ACEPT-AIR

LIFE+ 09 ENV/GR/000289

Action 4

Deliverable D10.

TITLE: Emission inventories for the three urban areas (AMA, TMA, GVA), for anthropogenic and natural sources, for the past decade (2000-2010)

August 2012

Coordinated by:

Technical
University
of Crete

D10. Emission inventories for the three urban areas (AMA, TMA, GVA), for anthropogenic and natural sources, for the past decade (2000-2010)

EXECUTIVE SUMMARY

One of the aims of this job is to compile anthropogenic emission inventories with focus on particulate matter ($PM_{2.5}$ and PM_{10}) for the Athens and Thessaloniki metropolitan areas (AMA and TMA respectively) and the greater area of Volos (GVA). Emissions from all major sources were calculated including navigation and aviation emissions.

Road transport has become by far the major source of atmospheric pollution and traffic congestion in urban areas. For the estimation of emissions from road transportation, the newer version of COPERT IV code, a user-friendly MS Windows software application (Ntziachristos et al., 2009) was applied. Despite the increase of the population of circulating vehicles, as there is a remarkable increase of less polluting vehicles, NOx, NMVOC and PM emissions decrease substantially. Finally, another considerable reduction of SO_2 emissions attributed to the improvement of the fuels characteristics (i.e. the reduction of their sulphur content) is obsereved in 2010. From the results for the period 2000-2010, it seems that PM10 and NOx concentrations are strongly associated with the corresponding road traffic related emissions. Moreover, PM10 concentrations show a considerable long term similarity with the corresponding emissions of NOx and NMVOCs.

The air pollution inventory for the Industrial Sector includes emissions deriving from Fuel Combustion and emissions related to the actual (Industrial) Production Processes themselves. The estimated emissions reflect the overall contribution of the Industry to the deterioration of air quality in the areas of study. The emission factors used are provided by the EMEP/EEA air pollutant emission inventory guidebook that has been released by the European Environmental Agency (EEA) in 2009. Fuel consumption data were used as well as more detailed emission data if available (especially for large industrial installations). Industrial emissions present a considerable decrease of all pollutants emissions in 2009 and 2010. This decrease is associated with the economic crisis.

The residential and commercial sector considers non-industrial (stationary) combustion processes such as residential activities in households and in institutional and commercial buildings (e.g. heating and cooking with fireplaces, stoves, cookers and small boilers), having a thermal capacity equal or lower than 50 MW. According to the calculation results, a considerable decrease in all pollutants emissions is observed in 2009 and 2010 again as a result of the fuel consumption decrease because of the economic crisis.

Navigation air pollutants emissions in 2010 were higher (43%) than the emissions in 2000, on the basis of fuel consumption data from this sector. Finally, air pollutant emissions from aviation increased by 82% since 1990 with an average annual increase rate of approximately 4% (MEEC, 2012). Although total emissions from aviation in Greece have a significant increasing trend from 1990, in the Athens airport as well as in the Thessaloniki airport, for the decade 2000-2010, only small fluctuations, not exceeding 20%, occur.

In addition, the objective of this work was to estimate the contribution from natural sources to particulate matter ($PM_{2.5}$ and $PM_{2.5\text{-}10}$) primary and secondary emissions over the Athens and Thessaloniki metropolitan areas (AMA and TMA respectively) and the greater area of Volos (GVA) and study the temporal trend in emissions during the period 2000 - 2010. Therefore emission inventories for PM from natural sources were created and compared to anthropogenic emissions in the areas. 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 results showed that the contribution from natural sources was significant, especially in the case of coarse particles (64.9 Gg per year for AMA, ~79%; 4.99 Gg per year for TMA, ~46%; 5.3 Gg per year for GVA, ~83%). In particular, the average contribution from the sea surface to the total particulate pollution over the AMA, TMA and GVA during the decade was approximately 37%, 10% and 44% for $PM_{2.5}$, respectively, while it was approximately 85%, 65% and 84% for $PM_{2.5\text{-}10}$. Windblown dust accounted for a relatively small fraction of total natural PM emissions in AMA, TMA and GVA (~8%; ~12.5%; ~9%). In addition, BVOCs emissions accounted for approximately 0.3%, 1.6% and 1.1% of total PM emitted from the AMA, TMA and GVA, respectively. It was also found that except for AMA natural PM emissions have increased from the beginning to the end of the studied period whereas their relative contribution to total PM_{10} emissions has increased in all areas (from 0.9% in AMA to 88% in GVA).

ΠΕΡΙΛΗΨΗ

Στόχος της παρούσας εργασίας είναι η απογραφή των εκπομπών ανθρωπογενών πηγών στις ευρύτερες περιοχές της Αθήνας, της Θεσσαλονίκης και του Βόλου. Υπολογίσθηκαν οι εκπομπές από όλες τις κύριες πηγές συμπεριλαμβανομένων των εκπομπών από τις εναέριες και θαλάσσιες μεταφορές.

Οι οδικές μεταφορές θεωρούνται ως η κύρια πηγή ατμοσφαιρικής ρύπανσης αλλά και κυκλοφοριακής συμφόρησης στις αστικές περιοχές. Για τον υπολογισμό των εκπομπών από τις οδικές μεταφορές εφαρμόσθηκε η νεότερη έκδοση του μοντέλου COPERT IV

(Ntziachristos et al., 2009). Παρά την σημαντική αύξηση του αριθμού των κυκλοφορούντων οχημάτων, λόγω της εισαγωγής στην κυκλοφορία νέας τεχνολογίας, λιγότερο ρυπογόνων οχημάτων, οι εκπομπές όλων των ρύπων μειώνονται σημαντικά. Ειδικότερα το διοξείδιο του θείου παρουσιάζει σημαντικές μειώσεις οι οποίες οφείλονται στην βελτίωση των χαρακτηριστικών των καυσίμων. Από τα αποτελέσματα προκύπτει ότι οι συγκεντρώσεις PM10 και NOx παρουσιάζουν σημαντική συσχέτιση με τις αντίστοιχες εκπομπές από την κυκλοφορία, ενώ ειδικότερα οι συγκεντρώσεις PM10 παρουσιάζουν συσχέτιση και με τις εκπομπές NOx και NMVOCs.

Η απογραφή των εκπομπών από την βιομηχανία περιλαμβάνει εκπομπές λόγω καύσης και εκπομπές που προέρχονται από την παραγωγική διαδικασία. Οι συντελεστές εκπομπής που χρησιμοποιούνται προέρχονται από την έκθεση «EMEP/EEA Air pollutant emission inventory guidebook» η οποία εκδόθηκε από την Ευρωπαϊκή Υπηρεσία Περιβάλλοντος το 2009. Για τους υπολογισμούς χρησιμοποιήθηκαν δεδομένα κατανάλωσης καυσίμου καθώς και πιο αναλυτικά στοιχεία της δραστηριότητας, στην περίπτωση που ήταν διαθέσιμα (κυρίως στις μεγαλύτερες εγκαταστάσεις). Οι εκπομπές από την βιομηχανία παρουσιάζουν σημαντική μείωση το 2009 και το 2010 ως αποτέλεσμα της οικονομικής κρίσης.

Οι εκπομπές του οικιακού και εμπορικού τομέα περιλαμβάνουν διεργασίες καύσης σταθερών πηγών όπως η θέρμανση σε κτίρια κατοικιών αλλά και επαγγελματικών ή εμπορικών χρήσεων. Σύμφωνα με τα αποτελέσματα των υπολογισμών, σημαντική μείωση των εκπομπών παρατηρείται κατά τα έτη 2009 και 2010 και πάλι ως συνέπεια της οικονομικής κρίσης.

Οι εκπομπές από τις θαλάσσιες μεταφορές κατά το έτος 2010 ήταν υψηλότερες κατά 43% από τις εκπομπές του 2000, σύμφωνα με τα στατιστικά στοιχεία κατανάλωσης καυσίμου. Τέλος, οι εκπομπές από τις εναέριες μεταφορές στην Ελλάδα, αυξήθηκαν κατά 82% από 1990 με μέσο ρυθμό αύξησης 4% (MEEC, 2012). Αν και οι συνολικές εκπομπές στην Ελλάδα παρουσιάζουν σημαντική αύξηση από το 1990, στο αεροδρόμιο των Αθηνών και αντίστοιχα στο αεροδρόμιο Θεσσαλονίκης, για την δεκαετία 2000-2010 μόνο μικρές διακυμάνσεις παρατηρούνται οι οποίες δεν υπερβαίνουν το 20%.

Ένας άλλος στόχος της παρούσας εργασίας είναι η μελέτη της ετήσιας μεταβολής των εκπομπών αιωρούμενων σωματιδίων ($ΑΣ_{2.5}$ και $ΑΣ_{2.5\text{-}10}$) από φυσικές πηγές στις μητροπολιτικές περιοχές Αθηνών (AMA), Θεσσαλονίκης (TMA) και Βόλου (GVA) και της συνεισφοράς των εκπομπών από φυσικές πηγές στις συνολικές εκπομπές των περιοχών ενδιαφέροντος την περίοδο 2000-2010. Για το σκοπό αυτό κατασκευάστηκαν μητρώα εκπομπών ΑΣ από φυσικές πηγές και συγκρίθηκαν με μητρώα ανθρωπογενών εκπομπών στις περιοχές. Τα μητρώα περιλαμβάνουν τις εκπομπές ΑΣ από το έδαφος αγροτικών και κενών εκτάσεων εξαιτίας της αιώρησής 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. Τα αποτελέσματα έδειξαν πως η συνεισφορά από τις φυσικές πηγές στις εκπομπές ΑΣ ήταν σημαντική, ειδικά στην περίπτωση των ΑΣ$_{2.5\text{-}10}$ (64.9 Gg ανά έτος στην AMA, ~79%; 4.99 Gg ανά έτος στην TMA, ~46%; 5.3 Gg ανά έτος στην GVA, ~83%). Ειδικότερα, η μέση συνεισφορά των σταγονιδίων θαλάσσιου άλατος στις συνολικές εκπομπές ΑΣ στις περιοχές AMA, TMA και GVA κατά την δεκαετία ήταν περίπου 37%, 10% και 44% για τα ΑΣ$_{2.5}$, αντίστοιχα, ενώ για τα ΑΣ$_{2.5\text{-}10}$ ήταν περίπου 85%, 65% και 84%. Επιπρόσθετα, η σκόνη από το έδαφος αποτελεί μικρό ποσοστό των φυσικών εκπομπών ΑΣ στις περιοχές AMA, TMA και GVA (~8%; ~12.5%; ~9%) όπως και οι εκπομπές Β.Π.Ο.Ε στις συνολικές εκπομπές πρωτογενών και δευτερογενών ΑΣ στις περιοχές ενδιαφέροντος (0.3%, 1.6% και 1.1%). Τέλος, οι εκπομπές ΑΣ από φυσικές πηγές βρέθηκαν, με εξαίρεση την AMA, αυξημένες σε σχέση με την αρχή της δεκαετίας ενώ η σχετική συνεισφοορά τους στις ολικές πρωτογενείς και δευτερογενείς εκπομπές ΑΣ αυξήθηκε σε όλες τις περιοχές (από 0.9% στην AMA ως και 88% στην GVA).

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.

In a national scale, an emission inventory from industrial as well as from residential/institutional/commercial activities was carried out from the Ministry of Environment, Energy and Climate Change (MEECC) in the beginning of the '90s. In the same period, another inventory from mobile sources was accomplished (Symeonidis et al., 2003, 2004). Moreover, several emission inventories have been developed for Greece using both the bottom-up and top-down approaches. In particular, Aleksandropoulou and Lazaridis (2004) created an emission inventory of anthropogenic and natural sources in Greece. Poupkou et al. (2007) developed a spatially and temporally disaggregated anthropogenic emission inventory in the Southern Balkan region and Symeonidis et al. (2008) estimated biogenic NMVOCs emissions in the same region. Also Markakis et al. (2010) presented an anthropogenic emission inventory for gaseous species for Greece, Poupkou et al. (2008) studied the effects of anthropogenic emissions over Greece to ozone production, Sotiropoulou et al. (2004) estimated the spatial distribution of agricultural ammonia emissions in the AMA and Hayman et al. (2001) developed spatial air emission inventories using the top-down approach for large urban agglomerations in southern Europe including the AMA. Finally, Progiou and Ziomas (2011a) presented a road traffic emissions inventory for Greece for the period 1990-2009 as well as they associated road traffic emissions with the corresponding air pollutants levels for the greater Athens area (Progiou and Ziomas, 2011b).

The aim of this job is to compile anthropogenic emission inventories with focus on particulate matter ($PM_{2.5}$ and PM_{10}) for the Athens and Thessaloniki metropolitan areas (AMA and TMA respectively) and the greater area of Volos (GVA). Emissions from all major sources were calculated including navigation and aviation emissions.

1.2 Road Transportation

Road transport has become by far the major source of atmospheric pollution and traffic congestion in urban areas. Additionally, road traffic emissions contribute to global warming as they are also associated with CO_2, NH_3, CH_4 and N_2O emissions (Uherek et al, 2010; Smit et al., 2010).

Especially for the area of Greece, the total national emissions of the main pollutants as well as those associated with road transport emission data officially submitted by the country in the framework of the Convention on Long Range Transboundary Air Pollution (CLRTAP) and the European Monitoring and Evaluation *Programme* (EMEP) via the UNECE secretariat (CEIP, 2010), are presented in Table 1. As shown in the table, road transport plays a major role in CO emissions, contributing with 67% of total emissions, and represents an important part of NO_x and NMVOCs emissions with a contribution of 29 and 23% correspondingly. Obviously, in the scale of a city these contributions become even higher resulting in high air pollutants levels. This is the case of Athens, the capital and largest city of Greece, whose complex geomorphology along with its Subtropical Mediterranean climate are considered responsible for the air pollution problems the city is facing (Ziomas, 1998). Nonetheless, high particulates levels have been reported in other urban areas: the Thessaloniki Metropolitan Area (TMA) which is the second larger urban agglomeration in Greece and the Greater Volos Area (GVA) which is a city of medium size with emissions coming from road traffic and residential/institutional/commercial sector as well as from a considerable industrial activity.

Table 1. National emissions versus road transport emissions in Greece for the year 2008 (as reported under the CLRTAP)

2008 Emissions (Gg)	**NO_X**	**NMVOC**	**SO_2**	**NH_3**	**CO**
TOTAL NATIONAL EMISSIONS	356.87	218.66	447.55	63.1	685.01
ROAD TRANSPORT EMISSIONS	104.41	50.33	2.03	2.67	461.11
% CONTRIBUTION OF ROAD TRANSPORT	29.26%	23.02%	0.45%	4.23%	67.31%

1.2.1 Methodology

Emissions from road transport are calculated either from a combination of total fuel consumption data and fuel properties or they result from a combination of specific emission factors and road traffic data.

For the estimation of emissions from road transportation, the newer version of COPERT IV code, a user-friendly MS Windows software application (Ntziachristos et al., 2009) was applied. The software contains all the algorithms and the necessary input parameters to estimate total road transport emissions on a national, regional or local/urban scale with the possibility of a year to day-long resolution.

COPERT 4 is an MS Windows software program aiming at the calculation of air pollutant emissions from road transport. The technical development of COPERT is financed by the European Environment Agency (EEA), in the framework of the activities of the European Topic Centre on Air and Climate Change. Since 2007, the European Commission's Joint Research Centre has been coordinating and financing the further scientific development of the model. In principle, COPERT has been developed for use from the National Experts to estimate emissions from road transport to be included in official annual national inventories. In this version of COPERT hybrid vehicle fuel consumption and emission factors were introduced as well as N_2O/NH_3 emission factors for PCs and LDVs and heavy duty vehicle emissions calculation methodology.

The major revisions made since previous version of the methodology are the following:

- New emission factors for diesel Euro IV PCs
- Revised emission factors for LDVs
- New emission factors for Euro V and VI PCs, LDVs and HDVs
- Emission factors for urban CNG buses
- Hybrid fuel consumption and emission factors
- New corrections for emission degradation due to mileage
- Revised CO_2, N_2O, NH_3 and CH_4 calculations
- Effect of biodiesel blends on emissions from diesel cars and HDVs
- Split of NOx emissions to NO and NO_2
- Developments on the cold start emission front
- Developments on evaporation losses

The methodology applied is also part of the EMEP/CORINAIR Emission Inventory Guidebook. The Guidebook, developed by the UNECE Task Force on Emissions Inventories and Projections, is intended to support reporting under the UNECE Convention on Long-Range Transboundary Air Pollution (CLRTAP) and the EU directive on national emission ceilings. The COPERT 4 methodology is fully consistent with the Road Transport chapter of the Guidebook. The use of a software tool to calculate road transport emissions allows for a transparent and standardized, hence consistent and comparable data collecting and emissions reporting procedure, in accordance with the requirements of international conventions and protocols and EU legislation.

Basic data requirements for the application of the model include: (a) energy consumption by fuel type, (b) fuel characteristics, (c) the number of vehicles per vehicle category, engine size or weight and emission control technology, (d) other parameters such as: the mileage per vehicle class and per road class, the average speed per vehicle type and per road (urban, rural and highway) and (e) climatic conditions. The energy consumption as well as the associated emissions are calculated based on those data and a number of equations described in Ntziachristos and Samaras (2000).

It should be noted here that COPERT IV, is a simulation model for road transport sector and not an optimization one. The solution algorithm is based on the minimisation of differences between energy consumption as reported in the national energy balance account and the estimated (by the model) energy consumption. This is achieved by adjusting appropriately the mileage driven by each vehicle category.

Especially for PM10 emissions calculations, two categories were taken into account: a) exhaust emissions (resulting from the fuel combustion in the vehicles engines) and b) emissions resulting from tyre, brake and road surface wear.

1.2.2 Results

In the last two years, as a result of economic crisis, the traffic characteristics applied for each vehicle type and category had to be further investigated. More specifically, the annual mileage driven was reconsidered for all vehicle categories as a result of economic crisis. The annual mileage was reassessed taking into account fuel consumption data. The different vehicle categories population along with the total annual kilometres driven by each category as well as fuel consumption data are presented in ***Figures 1-4***.

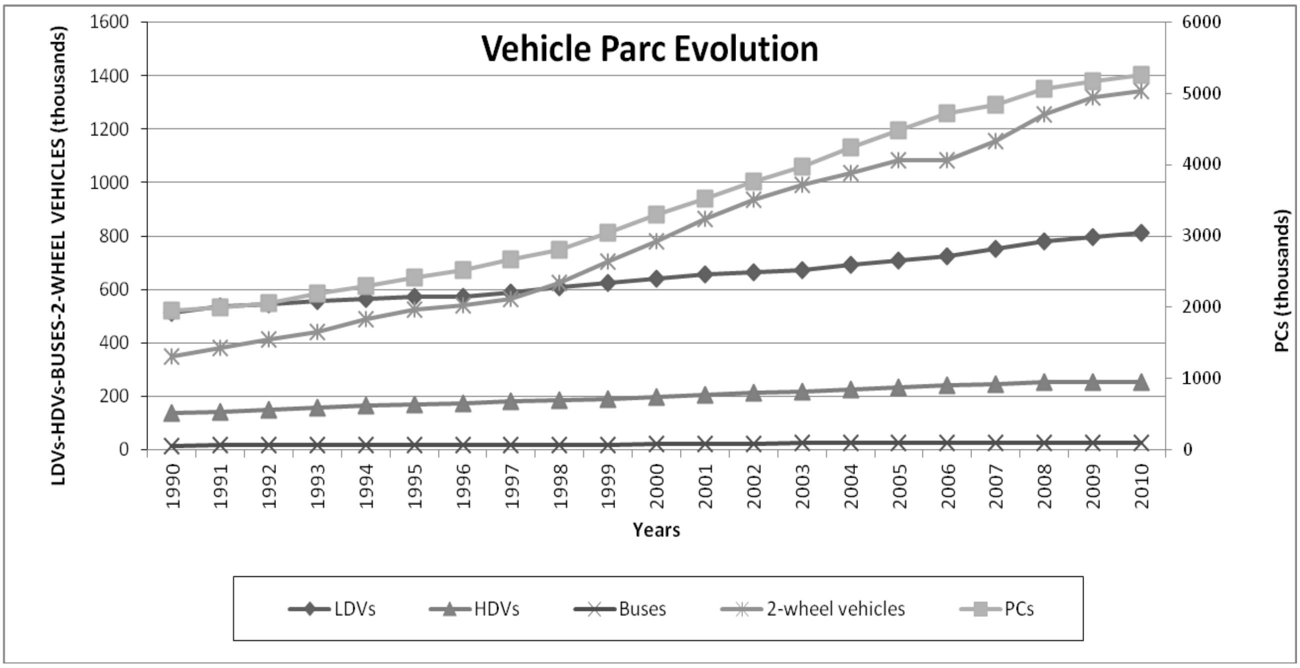


Figure 1 ***Vehicles population evolution for all vehicles categories during the whole time period 1990 – 2010***

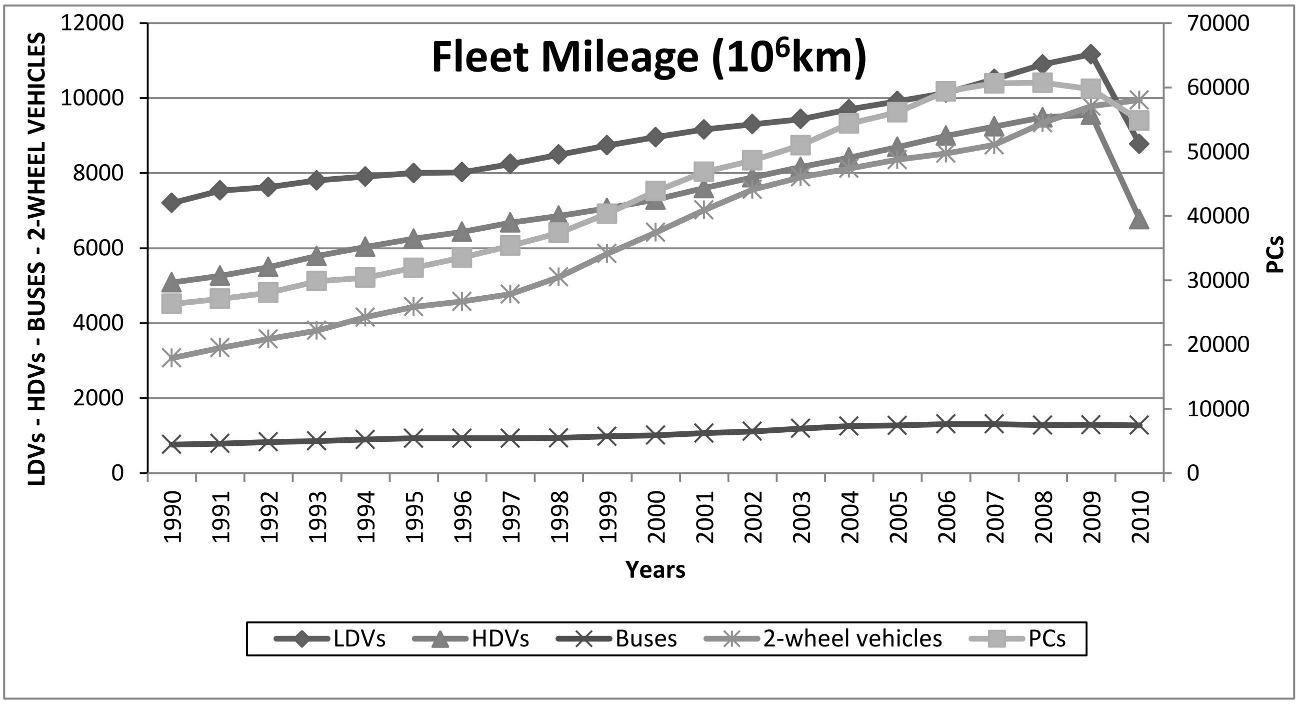


Figure 2 *Annual mileage driven by all vehicles categories during the whole time period 1990 – 2010*

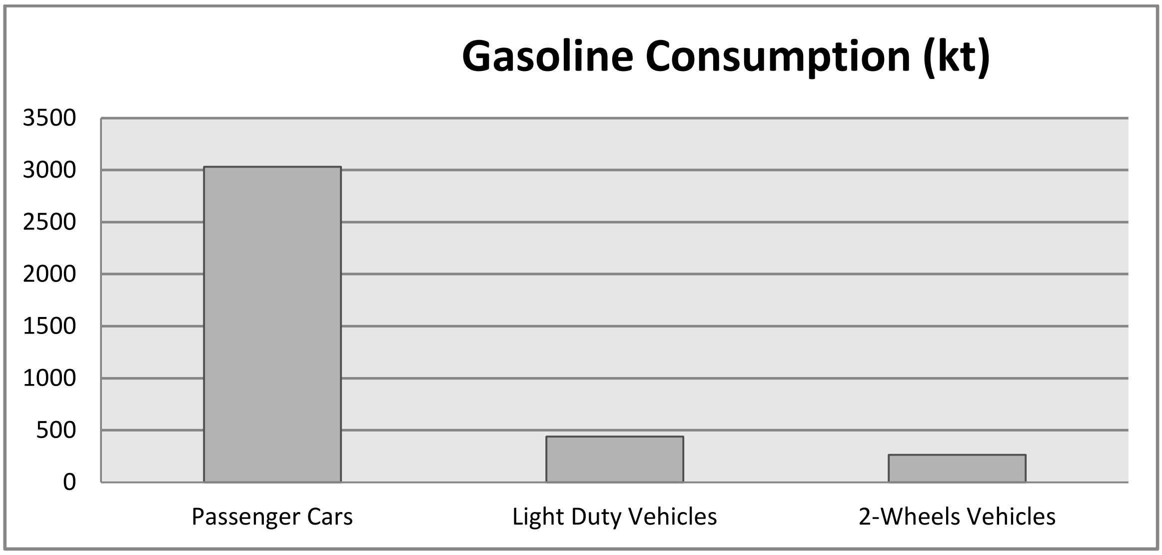


Figure 3 *Gasoline consumption (kt) by all vehicles categories for 2010*

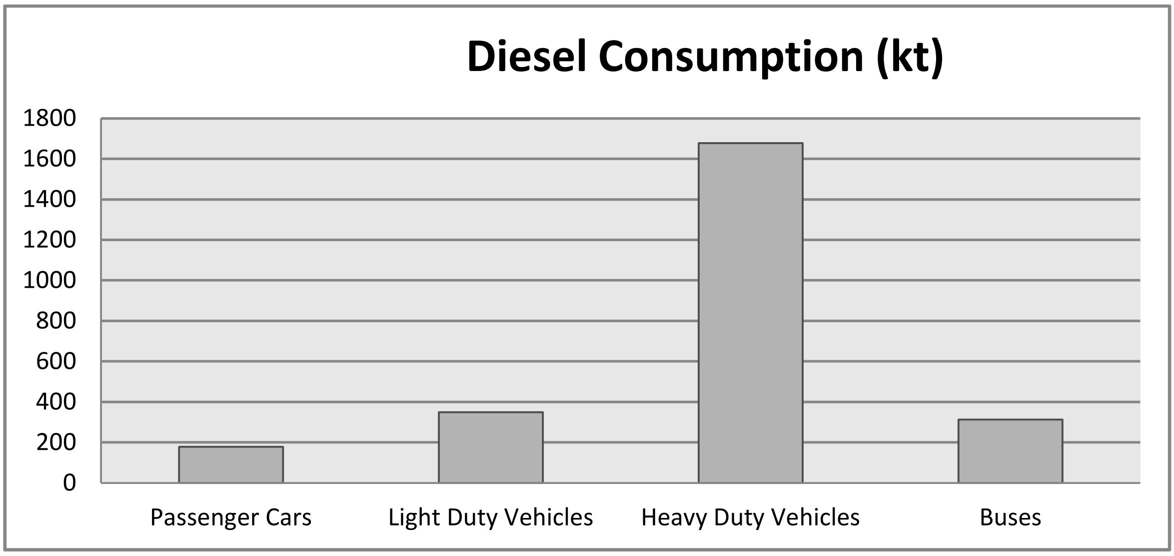


Figure 4 ***Diesel consumption (kt) by all vehicles categories for 2010***

In the last years, the vehicle fleet has increased by 265% compared to 1990 levels, while an increase of the share of medium and larger size passenger vehicles is observed (from 27% in 1990, to 36% in 2008). However this situation tends to change as a result of the high taxation imposed on vehicles with engines over 2000cm^3.

It should be noted that, despite the increase of the population of circulating vehicles, as there is a remarkable increase of less polluting vehicles, NOx, NMVOC and PM emissions decrease (***Table 2***). Finally, another considerable reduction of SO_2 emissions attributed to the improvement of the fuels characteristics (i.e. the reduction of their sulphur content) is obsereved in 2010.

In ***Figures 5-6*** the trend of PM10 and NOx emissions is presented for the period 2000-2010 along with the corresponding PM10 and NOx concentrations. Finally, in ***Figure 7*** the trend of PM10, NOx and NMVOCs emissions is presented versus PM10 mean yearly concentrations for the period 2000-2010. As shown in these figures, it seems that PM10 and NOx concentrations are strongly associated with the corresponding road traffic related emissions. Moreover, PM10 concentrations show a considerable long term similarity with the corresponding emissions of NOx and NMVOCs.

Table 2 ***Air pollutants emissions from road transportation in the three cities for the period 2000-2010 (t/y)***

PM10 TOTAL	**2000**	**2001**	**2002**	**2003**	**2004**	**2005**	**2006**	**2007**	**2008**	**2009**	**2010**
ATHENS	1561	1572	1556	1549	1501	1427	1400	1366	1273	1526	1224
THESSALONIKI	335	339	337	338	333	316	312	305	290	346	283
VOLOS	49	49	49	49	48	45	45	44	41	48	40
NOx											
ATHENS	36169	36354	34471	34505	34400	32040	32021	31531	29971	29290	22254
THESSALONIKI	8418	8458	8062	8069	8058	7533	7528	7407	7075	6916	5320
VOLOS	1112	1118	1082	1084	1083	1028	1028	1013	978	960	762
NMVOCs											
ATHENS	37910	37279	33509	32268	30636	25929	23722	22510	20124	19001	15118
THESSALONIKI	7867	7701	6894	6647	6350	5423	5044	4806	4309	4067	3472
VOLOS	1007	984	900	860	810	697	631	598	549	522	499
SO2											
ATHENS	796	792	823	853	195	203	207	204	42	34	27
THESSALONIKI	113	114	118	121	25	26	26	26	5	5	4
VOLOS	11	11	11	11	2	2	2	2	0	0	0

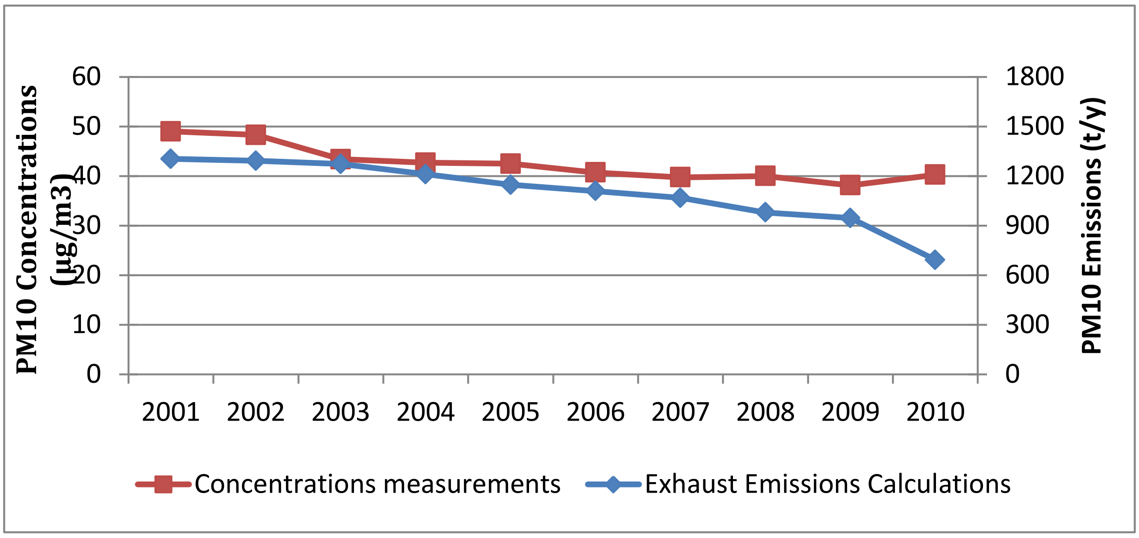


Figure 5 ***Yearly PM10 exhaust emissions (t/y) and PM10 mean yearly concentrations for the period 2001-2010 in the Athens Metropolitan Area (AMA)***

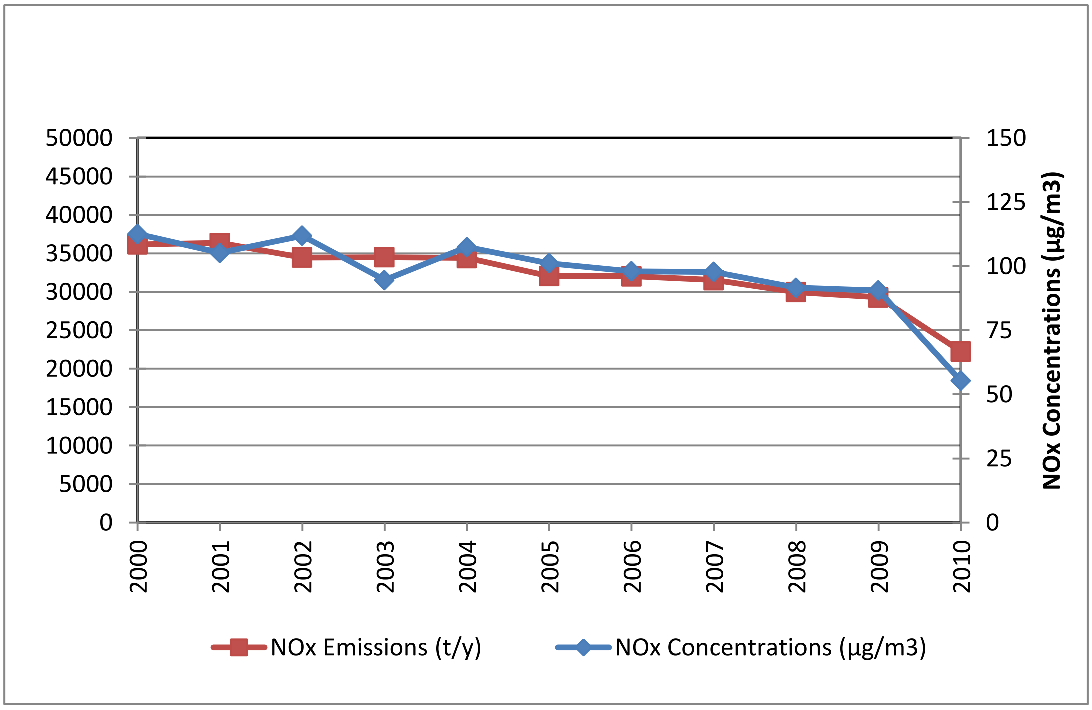


Figure 6 ***Yearly NOx emissions (t/y) and NOx mean yearly concentrations for the period 2000-2010 in the Athens Metropolitan Area (AMA)***

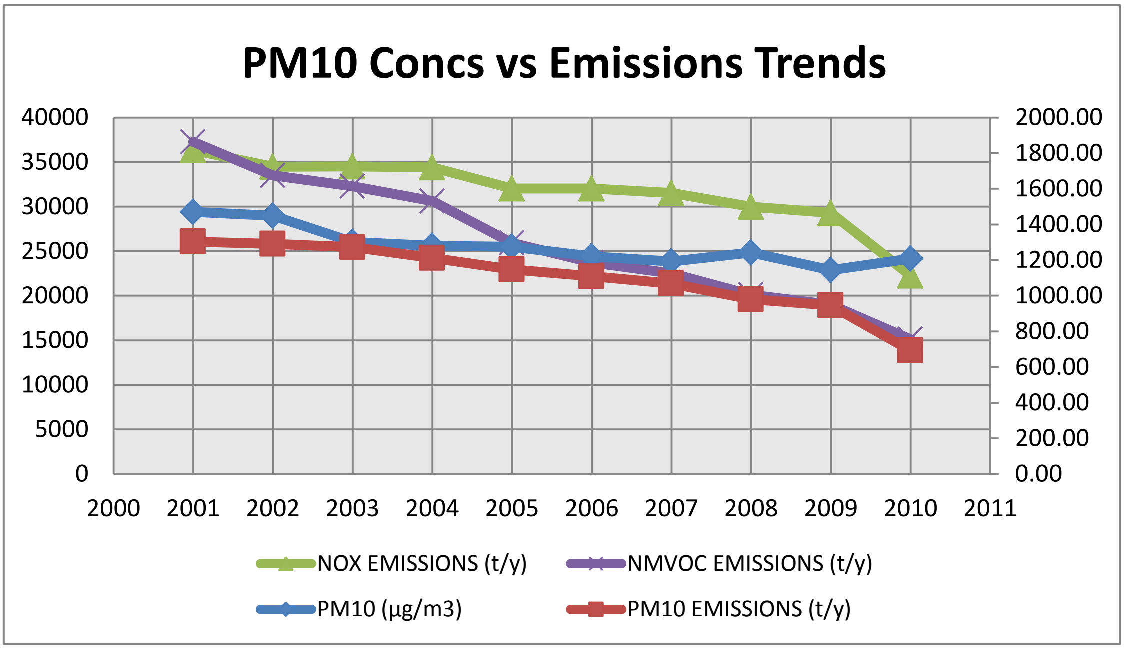


Figure 7 ***Yearly emissions (t/y) and PM10 mean yearly concentrations for the period 2000-2010 in the Athens Metropolitan Area (AMA)***

1.3 Industrial Sector

1.3.1 Methodology

The air pollution inventory for the Industrial Sector includes emissions deriving from Fuel Combustion and emissions related to the actual (Industrial) Production Processes themselves. The estimated emissions reflect the overall contribution of the Industry to the deterioration of air quality in the Greek territory.

The general equation supporting the estimation of emissions is described as follows:

$$E= AD*EF,$$

where E: Emissions, AD: Activity Data and EF: Emission Factors.

In general, the implementation of the above equation follows three patterns:

The simpler methodology is based on the use of readily available statistical data on the intensity of processes (activity rates) and default emission factors (Tier 1 methodology). These emission factors assume a linear relation between the intensity of the process and the resulting emissions. The Tier 1 default emission factors also assume an average or typical process description.

Due to the high uncertainty accompanied by the ignorance of technologies and abatement applied, the Tier 2 methodology is also used whenever available. In this case more specific, though still default, emission factors are being selected and used based on the knowledge of the types of processes and specific process conditions that are applied by the industrial plants. Tier 2 methods are more complex, reduce the level of uncertainty, and, as a result are preferred whenever data for their implementation are available.

Whenever more detailed data are available higher Tier 3 methodologies are being implemented. This is the case when plant specific, highly disaggregated data are being provided to the inventory team, including results of chemical content analysis and/or measurements. In this case the uncertainty introduced to the estimation is significantly lower.

With respect to the activity data (AD) used, their type depends on the inventory category. For the emissions deriving from Fuel Combustion, in energy and other type industry, the AD term refers to the quantity of fuel used by the industry. For emissions regarding the industrial process itself AD refer mainly to production data.

As it is obvious, activity data play a crucial role to the successful conduction of the inventory. The National Technical University of Athens (NTUA) team has in its possession a detailed database including all the high emissive industrial plants in Greece, including energy power plants. Production quantities, consumption of fuels and also of raw material data are included in the database, whereas information on the exact location of each plant is also recorded, based on the information provided by the plants in the framework of their reporting obligations under the EU Emissions Trading Scheme. Aggregated statistical data (including confidential data in some cases) provided by national and international sources such as the Hellenic Statistical Authority (Prodcom), the Ministry of Development (national energy balance), EUROSTAT etc are also introduced in the database, whereas information provided by the plants on the basis of personal communication with the NTUA team is also included.

The collected data permit the estimation of emissions from the following main air pollutants:

PM10, CO, NOx, SO2

The emission factors used are provided by the EMEP/EEA air pollutant emission inventory guidebook that has been released by the European Environmental Agency (EEA) in 2009. The specific guidebook is the most recent one, and is designed to facilitate the reporting of emission inventories by countries to the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP) and the EU National Emission Ceilings Directive. The emissions data coincide with the officially submitted emission data sets to UNECE under the Convention on Long-range Transboundary Air Pollution (CLRTAP, 2010).

With regards to the structure of the Industrial Inventory, the sector is divided in the subcategories described below. The applied methodology in each subcategory is based on the data available in the inventory's database.

1. Combustion in energy and processing industries

Refers to emissions procured through the combustion and conversion of fuels to produce energy. Emissions from public electricity and heat production, petroleum refining and manufacture of solid fuel and other energy industries are included in this category. These activities are closely connected to the emission of air pollutants, though the technology used plays an important role in the resulting emissions, especially for SO2 and NOx. Depending on the data availability Tier 2 or Tier 3 methodologies are broadly usually used for the estimation of emissions.

2. Mineral Production

Includes emissions deriving from fuel combustion and productive processes for the production of mineral products. Cement production is the most important subcategory, though other subcategories (lime production, limestone & dolomite use etc.) are also reported. Tier 2 methodology is being widely applied depending on the availability of plant-specific information by the plants. Emissions from Particulate Matter are the most important emissions from this category.

3. Chemical Production

Concerns emissions from chemical industries due to fuel combustion and productive processes. Ammonia production, nitric acid and sulphur acid are the main components of the subcategory. Emissions are closely connected to the consumption of natural gas and also to production levels. Detailed information is provided by the one plant operating in Greece, including NOx measurements whenever available. The main pollutants are sulphur and nitric oxides.

4. Metal Production

The main components of metal production subcategory are Iron and Steel, Aluminium and Ferroalloys industries. All main pollutants are emitted during metal production processes. For iron and steel and aluminium production Tier 2 methodologies are being applied, while for the rest Tier 1 methodologies are usually used.

5. Other Production

Refers to emissions of pollutants during the other production, mainly from Pulp & Paper and Food industries. NMVOC are the main pollutants and Tier 1 methodologies are used based on national statistical data.

6. Solvents

Includes NMVOC emissions from various applications of solvents such as paint application, degreasing, dry cleaning etc. The main activity data introduced in the calculations is the population of the country, as provided by the Hellenic Statistical Authority.

1.3.2 Results

The PM10 industrial emissions are presented for the three areas of interest in ***Figure 8*** for the period 2000-2010. In all three cities a decrease occurs in the two last years as a result of recession and the economic crisis.

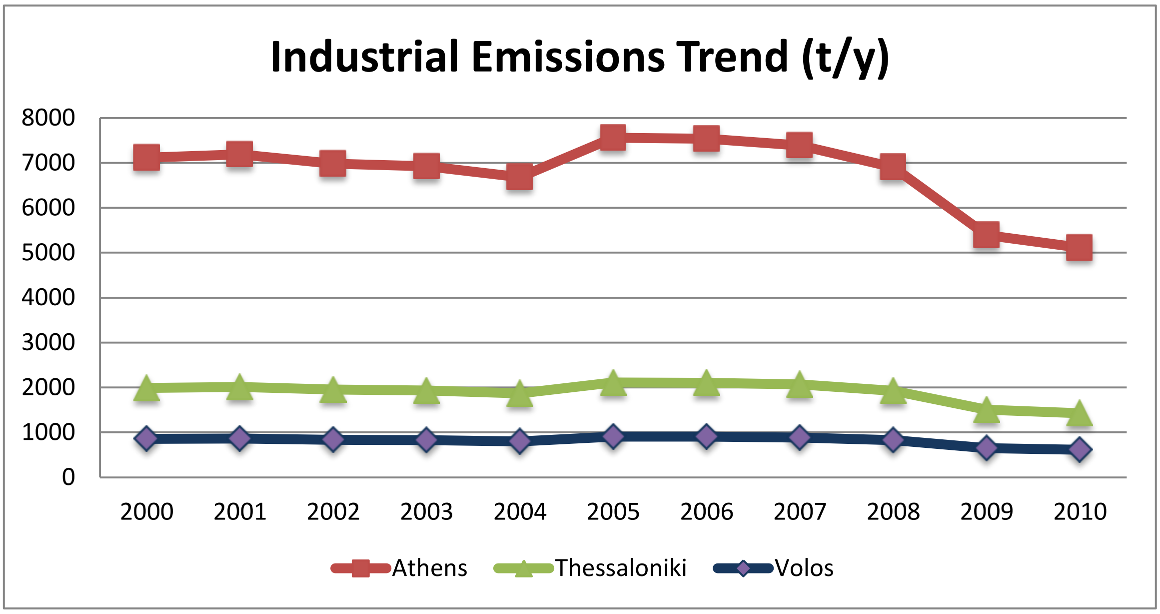


Figure 8 ***Yearly PM10 emissions (t/y) for the period 2000-2010 in the Athens Metropolitan Area (AMA), the Thessaloniki Metropolitan Area (TMA) and Volos***

In Table 3, all air pollutants emissions are presented for the whole time period and for PM10, NOx, SO2 and CO. A considerable decrease in all pollutants emissions is observed in 2009 and 2010. This decrease is associated with the economic crisis.

Table 3 Industrial emissions trend for the period 2000-2010 in the three cities (t/y)

ATHENS	**2000**	**2001**	**2002**	**2003**	**2004**	**2005**	**2006**	**2007**	**2008**	**2009**	**2010**
PM10	7115	7190	6980	6925	6684	7562	7535	7390	6900	5391	5116
NOX	21117	21341	20717	20553	19838	22444	22365	21932	20480	16002	15183
SO2	23770	24022	23320	23136	22330	25265	25175	24688	23053	18012	17091
CO	8920	9015	8751	8682	8380	9481	9447	9265	8651	6759	6414
THESSALONIKI											
PM10	1989	2010	1952	1936	1869	2114	2107	2066	1929	1507	1430
NOX	6136	6201	6020	5972	5764	6522	6499	6373	5951	4650	4412
SO2	6452	6521	6330	6280	6061	6858	6833	6701	6258	4889	4639
CO	4079	4122	4001	3970	3831	4335	4319	4236	3955	3091	2933
VOLOS											
PM10	854	863	838	831	802	908	904	887	828	647	614
NOX	882	891	865	858	828	937	934	916	855	668	634
SO2	1581	1598	1551	1539	1485	1681	1675	1642	1534	1198	1137
CO	2183	2206	2142	2125	2051	2320	2312	2267	2117	1654	1570

1.4 Residential/Commercial Sector

This sector considers non-industrial (stationary) combustion processes such as residential activities in households and in institutional and commercial buildings (e.g. heating and cooking with fireplaces, stoves, cookers and small boilers), having a thermal capacity equal or lower than 50 MW. The related NFR/CRF codes are commercial and institutional stationary plants (NFR 1A4ai) and residential stationary plants (NFR 1A4bi). The following pollutants are taken into account: NOx, PM10, NMVOCs and SO2.

The input data required (fuel consumption by fuel type) were provided by the Hellenic Statistical Authority and the Ministry of Development for all three areas of interest for year 2010.

Air pollutants emissions from the residential – tertiary sector result from energy consumption for heat in order to cover the needs for the space heating, water heating etc. Thermal needs in these sectors are covered mainly by liquid fossil fuels. The penetration of natural gas to the fuel mixture has an increasing trend.

Two basic technologies are considered: central heating boilers, and other stationary equipment (e.g. oil stoves). Fireplaces and other equipment that use biomass as fuel are not taken into account. For the allocation of fuel consumption by technology, it is assumed that the consumption of diesel, heavy fuel oil and natural gas concern central heating boilers, as no specific data for fuel consumption by other stationary equipment are available. It should be noted that due to economic crisis and the significant tax increase in diesel, there was a considerable turn to wood and wood products burning for heating purposes. This change has led to increased PM levels during the cold winter days, especially in the evening hours and during the week-ends. Unfortunately, these emissions cannot be assessed as no fuel consumption data are available for biomass burning.

Air pollutants emissions are calculated on the basis of fuel consumption and default EF are used from the EMEP/EEA emission inventory guidebook 2009.

Air pollutants emissions from the residential and the commercial/institutional sector in 2010 increased substantially compared to 1990 levels (42% and 116% respectively), as a result of the great increase of liquid fuel consumption since 1996, according to the national energy balance. A decreasing trend of the last years is attributed to the penetration of natural gas to the fuel mixture and economic recession (years 2009-2010).

The small combustion installations included are mainly intended for heating and provision of hot water in residential and commercial/institutional sectors. Some of these installations are also used for cooking (primarily in the residential sector).

In some instances, combustion techniques and fuels can be specific to an NFR activity category; however most techniques are not specific to an NFR classification. The applications can be conveniently sub-divided considering the general size and the combustion techniques applied:

- residential heating — stoves, cookers, small boilers (< 50 kW);
- institutional/commercial/agricultural/other heating including:
- heating — boilers, spaceheaters (> 50 kW),
- smaller-scale combined heat and power generation (CHP).

Emissions from smaller combustion installations are significant due to their numbers, different type of combustion techniques employed, and range of efficiencies and emissions. Many of them have no abatement measures nor low efficiency measures. In some countries, particularly those with economies in transition, plants and equipment may be outdated, polluting and inefficient. In the residential sector in particular, the installations are very diverse, strongly depending on country and regional factors including local fuel supply.

In small combustion installations a wide variety of fuels are used and several combustion technologies are applied. In the residential activity, smaller combustion appliances, especially older single household installations are of very simple design, while some modern installations of all capacities are significantly improved. Emissions strongly depend on the fuel, combustion technologies as well as on operational practices and maintenance.

For the combustion of liquid and gaseous fuels, the technologies used are similar to those for production of thermal energy in larger combustion activities.

1.4.1 Methodology

Relevant pollutants are SO2, NOx, CO, NMVOC, particulate matter (PM), black carbon (BC), heavy metals, PAH, polychlorinated dibenzo-dioxins and furans (PCDD/F) and hexachlorobenzene (HCB). For solid fuels, generally the emissions due to incomplete combustion are many times greater in small appliances than in bigger plants. This is particularly valid for manually-fed appliances and poorly controlled automatic installations. However, as already mentioned, there are no fuel consumption data available and, hence, no calculations for these appliances.

For both gaseous and liquid fuels, the emissions of pollutants are not significantly higher in comparison to industrial scale boilers due to the quality of fuels and design of burners and boilers, except for gaseous- and liquid-fuelled fireplaces and stoves because of their simple organization of combustion process. However, ‘ultra-low’ NOx burner technology is available for gas combustion in larger appliances. In general, gas- and oil-fired installations generate the same type of pollutants as for solid fuels, but their quantities are significantly lower.

Emissions caused by incomplete combustion are mainly a result of insufficient mixing of combustion air and fuel in the combustion chamber (local fuel-rich combustion zone), an overall lack of available oxygen, too low temperature, short residence times and too high radical concentrations (Kubica, 1997/1 and 2003/1). The following components are emitted to the atmosphere as a result of incomplete combustion in small combustion installations: CO, PM and NMVOCs, NH3 , PAHs as well as PCDD/F.

NH3 — small amounts of ammonia may be emitted as a result of incomplete combustion process of all solid fuels containing nitrogen. This occurs in cases where the combustion temperatures are very low (fireplaces, stoves, old design boilers). NH3 emissions can generally be reduced by primary measures aiming to reduce products of incomplete combustion and increase efficiency.

TSP, *PM10*, *PM2.5* — particulate matter in flue gases from combustion of fuels (in particular of solid mineral fuels and biomass) may be defined as carbon, smoke, soot, stack solid or fly ash. Emitted particulate matter can be classified into three groups of fuel combustion products.

The first group is formed via gaseous phase combustion or pyrolysis as a result of incomplete combustion of fuels (the products of incomplete combustion (PIC)): soot and organic carbon particles (OC) are formed during combustion as well as from gaseous precursors through nucleation and condensation processes (secondary organic carbon) as a product of aliphatic, aromatic radical reactions in a flame-reaction zone in the presence of hydrogen and oxygenated species; CO and some mineral compounds as catalytic species; and VOC, tar/heavy aromatic compounds species as a result of incomplete combustion of coal/biomass devolatilization/pyrolysis products (from the first combustion step), and secondary sulphuric and nitric compounds. Condensed heavy hydrocarbons (tar substances) are an important, and in some cases, the main contributor, to the total level of particles emission in small-scale solid fuels combustion appliances such as fireplaces, stoves and old design boilers.

The next groups (second and third) may contain ash particles or cenospheres that are largely produced from mineral matter in the fuel; they contain oxides and salts (S, Cl) of Ca, Mg, Si, Fe, K, Na, P, heavy metals, and unburned carbon formed from incomplete combustion of carbonaceous material; black carbon or elemental carbon — BC (Kupiainen, et al., 2004).

Particulate matter emission and size distribution from small installations largely depends on combustion conditions. Optimization of solid fuel combustion process by introduction of continuously controlled conditions (automatic fuel feeding, distribution of combustion air) leads to a decrease of TSP emission and to a change of PM distribution (Kubica, 2002/1 and Kubica et al., 2004/4). Several studies have shown that the use of modern and 'low-emitting' residential biomass combustion technologies leads to particle emissions dominated by submicron particles (< 1 μm) and the mass concentration of particles larger than 10 μm is normally < 10 % for small combustion installations (Boman et al., 2004 and 2005, Hays et al., 2003, Ehrlich et al, 2007).

Note that there are different conventions and standards for measuring particulate emissions. Particulate emissions can be defined by the measurement technique used including factors such as the type and temperature of filtration media and whether condensable fractions are measured. Other potential variations can include the use of manual gravimetric sampling techniques or aerosol instrumentation. Similarly, particulate emission data determined using methodology based on a dilution tunnel may differ from emission data determined by a direct extractive measurement on a stack. The main difference is whether the emission measurement is carried out in the hot flue gas, either in-stack or out-stack, or if the measurements is carried out after the semi-volatile compounds have condensed.

Typically the Swedish laboratory measurements (e.g. Johansson et al., 2004) are based on Swedish Standard (SS028426) which is an out-stack heated filter, meaning that the semi-volatile compounds will not have condensed. In the field measurements an in-stack filter was used to measure PM (Johansson et al., 2006).

CO — carbon monoxide is found in gas combustion products of all carbonaceous fuels, as an intermediate product of the combustion process and in particular for under-stoichiometric conditions. CO is the most important intermediate product of fuel conversion to CO2; it is oxidized to CO2 under appropriate temperature and oxygen availability. Thus CO can be considered as a good indicator of the combustion quality. The mechanisms of CO formation, thermal-NO, NMVOC and PAH are, in general, similarly influenced by the combustion conditions. The emissions level is also a function of the excess air ratio as well as of the combustion temperature and residence time of the combustion products in the reaction zone. Hence, small combustion installations with automatic feeding (and perhaps oxygen 'lambda' sensors) offer favourable conditions to achieve lower CO emission. For example, the emissions of CO from solid fuelled small appliances can be several thousand ppm in comparison to 50–100 ppm for industrial combustion chambers, used in power plants.

NMVOC — for small combustion installations (e.g. residential combustion) emissions of NMVOC can occur in considerable amounts; these emissions are mostly released from inefficiently working stoves (e.g. wood-burning stoves). VOC emissions released from wood-fired boilers (0.510 MW) can be significant. Emissions can be up to ten times higher at 20 % load than those at maximum load (Gustavsson et al, 1993). NMVOC are all intermediates in the oxidation of fuels. They can adsorb on, condense, and form particles. Similarly as for CO, emission of NMVOC is a result of low combustion temperature, short residence time in oxidation zone, and/or insufficient oxygen availability. The emissions of NMVOC tend to decrease as the capacity of the combustion installation increases, due to the use of advanced techniques, which are typically characterized by improved combustion efficiency.

Sulphur oxides — in the absence of emission abatement, the emission of SO2 is dependent on the sulphur content of the fuel. The combustion technology can influence the release of SO2 with (for solid mineral fuels) higher sulphur retention in ash than is commonly associated with larger combustion plant.

Nitrogen oxides — emission of NOx is generally in the form of nitric oxide (NO) with a small proportion present as nitrogen dioxide (NO2). Although emissions of NOx are comparatively low in residential appliances compared to larger furnaces (due in part to lower furnace temperatures), the proportion of primary NO2 is believed to be higher.

The Tier 1 approach for process emissions from small combustion installations uses the general equation:

$$E_{pollutant} = AR_{fuelconsumption} \times EF_{pollutant} \quad (1)$$

where:
Epollutant = the emission of the specified pollutant,
ARfuelconsumption = the activity rate for fuel consumption,
EFpollutant = the emission factor for this pollutant.
This equation is applied at the national/local level, using annual national/local fuel consumption for small combustion installations in various activities.

1.4.2 Results

The PM10 residential/commercial emissions are presented for the three areas of interest in ***Figure 9*** for the period 2000-2010. In all three cities a constant decrease of PM10 emissions occurs as a result of the penetration of natural gas.

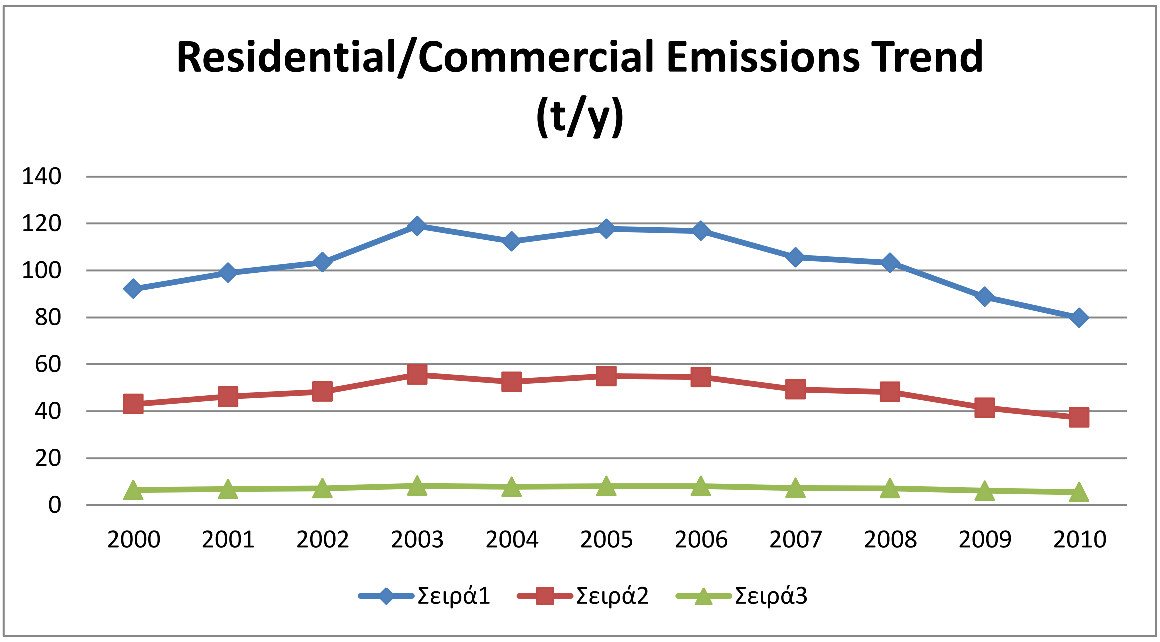


Figure 9 ***Yearly PM10 emissions (t/y) for the period 2000-2010 in the Athens Metropolitan Area (AMA), the Thessaloniki Metropolitan Area (TMA) and Volos***

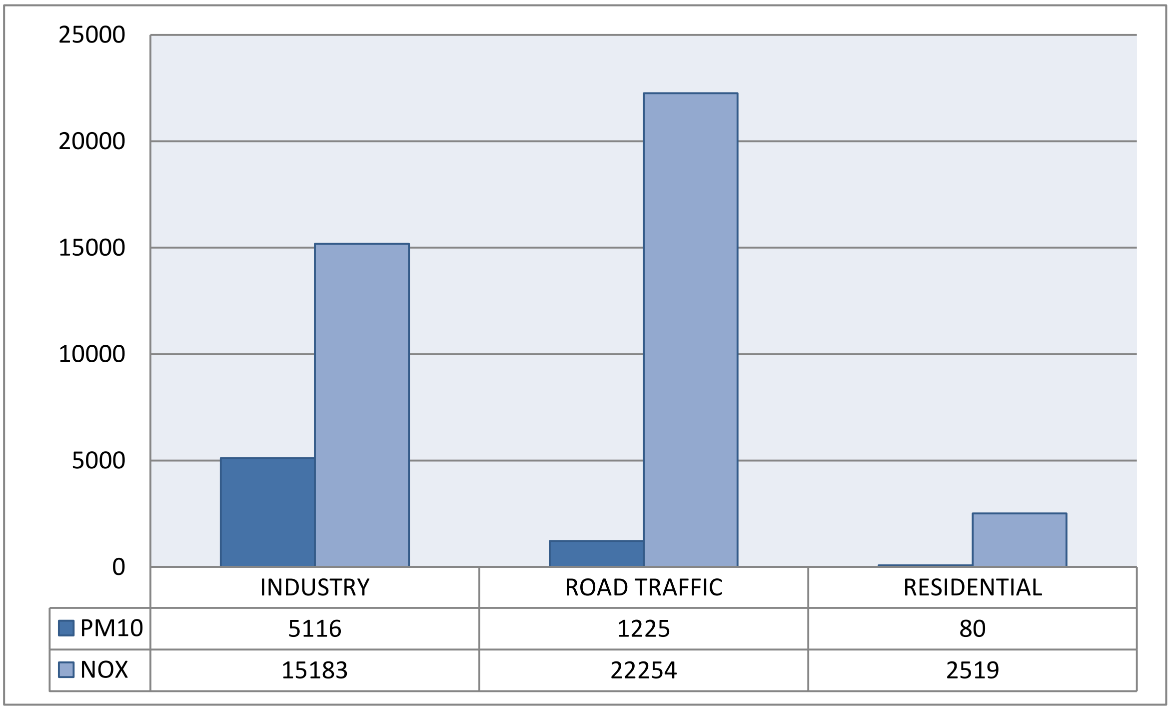
In Table 4, all air pollutants emissions are presented for the whole time period and for PM10, NOx, NMVOCs and SO2. A considerable decrease in all pollutants emissions is observed in 2009 and 2010 again as a result of the fuel consumption decrease because of the economic crisis.

Table 4 Air pollutants emissions trend from the residential/commercial sector for the period 2000-2010 in the three cities (t/y)

Athens	**2000**	**2001**	**2002**	**2003**	**2004**	**2005**	**2006**	**2007**	**2008**	**2009**	**2010**
PM10	92	99	103	119	112	118	117	106	103	89	80
NOx	2909	3125	3266	3756	3549	3715	3688	3332	3260	2802	2519
NMVOC	558	600	627	721	681	713	708	639	626	538	484
SO2	1334	1433	1498	1723	1628	1704	1692	1528	1495	1285	1155
Thessaloniki											
PM10	43	46	48	56	53	55	55	49	48	41	37
NOx	1219	1309	1368	1574	1487	1557	1545	1396	1366	1174	1056
NMVOC	243	261	273	313	296	310	308	278	272	234	210
SO2	641	688	719	827	782	818	812	734	718	617	555
Volos											
PM10	6	7	7	8	8	8	8	7	7	6	6
NOx	186	200	209	240	227	238	236	213	209	179	161
NMVOC	37	39	41	47	45	47	46	42	41	35	32
SO2	94	101	105	121	115	120	119	108	105	90	81

1.5 Comparative emissions results from the main anthropogenic sources

In the following ***Figures 10-12***, total PM10 and NOx emissions from the main pollutant sources are presented. It should be noted that, at a first glance, it appears that industrial PM10 emissions play the most important role for all three areas of interest. On the contrary, NOx emissions seem to be mostly associated with road traffic emissions. However, it is to be stressed that the occurring air pollutant levels are associated with other parameters too except from the amount of pollutants emitted from each source. Such parameters are: i) geographic location and distance from the area of interest, ii) spatial density of emissions and iv) topography and meteorology. These parameters are taken into account in the source apportionment models results.



	INDUSTRY	ROAD TRAFFIC	RESIDENTIAL
PM10	5116	1225	80
NOX	15183	22254	2519

Figure 10 ***Yearly PM10 and NOx emissions (t/y) for 2010 in the Athens Metropolitan Area (AMA) and for the main emission sources.***

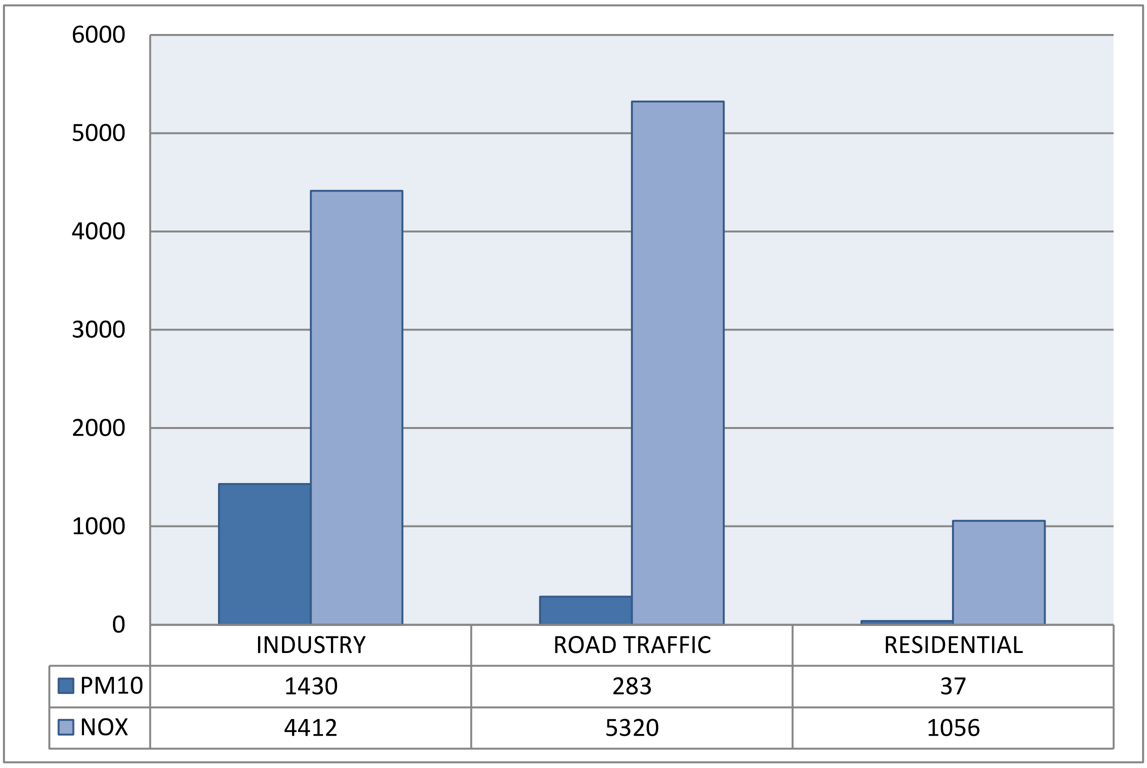


Figure 11 *Yearly PM10 and NOx emissions (t/y) for 2010 in the Thessaloniki Metropolitan Area (TMA) and for the main emission sources.*

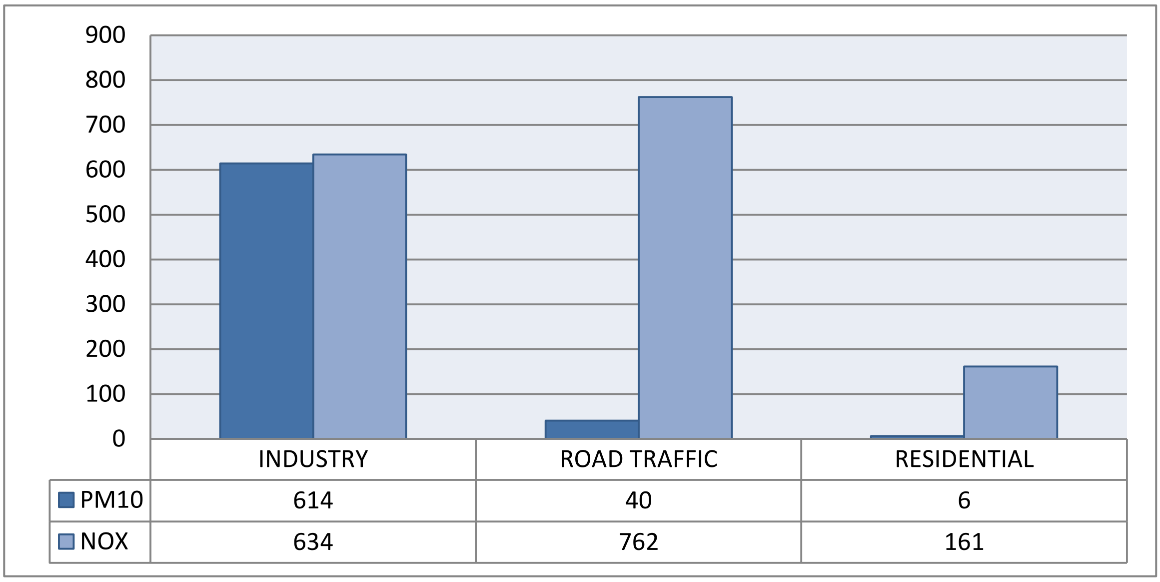


Figure 12 *Yearly PM10 and NOx emissions (t/y) for 2010 in Volos and for the main emission sources.*

1.6 Navigation

Air pollutant emissions from navigation are calculated according to the EMEP CORINAIR Tier 1 default methodology, which is based on the relative consumption of energy per fuel and default emission factors (SNAP 0804 – EEA 2001). Fuel consumption data were taken from the Hellenic Statistical Authority, whereas activity data for the yearly distribution of emissions were taken from local relevant agencies. Finally, activity data as well as emissions data for comparison reasons were taken from Tzannatos (2010).

It should be noted that the application of a higher Tier methodology requires detailed data for the composition of the fleet which are not available at present.

Air pollutants emissions are presented in ***Table 5*** for 2010 and for the three areas of interest. As shown in ***Table 6***, navigation emissions in 2010 were higher (43%) than the emissions in 2000, on the basis of fuel consumption data from this sector (***NIR, MEEC, 2012)***.

Table 5 ***Air pollutants emissions from navigation for the three areas (2010)***

t/y	NOx	SOx	PM10	PM2.5
PIRAEUS	40471	16838	1121	1121
THESSALONIKI	10881	4529	288	288
VOLOS	6043	2506	161	161

Table 6 ***PM emissions from navigation for the period 1990 – 2010***

t/y	2000	2001	2002	2003	2004	2005	2006	2007	2008	2009	2010
PIRAEUS	785	1062	962	957	1065	1017	1125	1044	933	1370	1121
THESSALONIKI	202	273	247	246	274	261	289	268	240	352	288
VOLOS	113	153	138	137	153	146	162	150	134	197	161

1.7 Aviation

Air pollutants emissions from aviation are calculated according to the Tier 2a methodology suggested by the IPCC Guidelines, which is based on the combination of energy consumption data and air traffic data (Landing and Take off cycles, LTOs). The emission factors used and the distribution of consumption in LTOs and cruise are the suggested CORINAIR values (SNAP 080501 & 080503 – EEA 2001) for average fleet.

The data on energy consumption derive from the national energy balance, while data on LTOs are provided by the Civil Aviation Organisation.

In Greece, air pollutant emissions from aviation increased by 82% since 1990 with an average annual increase rate of approximately 4% (MEEC, 2012). Air pollutants emissions for 2010 and for the two cities airports are presented in ***Table 7*** below. PM10 emissions trend of the Athens airport are presented in ***Figure 12***. Although, total emissions from aviation in Greece have a significant increasing trend, in the Athens airport for the decade 2000-2010 only small fluctuations, not exceeding 20% occur.

Table 7 ***Air pollutants emissions from aviation for the three areas (2010)***

t/y	CO	NMVOC	NOx	SOx	PM-10
Thessaloniki	451	52	219	19	4
Athens	1785	206	867	76	14

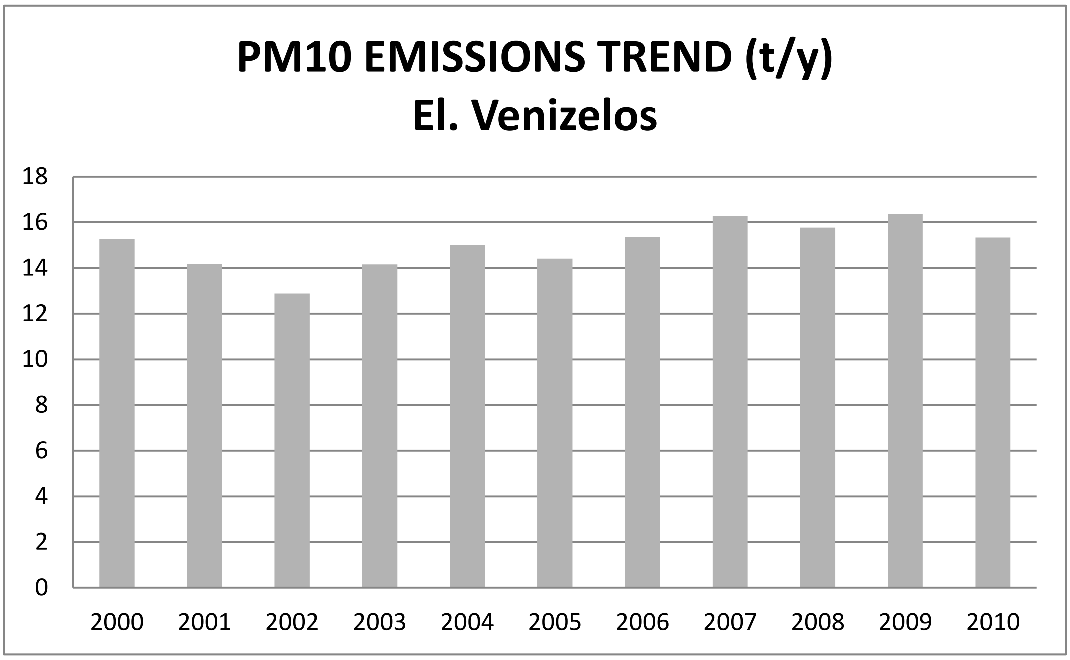


Figure 12 ***PM10 emissions trend (t/y) for the period 2000-2010 in Eleftherios Venizelos airport (Athens).***

1.8 Railways

Air pollutants emissions from railways are calculated according to the default methodology proposed in CORINAIR, which is based on the relative consumption of energy per fuel and the typical emission factors (SNAP 0802 – EEA 2001). Railways emissions are generally minor and they are mostly allocated across the railways network. Moreover, emissions from railways decreased by 69% from 1990 to 2010 (MEEC, 2012). Thus, they are considered as negligible in the areas of the train stations in the three cities and, consequently, they are not taken into account.

2. EMISSIONS FROM NATURAL SOURCES

2.1 Introduction

Exposure to particulate matter (PM) has been associated with increased human morbidity and mortality by many epidemiological studies (e.g. Dockery et al. 1993; Katsouyanni et al. 1995 and 2001; Pope et al. 1999 and 2002). 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 (Winiwarter et al. 2009). The construction of an emission inventory is an important tool in air quality management and can be also used for the development and assessment of the results of specific mitigation strategies (Placet et al. 2000; Karl et al. 2009).

Except for anthropogenic emissions, emissions from natural sources can be a significant contributor to air quality deterioration in urban areas. They are usually emitted in less populated areas away from the urban centres and built up areas and then are usually transported over the urban areas. Natural primary PM emissions can result from a number of sources, including the sea surface, soil, flora and biota, and can occur in the forms of e.g. windblown dust, sea-salt particles, fungal spores and plant debris, smoke from wildfires.

Several emission inventories have been developed for Greece using both the bottom-up and top-down approaches. In particular, Aleksandropoulou and Lazaridis (2004) created an emission inventory of anthropogenic and natural sources in Greece. Symeonidis et al. (2004) developed an emission inventory system from transport in Greece. Poupkou et al. (2007) developed a spatially and temporally disaggregated anthropogenic emission inventory in the Southern Balkan region and Symeonidis et al. (2008) estimated biogenic NMVOCs emissions in the same region. Also Markakis et al. (2010) presented an anthropogenic emission inventory for gaseous species for Greece, Poupkou et al. (2008) studied the effects of anthropogenic emissions over Greece to ozone production, Sotiropoulou et al. (2004) estimated the spatial distribution of agricultural ammonia emissions in the AMA and Hayman et al. (2001) developed spatial air emission inventories using the top-down approach for large urban agglomerations in southern Europe including the AMA.

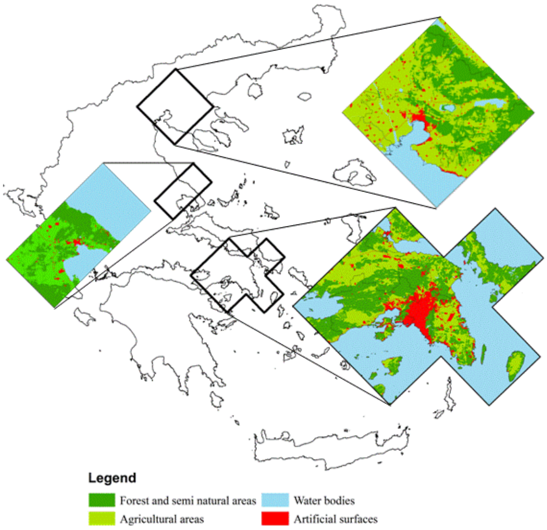


Figure 1 Areas of interest and distribution of landcover (main classes; EEA CLC 2009)

In this study emission inventories of particulate matter ($PM_{2.5}$ and $PM_{2.5\text{-}10}$) and gaseous pollutants (BVOCs) from natural sources for the Athens and Thessaloniki metropolitan areas (AMA and TMA respectively) and the greater area of Volos (GVA) are presented for the period 2001-2010 (Figure 1). In particular, the inventories include the emissions of primary particles from soil surface i.e. windblown dust (WB) emissions from agricultural and vacant lands and from the ocean/sea surface (sea salt particles) by waves breaking at the Sea Shore-surf zone (SS_SS) and from the bursting of bubbles from oceanic whitecaps - Open Ocean (SS_OO). Additionally, emissions of BVOCs (Biogenic Volatile Organic Compounds) from vegetation during photosynthesis, plant respiration and vaporization from stores within the plant tissue, precursor to PM, were estimated. The temporal evolution of natural PM emissions in the metropolitan areas of Athens, Thessaloniki and Volos is examined together with their contribution to total primary and secondary PM emissions in the study areas.

2.2 Methodology

2.2.1 Input data, assumptions and their implications to results

The variation in natural emissions is determined by changes in meteorological conditions and landcover.

The necessary meteorological data used in the calculations were retrieved from the FOODSEC Meteodata distribution page (action developed in the framework of the EC Food Security Thematic Programme; European Centre for Medium-Range Weather Forecast (ECMWF) ERA INTERIM reanalysis model data and ECMWF OPERATIONAL data from 01/01/2011 on; temporal analysis 10-days; spatial resolution 0.25 degree). Monthly averages of temperature and air velocity were calculated from the above data.

In addition, data on the monthly averaged days with rainfall used in the calculations were retrieved from the HNMS (Hellenic National Meteorological Service) database on climatology for meteorological stations in and round the area of interest (the values therefore correspond to a period spanning over at least 40 years).

Data on the soil texture were available by the European Soil Database (ESDB v2.0 2004) either in the form of the soil type as in FAO-UNESCO, 1974 classification (used in modified CEC 1985) texture classes (the dominant and secondary surface textural classes are provided) or as a textural profile containing the fractions of clay, silt and sand in the soil horizon.

In the calculations were used landuse data from the Land Cover 2000 database of the European Commission programme to COoRdinate INformation on the Environment across Europe (EEA CLC 2000, v2009) (Level 1 classification is depicted in Figure 1). Changes in landcover as regards areas burnt from forest fires during the period 2000-2008 in the Athens Larger Urban Zone have been incorporated in the emissions presented in Aleksandropoulou et al. 2013. The effects in PM_{10} windblown dust emissions and emissions of BVOCs during 2008 from more than 2230ha of forests and 1841 ha of woodlands burnt during the period 2000-2008 were an increase by approximately 1.7% and a decrease decrease by 3.5%, respectively. Based on the above results and due to the absence of relevant data changes in landcover in the three areas during the period 2000-2010 were not taken into account in the calculations. Therefore any observed trend arises solely from changes in meteorological conditions which vary from year to year.

An important source of uncertainty in windblown dust emission calculations is the assumption that wind erosion and suspension of particles occurs at any air velocity as long as the vegetation cover, soil properties and meteorological conditions allow the wind erosion of the surface. Also it has been assumed that monthly averaged wind speed values can be used in predicting the emissions of dust due to wind erosion and of sea salt particles. The effects of the assumptions in the calculation of windblown dust emissions 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). As regards the sea salt emissions from open ocean and sea shore, the same methodology applied in Aleksandropoulou et al. (2012) was used to justify the assumptions. In particular, the emissions from the sea surface were calculated using both monthly averaged values of meteorological parameters and 3h instantaneous values derived from the EMEP UNIFIED model input files (are based on forecast experiment runs with the Integrated Forecast System, a global operational forecasting model from the European Centre for Medium-Range Weather Forecasts; 3h instantaneous values; EMEP/MSC-W 2011) for one month (August 2008). It was assumed in the calculations that the 3h instantaneous value occurs throughout the 3h period. It was found that 446 Mg of sea salt $PM_{2.5}$ and 2050 Mg of sea salt $PM_{2.5-10}$ were emitted from the open ocean during August 2008, whereas based on the 3h instantaneous values during August 2008 are emitted 547 Mg and 2495 Mg of sea salt $PM_{2.5}$ and $PM_{2.5-10}$, respectively, with an emission rate approximately 10^{-10} g/m^{2}s. Likewise, during August 2008 are emitted 501 Mg $PM_{2.5}$ and 3903 Mg $PM_{2.5-10}$ of sea salt at sea shore, whereas based on the 3h instantaneous values are emitted 540 Mg and 4210 Mg of sea salt $PM_{2.5}$ and $PM_{2.5-10}$, respectively, with an emission rate of 10^{-14} g/m^{2}s.

Finally, the use of monthly averaged data in the calculations of BVOCs emissions introduces uncertainty in the results. In particular, using monthly averaged daytime temperature leads to large errors in the calculations, but only of order 20%, which is much less than the uncertainties in the emission potentials (EMEP/CORINAIR 2007). On the other hand, the use of ambient temperature and light-intensity provides a reasonable approximation to leaflevel light and temperature (moderate uncertainty for European conditions, EMEP/CORINAIR 2007).

2.2.2 Emissions of fugitive windblown dust

Dust is injected into the atmosphere as a result of natural wind erosion from soil grain abrasion and the deflation of the abrasion products and other materials deposited on the earth's surface (Korcz et al. 2009). The underlying physical processes that move particles upward from the surface are aerodynamic lift, saltation and sandblasting (Shaw et al. 2008 and references therein). The emissions of dust are modulated by the soil characteristics (composition, texture and type), surface characteristics (vegetation cover, moisture, aerodynamic roughness length) and the meteorological conditions (e.g. Alfaro and Gomes 2001; Draxler et al. 2001; Shao 2001; Zender et al. 2003). Many models have been developed to describe soil erosion by wind and the subsequent emissions of dust to the atmosphere (e.g. Gillette and Passi 1988; Ginoux et al. 2001; Marticorena et al. 1997; Nickovic and Dobricic 1996; Shao 2001; Westphal et al. 1987; Zender et al. 2003).

Dust emissions from wind erosion of agricultural and vacant lands were estimated using the method presented by Choi et al. (2008). The emissions of windblown dust depend on the land cover, soil texture, wind friction velocity and threshold friction velocity at the study area during the study period. The vertical dust emission flux (F_a; g/cm^{2}s) was estimated using the formulae of Westphal et al. (1987) modified by the results of Park and In (2003) and Liu and Westphal (2001) with the equation:

$$F_a = \begin{array}{ll} f \times 0.13 \times (1 - R) \times 10^{-14} \times U_*^4 & \text{when } U_* \geq U_{*t} \text{ for predominatly silt and clay soils} \\ f \times 0.13 \times (1 - R) \times 10^{-13} \times U_*^3 & \text{when } U_* \geq U_{*t} \text{ for predominatly sandy soils} \\ 0 & \text{when } U_* < U_{*t} \end{array} \qquad \text{(Eq. 1)}$$

where U_* is the friction velocity (cm/s), U_{*t} is the threshold friction velocity (cm/s), R is a reduction factor which depends on land cover, and f is the fraction of small silt and clay in the surface layer of the soils used in order to account for PM_{10} emissions. The friction velocity U_* is estimated with the wind velocity profile equation (Prandtl 1935) which relates the slope of the velocity to the natural logarithm of height. The threshold friction velocity U_{*t} (Marticorena et al. 1997) was calculated with the equation:

$$U_{*t,1} = 0.3\exp(7.22Z_0) \quad \text{(Eq. 2)}$$

where, Z_0 is the aerodynamic roughness length (cm). The ratio of $PM_{2.5}/PM_{10}$ is 0.1 (Choi et al. 2008).

Based on the ESDB data, soils were classified to predominately sandy and predominately silt and clay, and additionally the maximum fraction f, of clay and small silt, was estimated in order to account for the maximum PM_{10} emissions (worst case scenario). For the fraction of clay and small silt (f) for each soil category an averaged value equal to 0.55 on clay and clay loam soils and 0.4 for loam and sandy clay loam soils was considered (the fraction of small silt is considered 50% of silt as in Choi et al. 2008). In addition, surface roughness length values were assigned to each land cover type based on the values presented by Mansell et al. (2004) (Table 1).

It was assumed that emissions occur at any air velocity as long as the vegetation cover, soil properties and meteorological conditions allow the wind erosion of the surface (i.e. $U_* \geq U_{*t}$ always in Eq. 1). Although this is not correct physically it was essential as the values of critical velocity estimated by the available wind speed, never exceeded the threshold velocity for wind erosion due to the time averaging of the data values (monthly averages). The validity of predicted windblown dust emissions using this assumption was examined by applying the Eq. 1 using a meteorological dataset with finer time resolution (Eq. 2 was used for the calculation of the threshold friction velocity for wind erosion) (Aleksandropoulou et al. 2013).

Table 1 Values of windblown dust emissions for the reduction factor R and aerodynamic roughness length Z_0 (m) for different landcover types

LABEL1	LABEL2	LABEL3	Z0	R
Artificial surfaces	Artificial. non-agricultural vegetated areas	Green urban areas	1	1
		Sport and leisure facilities	1	1
Agricultural areas	Arable land	Non-irrigated arable land	0.031	0.4
		Permanently irrigated land	0.031	0.6
	Permanent crops	Vineyards	0.031	0.7
		Fruit trees and berry plantations	0.031	0.7
		Olive groves	0.031	0.7
	Pastures	Pastures	0.1	0.5
	Heterogeneous agricultural areas	Annual crops associated with permanent crops	0.031	0.5
		Complex cultivation patterns	0.031	0.5
		Land principally occupied by agriculture. with significant areas of natural vegetation	0.031	0.5
Forest and semi natural	Forests	Broad-leaved forest	1	0.9
		Coniferous forest	1.3	0.9

areas		Mixed forest	1	0.9
	Scrub and/or herbaceous vegetation associations	Natural grasslands	0.1	0.6
		Sclerophyllous vegetation	0.05	0.7
		Transitional woodland-shrub	0.05	0.8
	Open spaces with little or no vegetation	Beaches, dunes, sands	0.002	0.1
		Bare rocks	0.002	0.1
		Sparsely vegetated areas	0.002	0.1
		Burnt areas	0.002	0.1
Water bodies	Marine waters	Sea and ocean	1	0.1

Finally, it must be noted that data on the gravimetric soil moisture were not available therefore the calculated emissions using the above equations correspond to lower than the actual threshold friction velocities i.e. are overestimated (correspond to dry conditions). In order to account for the effect of soil moisture on emissions of windblown dust, the emissions evaluated were downscaled by adopting the assumptions previously used by Korcz et al. (2009) in their calculations of windblown dust emissions over Europe were adopted. Specifically, according to Mansell et al. (2004) dust reservoirs can be classified to those with unlimited potential which can emit for 10 successive hours and to those with limited potential which can emit dust for only one hour. Following these periods the reservoir needs 24 hours to recover its dust emission potential. Also a reservoir does not emit dust during and for 72 hours after each rain, when it is covered with snow or the temperature is below zero. In the calculations, periods with rain were considered as inactive for dust emissions (monthly average number of days with rain; mean climatology data were retrieved from the H.N.M.S.). In addition, surfaces were classified based on their landcover to those that have stable soil (limited potential) and unstable soil (unlimited potential).

2.2.3 Emissions of sea salt particles

Sea shore emissions

The sea salt emissions at sea shore were estimated using the source function provided by de Leeuw et al. (2000) modified according to Zhang et al. (2005) in order to account for the effect of relative humidity (80% in our calculations) on the size distribution of emissions. In particular, emission flux (in g/m^{2}s μm) of particles by the breaking of waves in the surf zone is estimated with the equation (Eq. 3):

$$\frac{dF_{M-surf}}{dD_p} = 10^{-15}\frac{dF_{N-surf}}{dD_{\rho_0}}\frac{\pi}{6}D_p^3\rho_p x$$

where D_p is the particle diameter, ρ_p is the particle density and x is the solute weight fraction in natural sea-salt solutions. The particle density and the solute weight fraction depend on the relative humidity and their values can be calculated by the polynomial equations:

$x(RH) = {}_{i=0}^{5} W_i RH^i$ (Eq. 4)

$\rho_p(RH) = 1000 \sum_{i=0}^{5} D_i RH^i$ (Eq. 5)

where W_i and D_i values are polynomial coefficients and their values are given in Table 2.

Table 2 Polynomial coefficients for solute weight fraction and density (Zhang et al., 2005)

i	Wi	Di
0	3.1657	3.8033
1	-19.079	-16.248
2	55.72	46.085
3	-83.998	-68.317
4	63.436	50.932
5	-19.248	-15.621

The emission flux of particles (in g/m^2s μm) is calculated by the equation (Eq. 6):

$$\frac{dF_{N-surf}}{dD_{p_0}} = 1.1 \times 10^7 e^{0.23U_{10}} D_{p_0}^{-1.65}$$

where U_{10} is the air velocity at 10 m height (m/s)and D_{p0} the sea salt particle diameter at formation. This diameter is calculated as:

$D_{p_0} = C^0 D_p$ (Eq. 7)

Where C^0 is a function of relative humidity and approximately equal to 2 when the relative humidity is 80%. Equation 7 can be substituted with:

$r_{80} = \frac{1}{2} C^{80} D_p$ (Eq. 8)

where r is the particle radius. Equation 3 is applicable for U_{10}<9 m/s and 1.6<D_{p0}<20 μm.

The length of the coastline in each cell covering sea shore was calculated from relevant spatial data available from the Hellenic Ministry for the Environment, Energy and Climate Change (geodata.gov.gr) and the surf zone width was assumed equal to 50 m (mean surf-zone).

Open-ocean sea emissions

Sea-salt particles are emitted directly from the sea surface as spume from the whitecap cover of waves at high wind speeds (u >10 m/s) and in the form of film and jet drops during the breaking of waves on the sea and other surfaces at lower wind speeds (Singh 1995). Hourly emissions, disaggregated in 8 size bins (PM_{10}; 0.04-0.08, 0.08-0.16, 0.16-0.31, 0.31-0.62, 0.62-1.25, 1.25-2.5, 2.5-5.0, 5.0-10.0 μm), were computed per grid cell according to Grini et al. (2002). The flux F (particles/m^2s) of sea salt particles to the thin atmospheric layer above the sea surface is estimated as a function of the wind speed and the size of particle which depends on the relative humidity (RH) of the atmospheric layer above the sea surface. The radius r of wet particles is estimated by the equation $r = \alpha r_d^{\beta}$ (Eq. 9), where r_d is the radius of the dry particle and α and β are coefficients which depend on the RH and are calculated by the equations (Fitzgerald 1975):

$\beta = e\ \frac{0.00077\,(RH)}{1.009-(RH)}$ (Eq. 10), and $\alpha = 1.62e\ \frac{0.066\,(RH)}{\Phi-(RH)}$ (Eq. 11)

where, $\Phi = 1.2$ for NaCl particles at RH<97%. We assumed that the RH at the thin atmospheric layer above the sea surface has a constant value equal to 80% (Aleksandropoulou et al. 2004). Therefore, according to the above equations, the radius for dry particles equals to half of their wet radius, $r_d = 0.5\ r$ (Fitzgerald 1975).

For particles with $r < 7$ μm, we used the equations by Monahan et al. (1986), which empirically estimates the emissions of sea salt particles from the whitecap cover as a function of wind speed. Specifically, for $u < 10$ m/s the production of film and jet drops is estimated by (Eq. 12):

$$\frac{\partial F}{\partial r} = 1.373u^{3.41}r^{-3}\ \ 1+0.057r^{1.05}\ \ 10^{1.19e^{-B^2}}$$

where, u is the wind speed at 10m height above the sea surface and B an empirical coefficient $B = 0.380 - \log r/0.650$ (Eq. 13).

The direct spume mechanism for $u > 10$ m/s was not taken into account due to high uncertainty (the equations for this mechanism are not presented; Monahan et al. 1986). The emissions of seasalt particles with r<7 μm at wind speeds greater than 10m/s were rather estimated as film and jet drops using the above equations.

For particles with r > 7μm we used the equations by Smith et al. (1993) (Eq. 14):

$$\frac{\partial F}{\partial r} = \overset{2}{\underset{i=1}{}}\ A_i e^{f\ \ln\frac{r}{R_{0i}}^{\,2}}$$

where, $f_1 = 3.1$, $f_2 = 3.3$, $R_{01} = 2.1$ μm, $R_{02} = 9.2$ μm, $\log A_1 = 0.0676u + 2.43$ (Eq. 15) and $\log A_2 = 0.0959u^{1/2} - 1.476$ (Eq. 16).

For each size bin the particles flux F was converted to mass $M_{r,tot}$ (g/h) by the equation: $M_{r,tot} = F\,A\,M_d\,3.6\times10^{6}\,dr$ (Eq. 17) where, A is the area of the grid cell covered by sea and M_d (kg) the mass of dry particles $M_d = 4/3\,\pi\rho r_d^3\,10^{-18}$ (Eq. 18). The density ρ of dry seasalt particles is ~2200 kg/m^3 (Hess 1998).

Table 3 Emission factors for isoprene (EIS), monoterpenes (ETE) and other biogenic VOCs (EOV) and biomass densities (BIS, BTE, BOV) per land cover type (only with non zero values) for summer (S) and winter (W) periods

LABEL1	LABEL2	LABEL3	BIS		EIS		BTE		ETE		BOV		EOV	
			S	W	S	W	S	W	S	W	S	W	S	W
Artificial surfaces	Artificial. non-agricultural vegetated areas	Green urban areas	0	0	0	0	400	200	0.1	0.1	400	200	1.5	1.5
		Sport and leisure facilities												
Agricultural areas	Arable land	Non-irrigated arable land	818	409	1.541	1.54	818	409	0.225	0.225	818	409	1.5	1.5
		Permanently irrigated land	668	334	0.09	0.09	868	434	0.123	0.123	868	434	1.5	1.5
	Permanent crops	Vineyards[1]	533	0.1	6.623	0.1	641	109	0.392	0.8	641	109	1.482	1.5
		Fruit trees and berry plantations	30	30	60	60	30	30	0.2	0.2	30	30	1.5	1.5
		Olive groves	200	200	0	0	0	100	0	0	200	200	1.5	1.5
	Pastures	Pastures	424	0	0.103	0	524	0	0.186	0.1	524	50	1.478	1.5
	Heterogeneous agricultural areas	Annual crops associated with permanent crops	180	0	0.15	0	180	0	0.49	0	180	0	1.37	0
		Complex cultivation patterns												
		Land principally occupied by agriculture. with significant areas of natural vegetation	668	334	0.09	0.09	868	434	0.12	0.123	868	434	1.5	0.5
Forest and semi natural areas	Forests	Broad-leaved forest[2]	320	0	60	0	320	0	0.2	0	320	0	1.5	0
		Coniferous forest	900	500	0	0	900	0	3	3	900	500	1.5	1.5
		Mixed forest[3]	232	125	32	8	465	38	1.713	2.163	465	358	1.5	1.5
	Scrub and/or herbaceous vegetation associations	Natural grasslands	150	75	8	8	350	175	0.34	0.34	350	175	1.5	1.5
		Moors and heathland	350	175	8	8	350	175	0.6	0.65	350	175	1.5	1.5
		Sclerophyllous vegetation[4]	400	400	8	8	400	400	0.6	0.65	400	400	1.5	1.5
		Transitional woodland-shrub	552	223	11.674	0.09	685	289	0.135	0.123	685	289	1.5	1.5
	Open spaces with little or no vegetation	Beaches, dunes, sands	200	100	8	8	200	100	0.65	0.65	900	100	1.5	1.5
		Sparsely vegetated areas[5]	150	75	8	8	350	175	0.34	0.34	350	175	1.5	1.5
Wetlands	Inland wetlands	Inland marshes[6]	350	175	8	8	350	175	0.65	0.65	0	0	0	0

Mapping of current CLC classes to the BVOCs emission factors used in the estimation of BVOCs emissions over Greece: [1]cropland and pasture (wheat, orchards, vineyards) with woodland, (Yay,2005); [2]oak woodland, (Yay,2005); [3]woodland (oak, pine, gum), (Yay, 2005); [4]sparsely vegetated, (Yay,2005) - Corinair snap code 110403 (garrique);

---

[5] short grassland and sparse shrub, (Yay,2005); [6] moorland/heathland, Corinair snap code 110403

2.2.4 Emissions of Biogenic volatile organic compounds (BVOCs)

BVOCs are emitted from vegetation during photosynthesis, plant respiration and vaporization from stores within the plant tissue. They are primarily controlled by environmental conditions i.e. leaf level temperature and radiation fluxes (PAR - Photosynthetically Active Radiation). In particular, isoprene is newly synthesized in the leaves in plant chloroplasts during daytime thus its emissions depend on light and temperature. On the other hand monoterpenes are produced and stored in the plant tissue and their emissions are considered to be temperature dependent. Some studies however suggest that for some species monoterpene emissions are also light dependent (e.g. Steinbrecher et al. 2008; Tao and Jain 2005 and references therein). Finally the mechanism of OVOCs (oxygenated VOCs and sequiterpenes) emissions varies with different species, however they are generally considered to depend on temperature. Although the leaf-level radiation fluxes and temperatures are the most important parameters driving the emissions, other parameters such as the past environmental conditions (temperature, light) experienced by the leaves, the soil moisture stress, and the age of leaves have also significant influence on emissions (Guenther et al. 2006; Müller et al, 2008). Several models have been developed to quantify emissions of BVOCs from vegetation (e.g. MEGAN v2 – Guenther et al. 2006).

BVOCs emissions, from vegetation during photosynthesis, plant respiration and vaporization from stores within the plant tissue, were estimated using a modification of the methodology presented in the EMEP/CORINAIR Atmospheric Emission Inventory Guidebook (2007). Monthly calculations of light and temperature (isoprene emissions, $E_{L,T}$) or only temperature (monoterpenes and OVOCs emissions, E_T) dependent BVOCs emissions (in μg/month) can be performed with the equations (Eqs. 19 and 20):

$$E_{L,T} = \mathop{\qquad}\limits_{mm=mm1}^{mm2} \varepsilon AD\gamma_{iso}\ T_{mm}\ N_{d,mm}N_{L,mm}$$

$$E_{T} = \mathop{\qquad}\limits_{mm=mm1}^{mm2} \varepsilon AD\gamma_{mts}\ T_{mm}\ N_{d,mm}24$$

where ε (μg/gh) is the emission potential for any species at a standard temperature $T_s = 303$ K and a standard Photosynthetically Active Radiation (PAR) flux equal to 1000 μmol photons (400-700 nm)/m^{2}s, A is the area covered by a specific vegetation type (in m^2), D is the foliar biomass density (g of dry weight biomass foliage/m^2), $N_{d,mm}$ is the number of days per month mm, $N_{L,mm}$ is the number of standard daily light hours per month (depending on area's latitude), mm1 and mm2 are the start and the end of the growing season for a particular vegetation type, and γ is a unitless environmental correction factor representing the effects of temperature and solar radiation changes on emissions. A constant PAR flux of 1000 μmol/m^{2}s during light hours is assumed and monthly averaged ambient temperatures (T_{mm}) are used for the estimation of the environmental correction factors.

In contrast to the methodology proposed in EMEP/CORINAIR Guidebook (2007) different foliar biomass densities and emission potentials have been adopted for the growing (summer) and dormant (winter) season, therefore in our case mm1 is always 1 (January) and mm2 is always 12 (December). Emission potentials and foliar biomass densities (Table 3) were adapted from the EMEP/CORINAIR Guidebook (2007) and from Yay et al. (2005) and assigned to each land cover class separately for isoprene, monoterpene and OVOCs emissions in accordance with predominant species of Greek flora (Aleksandropoulou et al. 2011; FILOTIS database; Ministry of Rural Development and Food). The above values, implemented in the methodology, have been tested for their accuracy in estimating BVOCs emissions in Greece and good agreement between modelled and measured concentrations was found in a study performed by Spyridaki et al. (2007). In the current application the values of emission potentials and foliar biomass densities have been partly modified to better reflect flora in the area. In particular, emission potentials and foliar biomass densities for the dominant species and other vegetation classes present in the area of interest were retrieved from Steinbrecher et al. (2008) and Karl et al. (2009).

The environmental correction factors for light and temperature dependence of isoprene emissions are estimated with the equations (Guenther et al. 1991):

$$\gamma_{iso} = C_T C_L \quad \text{(Eq. 21)}$$

$$C_T = \frac{\exp\left(C_{T1}\left(T - T_S\right)/RT_S T\right)}{1 + \exp\left(C_{T2}\left(T - T_M\right)/RT_S T\right)} \quad \text{(Eq. 22)}$$

where C_T and C_L are the environmental correction factors for temperature-dependence and light-dependence of isoprene emissions, respectively, T is the leaf temperature (K), C_{T1} = 95000 J/mol, C_{T2} = 230000 J/mol, T_M = 314 K, T_S = 303 K, R is the gas constant (8.314 J/Kmol). According to the adapted methodology the value of C_L is considered equal to 1 through daylight hours. The environmental correction factor describing the effect of temperature on monoterpenes and OVOCs emissions is estimated by (Pierce et al., 1990):

$$\gamma_{mts} = \gamma_{OVOC} = \exp\left(0.0739\left(T - T_S\right)\right) \text{(Eq. 23)}$$

2.2.5 Anthropogenic emissions inventory

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 $PM_{2.5\text{-}10}$) anthropogenic emissions derived from the UNECE/EMEP database (EMEP/CLRTAP 2009; CEIP, Emission from Greece during 2000-2010 as used in EMEP models). The dataset includes the anthropogenic part of emissions from each country/area classified according to SNAP 97 (Selected Nomenclature for sources of Air Pollution) categories and additionally the emissions from ships (local and international sea traffic are included in sector 8; Lavender 1999). Emissions are given on annual basis and have a spatial resolution of 50×50 km^2. It must be noted that agricultural emissions of NMVOCs and NH_3 from fertilizer use, unfertilized crops and animals (manure management) were estimated as described in Aleksandropoulou et. al. (2011) since their values included in the UNECE/EMEP dataset have been found rather underestimated compared to the predictions of the bottom up emission inventory of agricultural NH_3 emissions during 1996 in the greater area of Athens, by Sotiropoulou et al. (2004). The location and emissions of LPS were derived from the E-PRTR database (European Pollutant Release and Transfer Register). PM_{10} emissions from the E-PRTR dataset were split to $PM_{2.5}$ and PM_{coarse} using the emission ratio $PM_{2.5}/PM_{10}$ from the EMEP database (≈0.64 for Sector 1, ≈0.23 for Sector 3, ≈0.37 for Sector 4, and ≈0.65 for Sector 10) and the TNO dataset (≈0.63 for Sector 6; Berdowski et al. 1998).

2.2.6 Aerosol formation

The contribution of BVOCs to aerosol formation was calculated according to the methodology of de Leeuw (2002). In particular, BVOCs emissions are multiplied by the aerosol formation potential and results are reported in PM_{10} equivalents. The aerosol formation potential is a weighting factor that accounts for the fraction of BVOCs emissions changing into aerosol and the molecular weight difference. The aerosol formation potential for NMVOCs on European level is 0.02. Additionally, the contribution of gaseous pollutants from anthropogenic sources to aerosol formation was calculated with weighting factors 0.54 for SO_2, 0.88 for NOx, 0.64 for NH_3.

2.3 Results

The annual anthropogenic and natural emissions of $PM_{2.5}$, and $PM_{2.5-10}$ from the AMA during the period 2000-2010 are presented in Figure 2. It was found that the annual amount of $PM_{2.5}$ emitted from the sea surface was in the range 6.5 to 9.1 Gg during 2000 - 2010. In particular, 3.0±0.5 Gg of $PM_{2.5}$ were emitted from open ocean and 4.8±0.2 Gg from the sea shore. Emissions of windblown dust ranged from 0.35 to 0.67 Gg per year. As regards $PM_{2.5-10}$, the annual averages of emissions during the period 2000 – 2010 were in the range 10.9 – 18.1 Gg and 34.1 – 40.6 Gg for sea salt open-ocean and sea-shore emissions, and 3.1 – 6 Gg for windblown dust. Overall, $PM_{2.5}$ emissions from natural sources in AMA in 2010 were decreased by approximately 9.6% compared to their 2000 values, while $PM_{2.5-10}$ emissions dropped by 8.5%. In particular, $PM_{2.5}$ and $PM_{2.5-10}$ emissions of windblown dust have decreased 22% while for sea salt particles the decrease was approximately 9% for the fine fractions of particles and 7.3% for the coarse.

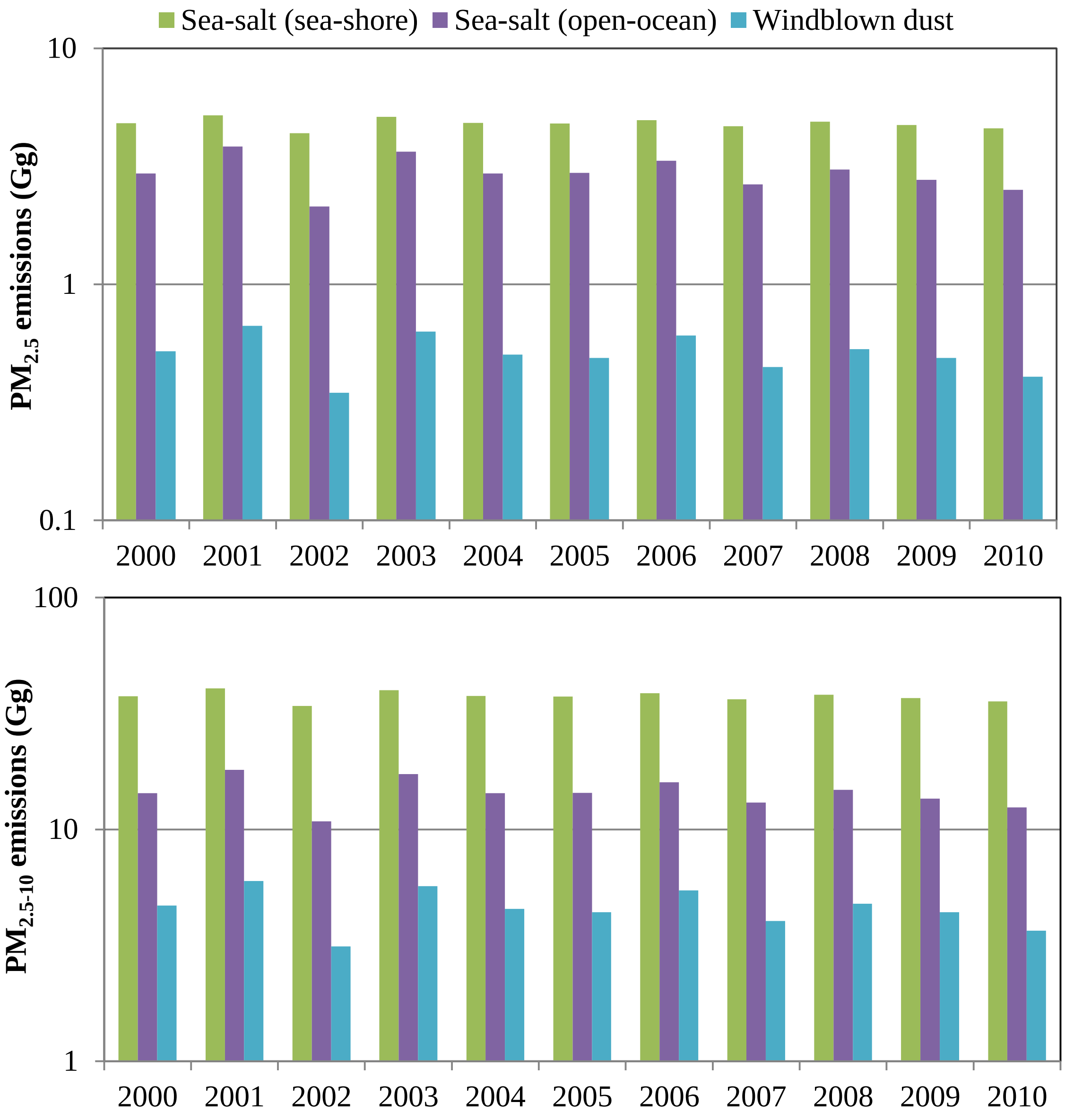


Figure 2 Annual emissions of natural $PM_{2.5}$ and $PM_{2.5\text{-}10}$ emissions from AMA for the period 2000-2010

Particulate matter emissions from TMA are depicted in Figure 3. Natural $PM_{2.5}$ emissions from the sea surface and agricultural and vacant lands increased approximately 8% during the period 2000 - 2010 The annual amount of $PM_{2.5}$ sea salt emitted from open ocean ranged from 0.05 to 0.07 Gg, at sea shore from 0.42 – 0.44 Gg whereas windblown dust $PM_{2.5}$ emissions ranged from 0.05 to 0.07 Gg. $PM_{2.5\text{-}10}$ emissions from natural sources were also increased from 2000 (4.2 Gg) to 2010 (4.6 Gg), approximately 7%. Their main source was sea salt particles (87.3±0.8%). In particular, 3.9±0.1 Gg of $PM_{2.5\text{-}10}$ sea salt particles were emitted annually during the period 2000-2010 whereas windblown dust emissions ranged from 0.48 to 0.63 Gg. The emissions of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ of windblown dust have increased 27% while for sea salt particles the increase was approximately 6% for the fine fractions of particles and 4.8% for the coarse.

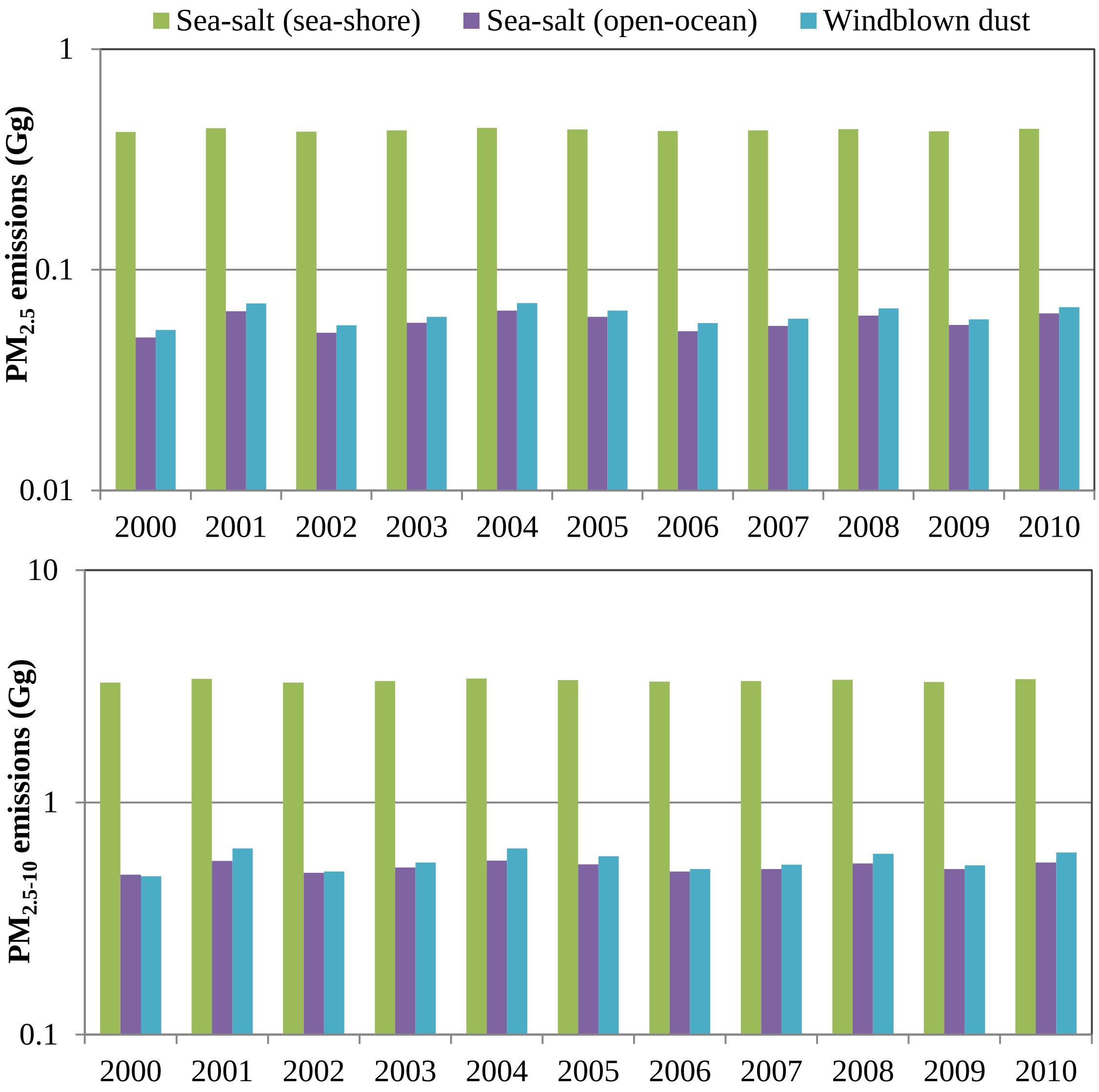


Figure 3 Annual emissions of natural $PM_{2.5}$ and $PM_{2.5\text{-}10}$ emissions from TMA for the period 2000-2010

Particulate matter emissions from GVA are depicted in Figure 4. Natural $PM_{2.5}$ emissions increased approximately 7.5% during the period 2000 – 2010. The annual amount of $PM_{2.5}$ sea salt emitted from open ocean ranged from 0.16 to 0.23 Gg, at sea shore from 0.36 – 0.39 Gg whereas windblown dust $PM_{2.5}$ emissions ranged from 0.04 to 0.06 Gg. $PM_{2.5\text{-}10}$ emissions from natural sources were also increased from 2000 to 2010 approximately 5.6%. Their main source was sea salt particles (92.4±0.5%). In particular, 4.2±0.2 Gg and 0.4±0.1 Gg of $PM_{2.5\text{-}10}$ sea salt and windblown dust particles, respectively, were emitted annually during the period 2000-2010. The emissions of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ of windblown dust have increased 17% while for sea salt particles the increase was approximately 6.8% and 4.5% for the fine and coarse fraction of particles, respectively.

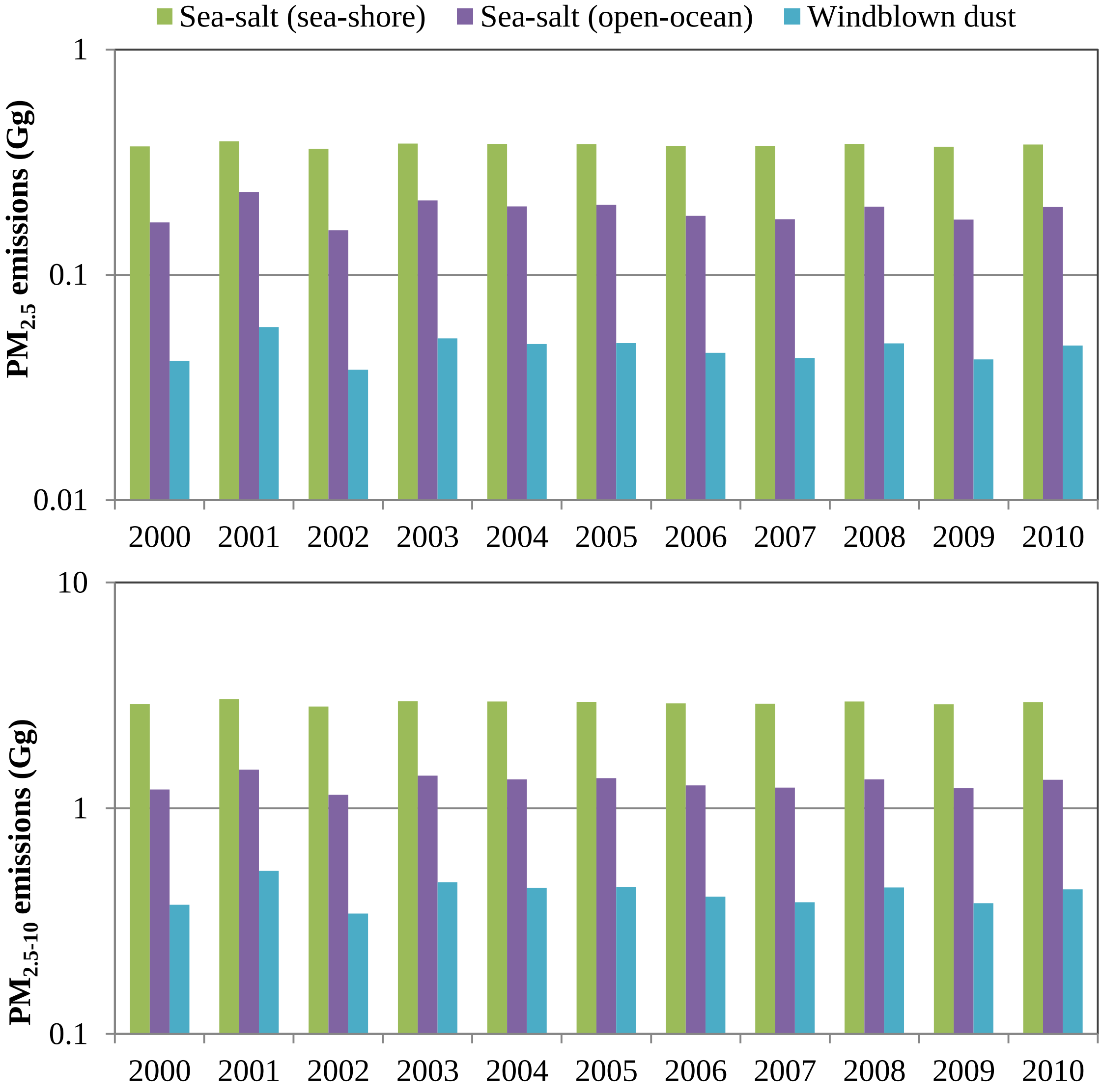


Figure 4 Annual emissions of natural $PM_{2.5}$ and $PM_{2.5-10}$ emissions from GVA for the period 2000-2010

In Figure 5 are shown the average emissions of BVOCs for the three areas of interest. It is observed that emissions of BVOCs are enhanced in 2010 compared to 2000. In particular, BVOCs emissions have increased since 2000 by 7.4% in AMA, by 3.6% in TMA and by 5.3% in GVA. The average emissions of BVOcs during the period 2000-2010 from AMA, TMA and GVA were 29.1±0.94 Gg, 40.7±2.35 Gg and 9.8±0.4 Gg , respectively. The annual variability in the emissions of BVOCs depends on the meteorological conditions.

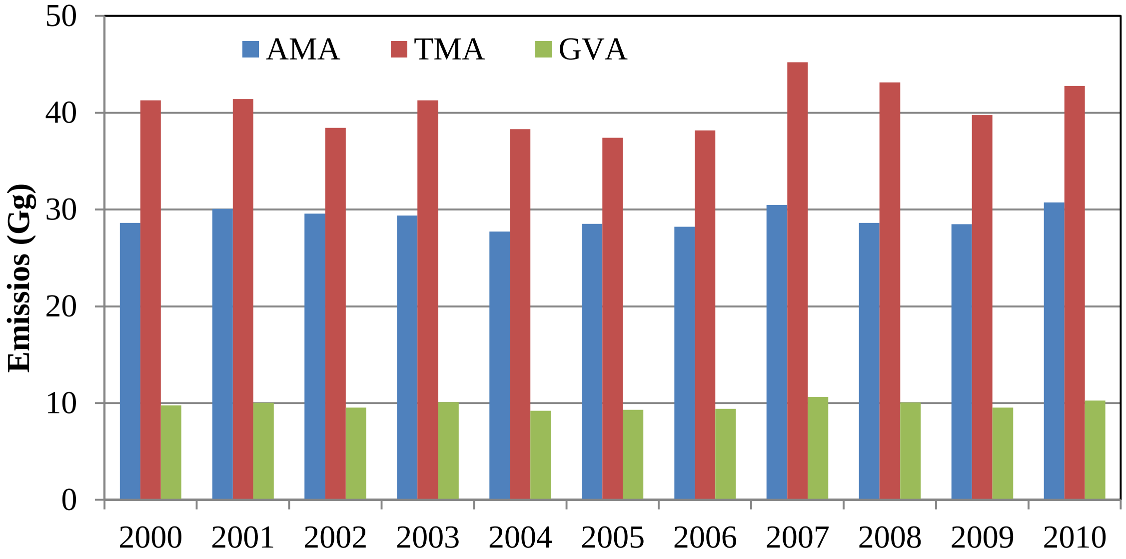


Figure 5 Annual emissions of BVOCs from AMA, TMA and GVA for the period 2000-2010

In Figures 6, annual natural PM_{10} emissions are depicted together with the anthropogenic emissions. For AMA a decreasing trend in natural PM emissions (8.6%) was observed for the period 2000 - 2010 while on the other side anthropogenic PM emissions were enhanced (51%), particularly during the last four years compared to the rest of the period. The contribution from natural sources to $PM_{2.5}$ and $PM_{2.5\text{-}10}$ emissions was approximately 39.9±4.2% and 92±1.7%, respectively. In particular, the contribution from the sea surface to the primary particulate pollution over the AMA was in the range of 32.3-42.8% for $PM_{2.5}$ while it was 84.5±1.4% for $PM_{2.5\text{-}10}$. Likewise, windblown dust emissions from agricultural and vacant lands accounted for approximately 2.5±0.4% and 7.4±0.8% of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ emissions in AMA, respectively. The annual variability in the relative contribution from natural sources to PM emissions depends on the meteorological conditions as well as on the variation of anthropogenic emissions.

As regard TMA, natural emissions of PM_{10} during the period 2000 – 2010 have increased by approximately 7.4%. $PM_{2.5}$ and $PM_{2.5\text{-}10}$ emissions from TMA were increased by 28% and 17%, respectively, during the period 2000-2010. Their main source was for $PM_{2.5}$ anthropogenic (4.3±0.4 Gg; ~88.6%) whereas for $PM_{2.5\text{-}10}$ natural. The contribution from natural sources to $PM_{2.5}$ and $PM_{2.5\text{-}10}$ emissions was approximately 11.4±0.9% and 74±2.2%, respectively. In particular, the contribution from the sea surface to the total particulate pollution over the TMA was 10.1±0.8% for $PM_{2.5}$ while it was 64.6±2.2% for $PM_{2.5\text{-}10}$. Likewise, windblown dust emissions accounted for approximately 1.3±0.1% and 9.4±0.6% of $PM_{2.5}$ and $PM_{2.5\text{-}10}$ emissions in TMA, respectively.

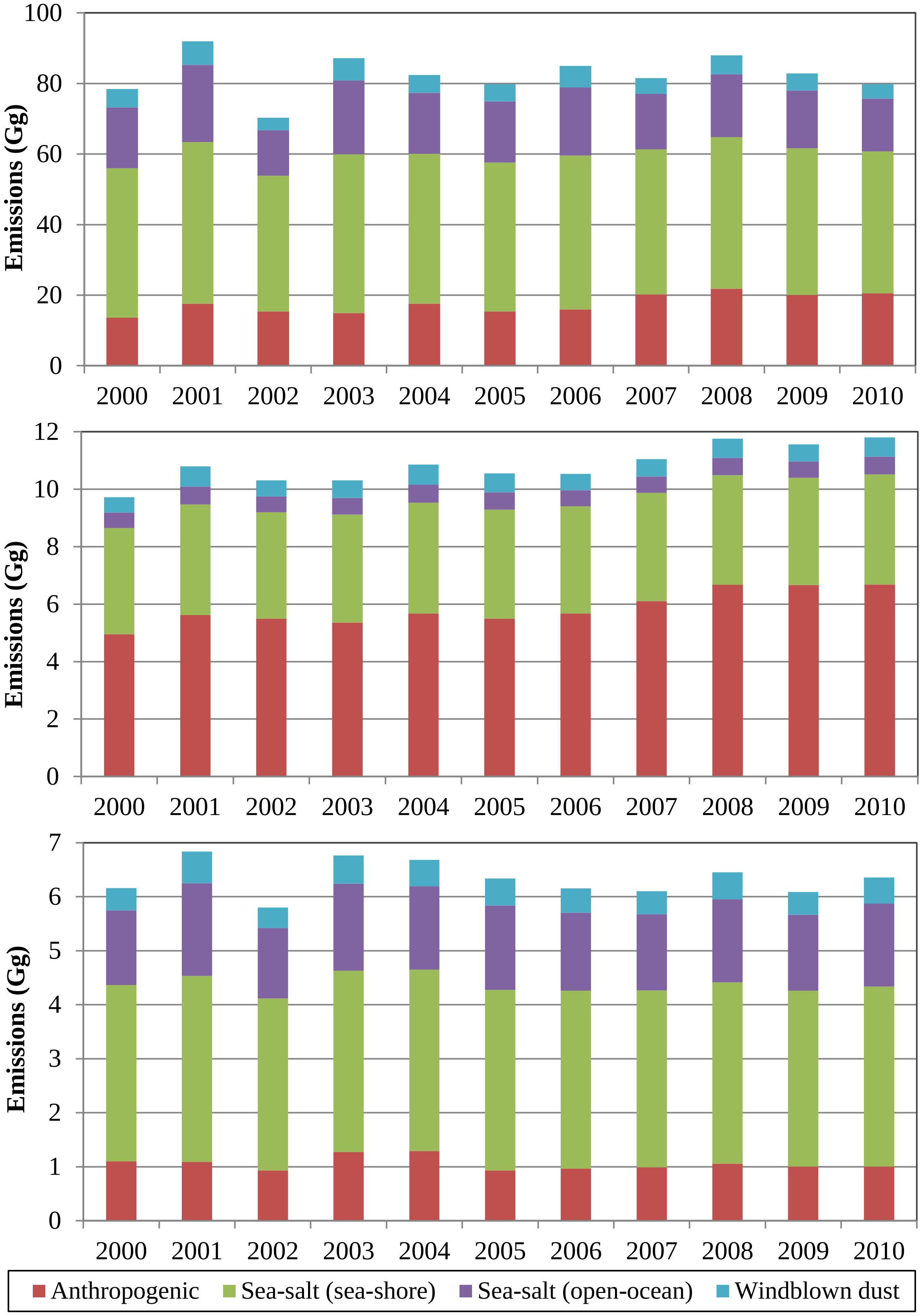


Figure 6 Annual emissions of natural and anthropogenic primary PM_{10} from AMA, TMA and GVA for the period 2000-2010

Figure 7 Contribution from natural sources to total annual primary and secondary particle emissions (PM_{10} equivalents) in AMA, TMA and GVA for the period 2000-2010

In GVA, natural emissions of PM_{10} during the period 2000 – 2010 have increased by approximately 5.8% while anthropogenic emissions decreased approximately 9%. The contribution from natural sources to $PM_{2.5}$ and $PM_{2.5-10}$ primary emissions from GVA changed -2.4% and 4.1%, respectively, during the period 2000-2010. The contribution from natural sources to $PM_{2.5}$ and $PM_{2.5-10}$ emissions was approximately 47.2±1.7% and 92.8±1.8%, respectively. In particular, the contribution from the sea surface to the total particulate pollution over GVA was 43.6±1.4% for $PM_{2.5}$ and 84.4±1.9% for $PM_{2.5-10}$. Windblown dust emissions accounted for approximately 3.6±0.3% and 8.4±0.6% of $PM_{2.5}$ and $PM_{2.5-10}$ emissions in GVA, respectively.

Overall the contribution from natural sources to total primary PM_{10} emissions were 78.7±3.04% for AMA, 46.1±2.2% for TMA and 83.4±1.3% for GVA.

In Figures 7 are shown the contributions from sea-salt, windblown dust, secondary PM formed by BVOCs and anthropogenic emissions to annual primary and secondary PM emissions for the three areas of interest. BVOCs and anthropogenic emissions are reported as PM equivalents using the aerosol formation factor (de Leeuw et al. 2002). It is observed that sea-shore is the most abundant source of natural PM emissions in all areas. In particular, sea salt particles emitted at the sea-shore account for approximately 64.8±2.3% of natural primary and secondary PM emissions and 20.4±1.8% of total primary and secondary PM emissions in AMA during the period 200 – 2010. The corresponding values are 65.1±0.9% and 7.8±1.1% for TMA and 60.6±1.4% and 18.5±3.6% for GVA. Overall the contribution from natural sources to total primary and secondary PM_{10} emissions were 31.5±2.8% for AMA, 11.97±1.63% for TMA and 30.5±6.1% for GVA.

References

Aleksandropoulou, V. and Lazaridis, M. (2004). Spatial Distribution of Gaseous and Particulate Matter Emissions in Greece. Water, Air & Soil Pollution 153, 15-34.

Aleksandropoulou, V., Torseth, K. and Lazaridis, M. (2011). Atmospheric emission inventory for natural and anthropogenic sources and spatial emission mapping for the Greater Athens Area. Water, Air & Soil Pollution 219, 507-526.

Aleksandropoulou, V., Torseth, K. and Lazaridis, M. (2013). The effect of forest fires in emissions of Biogenic Volatile Organic Compounds and windblown dust over urban areas. Air Quality, Atmosphere and Health 6, 277-294.

Alfaro SC, Gomes L (2001). Modeling mineral aerosol production by wind erosion: Emission intensities and aerosol size distributions in source areas. Journal of Geophysical Research 106(D16): 18075-18084

Berdowski, J.J.M., Mulder, W., Veldt, C., Visschedijk, A.J.H., and Zandveld, P.Y.J. (1998), Particulate emissions (PM_{10}-$PM_{2.5}$-$PM_{0.1}$) in Europe in 1990 and 1993. Bilthoven, RIVM Report.

Boman C., Nordin A., Öhman M., Boström D. (2005). 'Emissions from small-scale combustion of biomass fuels — Extensive quantification and characterization'. Energy

Technology and Thermal Process Chemistry Umeå University, STEM-BHM (P12648-1 and P21906-1), Umeå, February 2005.

Boman Ch., Nordin A., Boström D., and Öhman M. (2004). 'Characterization of Inorganic Particulate Matter from Residential Combustion of Pelletized Biomass Fuels'. Energy&Fuels 18, pp. 338–348, 2004.

Briggs, D. (2005), APMoSPHERE. Imperial College of Science, Technology and Medicine, London. www.apmosphere.org

CEC (1985), Soil Map of the European Communities, 1:1,000,000. Office for Official Publications of the European Communities, Luxembourg.

Choi, Y.J. and Fernando, H.J.S. (2008). Implementation of a windblown dust parameterization into MODELS-3/CMAQ: Application to episodic PM events in the US/Mexico border. Atmospheric Environment 42, 6039-6046.

CLRTAP CEIP (Centre on Emission Inventory and Projections): 2010 Submissions - Convention on Long Range Transboundary Air Pollution, Inventory Submissions 2010. http://www.ceip.at/submissions-under-clrtap/2010-submissions.

De Leeuw, F. (2002). A set of emission indicators for long-range transboundary air pollution. Environmental Science and Policy 5 (2), 135-145.

De Leeuw, G., Neele, F.P., Hill, M., Smith, M.H. and Vignali E. (2000). Production of sea spray aerosol in the surf zone. Journal of Geophysical Research-Atmospheres 105, 29397-29409.

Dockery DW, Pope CA, Xu X, Spengler JD, Ware JH, Fay ME, Ferris BG, Speizer FA (1993) An association between air pollution and mortality in six U.S. cities. New Engl J Med 329: 1753-1759.

Draxler RR, Gillette DA, Kirkpatrick JS, Heller J (2001) Estimating PM_{10} air concentrations from dust storms in Iraq, Kuwait and Saudi Arabia. Atmos Environ 35: 4315-4330.

EEA CLC2000 (2009). Corine land cover 2000 (CLC2000) 100 m - version 12/2009 (2009).http://www.eea.europa.eu/data-and-maps/data/corine-land-cover-2000-clc2000-100-m-version-12-2009/.

Ehrlich et al 2007. Ehrlich C, Noll G, Kalkoff W-D, Baumbach G, Dreiselder A. 'PM10 , PM2.5 and PM1.0 Emissions from industrial plants — Results from measurement programmes in Germany', *Atmospheric Environment* Vol. 41, No 29 (2007) pp. 6236–6254.

EMEP/CLRTAP (2009); EMEP Centre on Emission Inventories and Projections (CEIP), Emission used in EMEP models from Greece during 2007, http://www.ceip.at/emission-data-webdab/emissions-used-in-emep-models/

EMEP/CORINAIR (2007). EMEP/CORINAIR Atmospheric emission inventory guidebook – 2007. EEA Technical Report 16/2007.

EMEP/CORINAIR, Emission Inventory Guidebook (2009). Technical Report No 9, European Environmental Agency , http://reports.eea.eu.int/EMEPCORINAIR4/en.

EMEP/MSC-W (2011) EMEP/MSC-W Model.

https://wiki.met.no/emep/page1/emepmscw_opensource

E-PRTR (2009). European Pollutant Release and Transfer Register data base v1, as published on 09 Nov 2009. URL: http://prtr.ec.europa.eu.

E-PRTR-Dataset: EEA: E-PRTR-Dataset: The European Pollutant Release and Transfer Register (URL: http://www.eea.europa.eu/data-and-maps/data/member-states-reporting-art-7-under-the-european-pollutant-release-and-transfer-register-e-prtr-regulation-3, 05.08.2010)

ESDB v2.0 (2004), The European Soil Database distribution version 2.0, European Commission and the European Soil Bureau Network, EUR 19945 EN.

FAO–UNESCO (1974), FAO–UNESCO Soil Map of the World. Volume I: Legend. UNESCO, Paris.

FILOTIS: Database for the natural environment of Greece. http://www.itia.ntua.gr/filotis.

Fitzgerald, J.W. (1975). Approximation formulas for the equilibrium size of an aerosol particle as a function of its dry size and composition and the ambient relative humidity. Journal of Applied Meteorology 14, 1044-1049.

FOODSEC Meteodata Distribution Page. http://mars.jrc.ec.europa.eu/mars/About-us/FOODSEC/Data-Distribution.

Gillette DA, Passi R (1988) Modelling dust emission caused by wind erosion. J Geophys Res 93, 14233-14242.

Ginoux P, Chin M, Tegen I, Prospero JM, Holben B, Dubovik O, Lin S (2001) Sources and distributions of dust aerosols simulated with the GOCART model. J Geophys Res 106(D17), 20255-20273.

Guenther A, Karl T, Harley P, Wiedinmyer C, Palmer PI, Geron C (2006) Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature). Atmos Chem Phys 6, 3181-3210.

Guenther, A., Monson, R., Fall, R. (1991). Isoprene and monoterpene emission rate variability: Observations with eucalyptus and emission rate algorithm development. Journal of Geophysical Research 96, 10799-10808.

Gustavsson, L., Johansson, L, Leckner, B, Cooper, D, Tullin, C, Potter, A. 2004 b. 'Emission characteristics of modern and old-type residential boilers fired with wood logs and wood pellets', *Atmospheric Environment* Vol. 38, Issue 24, pp. 4183–4195, (2004).

Hayman, G., Bartzis, J., Dore, C., Ekstrand, S., Goodwin, J., Licotti, C., Olsson, B., Rabasco, C., Sjodin, A., Steinnocher, K., Tamponi. M., Vlachogiannis, D., and Winiwarter, W. (2001), IMPRESAREO - Improving the Spatial Resolution of Air Emission Inventories Using Earth Observation Data. Final Report. AEATyENVyRy0693. Culham, UK, AEA Technology.

Hays M.D., Smith N.D., Kinsey J., Dongb Y., Kariherb P. (2003). 'Polycyclic aromatic hydrocarbon size distributions in aerosols from appliances of residential wood combustion as determined by direct thermal desorption — GC/MS', *Aerosol Science*, 34, pp. 1061–1084, 2003.

Hellenic Civil Aviation Agency, http://www.ypa.gr/content/index.asp?tid=15

Hellenic Statistical Authority, PRODCOM 2008-2009, 2009-2010, Athens

Hess, M., Koepke, P. and Schult, I. (1998). Optical properties of aerosols and clouds: The software package OPAC. Bulletin of the American Meteorological Society 79, 831–844.

HNMS: Hellenic National Meteorological Service. http://www.hnms.gr.

Johansson, L., Persson H., Johansson, M., Tullin, C., Gustavsson, L., Sjödin, Å., Cooper, D., Potter, A., Paulrud, S., Lundén, E.B., Padban, N., Nyquist, L. & Becker, A., 2006: Fältstudie av metan och andra viktiga komponenter från vedpannor. Etapp 1. 2005. Slut-rapport för Energimyndighetsprojekt nr 21826-1.

Johansson, L.S., Leckner, B., Gustavsson, L., Cooper, D., Tullin, C. & Potter, A., 2004: Emissions characteristics of modern and old-type residential boilers fired with wood logs and wood pellets. Atmospheric Environment, 2004, 38.

Karl, M., Guenther, A., Köble, R., Leip, A. and Seufert, G. (2009). A new European plant-specific emission inventory of biogenic volatile organic compounds for use in atmospheric transport models. Biogeosciences 6, 1059-1087.

Katsouyanni K, Schwartz J, Spix C, Touloumi G, Zmirou D, Zanobetti A, Wojtyniak B, Vonk JM, Tobias A, Ponka A, Medina S, Bacharova L, Anderson HR (1996) Short-term effects of air pollution on health: a European approach using epidemiologic time series data: the APHEA protocol. J Epidemiol Commun H 50: S12-S18.

Katsouyanni K, Touloumi G, Samoli E, Gryparis A, Le Tatre A, Monopolis Y, Rossi G, Zmirou D, Ballester F, Boumghar A, Anderson H, Wojtyniak B, Paldy A, Barunstein R, Pekkanen J, Schnidler C, Schwartz J (2001) Confounding effect modification in the short-term effects of ambient particles on total mortality: results from 29 European cities within the APHEA 2 project. Epidemiology 12: 521-531.

Korcz, M., Fudala, J. and Klis, C. (2009). Estimation of wind blown dust emissions in Europe and its vicinity. Atmospheric Environment 43(7), 1410-1420.

Kubica K., (1997/1). 'Distribution of PAH generated in domestic fuels boilers'. Proc. of the ninth International Conference on Coal Science, Essen, Niemcy, 7–12.9.1997.

Kubica K., (2002/1). 'Emission of Pollutants during Combustion of Solid Fuels and Biomass in Small Appliances'. UN-ECE TFEIP Combustion and Industry Expert Panel Workshop on: 'Emissions from Small and Medium Combustion Plants', Ispra, April 2002, Procc. No I.02.87.

Kubica K., (2003/1). 'Environment Pollutants from Thermal Processing of Fuels and Biomass', and 'Thermochemical Transformation of Coal and Biomass' in Termochemical Processing of Coal and Biomass, pp. 145–232, ISBN 83-913434-1-3. Publication, copyright by IChPW and IGSMiE PAN, Zabrze-Kraków, 2003 (Polish).

Kubica K., Paradiz B., Dilara (2004/4). 'Toxic emissions from Solid Fuel Combustion in Small Residential Appliances'. Procc. sixth International Conference on Emission Monitoring CEM-2004, 9–11.6.2004, Milano Italy; www.cem2004.it .

Kupiainen, K., Klimont, Z., (2004). 'Primary Emissions of Submicron and Carbonaceous Particles in Europe and the Potential for their Control', IIASA Interim Report IR-04-079, www.iiasa.ac.at/rains/reports.html

Lavender K.A. (1999), Marine Exhaust Emissions, Quantification Study – Mediterranean Sea. Lloyds Register of Shipping Final Report, 99/EE/7044.

Liu, M., Westphal, D.L. (2001). A study of the sensitivity of simulated mineral dust production to model resolution. Journal of Geophysical Research 106, 18099-18112.

Mansell, G., Wolf, M., Gillies, J., Barnard, W. and Omary, M. (2004). Final Report: Determining Fugitive Dust Emissions from Wind Erosion. Prepared for Western Governors' Association (WRAP) by ENVIRON International Corporation.

Markakis, K., Poupkou, A., Melas, D., Tzoumaka,P., andPetrakakis, M. (2010), A Computational Approach Based on GIS Technology for the Development of an Anthropogenic Emission Inventory of Gaseous Pollutants in Greece, Water, Air, & Soil Pollution 207 (1), 157-180.

Markakis, K., Poupkou, A., Melas, D., Tzoumaka,P., and Petrakakis, M. (2010), A Computational Approach Based on GIS Technology for the Development of an

Anthropogenic Emission Inventory of Gaseous Pollutants in Greece, Water, Air, & Soil Pollution 207 (1), 157-180.

Marticorena, B., Bergametti, G., Gillette, D. and Belnap, J. (1997) Factors controlling threshold friction velocity in semiarid and arid areas of the United States. Journal of Geophysical Research 102, 23277-23287.

Ministry of Environment, Energy and Climate Change (2012): Annual Inventory Submission of Greece under the Convention and the Kyoto Protocol for Greenhouse and other Gases for the years 1990-2010 (NIR).

Ministry of Rural Development and Food. The current situation of Greek Forestry. available online at www.minagric.gr.

Monahan E.C., Spiel D.E. and Davidson, K.L. (1986), A model of marine aerosol generation via whitecaps and wave disruption. In Monahan E.C. and Mac Niocail G. (eds.), Oceanic Whitecaps, Reidel D., Norwell, MA, pp. 167-174.

Müller JF, Stavrakou T, Wallens S, De Smedt I, Van Roozendael M, Potosnak MJ, Rinne J, Munger B, Goldstein A, Guenther AB (2008) Global isoprene emissions estimated using MEGAN, ECMWF analyses and a detailed canopy environment model. Atmospheric Chemistry & Physics 8, 1329-1341.

National Statistical Service of Greece (NSSG), "Tourist Statistics", 1990 – 2001, Athens

National Statistical Service of Greece (NSSG), "Transport and Communications Statistics", 1990 – 2001, Athens

National Statistical Service of Greece (NSSG), PRODCOM, 1993-2007, Athens

Nickovic S, Dobricic S (1996) A model for long-range transport of desert dust. Mon Weather Rev 124: 2537-2544.

Ntziachristos L., Gkatzoflias D., Kouridis Ch., Samaras Z. COPERT: A European Road Transport Emission Inventory Model. Information Technologies in Environmental Engineering. Environmental Science and Engineering 2009; DOI 10.1007/978-3-540-88351-7_37, © Springer-Verlag Berlin Heidelberg

Ntziachristos L., Samaras Z., 2000, "COPERT III - Computer programme to calculate emissions from road transport. Methodology and emission factors (Version 2.1)", Technical Report No. 49, European Environmental Agency, Copenhagen

Park, S., In, H. (2003), Parameterization of dust emission for the simulation of the yellow sand (Asian dust) event observed in March 2002 in Korea, Journal of Geophysical Research 108 (D19), 4618.

Pierce, T., Lamb, B. and van Meter, A. (1990). Development of a biogenic emissions inventory system for regional scale air pollution models. Proceedings of the 83rd Air and Waste Manangement Association Annual Meeting.

Placet M. Mann CO, Gilbert RO, Niefer MJ (2000). Emissions of ozone precursors from stationary sources: a critical review. Atmospheric Environment 34, 2183-2204.

Pope CA, Burnett RT, Thun MJ, Calle EE, Krewski D, Ito K, Thurston GD (2002) Lung Cancer, Cardio-pulmonary mortality and Long-term exposure to fine particulate air pollution. J Am Med Assoc 287(9): 1132-1141

Pope CA, Thun MJ, Namboodiri MM, Dockery DW, Evans JS, Speizer FE, Heath Jr. DW (1995) Particulate air pollution as a predictor of mortality in a prospective study of U. S. adults. Am J Respir Crit Care Med 151: 669-674

Poupkou, A., Symeonidis, P., Lisaridis, I., Melas, D., Ziomas, I., Yay O. D. and Balis D. (2008). Effects of anthropogenic emission sources on maximum ozone concentrations over Greece. Atmospheric Research 89(4), 374-381.

Poupkou, A., Symeonidis, P., Lisaridis, I., Melas, D., Ziomas, I., Yay O. D. and Balis D. (2008). Effects of anthropogenic emission sources on maximum ozone concentrations over Greece. Atmospheric Research 89(4), 374-381.

Poupkou, A., Symeonidis, P., Ziomas, I., Melas, D., Markakis, K. (2007). A spatially and temporally disaggregated anthropogenic emission inventory in the southern Balkan region. Water, Air, and Soil Pollution 185 (1-4), 335-348.

Poupkou, A., Symeonidis, P., Ziomas, I., Melas, D., Markakis, K. (2007). A spatially and temporally disaggregated anthropogenic emission inventory in the southern Balkan region. Water, Air, and Soil Pollution 185 (1-4), 335-348.

Prandtl, L. (1935) Aerodynamic Theory Vol. III. In: Durand F (ed) Springer-Verlag, Berlin, pp 57-100.

Progiou Athena G., Ioannis C. Ziomas (2011b): Road traffic emissions impact on air quality of the Greater Athens Area based on a 20 year emissions inventory. Science of the Total Environment DOI information: 10.1016/ j.scitotenv.2011.09.050

Progiou Athena, Ioannis Ziomas (2011a): Twenty-Year Road Traffic Emissions Trend in Greece, Water Air Soil Pollut DOI 10.1007/s11270-011-0859-9.

Shao Y (2001) A model for mineral dust emission. J Geophys Res 106: 20239-20254

Shaw WJ, Allwine KJ, Fritz BG, Rutz FC, Rishel JP, Chapman EG (2008) An evaluation of the wind erosion module in DUSTRAN. Atmos Environ 42: 1907-1921

Smit R., Ntziachristos L., Boulter P. (2010) Validation of road vehicle and traffic emission models - A review and meta-analysis. Atmos Environ 2010; 44: 2943-53.

Smith M.H., Park P.M. and Consterdine I.E. (1993), Marine aerosol concentration and estimated fluxes over seas, Q. J. R. Meteorological Society, 809-824.

Sotiropoulou R.E.P., Tagaris E. and Pilinis C. (2004), An estimation of the spatial distribution of agricultural ammonia emissions in the Greater Athens Area, Sci. Tot. Environ., 318, 159–169.

Sotiropoulou R.E.P., Tagaris E. and Pilinis C. (2004), An estimation of the spatial distribution of agricultural ammonia emissions in the Greater Athens Area, Sci. Tot. Environ., 318, 159–169.

Spyridaki, A., Aleksandropoulou, V., Latos, M., Flatoy, F., Svendby, T. M. and Lazaridis, M. (2007). Contribution of natural emissions to ambient aerosol concentration levels in the eastern Mediterranean. Abstracts of the European Aerosol Conference 2007, Salzburg.

Steinbrecher, R., Smiatek, G., Köble, R., Seufert, G., Theloke, J., Hauff, K., Ciccioli, P., Vautard, R. and Curci, G. (2008). Intra- and inter- annual variability of VOC emissions from natural and semi-natural vegetation in Europe and neighbouring countries. Atmospheric Environment 43(7), 1380-1391.

Symeonidis Panagiotis Ioannis Ziomas, Athena Proyou (2004): Development of an Emission Inventory System from Transport in Greece, Environmental Modelling and Software, Vol. 19, Issue 4, pp 413-421.

Symeonidis Panagiotis, Ioannis Ziomas, Athena Proyou (2003): Emissions of Air Pollutants from the Road Transport Sector in Greece: Year to Year Variation and Present Situation., Environmental Technology Vol. 24, Num 6.

Symeonidis, P., Poupkou, A., Gkantou, A., Melas, D., Yay, D., Pouspourika, E., and Balis, D. (2008), Development of a computational system for estimating biogenic NMVOCs emissions based on GIS technology. Atmospheric environment 42(8), 1777-1789.

Symeonidis, P., Poupkou, A., Gkantou, A., Melas, D., Yay, D., Pouspourika, E., and Balis, D. (2008), Development of a computational system for estimating biogenic NMVOCs emissions based on GIS technology. Atmospheric environment 42(8), 1777-1789.

Symeonidis, P., Ziomas, I., and Proyou, A. (2004), Development of an emission inventory system from transport in Greece. Environmental Modelling and Software 19(4), 413-421.

Tao Z, Jain AK (2005) Modeling of global biogenic emissions for key indirect greenhouse gases and their response to atmospheric CO_2 increases and changes in land cover and climate. J Geophys Res 110: D21309

Tzannatos Ernestos (2010): Ship emissions and their externalities for the port of Piraeus-Greece, Atmospheric Environment Vol. 44, pp. 400-407

Uherek E., Halenka T., Borken-Kleefeld J., Balkanski Y., Berntsen T., Borrego C. et al.(2010) Transport impacts on atmosphere and climate: Land transport. Atmos Environ 2010; 44:4772-4816.

Westphal, D.L., Toon, O.B., Carlson, T.N. (1987), A two-dimensional numerical investigation of the dynamics and microphysical of Saharan dust storms. Journal of Geophysical Research 92, 3027–3049.

Winiwarter W, Kuhlbusch T.A.J, Viana M.and Hitzenberger R. (2009). Quality considerations of European PM emission inventories. Atmospheric Environment 43(25), 3819-3828.

Yay, O.D., Poupkou, A., Symeonidis, P., Gkantou, A., Melas, D. and Döğeroğlu, T. (2005). Biogenic Emissions of Volatile Organic Compounds from Turkey. Proceeding of the Third International Symposium on Air Quality Management at Urban, regional and Global Scales.

Zender CS, Bian H, Newman D (2003) The mineral Dust Entrainment And Deposition (DEAD) model: Description and 1990s dust climatology. J Geophys Res 108 (D14): 4416. doi:10.1029/2002JD002775

Zhang, K.M., Knipping, E.M., Wexler, A.S., Bhave, P.V. and Tonnesen, G.S. (2005). Size distribution of sea-salt emissions as a function of relative humidity Atmospheric Environment 39, 3373-3379.

Ziomas I.C.(1998) The Mediterranean Campaign of Photochemical Tracers-Transport and Chemical Evolution (MEDCAPHOT-TRACE): An outline. Atmos Environ 1998; 32, 12: 2045-2059.