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TITLE: Spatial and temporal disaggregation of emissions for the past decade (2000-2010)

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Coordinated by:

Technical University of Crete

D11. Spatial and Temporal Disaggregation of emissions for the past decade (2000-2010) – Anthropogenic and Natural Emissions

EXECUTIVE SUMMARY

In this work, the emissions of the three major anthropogenic sources, road traffic, industry and residential/institutional/commercial activities, are temporally and spatially disaggregated. Spatial disaggregation for navigation, aviation and railways emissions was not performed as these emissions are located in a specific area. For road transportation emissions, time disaggregation was based on previous works and information collected from transportation experts. The methodology applied to disaggregate spatially road traffic emissions was based on methodologies followed in similar jobs. For industrial emissions, with the aid of data available, it was considered that big installations work on a 24 hour basis whereas the smaller installations work on a 12 hour basis. The spatial allocation of the diffuse industrial emissions data was based on the E-PRTR database and CORINE land uses, in order to allocate diffuse emissions. Point emission sources were allocated according to their coordinates reported under the Convention on Long-range Transboundary Air Pollution (CLRTAP, 2010). Residential and commercial emissions are distributed equally in the course of the day from 7:00 to 21:00. The methodology for the spatial distribution of emissions from residential and commercial activities was based on population and land use data for each grid cell. Aviation emissions were disaggregated in time taking into account statistical data provided by the Hellenic Civil Aviation Agency. As for navigation, because of different activities taking place in the port area, no detailed data are available. Time disaggregation was accomplished taking into account statistical data for passenger ships movements daily variation and given that commercial shipping activities take place through the whole day. Emission inventories for particulate matter ($PM_{2.5}$ and $PM_{2.5-10}$) from natural sources were also created for the Athens and Thessaloniki metropolitan areas (AMA and TMA respectively) and the greater area of Volos (GVA) for the period 2000 - 2010. The inventories include primary windblown dust (WB) emissions from agricultural and vacant lands and primary sea salt particles emissions from the breaking of waves at the Sea Shore-surf zone (SS_SS) and the bursting of bubbles from oceanic whitecaps - Open Ocean (SS_OO). Additionally, emissions of BVOCs (Biogenic Volatile Organic Compounds), precursor to PM, are included. The objective of this work was to study the seasonal variation of natural PM emissions in the areas of interest and their spatial allocation during the period 2000-2010. Weighting factors, specific to each pollutant, source and area for the period 2000-2010, were calculated for the disaggregation of annual emissions to monthly and daily values and their allocation on high resolution grids. There was no significant seasonal variation observed in the natural PM emissions while BVOCs emissions were increased during the warm period due to the enhanced solar radiation and temperature. Moreover, the analysis focused on the seasonal

variation of natural sources' contribution to total emissions over the areas during the period 2000-2010. Therefore, the constructed emission inventories for $PM_{2.5}$, $PM_{2.5-10}$ and BVOCs from natural sources were spatially and temporally disaggregated and compared to anthropogenic emissions in the areas. There was no significant seasonal variation observed in the contribution of natural sources to total PM emissions.

ΠΕΡΙΛΗΨΗ

Στα πλαίσια αυτής της εργασίας, οι εκπομπές από τις κύριες ανθρωπογενείς πηγές (οδικές μεταφορές, βιομηχανία και οικιακό/εμπορικό τομέα) κατανεμήθηκαν χρονικά και χωρικά έτσι ώστε να έχουν ανάλυση χρονική ανάλυση 1 ώρας και χωρική ανάλυση 1X1km^2. Χωρική κατανομή των εκπομπών από τις θαλάσσιες, εναέριες και σιδηροδρομικές μεταφορές δεν έλαβε χώρα, δεδομένου ότι οι εκπομπές αυτές εκτείνονται σε συγκεκριμένη περιοχή. Η χωρική ανάλυση των οδικών μεταφορών βασίσθηκε σε προηγούμενες μελέτες και σε πληροφορία που συλλέχθηκε από ειδικούς σε θέματα μεταφορών. Η μεθοδολογία που ακολουθήθηκε για την χωρική ανάλυση των εκπομπών οδικής κυκλοφορίας βασίσθηκε σε μεθοδολογίες που εφαρμόσθηκαν και δημοσιεύθηκαν στα πλαίσια παρόμοιων μελετών. Οι βιομηχανικές εκπομπές αναλύθηκαν χρονικά με βάση στατιστικά στοιχεία για την λειτουργία τους. Η χωρική ανάλυση των εκπομπών επιφανειακών πηγών έγινε βάσει της μεθοδολογίας E-PRTR και της βάσης δεδομένων χρήσεων γης CORINE. Οι σημειακές πηγές τοποθετήθηκαν χωρικά σύμφωνα με τις γεωγραφικές τους συντεταγμένες. Οι εκπομπές οικιακού και εμπορικού τομέα κατανέμονται κατά τους μήνες λειτουργίας της θέρμανσης και κατά τις ώρες 7:00-21:00. Η χωρική κατανομή των εκπομπών στηρίζεται σε πληθυσμιακά στοιχεία και στοιχεία χρήσεων γης. Οι αεροπορικές εκπομπές αναλύθηκαν χωρικά βάσει στατιστικών στοιχείων κινήσεων από την Υπηρεσία Πολιτικής Αεροπορίας, ενώ ανάλογα αναλύθηκαν και οι εκπομπές από τις θαλάσσιες μεταφορές. Μητρώα εκπομπών σωματιδίων ($ΑΣ_{2.5}$ και $ΑΣ_{2.5-10}$) από φυσικές πηγές επίσης κατασκευάστηκαν για τις μητροπολιτικές περιοχές Αθηνών (AMA), Θεσσαλονίκης (TMA) και Βόλου (GVA) για την περίοδο 2000-2010. Τα μητρώα περιλαμβάνουν τις εκπομπές σωματιδίων από το έδαφος αγροτικών και κενών εκτάσεων εξαιτίας της αιώρησης τους από τον άνεμο (WB) καθώς και των σταγονιδίων θαλάσσιου άλατος που εκπέμπονται στην ζώνη κυματαγωγής όταν τα κύματα χτυπούν στην ακτή (SS_SS) ή από την επιφάνεια της ανοικτής θάλασσας με τη μορφή φυσαλίδων αφρού από τις κορυφογραμμές των κυμάτων (SS_OO). Επιπρόσθετα στα μητρώα περιλαμβάνονται οι εκπομπές ΒΠΟΕ (Βιογενείς Πτητικές Οργανικές Ενώσεις), που είναι πρόδρομες ενώσεις σωματιδίων. Ο στόχος της παρούσας εργασίας είναι η μελέτη της εποχιακής μεταβολής των εκπομπών ΑΣ από φυσικές πηγές στις περιοχές ενδιαφέροντος και της χωρικής κατανομής τους κατά την περίοδο 2000-2010. Έτσι υπολογίστηκαν συντελεστές βαρύτητας για τον διαμοιρασμό των ετήσιων εκπομπών σε μηνιαίες και ημερήσιες τιμές και την κατανομή τους σε πλέγματα υψηλής χωρικής ανάλυσης χωριστά για κάθε ρύπο,

πηγή και περιοχή για την περίοδο 2000-2010. Δεν βρέθηκε να υπάρχει σημαντική εποχιακή διακύμανση στις εκπομπές ΑΣ από φυσικές πηγές ενώ οι εκπομπές ΒΠΟΕ βρέθηκαν αυξημένες κατά τη θερινή περίοδο εξαιτίας της ενισχυμένης ηλιακής ακτινοβολίας και θερμοκρασίας. Επιπλέον, μελετήθηκε η εποχιακή μεταβολή της συνεισφοράς των εκπομπών από φυσικές πηγές στις συνολικές εκπομπές των περιοχών την περίοδο 2000-2010. Για το σκοπό αυτό τα μητρώα $ΑΣ_{2.5}$ και $ΑΣ_{2.5-10}$ από φυσικές πηγές και ΒΠΟΕ κατανεμήθηκαν χωρικά και χρονικά και συγκρίθηκαν με μητρώα ανθρωπογενών εκπομπών στις περιοχές. Δεν βρέθηκε να υπάρχει σημαντική εποχιακή κατανομή στην συνεισφορά των εκπομπών από φυσικές πηγές στις ολικές εκπομπές ΑΣ.

Table of Contents

1. EMISSIONS FROM ANTHROPOGENIC SOURCES

1.1 Introduction

The three major anthropogenic emission sources are road transport, industry and residential/institutional/commercial activities. All emissions from these sources were disaggregated in space and time. Spatial disaggregation for navigation, aviation and railways emissions was not performed as these emissions are located in a specific area. Time disaggregation for railways was not performed as railways emissions are minor and they are not taken into account.

1.2 Road Transportation

1.2.1 Time disaggregation

For road transportation emissions, time disaggregation was based on previous works as the Masterplan for the abatement of Atmospheric Pollution in the Greater Athens Area and in the Greater Thessaloniki Area (MEEC, 2003a; MEEC, 2003b). Moreover, information on daily traffic profiles was collected through personal communication with road traffic experts (Golias I., 2011). Yearly total emissions are disaggregated to daily emissions by dividing them by 330. These daily emissions correspond to the typical working day. The daily profile of traffic loads and, hence, of emissions is presented in the following ***Figure 1***.

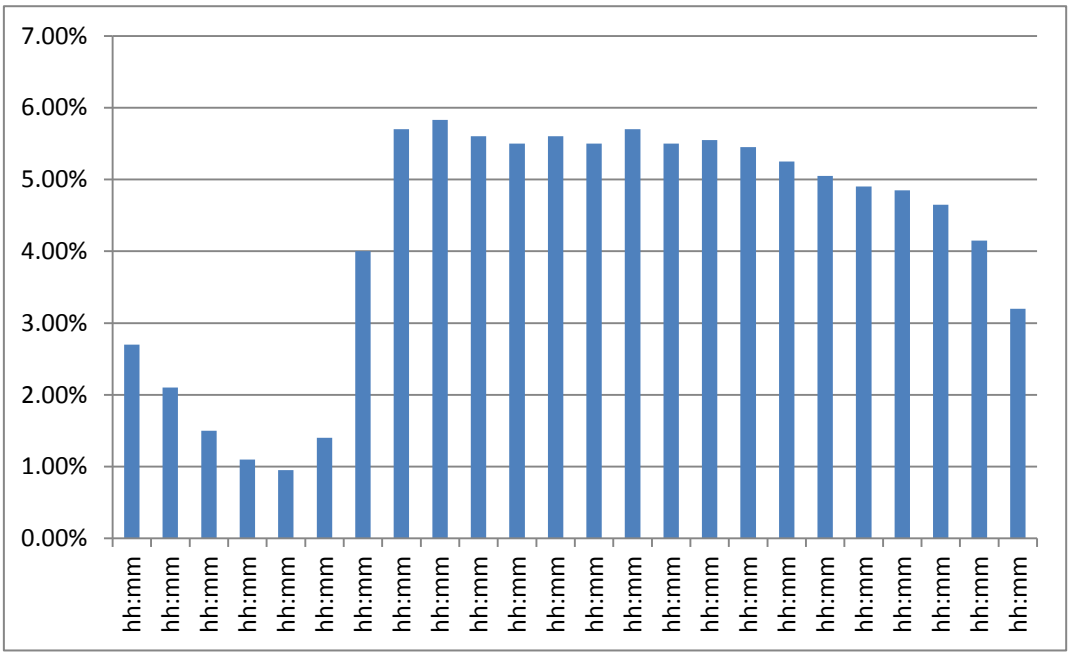


Figure 1 Daily profile of road traffic loads.

1.2.2 Spatial disaggregation

The methodology applied to disaggregate spatially road traffic emissions was based on methodologies followed in similar jobs (Saide et al., 2009; Tuia et al., 2007; Rose et al., 2009; Lindhjem et al., 2012). In order to allocate total emissions, the road network of each area was taken into account. The line sources cover more than one grid cell and therefore their shares for each grid cell were calculated before the spatial allocation has been performed. The road segments were divided in four different types, according their capacity, and total emissions were allocated according the density of each road type in each cell.

In the following figures (Figures 2-4) the spatial distribution of PM emissions is presented.

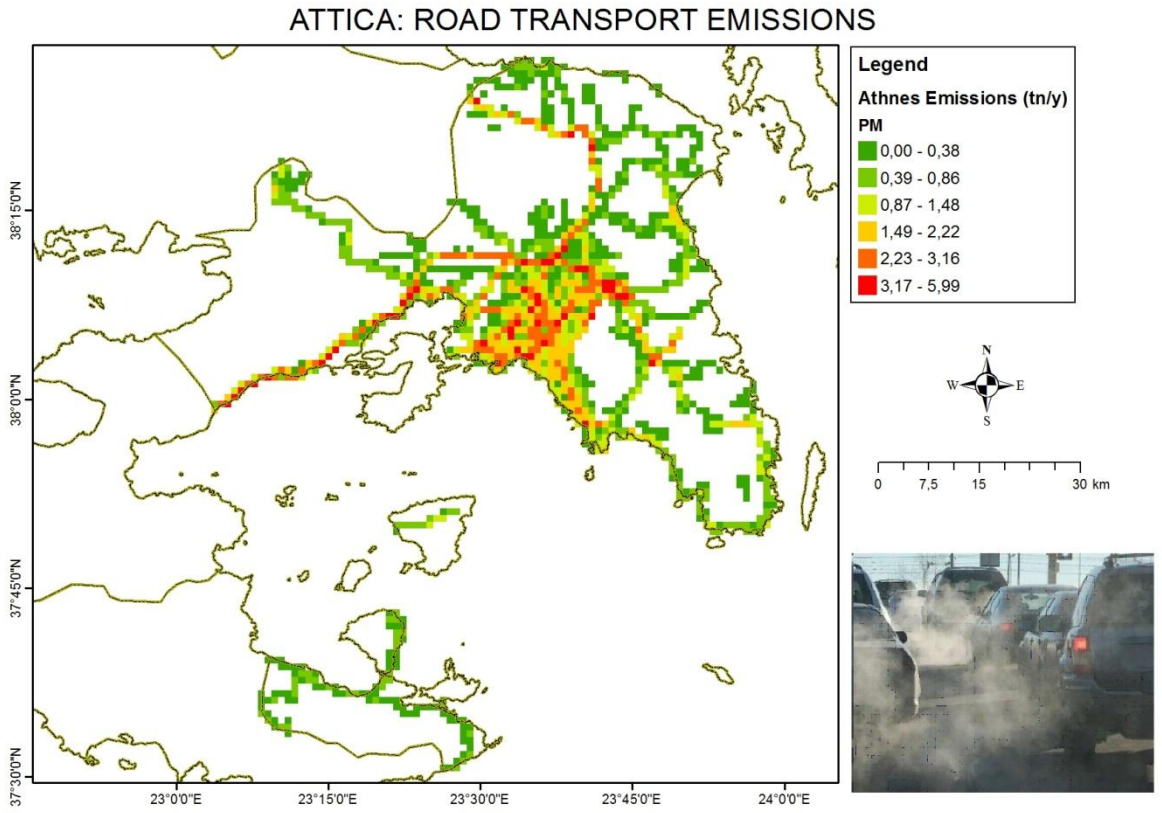


Figure 2 ***Spatial distribution of PM road traffic emissions in the Athens Metropolitan Area.***

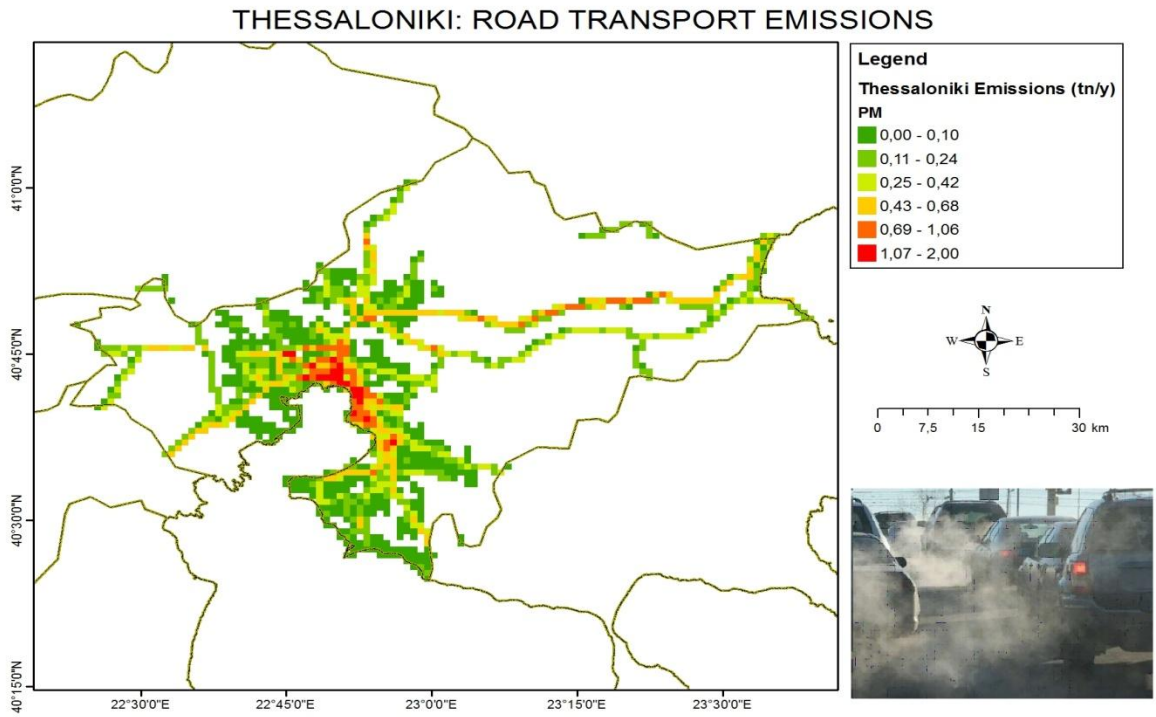


Figure 3 ***Spatial distribution of PM road traffic emissions in the Thessaloniki Metropolitan Area.***

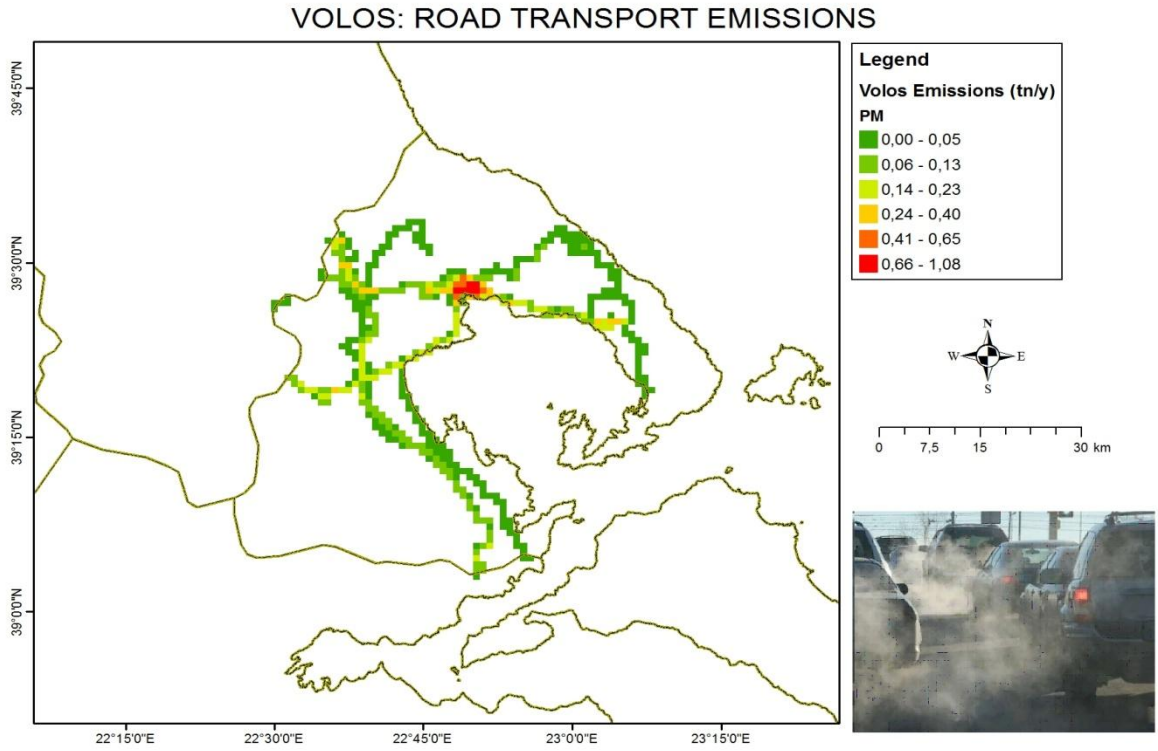


Figure 4 ***Spatial distribution of PM road traffic emissions in the Greater Volos Area.***

1.3 Industrial Sector

1.3.1 Time disaggregation

For industrial emissions, based on data available, it was considered that big installations work on a 24 hour basis. For the smaller installations, it was considered that they work on a 12 hour basis. All installations operate 12 months per year. In the figure below the daily profile of PM emissions is presented.

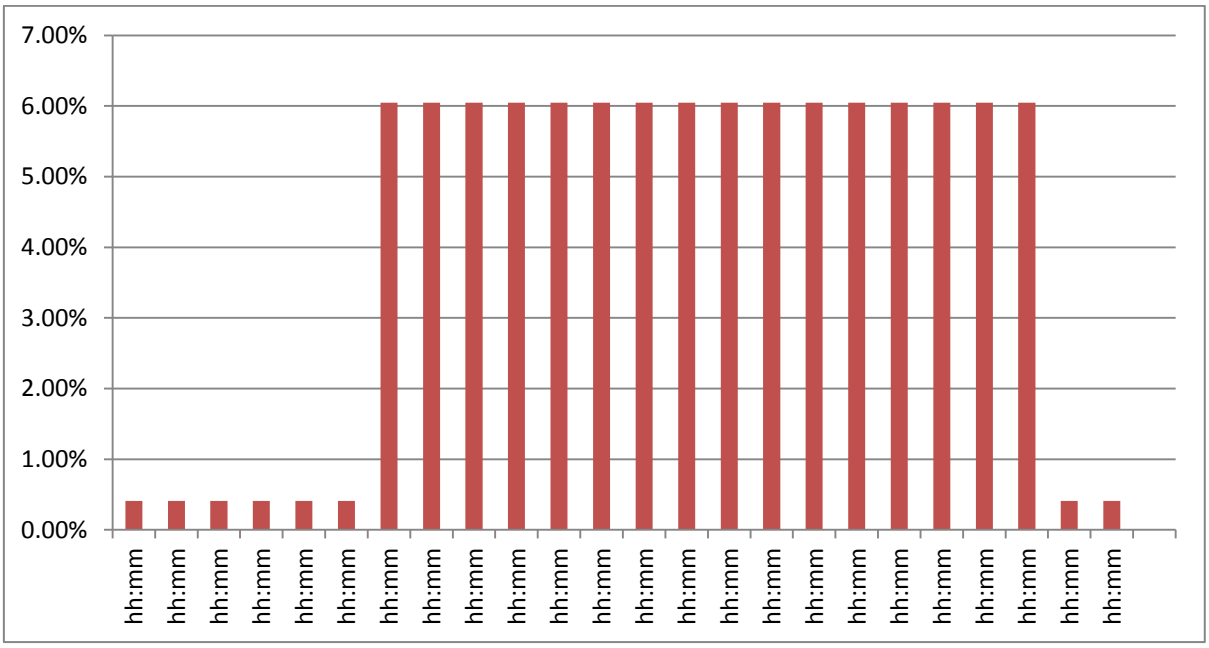


Figure 5 Daily profile of industrial emissions.

1.3.2 Spatial disaggregation

The methodology description includes the explanation of the selection of the distribution parameters, which are used for gridding of diffuse emissions for each pollutant. The target spatial resolution is a 5 km x 5 km grid cell size for maps covering all EU27 Member states and the EFTA countries (Switzerland, Liechtenstein, Norway and Iceland) for the selected sectors and pollutants. The emissions data of pollutants NOx, PM10, SO2 and CO are the officially submitted emission data sets to UNECE under the Convention on Long-range Transboundary Air Pollution (CLRTAP, 2010). Diffuse emissions are defined as emissions from sources excluding emissions from E-PRTR (*European Pollutant Release and Transfer Register)* related facilities. Therefore diffuse emissions, for the purpose of this programme, include small industrial point sources which fall below the Annex I and Annex II capacity thresholds of the E-PRTR regulation.

Hence, the spatial allocation of the diffuse industrial emissions data is given by the E-PRTR database. Nevertheless, the spatial resolution of 5X5 km^2, though sufficient for the scale of Europe, it is not reliable for the scale of a city, where more accurate spatial data are required. In this case, 1X1 km^2 resolution was applied.

To this aim, CORINE land uses were used in order to localise industrial areas (CODE 121), and diffuse emissions were allocated according to the location of these areas, resulting hence in the required resolution. In order to distribute the area sources the shares for each concerning grid cell were derived. Point emission sources were allocated according to their coordinates reported under the Convention on Long-range Transboundary Air Pollution (CLRTAP, 2010).

In the following figures (***Figures 6-8***) the spatial allocation of PM industrial emissions is presented for the three areas of interest.

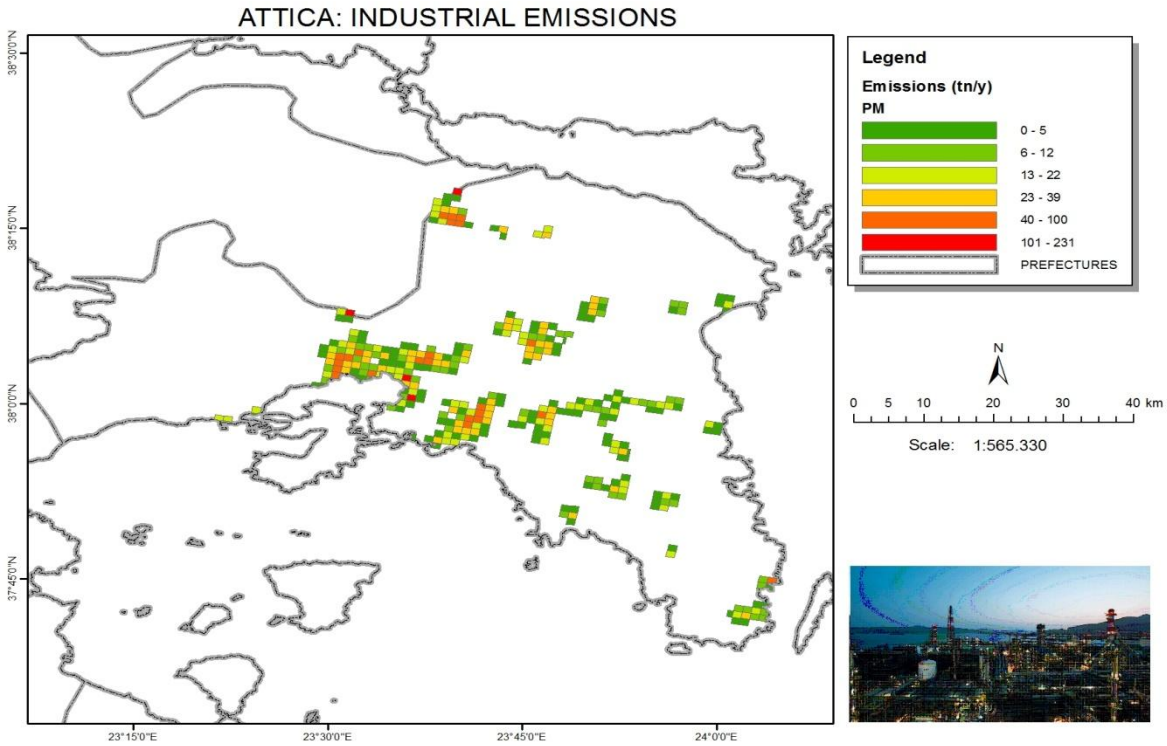


Figure 6 ***Spatial distribution of PM industrial emissions in the Athens Metropolitan Area.***

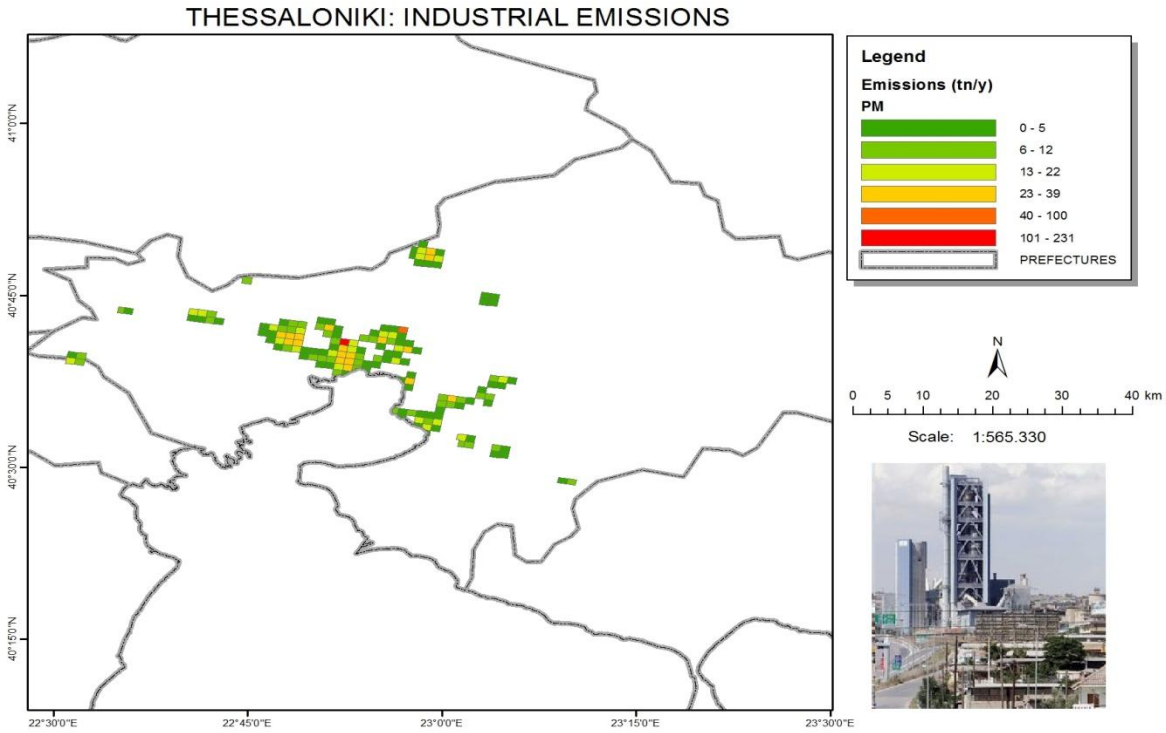


Figure 7 ***Spatial distribution of PM industrial emissions in the Thessaloniki Metropolitan Area.***

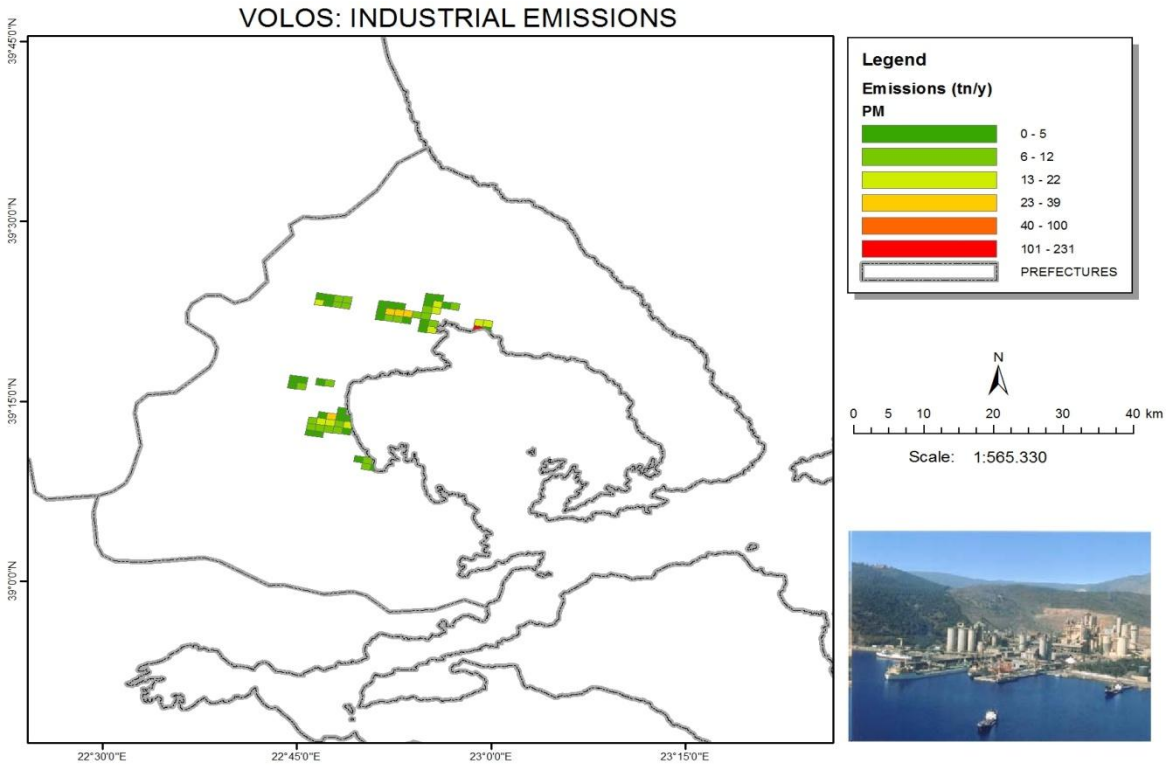


Figure 8 ***Spatial distribution of PM industrial emissions in the Greater Volos Area.***

1.4 Residential/Commercial Sector

1.4.1 Time disaggregation

According to the results of a survey contacted for the three areas of study (MEEC, 2012; Hellenic Statistical Authority, 2012), residential and commercial emissions are distributed equally in the course of the day from 7:00 to 21:00. However, these emissions do not cover the whole year. According to the meteorological measurements and fuel consumption statistical data, for the AMA and for Volos they cover a period from November to April whereas in the TMA they cover a period of 7 months from mid-October to mid-April.

1.4.2 Spatial disaggregation

The methodology for the spatial distribution of emissions from residential and commercial activities was based on the following data:

- The number of employees and the population data for each grid cell.
- Land use data in order to allocate emissions from commercial combustion and residential combustion

In the following figures (Figures 9-11), the spatial allocation of PM residential and commercial emissions is presented.

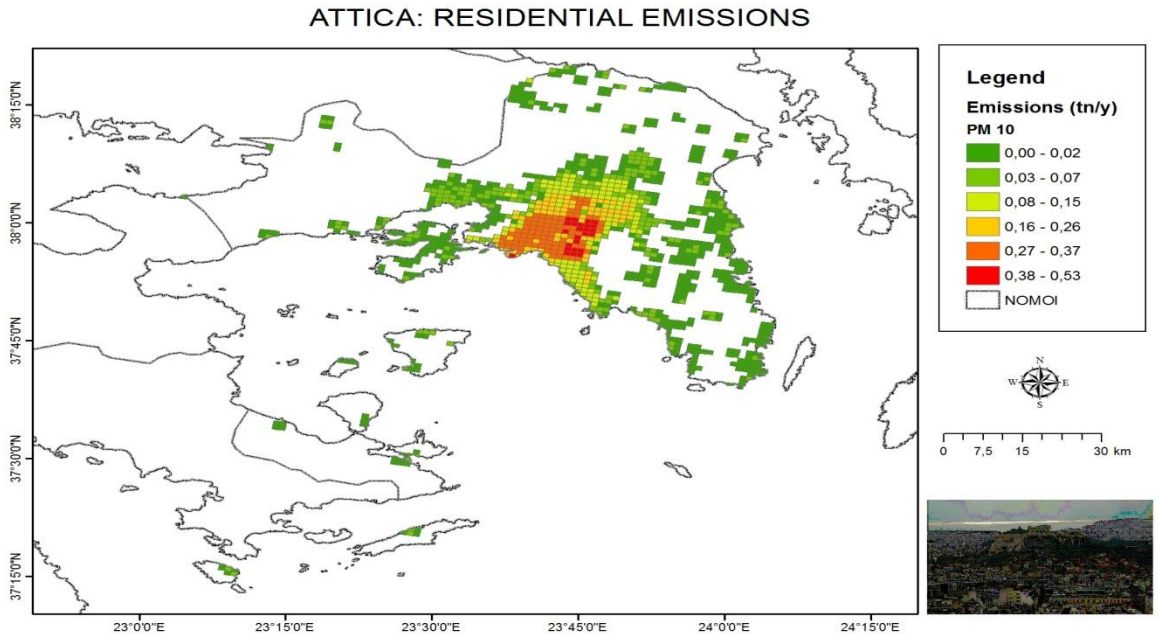


Figure 9 ***Spatial distribution of PM residential/commercial emissions in the Athens Metropolitan Area.***

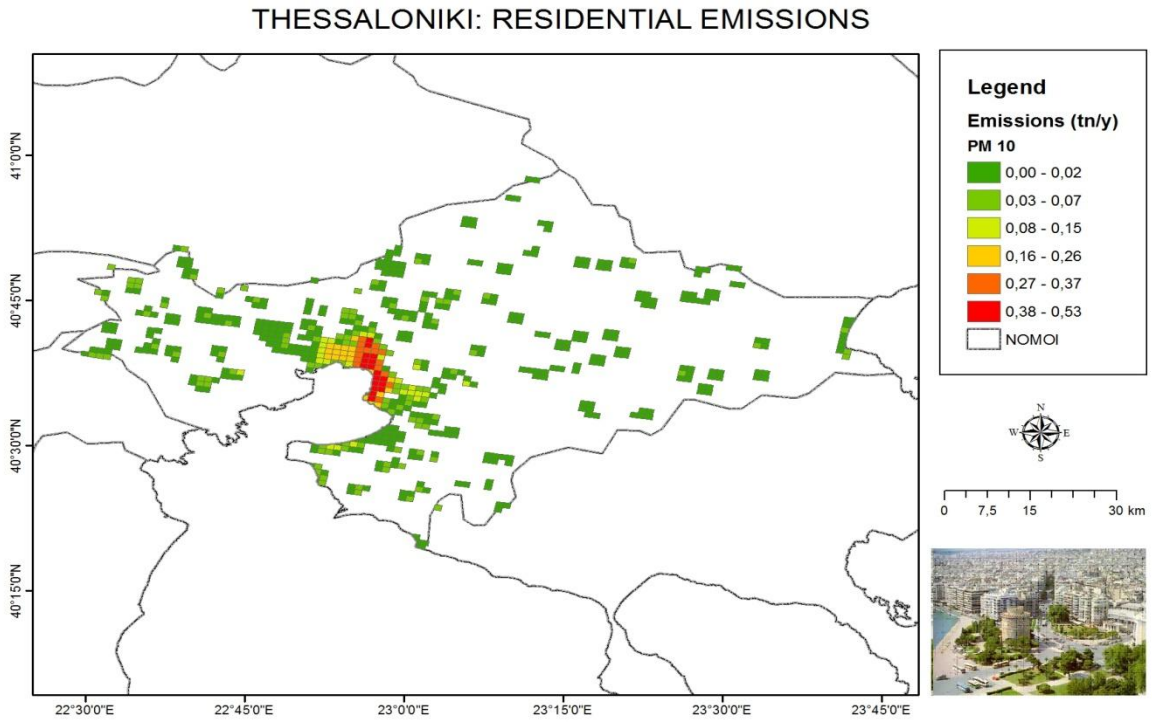


Figure 10 ***Spatial distribution of PM residential/commercial emissions in the Thessaloniki Metropolitan Area.***

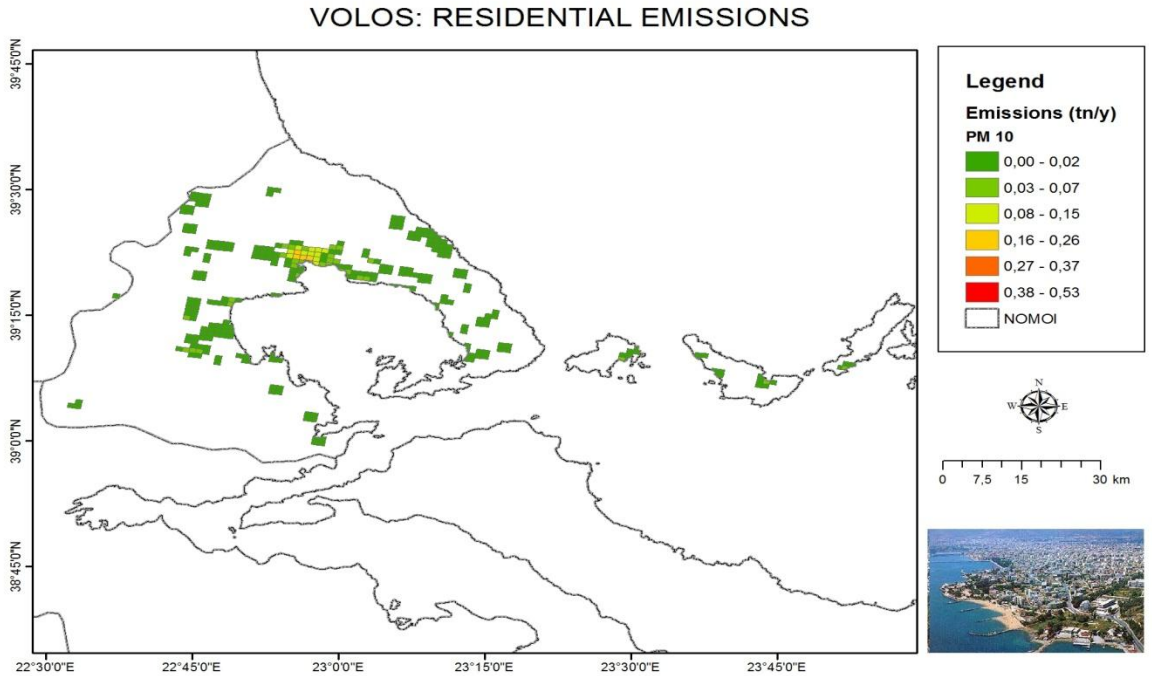


Figure 11 ***Spatial distribution of PM residential/commercial emissions in the Greater Volos Area.***

1.5 Aviation/Navigation

1.5.1 Temporal disaggregation

Aviation emissions were disaggregated in time taking into account the daily distribution of the number of flights as given by the Hellenic Civil Aviation Agency. The monthly variation of the aviation emissions is presented in ***Figure 12*** and the daily flights distribution is presented in ***Figure 13***. The busiest month is August whereas the busiest hours are the morning (06:00-11:00) and the evening hours (16:00-20:00). However, as the airport emissions take place far away from the densely populated areas, they are not considered to influence the PM levels occurred.

As for navigation, because of different activities taking place in the port area, no detailed data were available. Taking into account statistical data for passenger ships movements daily variation and the fact that commercial shipping activities take place through the whole day, the diurnal variation of emissions was calculated and it is presented in ***Figure 14***.

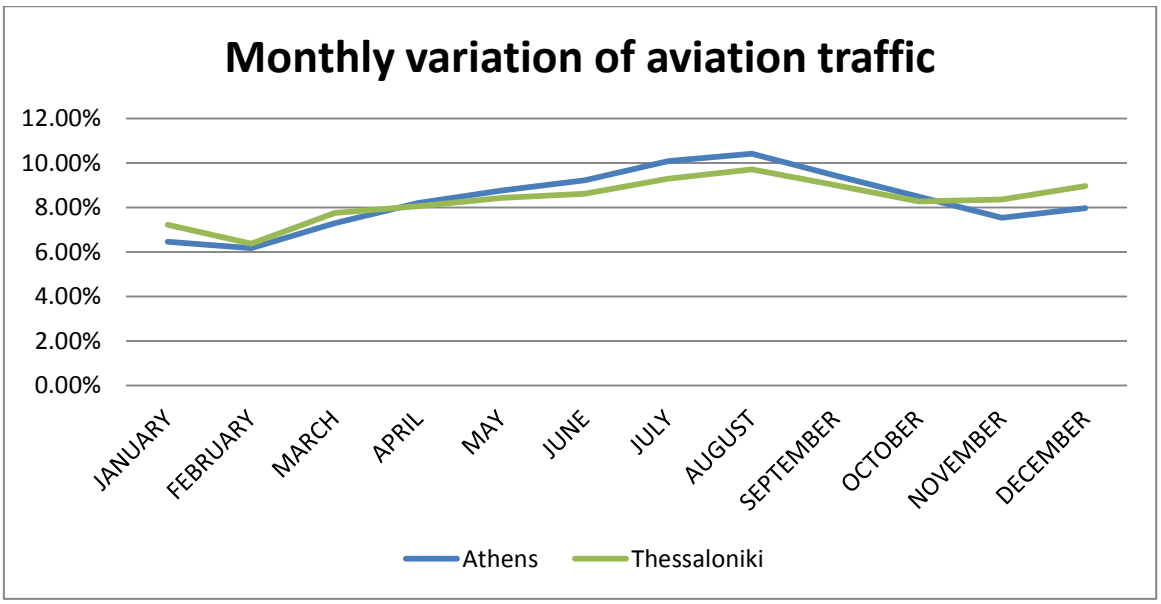


Figure 12 Monthly variation of aviation traffic.

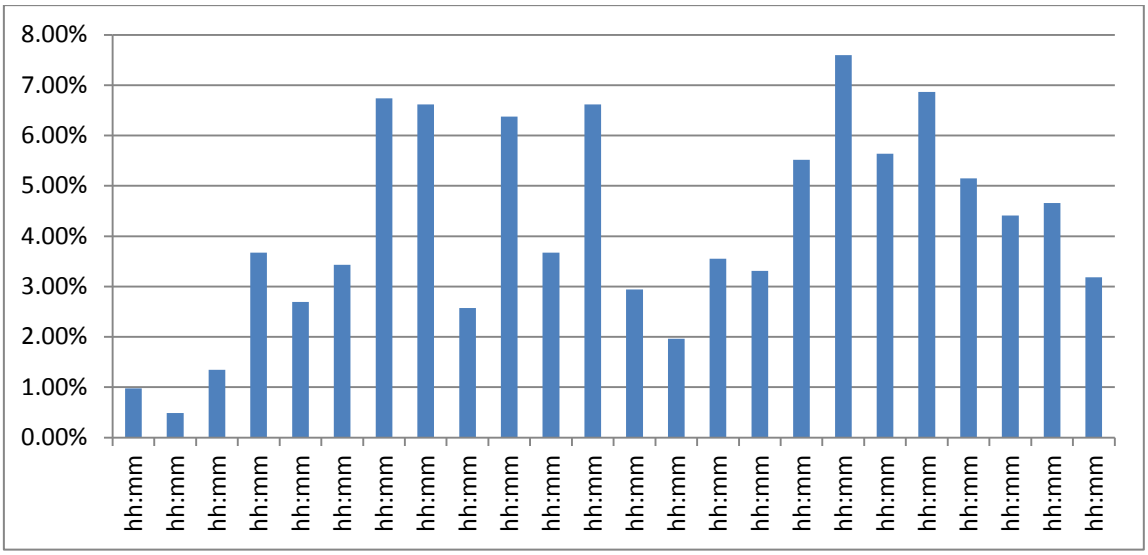


Figure 13 Daily profile of aviation traffic.

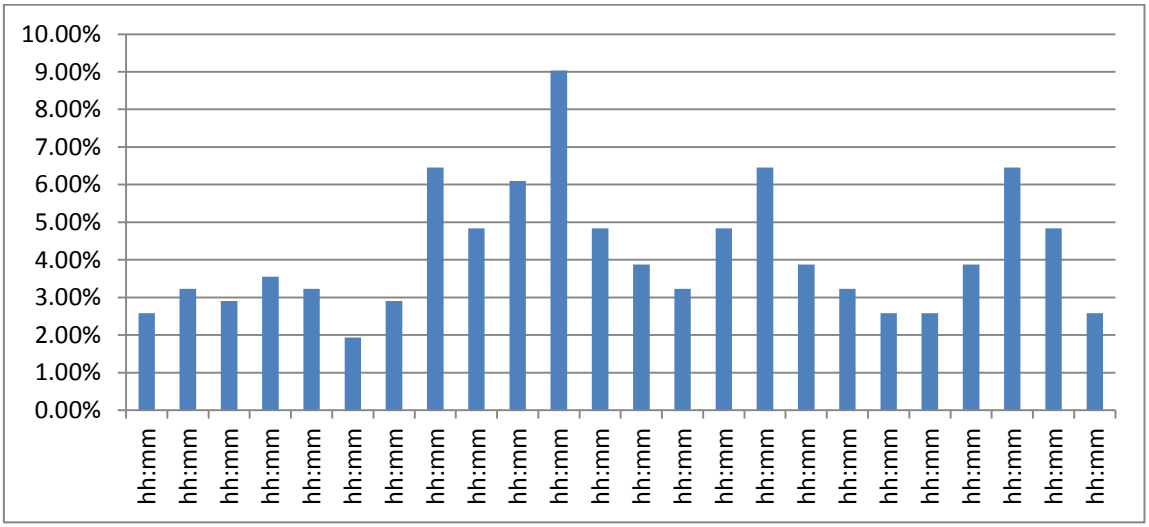


Figure 14 Daily profile of navigation emissions.

2. EMISSIONS FROM NATURAL SOURCES

2.1 Introduction

An important step in improving air quality in an area is to assess the impact of specific human activities and natural sources responsible for air quality deterioration through the quantification of pollutants emissions (e.g. Winiwarter 2009). Spatiotemporal emission data are important inputs for air quality models. The spatial and temporal evolution of emissions determines their atmospheric dispersion and their impact area.

Several emission inventories have been developed for Greece and its major metropolitan areas but only a few regard or include spatiotemporally allocated emissions from natural sources. In particular, Aleksandropoulou and Lazaridis (2004) created an emission inventory of natural sources in Greece with a spatial distribution of 5×5 km^2 for a summer and a winter period in 2000-2001. Also, Symeonidis et al. (2008) estimated biogenic NMVOCs emissions in the Southern Balkan region with a spatial resolution of 1km using mean monthly climatic data for the period 1961-1990. The emissions of sea salt particles were presented by Athanasopoulou et al. (2008) in two domains with resolutions of 6km and 2km over Greece and Athens for the period 21-24 June 2008 whereas the emissions of windblown dust in the same domains for some days during May and April 2005-2007 were presented in Athanasopoulou et al. (2010). Moreover, Aleksandropoulou et al. (2011) presented natural emissions over Athens for 14 January 2008 with 1km spatial resolution. In addition, studies on natural emissions on a European scale include also emissions from Greece (for BVOCs Oderbolz et al. 2013; Karl et al. 2009; Steinbrecher et al. 2008, for WB Korcz et al. 2009, for SS).

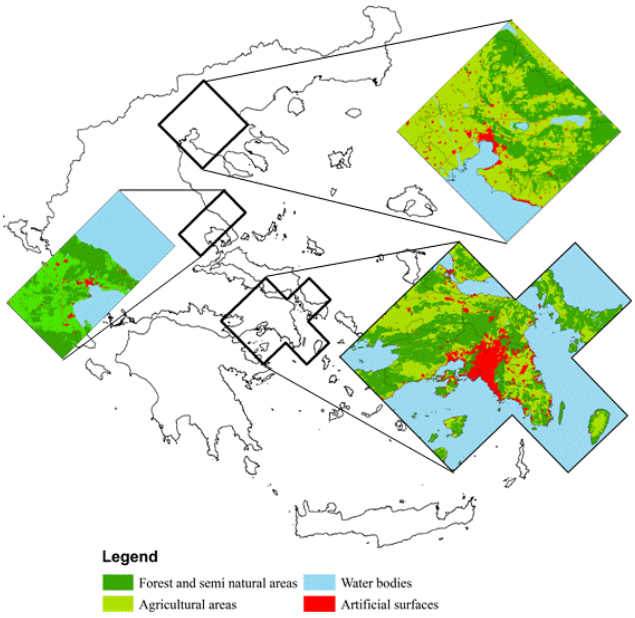


Figure 1 Areas of interest and distribution of landcover (main classes; EEA CLC 2009)

In this study a spatially and temporally resolved emission inventory was used to examine the temporal evolution of PM emissions in the metropolitan areas of Athens, Thessaloniki and Volos for the period 2000-2010 (Figure 1). The inventory includes the emissions of $PM_{2.5}$ and $PM_{2.5-10}$ from natural sources i.e. emissions of windblown dust from agricultural and vacant lands and sea salt particles emissions from the breaking of waves at the Sea Shore-surf zone (SS_SS) and the bursting of bubbles from oceanic whitecaps - Open Ocean (SS_OO). In addition, the inventory includes potential secondary organic particle emissions from natural sources, in particular biogenic gaseous pollutants (BVOCs) emissions from vegetation during photosynthesis, plant respiration and vaporization from stores within the plant tissue. The methodology for the spatial and temporal disaggregation of the emission inventory is described and the analysis then is focused on the seasonal/monthly variation of particulate matter and their precursor gases emissions from natural sources and their spatial distribution. Additionally, the inventory includes also the emissions of NO_x, SO_x, NMVOCs, CO, NH_3, $PM_{2.5}$ and $PM_{2.5-10}$ from anthropogenic sources, particularly combustion (industrial, non-industrial, commercial, residential), industrial production, transportation, agriculture, waste treatment and solvent use. Thus, the monthly variation of the contribution of primary $PM_{2.5}$ and $PM_{2.5-10}$ emissions and PM precursor gases from natural sources to the particulate matter emissions in the areas of interest is estimated.

2.2 Methodology

2.2.1 Areas of interest

Athens Metropolitan Area

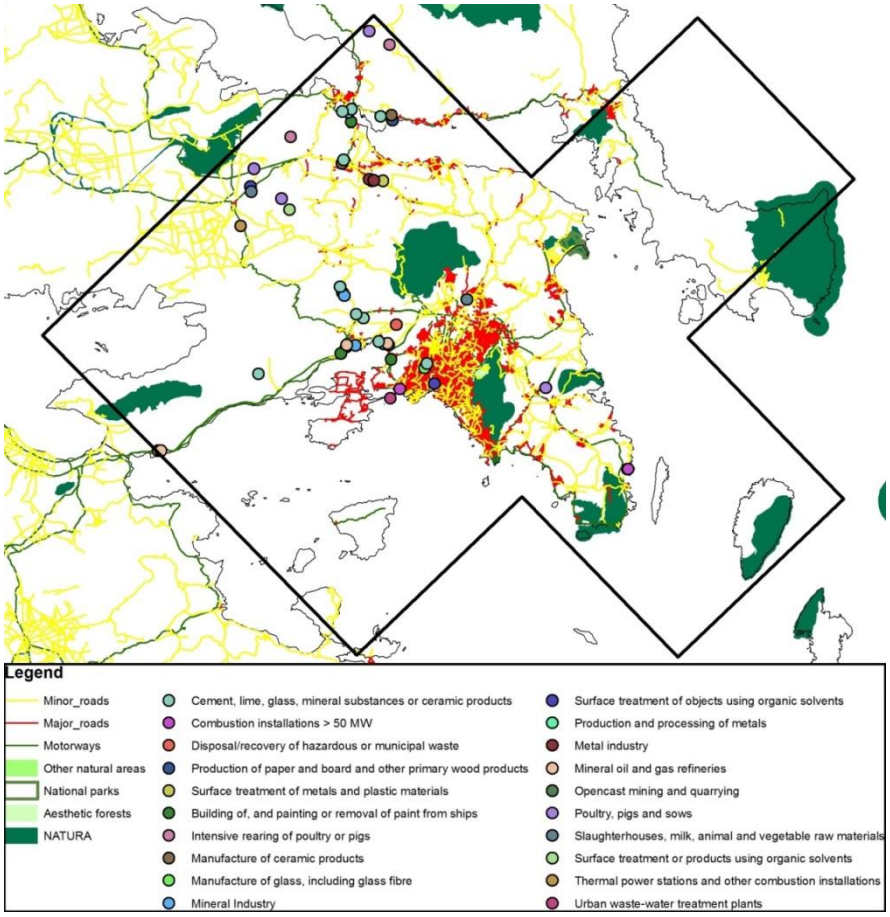


Figure 2 Athens Metropolitan Area (AMA) with road network, large point sources and green areas

The Athens Metropolitan Area (AMA) suffers air pollution problems like most metropolitan areas in the world due to high population density and the accumulation of major economic activities in the region. Athens is located along a basin of approximately 400 km² oriented SW-NE, surrounded by high mountains and open to the sea only towards the south. This topography results in wind blowing mostly from SW and NE over the area and hinders the dispersion of air pollutants (Hellenic Ministry for the Environment, Physical Planning and Public Works 2007). The area also includes several municipalities in West and East Attica prefectures, and the city of Piraeus. In Figure 2 the domain used in the calculations of emissions in the AMA is presented together with the location of large point sources with pollutant releases to air (LPS - with air emissions of the primary air

pollutants studied: NO_x, SO_x, NMVOCs, CO, NH_3, and PM_{10}; European Pollutant Release and Transport Register E-PRTR 2009 and European Pollutant Emission Register EPER 2008). Most of the LPS are situated in West Attica prefecture and Piraeus and one is located in East Attica. In addition, the main road network in the area is depicted in Figure 2 and areas of interest with regards to natural emissions (forests and parks).

Thessaloniki Metropolitan Area

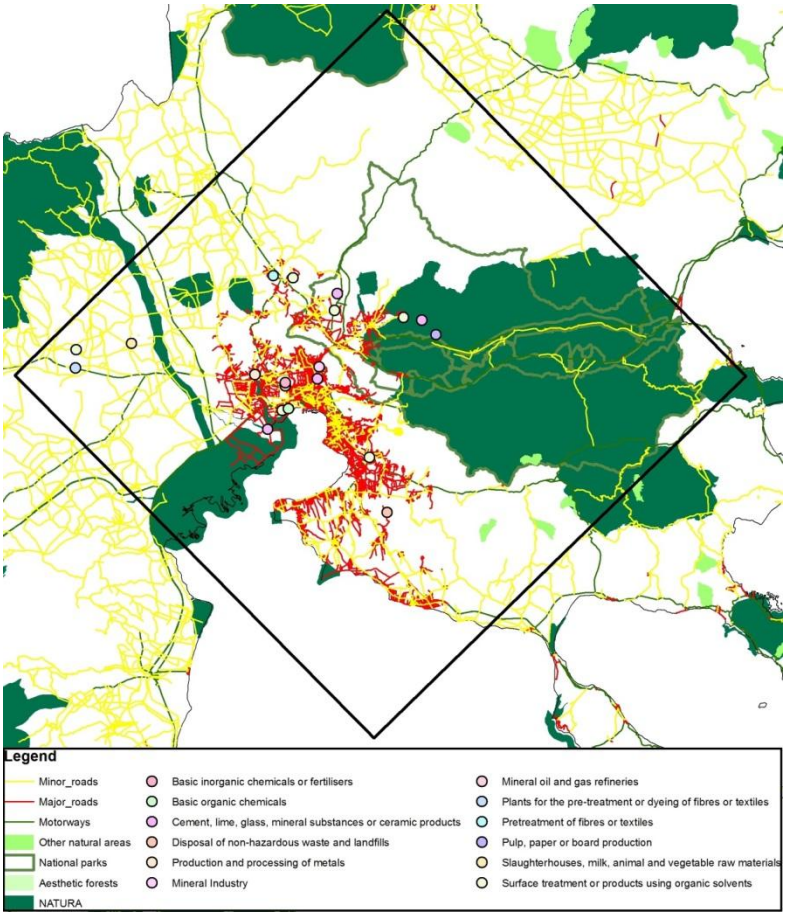


Figure 3 Thessaloniki Metropolitan Area (TMA) with road network, large point sources and green areas

Thessaloniki is the second largest city in Greece with a population (in the greater area of the city) of 1054000 inhabitants (El.STAT. 2002). Economic activities, mainly in the fields of manufacturing, commerce and culture are concentrated in the area. These activities along with heavy traffic, industrial units (mainly in the northern and northwestern part) and a very busy port and airport contribute to the air pollution in the area. In Figure 3 the domain used in the calculations of emissions from the greater area of Thessaloniki is presented together with the location of large point sources with pollutant releases to air (EPER 2008; E-PRTR 2009), the main road network and green areas.

Greater Volos Area

Volos is a coastal city in central Greece, in particular in Thessaly located at the north coast of Pagasitikos gulf. The city has a dense road network with occasionally intense traffic due to city activities, tourism and port operations (Papanastasiou et al. 2010). Two industrial areas, in the west and northwest part of the city, a large cement plant at the eastern coast of the city, central heating and the port located to the west of the city are the main stationary pollution sources in the area (Papaioannou et al. 2010). The above sources together with the topographic features of the area (surrounded by hills to the northwest and by Pelion Mountain to the northeast and east) favor the accumulation of air pollutants over the city. In Figure 4 the domain used in the calculations of emissions in the greater area of Volos is presented together with the location of large point sources with pollutant releases to air (EPER 2008; E-PRTR 2009), the main road network and green areas.

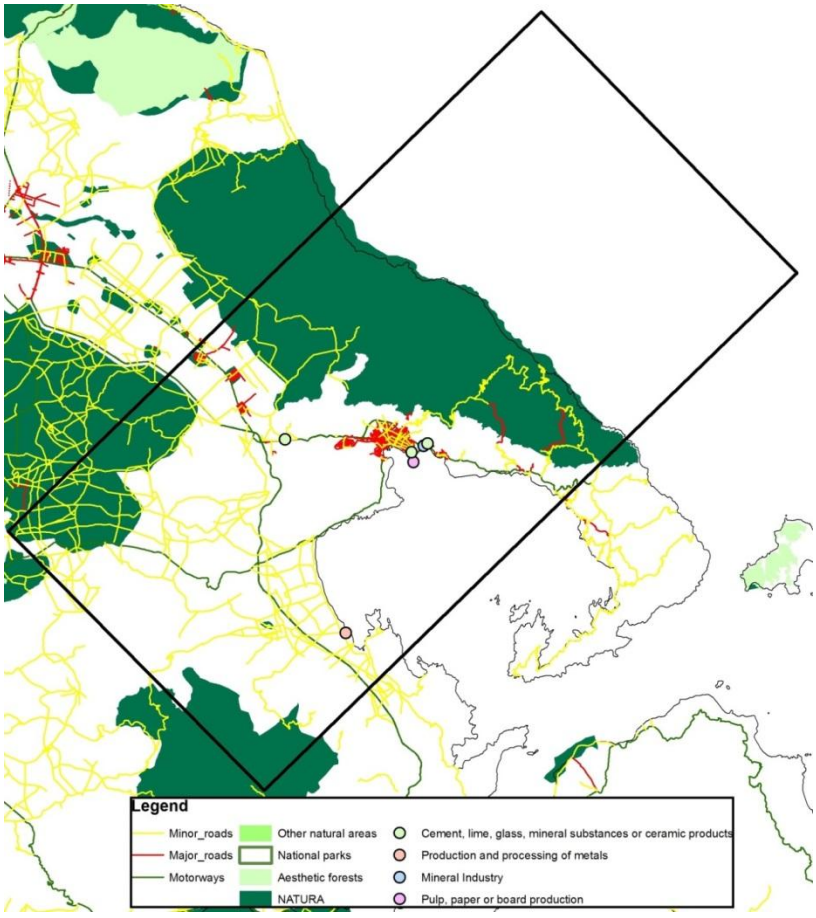


Figure 4 Greater Volos Area (GVA) with road network, large point sources and green areas

2.2.2. Input data and assumptions affecting the spatial and temporal variation of natural emissions

The temporal disaggregation is performed using calculated disintegration coefficients based on monthly and daily averages of meteorological conditions (monthly meteorology from FOODSEC Meteodata distribution page: action developed in the framework of the EC Food Security Thematic Programme; European Centre for Medium-Range Weather Forecast data ECMWF ERA INTERIM reanalysis model data; temporal analysis 10-days; spatial resolution 0.25 degree; daily meteorology from NCEP Reanalysis Derived data provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA, from their Web site at http://www.esrl.noaa.gov/psd/; Kalnay et al., 1996). The temporal resolution of meteorological data can affect the emission estimates especially on local scales as it has been previously shown by Ashworth et al. (2010) for global isoprene emission estimates.

The effects of using monthly averaged wind speed values instead of hourly data (particularly 3h averages) in predicting the emissions of dust due to wind erosion and of sea salt particles in the calculations have been examined in Aleksandropoulou et al. (2013). It was found that although emission rates can differ substantially from the actual ones, the results as regards monthly emissions are acceptable since the error introduced by the above assumptions can be considered the same to that introduced by uncertainty in other parameters (i.e. the soil moisture content and texture, the surface roughness length, and constraining factors like the vegetation coverage and the presence of non-erodible elements). Moreover, the use of daily temperature data instead of monthly (NOAA/OAR/ESRL PSD data vs. ECMWF ERA INTERIM reanalysis model data) resulted in a difference in annual BVOCs emissions of -4.3% from AMA, -23% for TMA and 14% for GVA. With regards to the seasonal variation in BVOCs emissions the use of daily temperature values resulted in differences in the range of 4% and -58% for the warm and cold period in AMA, respectively while the corresponding values were -18% and -82% for TMA and 22% and -79% for GVA.

The spatial resolution of meteorological data can also affect the emission estimates and their spatial distribution especially on local scales as it has been previously shown by Pugh et al (2013) for global isoprene emission estimates. In this study we used meteorological data averaged over the domains therefore the spatial resolution of emissions is depends only on the landuse, the soil characteristics and the vegetation type.

Additionally, the effects from snow cover and rain on daily PM and BVOCs emissions could not be taken into account in the calculations due to the spatial and temporal resolution of the meteorological data. Their effects on monthly emissions were incorporated in the calculations by downscaling the results by considering periods with rain and snow as inactive for WB emissions.

In the absence of other relevant data it was assumed that the landcover remains unchanged throughout the period (Land Cover 2000 database of the European Commission

programme to COoRdinate INformation on the Environment across Europe; EEA CLC 2000, v2009). In addition, based on the conclusions in Aleksandropoulou et al. (2013), changes in landcover due to forest fires have not been taken into account in the calculations.

2.2.3 Temporal disaggregation of emissions

The variation in natural emissions depends on the meteorological conditions, the emission factors and changes in landuse. Since landuse data remained unchanged throughout the period, temporal disintegration coefficients depend solely on the meteorological conditions and the seasonal and hourly variation in emission factors.

The annual PM emissions are temporally disaggregated to hourly emissions using the function (Eq. 1):

$$Em_{h,m,y,j,k} = E_{y,j,k} \frac{M_{y,m,j,k}}{12} D_{y,m,j,k} H_{y,m,j}$$

where $Em_{h,y,j,k}$ is the emission for hour h, month m, and year y, of pollutant j from source k; $E_{y,j,k}$ is the emission for year y, of pollutant j from source k; $M_{y,m,j,k}$, $D_{y,m,j,k}$ and $H_{y,m,j}$ are the monthly, daily and hourly disintegration coefficients for month m and year y, of pollutant j from source k; j = [$PM_{2.5}$; $PM_{2.5\text{-}10}$; BVOCs]; k = [SS_SS; SS_OO; WB; BVOCs]; y = (2000, 2010); m = (January, December); h = (1, 24).

The emission estimation methodology used in this study resulted in the calculation of emissions either in the form of hour/day averages (BVOCs) or as average emissions per second (SS, WB) for specific month and year which were then combined to produce annual emissions. Consequently, the monthly coefficients, $M_{y,m,j,k}$, for month m and year y, for pollutant j from source k were calculated based on the equation (Eq. 2):

$$M_{y,m,j,k} = \frac{E_{y,j,k}}{12} E_{y,m,j,k}$$

where $E_{y,m,j,k}$ is the emission for year y, of pollutant j from source k and month m.

The emissions are equally distributed to each day of the month except for WB and BVOCs emissions of the period 2010-2013, for which daily disintegration coefficients have been calculated based on the intermonth variation in daily temperature and windspeed values (data from NOA stations and NCEP Reanalysis Derived data provided by the NOAA/OAR/ESRL PSD, Boulder, Colorado, USA, from their Web site at http://www.esrl.noaa.gov/psd/; Kalnay et al., 1996). In particular, daily averages of BVOCs emissions have been calculated. Accordingly, the daily coefficients, $D_{y,m,j,k,}$ for month m and year y, for pollutant j from source k in equation 1equals to (Eq. 3):

$$D_{y,m,j,k} = \begin{cases} \frac{1}{N_{m,y}}, \text{ for SS emissions and WB and BVOCs emissions (2000-2009)} \\ \frac{U_{d,m,y}\ U_{m,y}}{N_{m,y}}, \text{ for WB emissions in 2010} \\ \frac{E_{d,y,m,j,k}}{E_{y,m,j,k}}, \text{ for BVOCs in 2010} \end{cases}$$

where $N_{m,y}$ are the number of days in month m of year y, $U_{d,m,y}$ is the day average of windspeed for day d of month m in year y, $U_{m,y}$ is the monthly average of windspeed in year y, $E_{d,y,m,j,k}$ is the daily emission for month m of year y for pollutant j from source k and $E_{y,m,j,k}$ is the emission for year y, of pollutant j from source k and month m.

In addition, emissions can be equally distributed to each hour of the day, i.e. $H_{y,m,j}$ equals to (1/24). The latter applies only to PM emissions since BVOCs emissions depend also on light.

The final temporal resolution of emissions is 1 h for $PM_{2.5}$ and $PM_{10-2.5}$ and 1 d for BVOCs.

2.2.4 Spatial disaggregation of emissions

The domain for each area of interest was created based on the availability and spatial resolution of the officially reported emission data in the UNECE/EMEP database (EMEP/CLRTAP 2009; for comparison with anthropogenic emissions). Each domain was covered with a high resolution grid (1×1 km^2; depicted in Figure 5) for the spatial distribution of emissions (European Terrestrial Reference System (ETRS) 1989 Lambert Azimuthal Equal Area (LAEA) projection, Central Meridian: Greenwich, false easting: 4321000 m, false northing: 3210000 m, central meridian: 10.0, latitude of origin: 52.0, D_ETRS_1989).

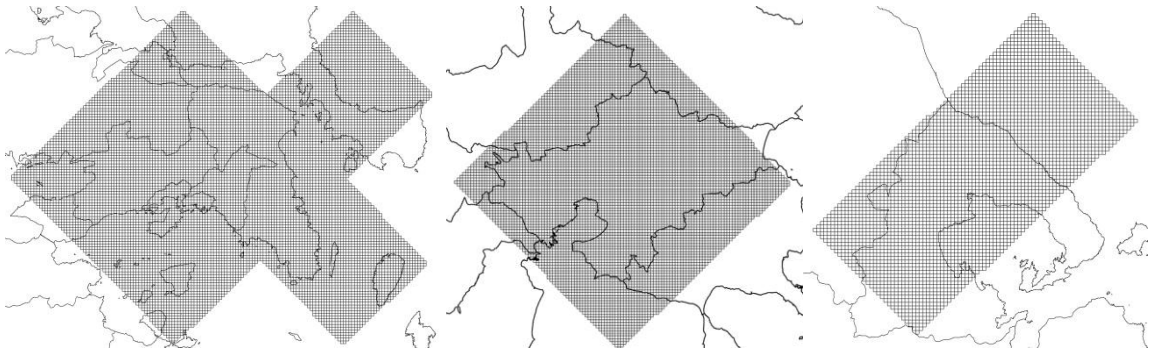


Figure 5 Grids covering the AMA (10896 cells), TMA (7596 cells) and GVA (3658 cells)

The annual emissions were spatially disaggregated and allocated to the grid covering each area of interest using the function:

$$Em_{i,k,y} = E_{k,y} WF_{i,k,y} \text{ (Eq. 4)}$$

where $Em_{i,k,y}$ is the emission from cell i from source k during year y; $E_{k,y}$ is the year's y emissions from the area of interest from source k; $WF_{i,k,y}$ is the spatial disaggregation weighting factor of cell i for emissions from source k in year y; i = [AMA (0, 10895); TMA (0, 7595); GVA (0, 3657)]; k = [SS_SS; SS_OO; WB; BVOCs].

The emission estimation methodology used in this study resulted in the calculation of emissions either in the form of emissions per cm^2 of area with specific landcover and soil characteristics (SS, WB) or as emissions over areas (in m^2) of specific vegetation type which were then combined to produce emissions per grid cell (1 km^2). The spatial distribution of the calculated emissions depends on the landcover and soil characteristics, the variation in emission factors and the spatial distribution of meteorological parameter values. Since landuse and soil characteristics data remained unchanged throughout the period and the meteorological data were averaged over the whole areas of interest, the calculated spatial disaggregation coefficients (weighting factors) depend solely on the meteorological conditions and the seasonal and hourly variation in emission factors. Equation 4 can also be used for the spatial distribution of monthly and daily emissions but with weighting coefficients, $WF_{i,k,m,y}$, specific for each cell i and source k during year y and month m. The weighting factors WF have been calculated separately for each year and month and the results are presented later in the text.

2.2.5 Anthropogenic emissions inventory - spatial and temporal allocation of anthropogenic emissions

In order to calculate the contribution from natural sources to total primary and secondary particle emissions in the areas of interest a spatiotemporally disaggregated anthropogenic emissions inventory was used. The inventory includes the annular (tn/yr) gaseous pollutants (NO_x, SO_x, NMVOCs and NH_3) and particulate matter ($PM_{2.5}$ and $PM_{2.5-10}$) anthropogenic emissions derived from the UNECE/EMEP database (EMEP/CLRTAP 2009; CEIP, Emission from Greece during 2000-2010 as used in EMEP models). The anthropogenic emissions were allocated on the grids covering the areas of interest using surrogate spatial datasets according to the methodology on spatial emissions mapping presented in the EMEP/EEA Emission Inventory Guidebook (Goodwin et al. 2009; Maes et al. 2009). Additionally, the annual anthropogenic emissions were temporally disaggregated. More details on the spatial and temporal disaggregation of the anthropogenic emissions inventory can be found in Aleksandropoulou et al. (2011).

2.2.6 Aerosol formation

The contribution of gaseous pollutants to aerosol formation was calculated for each source sector according to the methodology of de Leeuw (2002). According to the above methodology emissions of each precursor gas can be weighted to account for potential secondary aerosol formation. The weighting factors account for the fraction of emissions of pollutant changing into aerosol and the molecular weight difference. Their values have been derived on European level and are 1 for primary PM, 0.54 for SO_2, 0.88 for NOx, 0.64 for NH_3 and 0.02 for NMVOCs. Emissions of each pollutant are multiplied by the aerosol formation potential and results are reported in PM_{10} equivalents.

2.3 Results

2.3.1 Monthly variation of natural emissions

The monthly emissions of primary PM from natural sources in the three areas of interest during the period 2000 – 2010 are depicted in Figures 6. Based on the assumptions made in the calculations it was found that the monthly variation of $PM_{2.5-10}$ windblown dust and sea-salt sea-shore emissions is the same as to $PM_{2.5}$ emissions but with higher values. In addition, it is observed that most of the emissions of natural PM occur during the cold period of the year at all areas. In particular, in AMA natural $PM_{2.5}$ and $PM_{2.5-10}$ emissions exhibit their lowest value in May ($PM_{2.5}$ SS_SS: 344.3±28.8 t, SS_OO: 131.2±42.1 t, WB: 21.9±7.9 t; $PM_{2.5-10}$ SS_SS: 2682.2±224.3 t, SS_OO: 704.9±182.7 t, WB: 197.1±71.2 t) and the highest in December for sea- salt particles ($PM_{2.5}$ SS_SS: 459.8±75.4 t, SS_OO: 362.1±191 t; $PM_{2.5-10}$ SS_SS: 3582.1±587.2 t, SS_OO: 1691.8±808.7 t) and in August for windblown dust (WB $PM_{2.5}$: 63.7±35.1 t; $PM_{2.5-10}$:

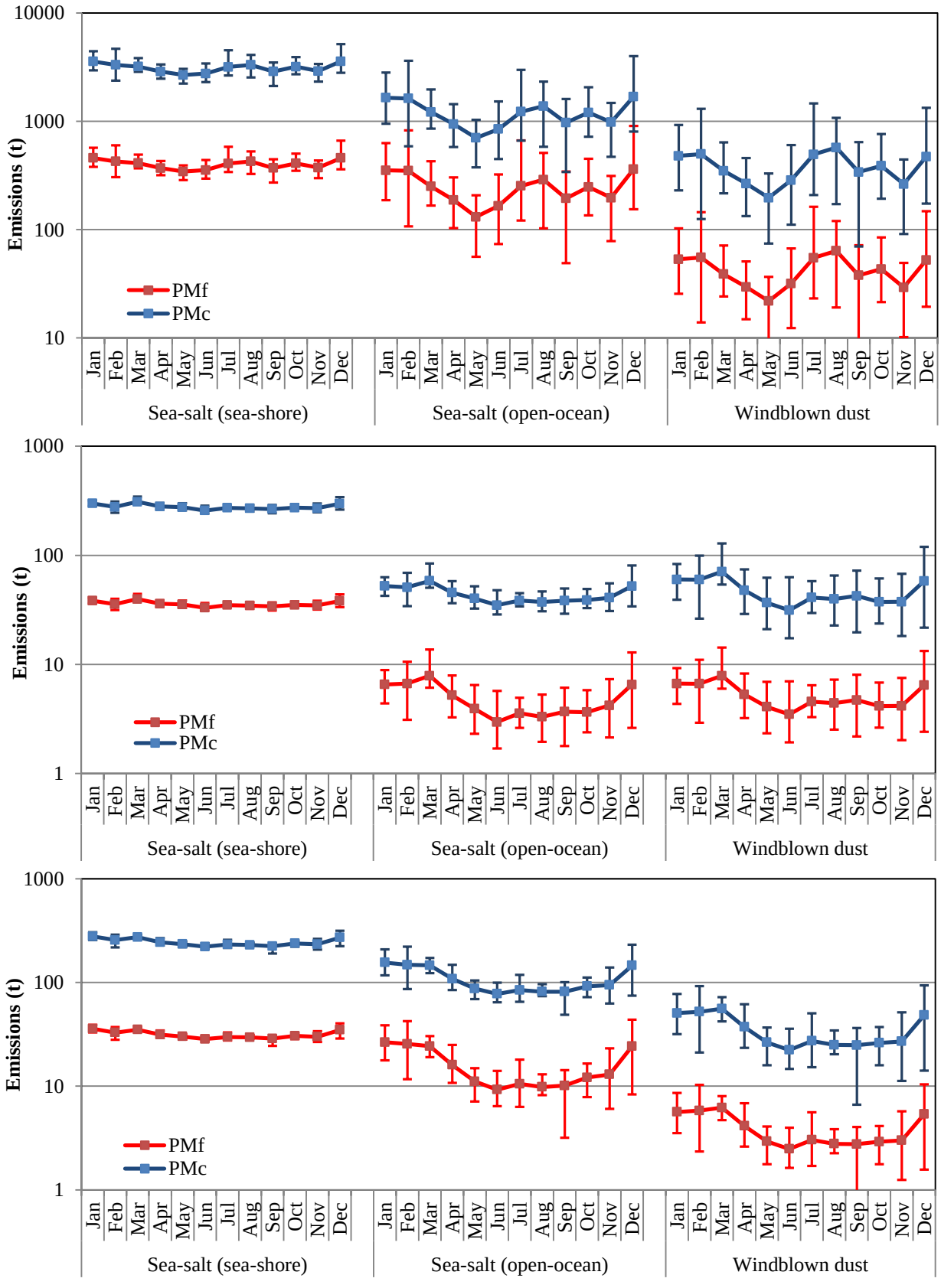

10000
1000
100
10
Emissions (t)
PMf
PMc
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec
Sea-salt (sea-shore)
Sea-salt (open-ocean)
Windblown dust
1000
100
10
1
Emissions (t)
PMf
PMc
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec
Sea-salt (sea-shore)
Sea-salt (open-ocean)
Windblown dust
1000
100
10
1
Emissions (t)
PMf
PMc
Jan Feb Mar Apr May Jun Jul Aug Sep Oct Nov Dec
Sea-salt (sea-shore)
Sea-salt (open-ocean)
Windblown dust

Figure 6 Monthly variation of $PM_{2.5}$ and $PM_{2.5-10}$ emissions in AMA, TMA and GVA during the period 2000-2010. Average, minimum and maximum monthly emission values are depicted.

573.7±316.1 t) with a Warm/Cold period emission ratio equal to 0.9 for $PM_{2.5}$ and $PM_{2.5-10}$ SS_SS emissions, 0.88 for $PM_{2.5}$ and $PM_{2.5-10}$ WB emissions, 0.69 for $PM_{2.5}$ and 0.73 for $PM_{2.5-10}$ SS_OO emissions. Higher values are observed during the period December to February and lower during May and June.

Likewise, in TMA natural $PM_{2.5}$ and $PM_{2.5-10}$ emissions exhibit their lowest value in June ($PM_{2.5}$ SS_SS: 33±1.6 t, SS_OO: 3±1.1 t, WB: 3.5±1.4 t; $PM_{2.5-10}$ SS_SS: 257.4±12.1 t, SS_OO: 34.9± 5.2t, WB: 31.4±12.4 t) and the highest in March ($PM_{2.5}$ SS_SS: 39.7±1.6 t, SS_OO: 7.9±2 t, WB: 7.9±2.2 t; $PM_{2.5-10}$ SS_SS: 309.2±12.7 t, SS_OO: 58.4±8.9 t, WB: 70.9±19.7 t) with a Warm/Cold period emission ratio equal to 0.94 for $PM_{2.5}$ and $PM_{2.5-10}$ SS_SS emissions, 0.74 for $PM_{2.5}$ and $PM_{2.5-10}$ WB emissions, 0.64 for $PM_{2.5}$ and 0.8 for $PM_{2.5-10}$ SS_OO emissions. Higher values are observed during the period December to March. Finally in GVA natural $PM_{2.5}$ and $PM_{2.5-10}$ emissions exhibit their lowest value in June ($PM_{2.5}$ SS_SS: 28.4± 1.1t, SS_OO: 9.2±2.2 t, WB: 2.5±0.7 t; $PM_{2.5-10}$ SS_SS: 221.5±8.5 t, SS_OO: 77.7±10.2 t, WB: 22.3±6.2 t) and the highest in January for sea-salt particles ($PM_{2.5}$ SS_SS: 35.7±2 t, SS_OO: 26.5±6.7 t; $PM_{2.5-10}$ SS_SS: 278.3±15.8 t, SS_OO: 156±29.6 t) and in March for windblown dust (WB $PM_{2.5}$: 6.2±0.9 t; $PM_{2.5-10}$: 55.9±8.5 t) with a Warm/Cold period emission ratio equal to 0.89 for $PM_{2.5}$ and $PM_{2.5-10}$ SS_SS emissions, 0.63 for $PM_{2.5}$ and $PM_{2.5-10}$ WB emissions, 0.53 for $PM_{2.5}$ and 0.67 for $PM_{2.5-10}$ SS_OO emissions. Higher values are observed during the period December to March and lower in June – September

As previously mentioned, the monthly variation of natural PM emissions depends on the meteorological conditions. It is observed that sea salt emissions from the sea-shore do not exhibit significant seasonal variation. On the other hand sea salt emissions from the open sea are more enhanced during the cold period of the year. This is attributed to the higher wind speeds during the cold period of the year. In AMA enhanced emissions of sea salt particles in summer can be attributed to the Etesians. As regards WB dust emissions, it was found that cold period emissions are enhanced and particularly in GVA are almost twice the warm period values.

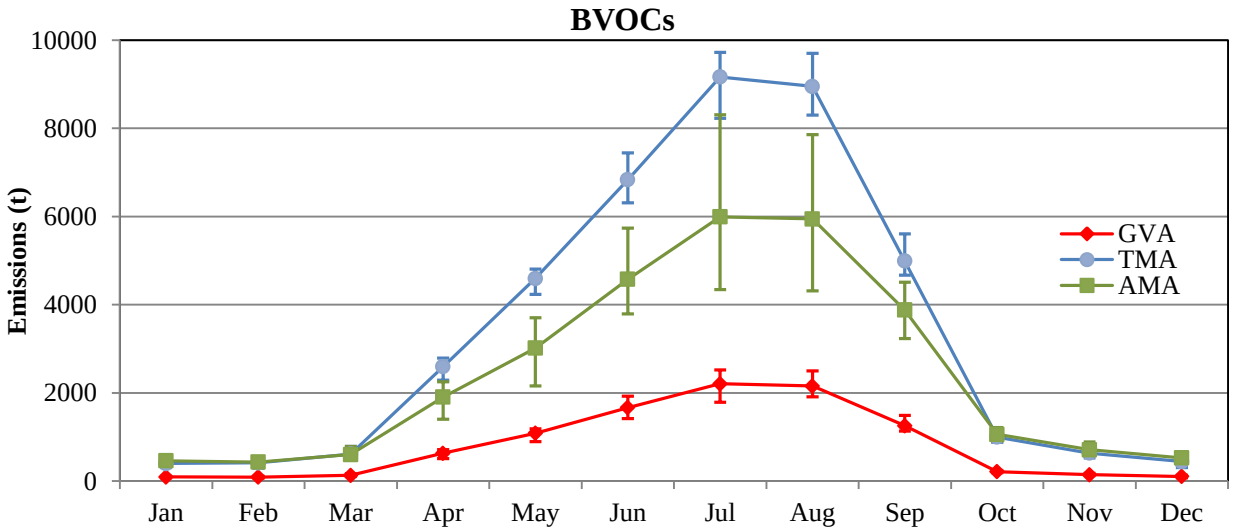
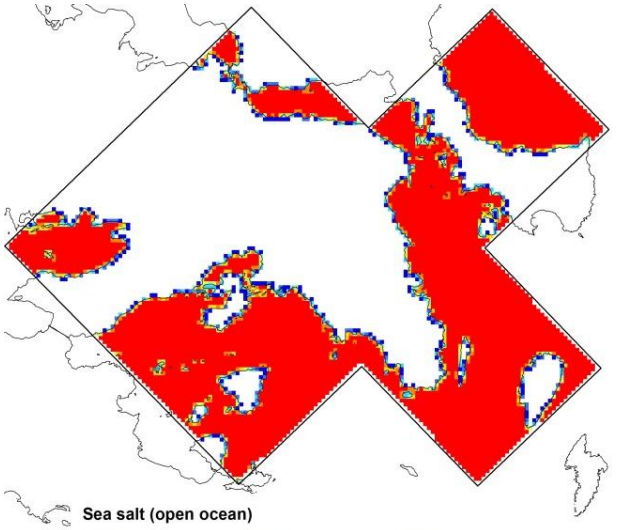
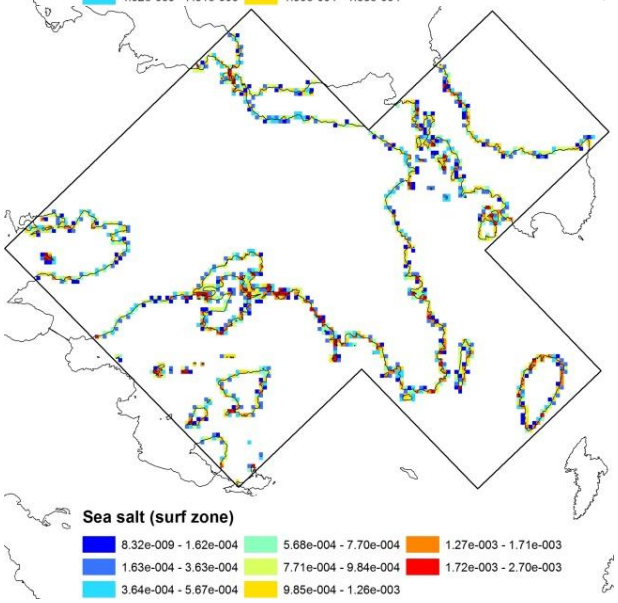
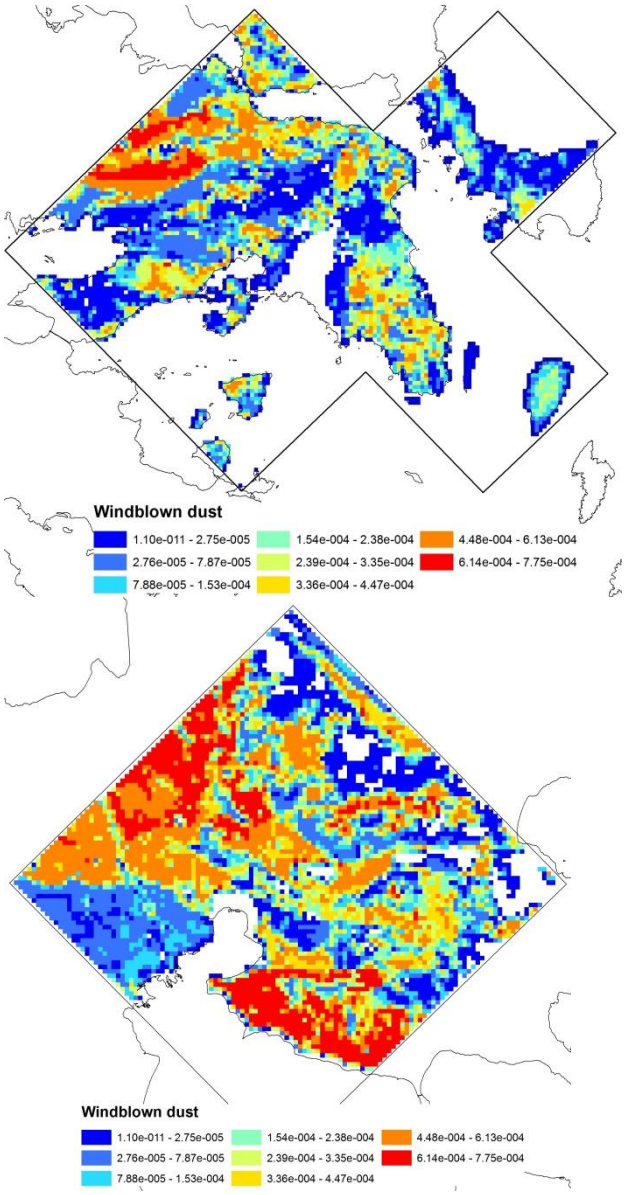


Figure 7 Monthly variation of BVOCs emissions in AMA, TMA and GVA during the period 2000-2010. Average, minimum and maximum monthly emission values are depicted.


Sea salt (open ocean)
2.93e-011 - 1.69e-005
1.70e-005 - 4.31e-005
4.32e-005 - 7.31e-005
7.32e-005 - 1.07e-004
1.08e-004 - 1.35e-004
1.36e-004 - 1.58e-004
1.59e-004 - 1.78e-004
1.79e-004 - 1.87e-004


Sea salt (surf zone)
8.32e-009 - 1.62e-004
1.63e-004 - 3.63e-004
3.64e-004 - 5.67e-004
5.68e-004 - 7.70e-004
7.71e-004 - 9.84e-004
9.85e-004 - 1.26e-003
1.27e-003 - 1.71e-003
1.72e-003 - 2.70e-003


Windblown dust
1.10e-011 - 2.75e-005
2.76e-005 - 7.87e-005
7.88e-005 - 1.53e-004
1.54e-004 - 2.38e-004
2.39e-004 - 3.35e-004
3.36e-004 - 4.47e-004
4.48e-004 - 6.13e-004
6.14e-004 - 7.75e-004
Windblown dust
1.10e-011 - 2.75e-005
2.76e-005 - 7.87e-005
7.88e-005 - 1.53e-004
1.54e-004 - 2.38e-004
2.39e-004 - 3.35e-004
3.36e-004 - 4.47e-004
4.48e-004 - 6.13e-004
6.14e-004 - 7.75e-004

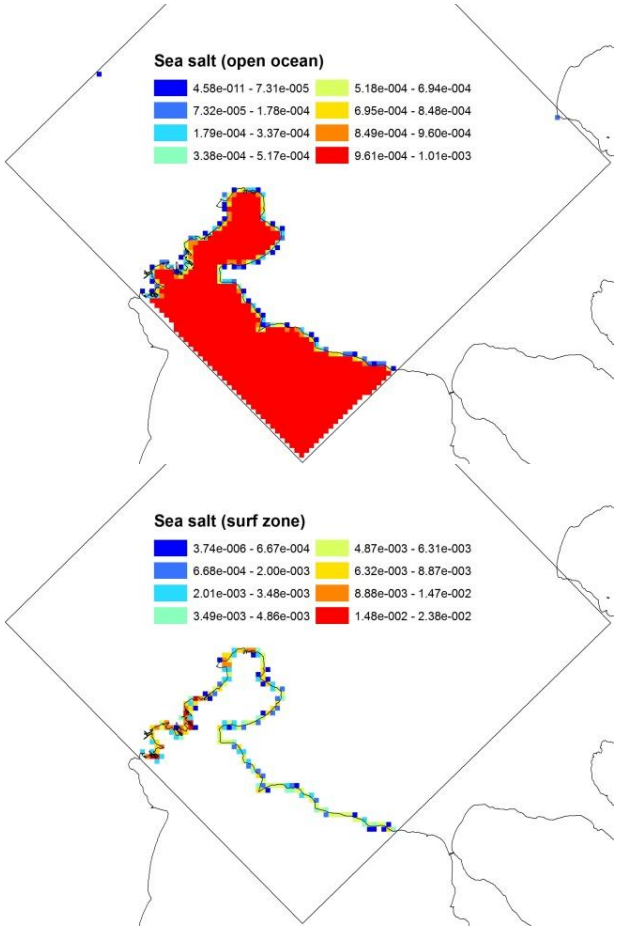
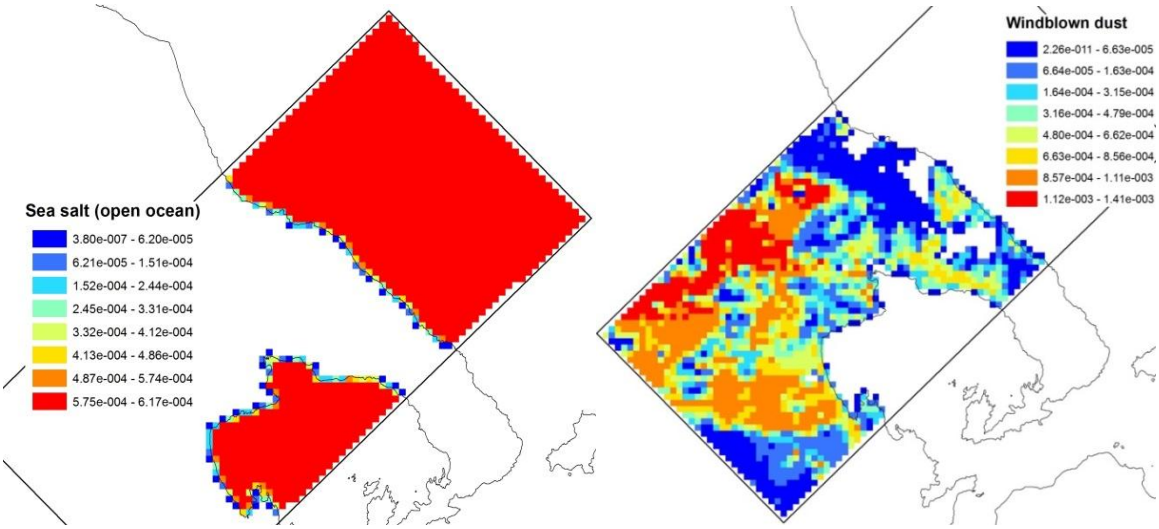


Figure 8 Spatial distribution coefficients for $PM_{2.5}$ and $PM_{2.5-10}$ emissions of sea salt (open-ocean), sea salt (surf zone) and windblown dust for AMA and TMA.



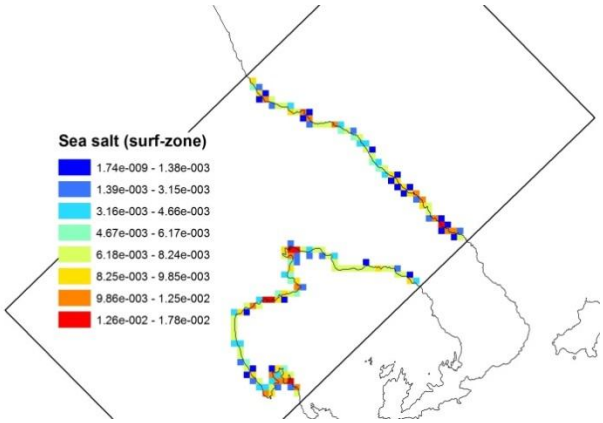


Figure 9 Spatial distribution coefficients for $PM_{2.5}$ and $PM_{2.5-10}$ emissions of sea salt (open-ocean), sea salt (surf zone) and windblown dust for GVA

The results of the calculations for monthly emissions of BVOCs during the period 2000-2010 are summarised in Figure 7. It was found that BVOCs emissions are increased from April to September (warm season) due to the enhanced solar radiation and temperature. In addition, the maximum monthly emissions were observed in July and the minimum during January at all areas. During the warm period are emitted 86.9±0.6%, 91.3±0.4% and 91.9±0.4% of annual BVOCs emissions in AMA, TMA and GVA, respectively.

2.3.2 Spatial distribution of natural emissions

Figures 8-9 present the weighting factors which were used for the spatial disaggregation of natural PM emissions for the period 2000-2010. Based on the assumptions the spatial distribution coefficients remain unchanged throughout the period 2000-2010. In particular, no landcover changes are incorporated in the calculations and the values of meteorological conditions are averaged over each area of interest thus the spatial disaggregation coefficients are the same for every year for PM emissions and the same for $PM_{2.5}$ and $PM_{2.5-10}$. Inter-annual changes were found only in the cells including coastal areas which were considered insignificant (up to 5.6×10^{-18}). It is observed in Figures 8-9 that natural PM emissions are scattered over arable land, areas with sclerophyllus vegetation and the sea. In particular, emissions of sea salt particles are equally distributed over the open sea whereas the variation of values at coastal cells corresponds to differences in the length and complexity of the shoreline. The spatial variation of windblown dust emissions is attributed to differences in the soil texture and landcover. With regards to landcover, the emission rates increase in the order of partly built-up areas to dense forest with higher values over agricultural land. The same trend applies also to the values of weighting factors with some discrepancy attributed to the aggregation of emissions in grid cells with variable landcover (see Figures 1-4). On the other hand, the weighting factors for BVOCs emissions spatial disaggregation vary seasonally and from year to year. In Figures 10 the

spatial weighting factors of BVOCs seasonal emissions are depicted. It is observed that BVOCs emissions in AMA are concentrated over the Northern part of the area, the Eastern part of the Attica peninsula, Evoia at the NE and the Gerania mountains at the W part of the domain where areas with significant natural vegetation occur (forests and semi-natural areas). In TMA BVOCs emissions are enhanced over the eastern part of the domain during both the warm and cold periods whereas in GVA emissions are mainly distributed over agricultural land. BVOCs emissions scattered over the whole domains are mainly attributed to OVOCs which are emitted from every non-artificial surface. Changes in seasonal emissions weighting factors are attributed to the seasonal variation in emission factors and foliar biomass densities. For example no emissions of BVOCs occur during the cold period over the deciduous forest in mountain Pilio in GVA.

2.3.3 Contribution from natural sources to primary emissions and total PM emissions in the areas of interest

The seasonal variability in contributions from natural sources to total $PM_{2.5}$ and $PM_{2.5-10}$ emissions in AMA is examined in Figures 11. Each box depicts the lower, the median and the upper quartile (bottom: 25^{th}, band: 50^{th} and top: 75^{th} percentile) of emission contributions for each month, whereas the point and the whiskers show the average, minimum and maximum values. It is observed that particles emitted at the sea shore have the largest contribution to PM emissions with monthly average values ranging from 17.3±2% to 27.5±1.7% for $PM_{2.5}$ and from 57.8±5.2% to 67.7±2.2% for $PM_{2.5-10}$. The minimum contribution values are depicted in February whereas the maximum in September for $PM_{2.5}$ and in May for $PM_{2.5-10}$. This is associated with the increase in anthropogenic PM emissions in AMA during the winter months. Additionally, the monthly contribution from sea salt particles emitted at the sea shore has relatively the smaller variation during the studied period as indicated by the box and whiskers plot (particularly for $PM_{2.5}$). On the other hand, the monthly contribution to $PM_{2.5}$ and $PM_{2.5-10}$ emissions from sea salt open-ocean particles and windblown dust shows large variation during the period 2000 -2010. The contributions from open ocean emissions of sea salt particles to total $PM_{2.5}$ and $PM_{2.5-10}$ emissions in AMA were in the range 10.4±3.1% - 17.9±6% and 17.4±2.7% - 26.4±3.3%, respectively. The monthly contribution values during the period 2000 – 2010 were ranged from 4.7% in May to 31.7% in July for $PM_{2.5}$ and from 11.9% in May to 36.4% in February for $PM_{2.5-10}$ emissions. For windblown dust emissions the corresponding values are 0.7% in May to 7.7% in July for $PM_{2.5}$ and from 2.3% in May to 15.7% in July for $PM_{2.5-10}$ emissions. In addition, the contributions from windblown dust emissions to total $PM_{2.5}$ and

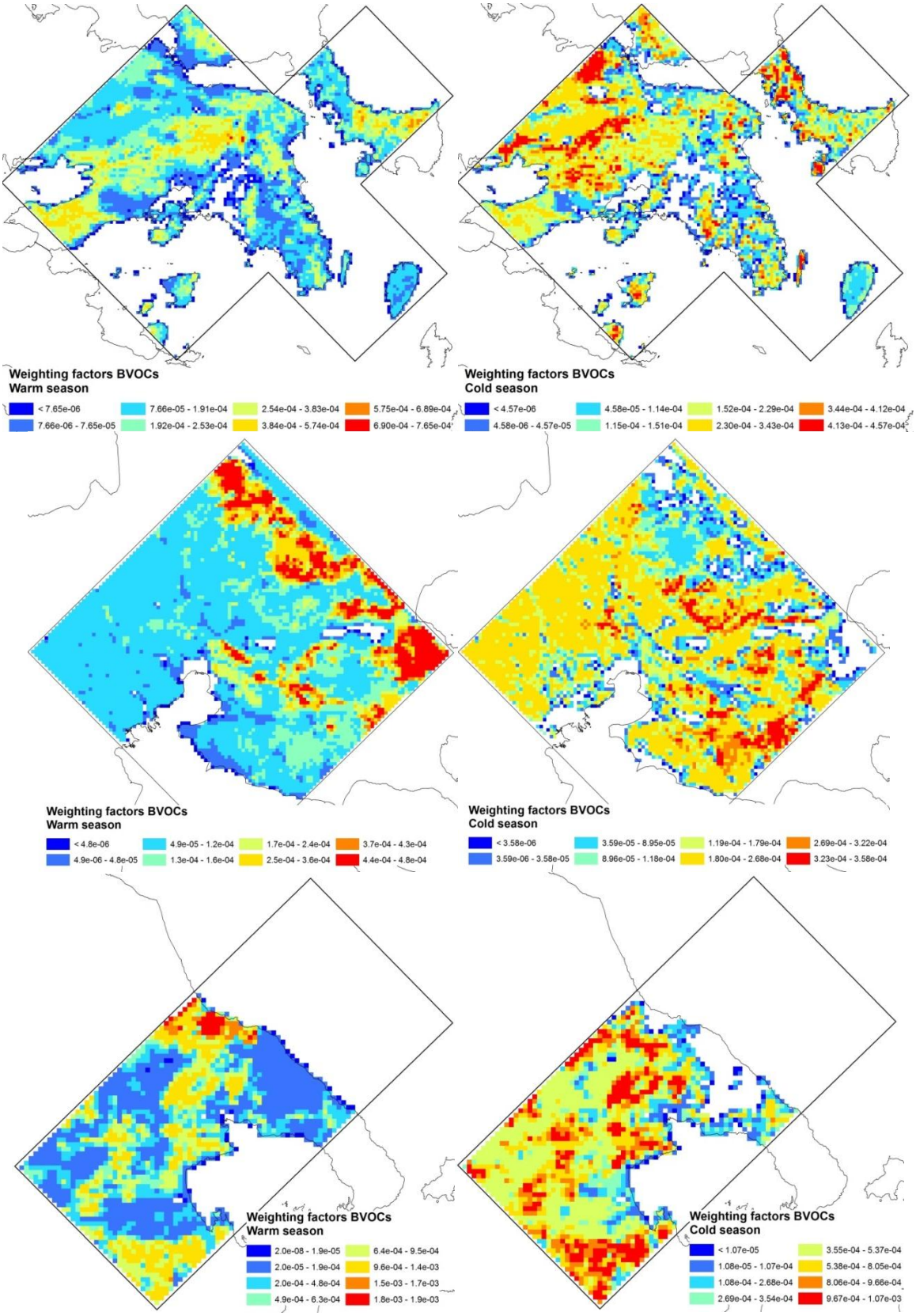
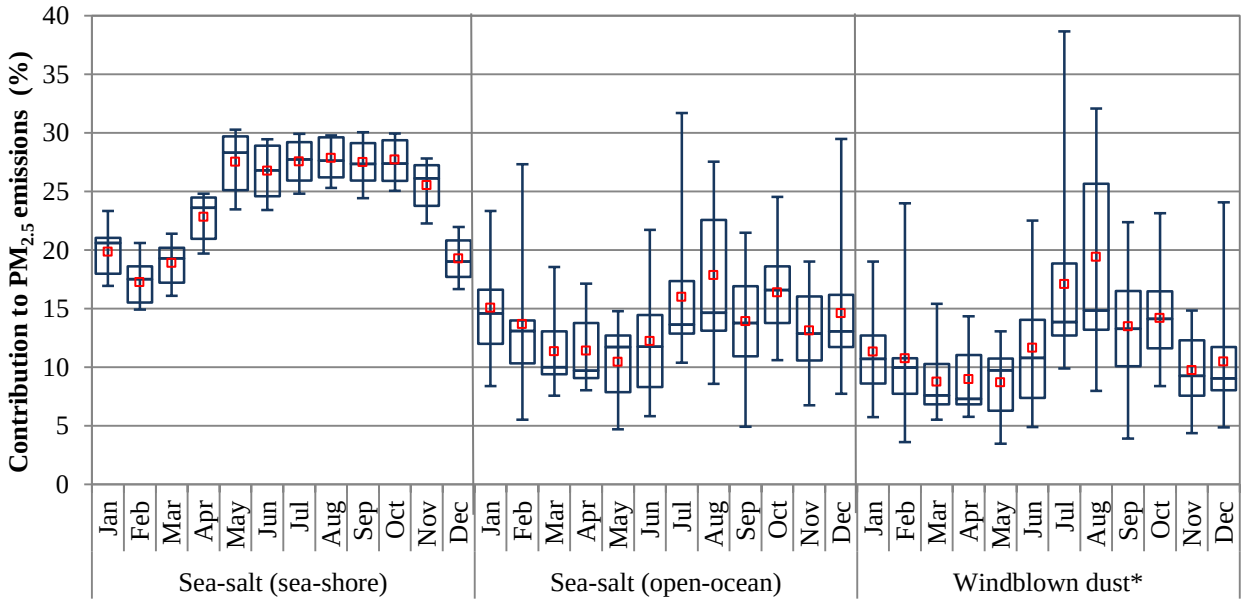


Figure 10 Comparison between warm and cold period spatial weighting factors for BVOCs emissions in AMA, TMA and GVA. The values in the legend correspond to the

1%, 10%, 25%, 33%, 50%, 75%, 90% and 100% percentile of weighting factor values for each period.

$PM_{2.5-10}$ emissions in AMA were in the range 1.7±0.6% - 3.9±1.6% and 4.8±1.3% - 9.4±3%, respectively.

The variability in monthly values of natural PM emissions' contribution to primary PM emissions in AMA is associated with the inter-annual differences in monthly wind-speed values and in anthropogenic PM emissions during the period 2000 - 2010. Overall, there was no significant difference in the relative contribution of natural $PM_{2.5-10}$ emissions to primary $PM_{2.5-10}$ emissions in AMA during the cold (October - March) and warm (April - September) period of the year (up to 1.96% for 2010; yearly average value of 91.97±1.69%). On the other hand the contribution of $PM_{2.5}$ emissions from natural sources to the total emissions over the area was generally enhanced during the warm period of the year (difference up to 9.53% for 2009; warm period average value of 43±3.9% compared to 37.6±4.4% during cold periods).



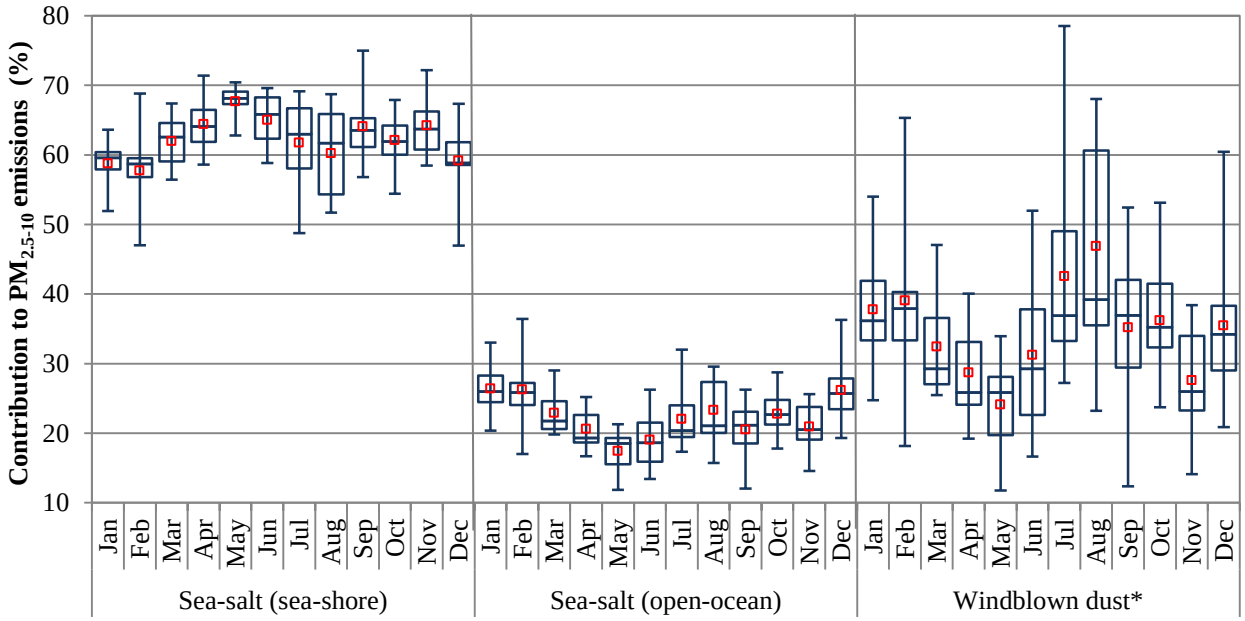


Figure 11 Variation of monthly natural emissions contribution to total primary $PM_{2.5}$ and $PM_{2.5-10}$ emissions in AMA during the period 2000-2010 (*Values multiplied by 5).

The seasonal variability in contributions from natural sources to total $PM_{2.5}$ and $PM_{2.5-10}$ emissions in TMA is depicted in Figures 12. It is observed that the monthly variation in contributions is similar to AMA, however with smaller values (due to land cover differences in the two areas). Particles emitted at the sea shore have the largest contribution to $PM_{2.5}$ emissions with monthly values in the range from 6.1±0.7% to 12.1±0.7%. The minimum contribution value is depicted in February (5.1%) whereas the maximum in August (13%). Additionally, the mean monthly contribution to total $PM_{2.5}$ emissions from sea-salt open-ocean particles during the period 2000-2010 in TMA were 1.2±0.1%, with the minimum monthly average observed in December and the maximum in November. Moreover the mean monthly contribution of windblown dust to total $PM_{2.5}$ emissions was 1.3 ±0.1% whereas the minimum and maximum values were observed in December and August-September, respectively. The monthly variation is mainly associated with seasonal changes in anthropogenic emissions rather than changes in meteorological conditions.

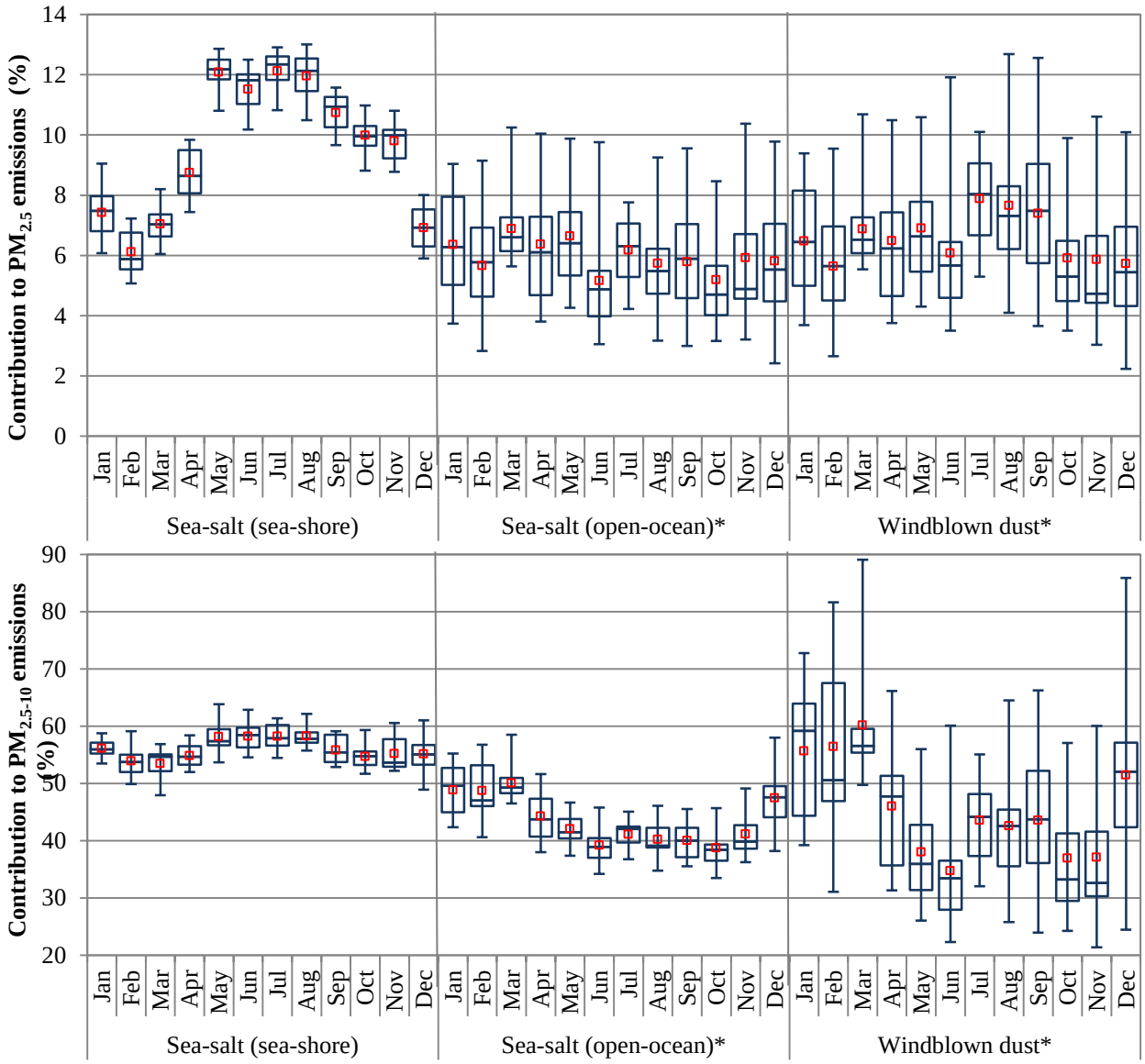


Figure 12 Variation of monthly natural emissions contribution to total primary $PM_{2.5}$ and $PM_{2.5-10}$ emissions in TMA during the period 2000-2010 (*Values multiplied by 5).

With regard to $PM_{2.5-10}$ emissions, the monthly contribution from natural sources was ranged from 7.8±0.7% to 10±0.6% for sea salt particles emitted from open-ocean, and from 6.9±2.2% to 12±2% for windblown dust. Monthly contributions of sea salt particles emitted at sea shore were large compared to the other sources, approximately 55.8±2.1%. Overall, there was no significant difference in the relative contribution of natural emissions to primary $PM_{2.5}$ and $PM_{2.5-10}$ emissions in TMA during the cold (October - March) and warm (April - September) period of the year (average values for: $PM_{2.5}$ warm period 13.8±1.0%, cold period 10.3±0.8%; $PM_{2.5-10}$ warm period 73.8±2.6%, cold period 73.8±2.1%).

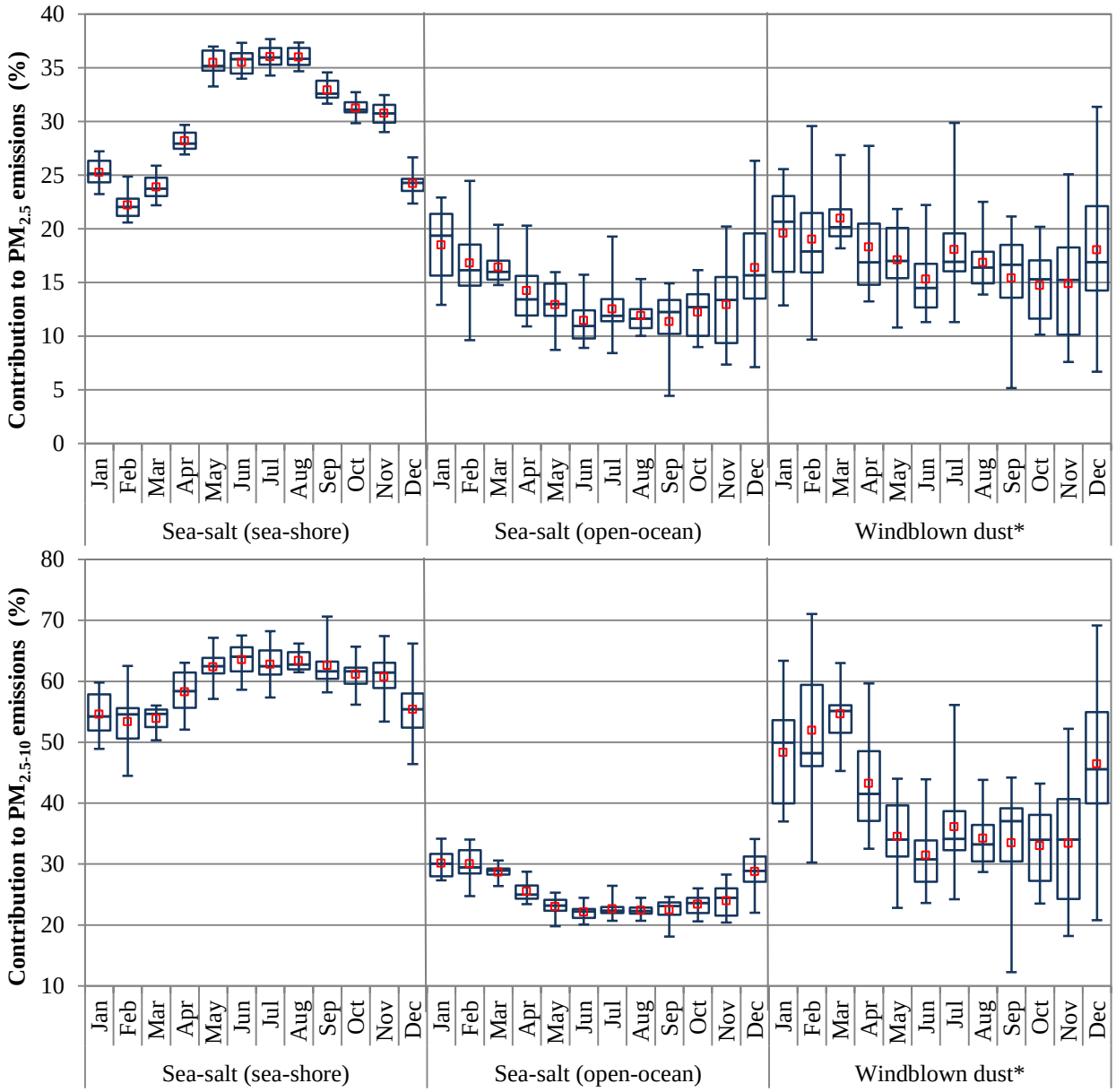


Figure 13 Variation of monthly natural emissions contribution to total primary $PM_{2.5}$ and $PM_{2.5\text{-}10}$ emissions in GVA during the period 2000-2010 (*Values multiplied by 5).

The results on the monthly contribution from natural sources to total $PM_{2.5}$ and $PM_{2.5\text{-}10}$ emissions in GVA during the period 2000-2010 are depicted in Figures 13. Similarly to the results for AMA and TMA, particles emitted at the sea shore have the largest contribution to $PM_{2.5}$ and $PM_{2.5\text{-}10}$ emissions with monthly values in the range from 22.2±1.3% to 36±1.1% and 53.3±4.5% to 63.5±2.6%, respectively. The minimum contribution values were depicted in February (20.6% and 44.5%) whereas the maximums in July (37.7%) and September (70.7%) for $PM_{2.5}$ and $PM_{2.5\text{-}10}$, respectively. The monthly average contributions from sea salt open-ocean particles and windblown dust during the period 2000 -2010 in GVA were 14.7±1.2% and 3.6±0.3% and 25.9±0.8% and 8.4±0.6% to total

$PM_{2.5}$ and $PM_{2.5-10}$ emissions, respectively. The minimum monthly averages were observed in September for both sources and size fractions and the maximum in December for $PM_{2.5}$ and in January and February for $PM_{2.5-10}$ for SS and WB, respectively. Overall, there was no significant difference in the relative contribution of natural emissions to primary $PM_{2.5}$ and $PM_{2.5-10}$ emissions in GVA during the cold (October - March) and warm (April - September) period of the year (average values for: $PM_{2.5}$ warm period 49.8±1.6%, cold period 45.4±2.2%; $PM_{2.5-10}$ warm period 92.3±2%, cold period 92.9±1.8%).

Overall, the monthly contribution from natural sources to total primary PM_{10} emissions during the period 2000-2010 ranged from 73.8±6.1% in March to 82.8±3.7% in July in AMA, 41.3±3.5% in February to 51.9±2.6% in July in TMA, and 82.7±1.9% in February to 85.1±2.1% in July in GVA. The lower variation in monthly PM_{10} contributions during the period 2000-2010 was found in GVA where the natural emissions are the most dominant PM source and also the anthropogenic emissions have the lower inter-annual variation (1075±113 t). The variation in annual anthropogenic emissions from GVA is mainly attributed to the steady increase of non-industrial combustion emissions (18% from 2000 to 2010).

Finally, the monthly variation of natural sources contribution to total primary and secondary PM_{10} emissions in AMA, TMA and GVA are depicted in Figure 14. Particles emitted at the sea shore have the largest, compared to other natural sources of emissions, contribution to PM_{10} equivalent emissions in all areas of interest with monthly values in the range from 19±1.8% to 21.9±1.7% in AMA, 7.2±1% to 8.6±1.2% in TMA and 17.5±3.6% to 19.6±3.4% in GVA. Sea-salt emitted at sea-shore are the second significant natural contributor to PM_{10} equivalent emissions in AMA and GVA with monthly average contribution values in the range of 5.2±1.1 to 10.9±3.2% and 6±1.1% to 11.4±3.3%, respectively. Moreover, the monthly averages of contribution values for WB dust range from 1.3±0.4% to 3.4±1.5% in AMA, 0.9±0.3% to 1.9±0.5% in TMA and 1.7±0.4% to 3.9±0.9% in GVA. The contribution of BVOCs to total PM_{10} equivalent emissions was increased during the warm period due to the enhanced solar radiation and temperature. The average contribution of BVOCs to total PM_{10} equivalent emissions (anthropogenic and natural) ranged from 0.05±0.01% - 0.7±0.1% in AMA, 0.2±0.03% - 4.4±0.9% in TMA and 0.12±0.03% - 3.02±0.86% in GVA.

Overall the contribution from natural sources to total primary and secondary PM_{10} equivalent emissions ranged from 25.8±2.8% in May to 35.6±6.9% in January in AMA, from 10.2±1.4% in November to 14±1.9% in July in TMA and from 27.5±5.3% in June to 34.6±7.7% in January in GVA. The contribution of natural emissions to total primary and secondary PM_{10} emissions during the cold and warm periods in AMA was 33.1±3.2% and 29.8±2.9%, respectively. Likewise, in TMA the contribution of natural emissions to total primary and secondary PM_{10}

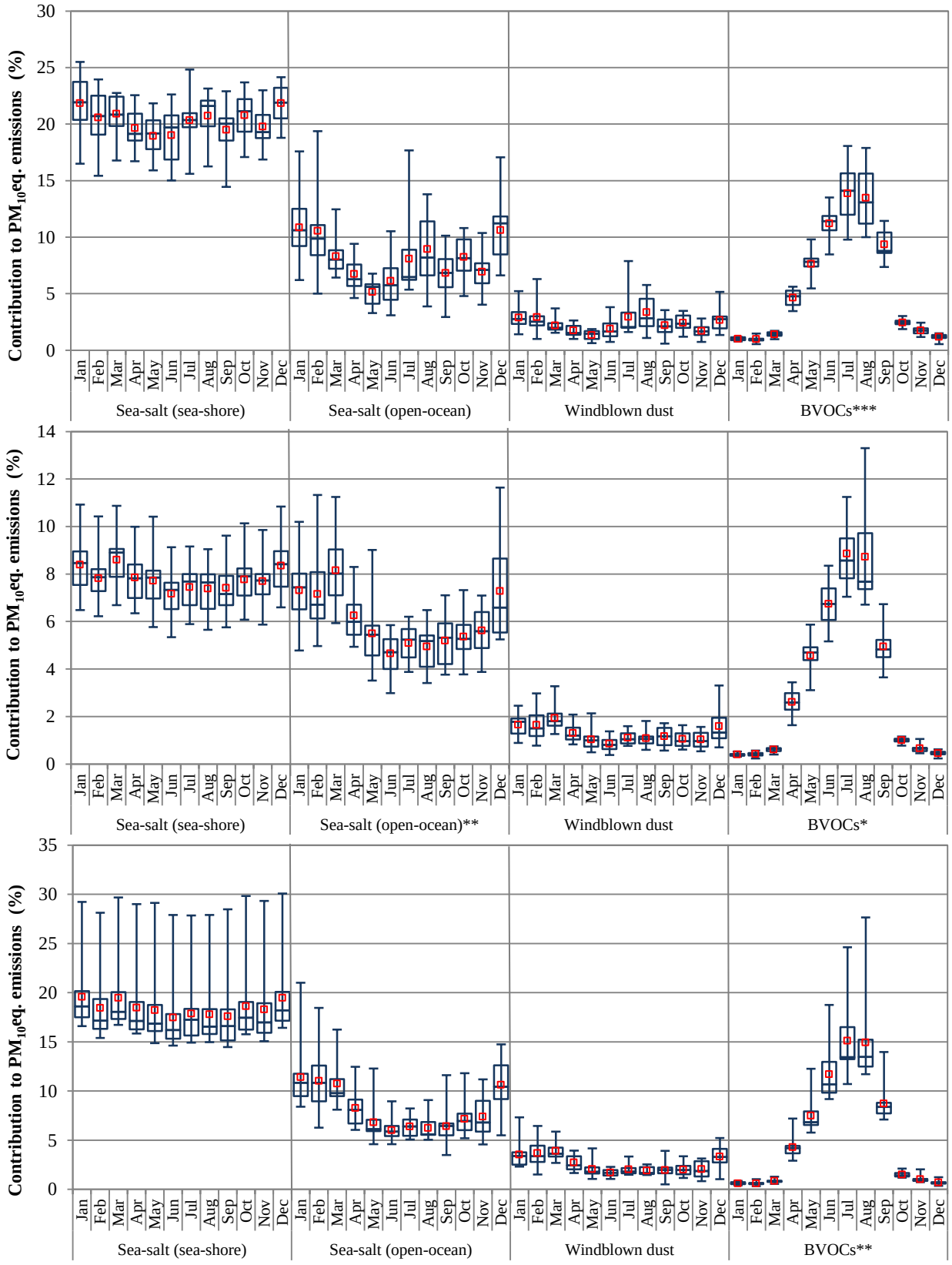


Figure 14 Variation of monthly natural emissions contribution to total primary and secondary PM_{10} emissions (PM_{10} equivalents) in AMA, TMA and GVA during the period

2000-2010 (*Values are multiplied by 2; **Values are multiplied by 5; ***Values are multiplied by 20).

emissions was 11.3±1.5% during the cold and 12.7±1.8% during the warm periods, whereas in GVA the respective seasonal contributions were 32.2±6.4% and 28.7±5.8%.

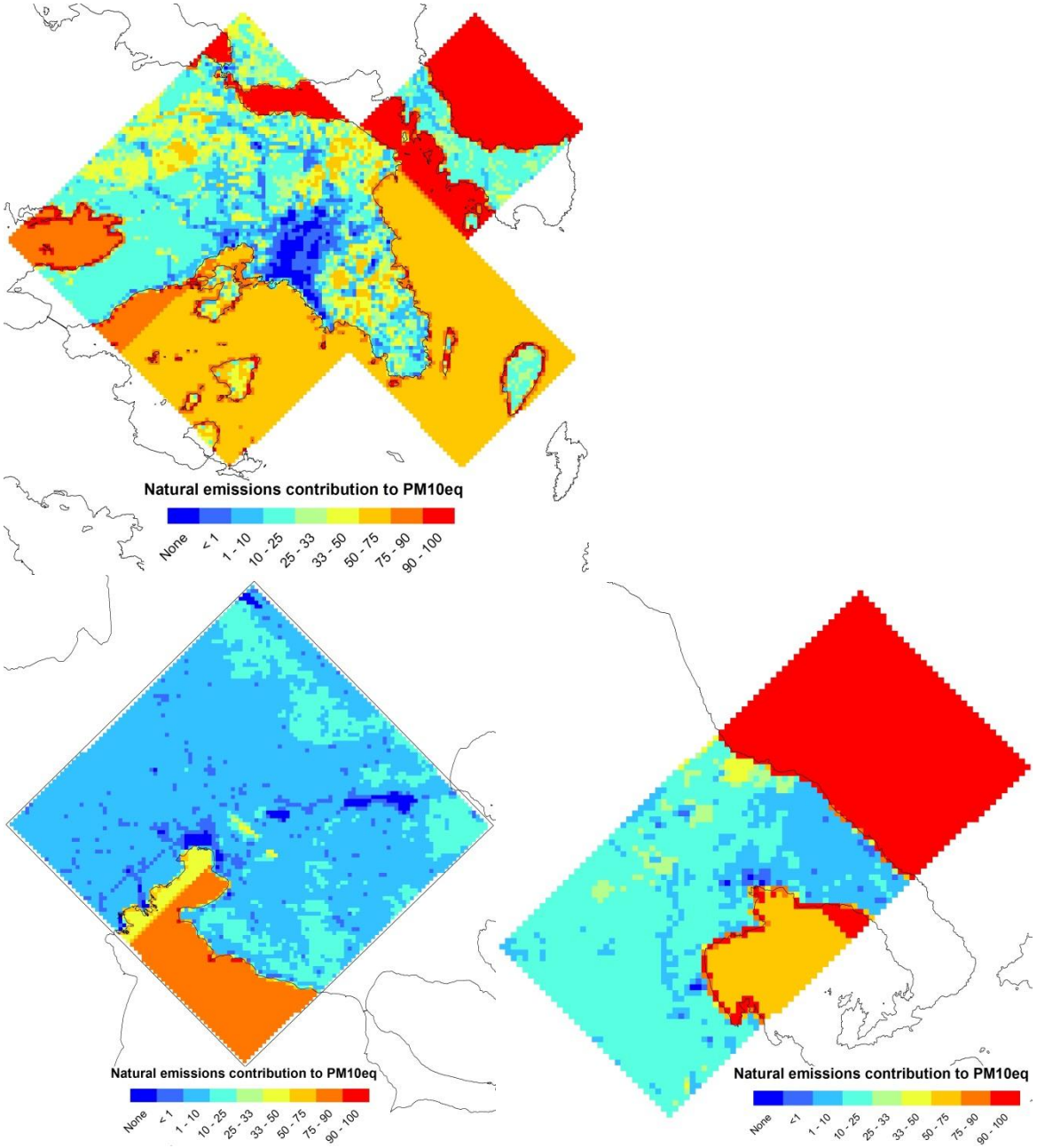


Figure 15 Spatial distribution of the average contribution from natural sources to total anthropogenic and natural primary and secondary PM_{10} equivalent emissions in the AMA, TMA and GVA* during the period 2000 – 2010 (*2008 values).

In Figure 15 is presented the spatial distribution of the average contribution from natural sources to total anthropogenic and natural primary and secondary PM_{10} equivalent

emissions in the AMA and TMA during the period 2000 – 2010. Also is depicted the spatial distribution of the contribution from natural sources to total anthropogenic and natural primary and secondary PM_{10} equivalent emissions in the GVA during 2008. It is observed that anthropogenic and natural emissions are concentrated over different areas. In particular, the average contribution from natural sources in AMA was enhanced over marine waters (75.5±23.5%; sea-salt particles), agricultural areas (31.2±24.7%) and forests (34.7±29.6%). The highest contributions over land were observed over open spaces with little or no vegetation (51.7±30.1%; windblown dust) in particular bare rocks (65.4±31.9%) and beaches (45.4±27%), over natural grasslands (43.9±34.7%) and areas with sclerophyllous vegetation (41.4±33.2%), and over pastures (43.3±32.6%). Over artificial surfaces the contribution, where it is not null, ranges from 0.1±0.2% for green urban areas to 42.7±30.6% for port areas (average 15.5±23.9%).

For TMA the contribution from natural sources was too low compared to anthropogenic sources except for marine areas (38.4±34.7%). In agricultural areas (5.7±6.9%) the contribution ranged from 2.6±2.7% for olive groves to 9.1±16.8% for complex cultivation patterns, whereas in forest and seminatural areas (7.3±5.2%) the contribution ranged from 4.6±3.3% for beaches to 11.2±5.4% for mixed forests. The contribution from natural sources to PM_{10} equivalent emissions over artificial surfaces in TMA, where it was not null, ranged from 1±0.8% for road and rail networks and associated land to 29.8±30.2% for airport areas (average 4.1±10.3%). Finally for GVA the contribution from natural sources was 77.2±29% for marine areas, 23.9±26.4% for agricultural areas, 23.4±26.5% for forest and seminatural areas and 19.7±28.3% over artificial surfaces. In particular over agricultural areas the contribution ranged from 11.5±3.9% for permanently irrigated land to 31.9±35.4% for land principally occupied by agriculture, with significant areas of natural vegetation, whereas in forest and seminatural areas the contribution ranged from 16.2±14.4% for coniferous forests to 50.8±36.5% for mixed forests. The contribution from natural sources to PM_{10} equivalent emissions over artificial surfaces in GVA, where it was not null, ranged from 2.8±4.1% for continuous urban fabric to 75.3±11% for sport and leisure facilities.

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