</td>
<td>forest, grass and mixed</td>
<td>boreal, temperate and tropical forests, residual crop, grass, shrub, peat</td>
</tr>
<tr>
<td>calibration period</td>
<td>fire cases per vegetation class</td>
<td>2002-2013</td>
</tr>
<tr>
<td>calibration data</td>
<td>remote sensing and ground-based measurements vs modelled AOD</td>
<td>remote sensing measurements vs modelled AOD</td>
</tr>
<tr>
<td>Plume injection height</td>
<td>Homogeneous from surface up to 1km</td>
<td>Parameterisation derived from MISR fire plume observations (Sofiev et al., 2012).</td>
</tr>
<tr>
<td>diurnal variation</td>
<td>night/day-time emissions = ± 50% (Saarikoski et al. 2007)</td>
<td>Parameterized for vegetation classes based on SEVIRI-FRP (Sofiev et al., 2013)</td>
</tr>
<tr>
<td>Masked data</td>
<td>-</td>
<td>High-energy sources</td>
</tr>
</tbody>
</table>

4.3 Anthropogenic emissions for urban scale applications

The most relevant PM anthropogenic emissions, for HMA, are traffic and energy production (Niemi et al, 2009). Most widely used methods for heating of residential buildings and domestic water were taken into consideration, with the exception of small-scale combustion, which mainly consists of wood burning. This source can contribute up to 25% of the national total emission (Karvosenoja et al., 2008) but the spatial distribution of the emission data was not known with sufficient accuracy. Energy
production and other industrial sources emissions were estimated by combining the activity data (Statistics Finland, 2012) and the emission factors (Karvosenoja et al., 2008) available for this sector.

Vehicular traffic emissions were estimated for each link of the HMA using the average speed-dependent functions, determined separately for each vehicle category (Laurikko et al., 2003), and the correspondent emission factors. The traffic information for each line source is obtained by EMME/2 (INRO, 1994). The emission factors are based on the European emission factors and taking into consideration the age distribution of the Finnish vehicle fleet (Laurikko et al., 2003; Kauhaniemi et al., 2011). Cold start and cold driving emissions were taken into account, using coefficients based on laboratory emission measurements (Laurikko, 1998). These emissions are dependent on the ambient air temperature and on the fraction of vehicles using pre-heating of the engine (Kauhaniemi et al., 2008).

Road dust emissions were estimated by FORE (Kauhaniemi et al., 2011). The emission factors for suspension of road dust are a product of the so-called reference emission factors, the reduction factor of the moisture content of the street, and a weighted sum of the contribution of particles from the wear of pavement and traction sand. Emissions from brake, tyre and clutch wear are not included in the model, due to their small contribution compared to suspension and road wear emissions in the Nordic countries. The baseline values for the suspension emission model were set by the reference emission factors that depend on the period (e.g. sanding or not), the mass fraction of particles, and the traffic environment.

Shipping emissions were estimated by STEAM (Jalkanen et al. 2009, 2012). The model combines vessel technical data with the positioning of ship, with a high spatial resolution (typically a few tens of metres), to obtain emission data. This model provides emissions from ships cruising, ships manoeuvring in harbour and while the ship is at berth.

5 Model applications and evaluation

5.1 Sea salt aerosol contribution to the atmospheric composition

SSA contributes from 30 to 75 % of the total production of natural aerosol (Lewis and Schwartz, 2004); with measurements showing SSA as the most important contributor to the total aerosol loading over the Oceans (e.g. Quinn et al., 1999). Understanding the magnitude and spatial variation of SSA is fundamental for assessing anthropogenic and continental impacts, to improve interpretation of satellite retrievals, and to understand the Earth’s radiation budget. This understanding has increased substantially, but process-based estimates of the total mass and size distribution of emitted sea spray particles continue to have large uncertainties (de Leeuw et al., 2011). CTMs and general circulation models (GCM) estimates of sea salt burden may vary over 2 orders of magnitude (Textor et al., 2006, Gantt et al., 2012; Grythe et al., 2014).
Paper I describes the development and evaluation of a new SSA parameterisation in order to unify data and existent parameterisations, available for different size ranges and seawater properties. The application of this parameterisation allowed the evaluation of the SSA emission flux and assessment of the SSA impact on the atmospheric composition. The emission module was evaluated by comparing results from simulations with observations: concentration, deposition and AOD. The simulations for this purpose are described below. In these papers, the SSA mass refers to the total mass of dry particles. If sodium (Na\textsuperscript{+}) concentrations are mentioned instead, it is assumed that Na\textsuperscript{+} mass fraction in PM is ~30\% (Seinfeld and Pandis, 2006).

- **Paper I:** European and Northern Atlantic runs for different time periods: May–October 2003 and years 2000, 2007, 2009 and 2010, driven by meteorological fields from ECMWF-IFS (ECMWF, 2015). The horizontal resolution was 30 km grid spacing and the vertical grid consists of 9 unevenly spaced layers, with the lowest layer being 50 m thick and the top reaching up to the tropopause.

- **Paper I:** Global runs for the years 2001 and 2008, driven by meteorological fields from ECMWF-IFS (ECMWF, 2015). The horizontal resolution was 1\(^\circ\) grid spacing and the vertical grid as described above was considered.

- **Paper II:** Europe and Northern Atlantic for a 20-year period (1990-2009), driven by the RCA3 meteorology (Samuelsson et al., 2011; Kjellstrom et al., 2011). RCA3 was driven by the global climate ECHAM5/MPIOM GCM (Roeckner et al., 2006) fields and by emissions from the SRES A1B scenario (Nakićenović, 2000). The horizontal resolution of 50*50 km\(^2\) and vertically similar to Paper I, but the lowest layer being 25 m thick.

The typical spatial distribution for this natural aerosol is depicted in Figure 5.1. Figure 5.1 shows the general pattern of SSA concentration following the emission areas (sea surfaces) with stronger winds and frequent storms. Global runs typically show higher concentration in the Southern Hemisphere, where winds are stronger, and in the equatorial belt due to higher seawater temperature. High resolution runs (Figure 5.1, right) show up to 10-times higher concentrations at open seas than at closed seas e.g. Atlantic vs Baltic Sea. These runs show that the European Seas also have concentrations gradients, e.g. at the Mediterranean Sea, where the model show pronounced maxima at the Balearic Sea and the Levantine Sea. In Figure 5.1, the transport of SSA over land is visible hundreds of km from the coast; near the European coast line it can contribute up to 6 µg m\(^{-3}\) to PM\(_{10}\), at annual level. With dry diameter lower than 1 µm, SSA can easily be transported for long distances in the atmosphere. This is confirmed by measurements campaigns with MARGA instrument (Makkonen et al., 2012), that revealed that PM measurements at continental sites with more than 200 km distance from the sea, such as Hyytiälä, have marine Na\textsuperscript{+} contribution. Figure 5.2 (left) shows the predicted contributions of the Atlantic and Baltic Sea on two measurements sites in Finland: Helsinki, close to the Baltic Sea; and Hyytiälä, an inland site (Figure 5.2, right). The results show that, even though the Atlantic Ocean is 700km away from the measurement sites, there is a contribution of the
Atlantic Ocean to these sites. Also, it is important to notice that the contribution is higher when concentrations are high. The latter is explained by the low salinity of the Baltic Sea: as seen in Figure 4.2, right, the amount of SSA produced is highly affected by salinity. But low salinity also allows the emissions of smaller particles that are likely to be transported for longer distances, reaching Hyytiälä measurement site. Table 5.1 shows how the model captured the concentration of Na⁺ in aerosol for the two sites. The model performs well for the coarse fraction but overestimates the fine fraction; correlation of daily time series for Na⁺ in PM₁₀ is good compared to PM₂.₅ daily time series.

Figure 5.1 SSA near-surface concentrations (µgPM₁₀ m⁻³) for simulations with the actual sea surface temperature. Left: annual mean over the year 2001; right: annual mean over the time period 1990-2009.

Figure 5.2 Left: predicted impact of Atlantic Ocean and Baltic Sea: Na⁺ concentrations (µg Na⁺ m⁻³) originated from Baltic Sea vs those originated from Atlantic Ocean for two measurements sites: Helsinki (pink) and Hyytiälä (blue). Right: location of the measurement sites: Helsinki (pink) and Hyytiälä (blue).
Table 5.1 Comparison of SILAM predictions of Na⁺ concentrations in PM_{2.5} and PM_{10} with observation at Hyytiälä (continental) and Helsinki (coastal).

<table>
<thead>
<tr>
<th>Sites in Finland</th>
<th>substance</th>
<th>Mean Observation</th>
<th>Mean Model</th>
<th>Correlation</th>
<th>RMS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hyytiälä</td>
<td>Na⁺ in PM_{2.5}</td>
<td>0.01</td>
<td>0.04</td>
<td>0.20</td>
<td>0.05</td>
</tr>
<tr>
<td>Hyytiälä</td>
<td>Na⁺ in PM_{10}</td>
<td>0.03</td>
<td>0.05</td>
<td>0.51</td>
<td>0.06</td>
</tr>
<tr>
<td>Helsinki</td>
<td>Na⁺ in PM_{2.5}</td>
<td>0.04</td>
<td>0.07</td>
<td>0.10</td>
<td>0.09</td>
</tr>
<tr>
<td>Helsinki</td>
<td>Na⁺ in PM_{10}</td>
<td>0.08</td>
<td>0.06</td>
<td>0.41</td>
<td>0.12</td>
</tr>
</tbody>
</table>

To evaluate the sea-salt size distribution inherent in these calculations, eight months in 2009 and 2010 were computed with high spectral resolution (18 bins over a $D_p$ size range from 0.01 to 10 µm range). These calculations were compared with a measurement campaign on a cruise, operating over the Northeast Atlantic, NEAT (O’Dowd et al., 1997), and with the work published by Pierce and Adams (2006). The time period selected for comparison between the model and the observations was different due to unavailability of meteorological data. The results of this comparison are shown in Figure 5.3.

Figure 5.3 Comparison of observed concentrations with predictions by SILAM (µg SSA m^{-3} m^{-1}). Top: observed mean under the NEAT campaign (blue dots) and predicted by SILAM (lines, monthly averages) SSA concentration spectra in the Northeast Atlantic, unit: (# µm^{-1} m^{-3}). Bottom: observed (black lines) published by Pierce and Adams (2006) and SILAM predicted (colour dots) SSA mass concentration over the globe (normalised with the mean diameter of the $D_p$ size range).

The first set of comparisons (Figure 5.3, top) show that the source function applied in SILAM has a close resemblance to the distribution showed by the NEAT campaigns. The closest representations is for the size range from 0.2 to 1 µm, where concentration for the current parameterisation peaks at
approximately 20 nm, then decreasing towards coarser particles. The decrease in concentrations below the 10 nm, contradicting the measurement results, can be a result of the removal of the aerosols due to water uptake or coagulation. The underestimation shown for sizes larger than 2 µm is harder to explain, and probably reflects specific local aerosol formation, such as at the surf-zone, where coarser particles are formed with wave breaking. The same pattern is seen in the work of Pierce and Adams (2006). SILAM’s predictions (Figure 5.3, top) shows that November tends to have higher production of SSA due to stronger storms, but coarser particles are emitted less due to low seawater temperature (Figure 4.2). This explains why fine-particle concentration was higher in the winter by few tens of percent, while the coarse-aerosol load remained constant.

Observations available from the EMEP network (Tørseth et al. 2012) were used to evaluate the SSA bulk mass (Figure 5.4). The observations consist of Na⁺ concentration in aerosol and ion analysis of precipitation including Na⁺. Typically, the surface concentrations are well reproduced by SILAM, the correlation coefficients for all the runs are higher than 0.5 and relatively low bias (0.14) and RMSE (0.71).

Figure 5.4 Model-measurement comparisons for monthly mean Na⁺ concentration (µg m⁻³) (left) and monthly wet deposition (mg m⁻²) (right), over the time period 1990-2009. The Pearson correlation (r), root mean square error (rmse), bias, standard deviation ration (stdRatio), p-value (p), 1:1 (red solid), 1:2 (green), and 2:1 (green) lines are shown.

The current model version underestimates wet deposition. This could mean that SSA is strongly scavenged at the source, reducing the availability of the aerosol at the measurement sites. Here is shown the comparison with full size range available (Dp from 0.01 to 30 µm) since the observed wet deposition does not cut-off the size of the particles, i.e. SSA coarser than 10 µm is accounted for, including the SSA produced in the surf zone. Considering the full size range strongly reduced the bias (400 times), correlation improved 1.5 times, and RMSE became slightly smaller. This mostly explains the large negative bias of the models, when reporting PM₁₀ only, and, to some extent, the low
correlation. In summer, the scores are slightly better than in winter, but the absolute values and importance of this removal process is smaller in summer time. Nevertheless, wet deposition is harder to evaluate since it depends highly on the quality of the precipitation fields.

The predictions of AOD, due to the presence of SSA in the atmosphere, were evaluated by comparing SILAM’s results with satellite observations. The AOD retrievals of MODIS instruments on-board NASA satellites Aqua and Terra (Collection 5, level-2 data), were first pre-processed to remove cloud contaminated pixels (Remer et al., 2008). Then the pixels were projected to the SILAM grid, hourly aggregated, and collocated in time and by grid-cell with the model results. Co-locating the datasets, however, is not enough: the MODIS AOD includes the contribution of aerosols of all types and origin, while the SILAM computations included only SSA. Hence the comparison was restricted to the areas where the SSA load is dominant, i.e. where the AOD is least influenced by non-SSA. These are the oceanic regions at mid-latitudes in the Southern Hemisphere, central part of the North Atlantic and North-Eastern part of the Pacific Ocean (Figure 5.5, black squares). Figure 5.5 shows that AOD predictions are on the same order of magnitude as MODIS, if only the SSA-dominated regions described above are considered. The results also show the regions least affected by the contribution from terrestrial aerosol sources: the Southern and Northern Pacific Oceans. The impact of non-SSA aerosol on AOD over the Northern Atlantic, parts of the Southern Atlantic and the Indian Oceans is noticeable bigger. Both of these findings are confirmed when comparing MODIS and SILAM AOD histograms for the Southern Pacific (area A, in Figure 5.5) and Southern Atlantic and Indian Ocean (area B, in Figure 5.5.) areas. The histograms for SILAM and MODIS for the Southern Pacific area are very similar and the mean-AOD differs by less than 2% (Figure 5.6, top), showing that it is a SSA-dominated area. The same histograms for Southern Atlantic and Indian Ocean show a shift of the predicted SSA AOD to the lower AOD values as compared to MODIS (Figure 5.6, bottom), indicating that other sources are influencing the observations.

Figure 5.5 Spatial distribution for predicted SSA-AOD by SILAM (left) and by AOD observed by MODIS (right).
Figure 5.6 AOD histograms for Southern Pacific (area A in Figure 5.5) and Southern Atlantic and Indian Ocean (area B in Figure 5.5) areas. Top: collocated SILAM (red), un-colloca ted SILAM (green), MODIS (blue), and fraction of SILAM cells observed by MODIS (black line, right-hand axis). Bottom: collocated SILAM (red) and MODIS (blue) data.

Figure 5.7 AOD histograms for SSA-dominated regions for the 2001 simulations, co-located SILAM and MODIS sets, fixed seawater temperature at 15 °C for Southern Pacific area (area A in Figure 5.5).

The model evaluation showed that the SSA concentration and AOD are reproduced well, especially for the summer period. In winter time, the wind-speed is higher and more prone for coarse particles production. The coarser particles are typically underestimated since the parameterisation is not accounting for spume production. Adding spume droplet formation, for winds higher than 6 m s⁻¹ based on the formulation of Andreas (1998), brings the results closer to the ground-based measurements – and further increases the total SSA load.
Finally, predictions of SSA were compared between SILAM and other CTMs widely used in Europe: DEHM, EMEP and MATCH (Paper II). Formally, these models use M86 and M03 parameterisations and differences in SSA emission should be attributed to the temperature and salinity dependencies. To scrutinize this, box-model calculations of the SSA mass flux as a function of temperature were made for seawater salinity of 10 and 35 ‰, representing the Baltic Sea and the Atlantic Ocean, respectively, and with wind-speed fixed at 15 m s\(^{-1}\) (Figure 5.8, left).

![Figure 5.8 SSA mass flux (gPM\(_{10}\) m\(^{-2}\) s\(^{-1}\)) box calculations (left) and coarse mode fraction of the mass flux (right) as a function of radius (dry for DEHM and SILAM and RH = 80 % for MATCH) and temperature, for wind speed 15 m s\(^{-1}\) and salinities 10 ‰ and 35 ‰.](image)

In general, the SSA flux can be substantially different between models, for the same seawater properties. All the models show an increase of mass flux of SSA with temperature and salinity, except EMEP that does not apply any correction for salinity. Both DEHM and EMEP mass fluxes show little difference between low and high temperatures; SILAM and MATCH show a substantial dependency of the mass flux on temperature throughout the size ranges. This difference is explained by how the seawater temperature dependency is implemented in each model. In DEHM and EMEP, only PM\(_{2.5}\) size range varies with seawater temperature, based on the M03 source function. SILAM has the dependency for all size ranges based on the formulation described in section 4.1 and Paper I. In MATCH, the implementation of seawater temperature correction is done by combining the temperature correction included in the M03 for size-range below \(D_p = 0.4 \mu m\) and the use of the temperature corrections from Paper I for coarser sizes.

Size distribution will affect production of SSA and will possibly affect the removal of SSA from the atmosphere. Figure 5.8 (right) shows how the different models distribute the mass between fine (PM\(_{2.5}\)) and coarse (PM\(_{5.10}\)) modes. Again there is a discrepancy between the models. Both DEHM and EMEP assume that the contribution of the coarser mode is reduced with temperature, since more SSA is produced with higher temperatures, for size ranges below 2.5 μm. Temperature-wise, EMEP has always the highest contribution for the coarse mode. For MATCH and SILAM, the contribution to
the coarser mode increases with temperature, though MATCH has a lower coarse mode contribution than SILAM. There is an agreement between the DEHM, MATCH and SILAM for very saline water, where coarser particles are emitted.

For more insight about the unified parameterisation for SSA flux, the reader is referred to Tsyro et al (2011), Grythe et al. (2014), Liora et al. (2015), and Witek et al (2016), where the parameterisation has been used and evaluated.

5.2 Sea salt aerosol and climate change: a European perspective

Roughly 70 % of the Earth’s surface is covered by water. Thus, SSA might play a major role for atmospheric processes that govern climate change, especially over remote water surfaces where no other source of aerosols exists. SSA can contribute to changes in cloud albedo and precipitation, since it can lead to cloud formation, and serves as a sink for condensable gases and smaller aerosol particles, changing the size distribution of the marine aerosol (e.g. Korhonen et al., 2010; Wang et al., 2011).

Currently, there is an improved understanding regarding the impact of aerosol on global climate than on regional scale (Vogel et al., 2009), with very few studies regarding the European region (Lundgren et al., 2013). The study presented in Paper II fills-in the knowledge gap that currently exists, by studying the climate impact on SSA production and fate over a 40 year period (1990-2009 and 2040-2059) over the European region. The paper also includes considerations about the radiative impact of SSA over sea surface and land. The runs are described in Section 5.1.

This study indicates that, in the future, there will be more emissions of SSA due to changes in wind speed and temperature (salinity was kept constant), see Figure 5.9. For instance, it is expected an increase of emissions over the east of Iceland where temperature is predicted to rise by almost 2 K, and over the Black and Aegean Seas, due to an increase of seawater temperature, supported by higher wind speed. The smallest absolute difference between future and past is predicted for the Baltic Sea; this was expected due to its low seawater salinity. But in relative terms, the model shows an increase up to 20% in Gulf of Bothnia, which is actually higher than the increase predicted for North Sea (5-15%), due to the temperature rise. Therefore, the increase or decrease of SSA emissions needs to be carefully analysed.

SSA directly scatters solar radiation back to space, resulting in a cooling effect on climate by decreasing the amount of radiation absorbed by the water surface. Over land, there can be both cooling over the low-reflectance surfaces, and warming over high-albedo surfaces (e.g., Haywood and Boucher, 2000). The direct radiative effect (DRE) due to SSA is based on the AOD predicted by SILAM for past and future periods. The past period simulation estimates the upward scattering by SSA, at TOA, to be up to 0.5 W m\(^{-2}\) over seawater surfaces. This value is within the estimates on upward scattering of radiation by SSA: ranging between 0.08 and 6 W m\(^{-2}\), at wavelengths in the range of 0.3-4 µm (Lewis and Schwartz, 2004). The predicted change in the DRE due to SSA suggests an
overall cooling (negative change) in the future for the Northern and warming for the Southern latitudes. The Mediterranean region is the most sensitive area in this study, where an overall warming was predicted both over sea and over land, and cooling is predicted to happen over the eastern basin of the Mediterranean Sea. The DRE pattern for the whole year is highly influenced by the summer period. The upward scattering in the summer time can be up to 1.7 times higher than in winter (Figure 5.10, left), due to lower cloudiness and prolonged daylight. This can be seen in Figure 5.10 (right), which shows the change between future and past periods but considering only the summer months (June, July and August). This study predicts a substantial seasonal variation for the DRE over the sea surface waters.

Figure 5.9 Absolute change between future (2040-2059) and past (1990-2009) periods. Top-left: wind speed (m s\(^{-1}\)), top-right: wind forcing (≈ U\(_{10}^{3.41}\)), bottom-left: seawater temperature (K), and bottom-right: SSA emissions (mgPM\(_{10}\) m\(^{-2}\)).
5.3 Fire emission estimation and contribution to atmospheric composition

Wildland fires have been recognised among the most-powerful sources of atmospheric tracers and particles, including precursors for secondary pollutants (Lamarque et al., 2010). Annual estimations of the globally consumed biomass usually range between 5 and 10 Gt (Scholes and Andreae, 2000). However, the estimates of fire emissions are highly uncertain, even if large-scale and long-term averages are considered: inaccuracies of input data and variations in the methodologies employed may lead to an uncertainty in emission estimates of at least 50% (French et al., 2004; Kasischke and Penner, 2004; Schultz et al., 2008).

Estimations for wildland fire emissions for total PM by IS4FIRES are currently on the higher end of the estimations, being at least a factor of 5 higher from other top-down approaches (van der Werf et al., 2010; Kaiser et al., 2012). Figure 5.11 shows the spatial distribution of PM emissions for IS4FIRESv1 and GFASv1 (Kaiser et al., 2012).

Considering the overestimation of the PM emissions by IS4FIRESv1, improvements to the system were undertaken. Section 4.2 and Paper III describe the implementation and optimization of the IS4FIRESv2 and the evaluation of the previous and the current version of the system. A global evaluation of the system was based on AOD. The evaluation considered MODIS AOD at 500 nm (Collection 5, level-2) and AOD predicted by SILAM simulations including primary and secondary inorganic gases and particulates. The simulations for this purpose included sea spray, wildland fire, wind-blown dust and anthropogenic emissions (described in Section 4). The chemical transformation module for secondary inorganic aerosol formation (Sofiev, 2000) was applied together with a bulk mass aerosol dynamics scheme that includes gas phase and heterogeneous oxidation of $SO_2$ to $SO_4$, dynamic equilibrium between $NH_4NO_3$ aerosol and $NH_3$ and $HNO_3$ in gas phase, and condensation of $HNO_3$ to the surface of the sea salt particles. The model was driven by ERA-Interim (Dee et al. 2011)
meteorological data with 3-hour temporal resolution and 0.72 ° horizontal resolution. The global simulations were set for a period between 2003 and 2012, with a horizontal resolution of 1*1 ° and the vertical grid consisting of 9 unevenly spaced layers, with the lowest layer being 25 m thick and the top reaching up to the tropopause. All simulations were made with a 15 min internal time-step, while the model results are provided as hourly averages. Observed and predicted AOD were spatially collocated, on an hourly basis, then averaged monthly and annually.

When comparing predicted and observed AOD in an annual level, only the mean emission factor for “typical” fires will be obtained. Nevertheless such comparison revealed how the refinement of the IS4FIRESv1 has a significant impact on the AOD estimations. Increasing the number of land-use types from three (IS4FIRESv1) to seven (IS4FIRESv2) improved the prediction scores by reducing the overestimation of the system, and improving mean values and RMSE (Figure 5.12).

Expectedly, the difference is not particularly substantial in Eurasia, since IS4FIRESv1 was initially calibrated for Eurasian fires. The substantial improvements are shown for pro-fire areas such as the African continent: RMSE is reduced by half if the number of land-use types is increased. Further 10% of AOD reduction is obtained by eliminating the misattribution of some specific GLCC land-uses to the selected classes.
Figure 5.12 Histograms (top), cumulative distribution (bottom-left) and statistical parameters (median, mean, RMSE and R, bottom-right) for fire dominating cells (daily averages) for Eurasia and Africa, in August 2008: AOD predicted (MODIS) and computed by SILAM with fire emissions estimated by IS4FIRESv1 (SILAM-IS4FIRESv1) and IS4FIRESv2 (SILAM-IS4FIRESv2).

There are clear cases of MODIS pixels that are persistently reported as fires (Figure 5.13, red crosses), in particular in areas such as Arabian Peninsula. These pixels are currently flagged as highly-energetic sources. The impact of such sources has been simulated with SILAM for August of 2010.

Figure 5.13 Relative AOD reduction due to masking out non-fire pixels (red crosses) from the MODIS database.

Simulations with and without these sources show a substantial impact on AOD values; the fractional difference is shown in Figure 5.13. When highly-energetic sources, such as oil extraction/production plants, are masked-out from the fire emission database, AOD can be reduced by ~80 % in the immediate vicinity of these sources but the effect quickly fades out when the distance from the sources increases. The most-significant difference was predicted for equatorial regions and part of Sahel,
where there is an accumulation of atmospheric aerosol in the Intertropical convergence zone, converging the fire plumes.

5.4 Human exposure to PM$_{2.5}$ in the Helsinki Metropolitan Area

Particulate matter mass concentration has been associated with hospital admissions and mortality for decades (e.g. Pope et al., 1995; Dominici et al., 2006; Kioumourtzoglou et. al., 2016). The World Health Organization suggested that for health effects, the fine fraction is more relevant than PM$_{10}$ and recommends the regulation of PM$_{2.5}$ instead of PM$_{10}$ (WHO, 2006). PM$_{10}$ is less suitable for exposure-response relationships assessment because it includes particles that cannot be inhaled. A comprehensive study on PM phenomenology in Europe was compiled (Putaud et al., 2010). This study shows that there is a clear difference throughout Europe regarding the physical and chemical characteristics of PM, and also when moving from kerbside to rural sites. Helsinki, per se, can be substantially different from other European cities. In Helsinki, the total particle concentration is very low compared to other sites with the same characteristics (e.g. kerbside) and the diurnal variation of local vehicular traffic flows is not always correlated with the PM$_{2.5}$ concentrations (e.g. Pohjola et al., 2002). Karppinen et al. (2005) estimated the contribution from regional and long range transported origins to be less than 50% in Helsinki centre (and nearly 100% in the outskirts of the HMA). This implies that when assessing urban scale atmospheric composition and, consequently, human exposure to PM$_{2.5}$ in HMA, not only local sources should be carefully considered but also accurate estimations for regional background concentrations.

An example of such studies is described in Paper IV. This study assesses the human exposure to PM$_{2.5}$ in the HMA and Helsinki for the years 2008 and 2009, respectively. The HMA comprises four cities; Helsinki, Espoo, Vantaa and Kauniainen. The total population in the HMA is approximately 1.1 million, while the population of Helsinki is about 0.6 million inhabitants. The integrated system EXPAND (Section 3.1) was chosen to perform such assessments. In order to include all the possible contributors to PM$_{2.5}$ surface concentrations, approximately 5000 line sources for vehicular traffic and shipping, and 40 stationary sources (power plants and industrial facilities) were considered. The sources are described in section 4.2.3. Background concentrations where estimated with the LOTOS-EUROS model (Schaap et al., 2008) to include the contribution from regional and long range transport. This contribution is derived from the grid cell including the regional background station Luukki, where the influence of local sources on PM$_{2.5}$ concentrations has been estimated to be on average less than 10 % (Karppinen et al., 2005). Since the exposure estimations included indoor and outdoor activities, an infiltration ratio of 0.57 for home and work microenvironments was applied; both traffic and other activities microenvironments considered that the population is fully exposed to the PM$_{2.5}$ levels (Hänninen et al., 2004, 2005 and 2013). Hourly concentration values were averaged over the whole year of 2008 and 2009. Both concentration and activity data were interpolated to a 50x50 m$^2$ grid-cell. Though the study for Helsinki represents a smaller area and a different year, it
should still be valid to evaluate how different sources impact the urban atmospheric composition and human exposure in HMA in 2008.

The results for 2008 show that the regional background is the main contributor to PM$_{2.5}$ concentrations in HMA, confirming the previous studies. Figure 5.14. (left) shows the predicted concentrations for vehicular emissions and background contribution for the HMA in 2008. The overall area is dominated by background concentrations, with a maximum of 7 µgPM$_{2.5}$ m$^{-3}$, with highest concentrations being located in the vicinity of the main roads and streets, and in the centre of Helsinki. Figure 5.14 (right) shows the contribution of anthropogenic sources to the PM$_{2.5}$ emissions in HMA, where vehicular traffic is the most relevant contributor for PM$_{2.5}$ emissions in HMA. The contribution from shipping is as relevant as stationary sources. How these concentrations reflect on human exposure in HMA is show in Figure 5.15 (left and right).

![Figure 5.14](image)

**Figure 5.14** Left: predicted annual average concentrations of PM$_{2.5}$ (µg m$^{-3}$), contribution from vehicular traffic and background for the HMA in 2008. Right: Source contribution to PM$_{2.5}$ emissions for Helsinki in 2009.

![Figure 5.15](image)

**Figure 5.15** Contribution of different sources of PM$_{2.5}$ to exposure (left), and of time-activity (middle) and human exposure in each microenvironment (right).

When exposure is analysed source-wise, it is clear that the background contribution is the most relevant, with vehicular traffic being the second contributor but only with a meagre 12 %. These
results relate to the concentration results discussed above. The exposure originated from major stationary sources is negligible, caused by the dispersion of pollutants to wide regions due to the high stacks for most of these installations. Although the average contribution of shipping to the total PM$_{2.5}$ concentrations within the area considered was a modest 3%, this contribution can be higher than 20% in the vicinity of the harbours (within a distance of approximately one kilometre). Figure 5.16 shows how the population exposure to vehicular traffic and shipping emissions can be different due to the spatial distribution of concentrations, which is linked with the predominant sources.

![Figure 5.16 Population exposure per year (µg m$^{-3}$ * # persons) to PM$_{2.5}$ in Helsinki in 2009 to emissions from vehicular traffic (right) and emissions from shipping (left).](image)

The microenvironmental population exposure analysis showed that exposure at home is responsible for most of the population’s exposure to PM$_{2.5}$, followed by work and other activities. Overall, people are less exposed while commuting, because it is where they spent less time (see activity data, Figure 5.15, middle-panel). Home, work and other activities microenvironments show similar patterns, indicating that there are no major relative differences in the average concentrations prevailing at those microenvironments. However, for traffic, the contribution to exposure is substantially higher than the corresponding contribution to time-activity. This is mainly caused by the relatively higher concentrations on the roads and their vicinity.
The spatial distribution of the predicted annual average population exposures in the HMA, in 2008, for home and work microenvironments, is shown in Figure 5.17. These distributions exhibit characteristics of both spatial concentration and time-activities distributions. There are elevated values in the Helsinki city centre, along major roads and streets, and in the vicinity of urban district centres. The high exposure values at home and work in the centre of Helsinki are caused both by relatively high concentrations and high population densities in the area.

![Figure 5.17 Annual average population exposure (µg m⁻³ * # persons) for 2008: home (left) and work (right) microenvironments.](image)

The exposure computations could not be evaluated at the time of this study, since there was no PM$_{2.5}$ personal exposure measurements available. Comparison between modelled personal exposure and monitored personal exposure under the ESCAPE campaign in Finland (Beelen et al., 2015) is currently ongoing at FMI. Paper V describes possible methods to evaluate exposure metrics through box model calculations and personal monitoring data.
6 Conclusions

The main focus of this thesis was to improve the modelling of aerosol emissions and its atmospheric dispersion at different spatial and temporal scales, and to integrate the dispersion modelling with the population activity data to obtain accurate exposure metrics. Emission modelling of SSA and wildland fires in the SILAM model was refined by combining and re-assessing widely used formulations and datasets. Both emission models were thoroughly evaluated with ground-based and remote sensing observations, for several years. Emissions for HMA were revised to bring up-to-date the emissions for traffic and energy sectors in use in urban-scale modelling. The EXPAND model, was revised and applied to bring concentrations and activity data together to compute parameters such as population exposure or intake fraction.

A new parameterisation for bubble-mediated SSA emissions has been developed. It takes into account the effects of wind speed and seawater salinity and temperature, and can be applicable to SSA particles with dry diameters raging between 0.01 and 10 µm. The parameterization is valid for low-to-moderate wind speed, seawater salinity ranging between 0 and 33 ‰ and seawater temperature ranging between -2 and 25 °C. The estimation for SSA emissions with this unified function, 6700-7400 Tg yr\(^{-1}\), are within the range of other global estimates (1000-20000 Tg yr\(^{-1}\), Schulz et al., 2009; Textor et al., 2006). The spatial distribution of SSA has typically higher concentrations where winds are stronger (Southern Hemisphere) and temperatures are high (equatorial belt). Contribution from oceans can be up to 10-times higher than other seas, e.g. the Atlantic vs Baltic Sea, due to higher salinity and stronger wind-gusts. European Seas also have pronounced concentrations gradients, e.g. at the Mediterranean Sea. Transport of SSA over land is significant, even for low salinity waters such as the Baltic Sea, and can contribute up to 6 µg m\(^{-3}\) to PM\(_{10}\), at annual level. Wet deposition is more challenging overall, since it relies heavily on the meteorology driving the models. Nevertheless, it is recommended to use full size range available for SSA, to be comparable with the observations.

The first climate impact study related to SSA over Europe was done by applying the unified SSA flux parameterisations. The European climate-change context shows that the difference between the current and future climatic conditions (1990-2009 vs 2040-2059) is driven by the trends in seawater temperature, as the near-surface wind speed is projected to stay nearly the same in the climate scenario used. These results are more accentuated over the Black and the Mediterranean Sea, where the increase of emission and concentration of SSA is significant. Simple calculations for the possible impact of SSA on the DRE show that the North of Iceland, the Norwegian and Baltic Seas are the most affected areas by cooling, and the Mediterranean Area (over land and sea) is mainly affected by warming. The prediction for the upward scattering by SSA, at TOA, can be up to 0.5 W m\(^{-2}\).

The methodology for estimating wildland fire PM emissions under IS4FIRES, shows similar spatial distribution when compared to other top-down methodologies, but can differ by an order of magnitude from the lowest estimation. This methodology shows that fires are very specific concerning vegetation and meteorology, reflecting the local conditions of where the fire is taking place. For this reason, the
number of vegetation types was increased, the diurnal variation per vegetation type was improved, and the emission model was re-calibrated. This reduces the overestimation substantially, especially for fire prone areas such as Africa (~50%). Additionally, FRP retrieved from MODIS should be carefully scrutinized so that the information coming to fire emission models will not take pixels with misleading fire information. Currently, MODIS-FRP classifies highly energetic sources as fires. This misattribution can cause an overestimation in AOD up to 80%, especially close to the misattributed sources. The estimation of fires based on the FRP data should be regularly calibrated with up-to-date remote sensing observations.

The description of regional sources and background contribution is crucial when assessing PM$_{2.5}$ concentrations. According to the study performed, background contribution is the most relevant to the PM$_{2.5}$ concentration in HMA. Traffic is the local source that contributes the most to the PM$_{2.5}$ concentrations (11%). Although the average contribution of shipping to the total PM$_{2.5}$ concentrations was modest (3%), this contribution can be higher in the vicinity of the harbours. Energy production does not contribute substantially, since the high stacks allow the dispersion of the emitted PM for longer distances and do not impact the study area. The outcome of this study shows that the population exposure in HMA mostly happens while at home (60%), where people spend most of their time. The lowest exposure will happen while driving or commuting (4%) but comparing the activity data with the exposure results shows that exposure will be more acute while at traffic. Background concentration was responsible the major fraction of the total exposure (86%), followed by vehicular emissions (12%). The shipping did not contribute substantially in the whole HMA (2%) but in harbour areas and their vicinity the contribution can be as high as 20%. There was no impact of stationary sources on human exposure in the study area.

7 Future of the research field

This study highlights several bottlenecks for emission modelling and what needs to be improved. The SSA parameterisation of SILAM showed high potential for integrated approaches bringing together major factors controlling emission, but also revealed a substantial lack of experimental data describing formation of e.g. coarse particles, strong wind speeds, spume drop formation mechanism, etc. Additionally, surf-zone processes are severely underrepresented. Studies such as Albert et al. (2015), based on satellite observations, claim that the whitecap-area based parameterisation typically used by CTMs is misrepresenting the absolute values, mostly underestimating the production of SSA. Therefore, parameterisations based on Monahan et al. (1986) should be reformulated.

DMS is the dominant volatile biogenic sulphur compound emanating from the ocean (Simó, 2001) and is typically oxidized to form sulfuric and methanesulfonic acids, which contribute to new particle formation and growth, affecting the radiation budget of the Earth (Liss and Duce, 1997). This natural source of aerosol is currently missing or being poorly represented in many CTMs. Global mean surface DMS concentration is quite robust because of the large data set used, but the estimates for


specific regions and seasons remain highly uncertain in many ocean regions where sampling has been sparse (Lana et al, 2011).

Representation of the wildland fire behaviour and emission for regional-to-global scales is still too rudimentary in current available CTMs. A better representation is needed to characterise the fire phase (flaming or smouldering) and relating it to a weighted mass spectrum derived from field studies such as Virkkula et al. (2014), Janhäll et al. (2010), and Chubarova et al. (2012). Representation of the fire relies on the type of vegetation burning, the analysis performed in this study shows that the current emission factors can hardly be used for all the regions with similar land-use type over the globe, and regionalised approaches are needed. More research is needed to identify major factors that influence seasonal and interannual variability in fire occurrence, for different ecosystems, to map the temporal and spatial extent of fires. Hao and Larkin (2014) describes the considerable recent progress in this area, but fires are not easily mapped by satellite-based sensors because they are typically of small size and duration and burn beneath forest canopy. On the other hand, the same sensors are detecting highly-energetic sources, misleading the fire emission estimates.

Small scale combustion is a ubiquitous source during the winter all over Europe, but these emissions have been severely misrepresented in the current inventories (Stohl et al., 2013) and, consequently, in dispersion modelling. As an example, domestic wood combustion in Finland is estimated to contribute around 23% to the PM$_{2.5}$ emitted (Karvosenoja, 2008).

A new challenge for human exposure modelling is the generalization of the approach and upscaling to regional scale. A significant problem for models such as EXPAND, is the lack of activity data of population. More should be done to create generic proxies for the population movements between microenvironments. This could result in an easier link between the dispersion models, such as SILAM, and exposure tools, such as EXPAND, which could be directly used to forecast and assess air quality together with health risk assessment.

There are very few studies that include impact of future air quality directives or climate on human exposure in regional or local scale. Future scenarios for emission and activity data should be compiled to obtain possible human exposure in the near future or climate impact on human exposure.

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