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A GIS based anthropogenic PM_{10} emission inventory for Greece

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ABSTRACT

An anthropogenic, chemically speciated PM_{10} emission inventory was compiled for Greece in 10 km spatial resolution. The inventory comprises of all anthropogenic particulate matter sources and it was compiled using a Geographical Information System (GIS) integrated with SQL programming language. Input data from the national and international databases were used for the calculation of spatially and temporally resolved emissions for the road transport and all the subsectors of the other mobile sources and machinery sector using top–down or bottom–up methodologies. Annual data from existing emission databases were also used and were temporally and spatially disaggregated using source relevant statistical data and high resolution maps. The sectoral emission totals are compared with other emission databases or studies conducted in the area. Total anthropogenic emissions in Greece were estimated to be 182 219 t for the base year 2003. The results indicate the industrial sector as the major PM_{10} emission source (39.9% contribution) with the major industrial units though to be situated inside the organised industrial areas of the country. The power generation sector (21.4%) is the second largest contributor in national level mostly derived from one specific industrial region at north. International cargo shipping activities (9.6%) is also an important source category for particles. Heat production and road transport are found to play a significant role inside the urban centres of the country.

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1. Introduction

The impact of air pollution in human health, welfare and natural ecosystems is a key environmental problem in Greece and worldwide. The calculation of reliable quantitative emission data (i.e. the development of emission inventories) is necessary to produce emission fields over areas of interest without having to measure emissions on a continuous basis. In addition, a reliable spatially and temporally resolved emission inventory is a crucial dataset needed to support the implementation of chemical transport models (Symeonidis et al., 2004; Zanis et al., 2007; Athanasopoulou et al., 2008; Im et al., 2008) allowing us to assess whether compliance with air quality limits is attained (Ma and Van Aardenne, 2004).

A limiting drawback, concerning the usability of emission inventories for air quality modelling, is always the accuracy of emission estimates (Passant, 2003). A prerequisite for compiling accurate emission inventories is to assemble detailed and updated emission data or input parameters, which are in most cases unavailable or incomplete. The data frequently originate from different sources (international, national and local authorities, public and private companies) resulting in inconsistencies and incompatibilities, needed to be overwhelmed (Schneider et al., 1997). Assumptions are often compulsory, reflected to the inventory reliability. In addition, high spatial and temporal resolution is another precondition for an inventory, to efficiently support the estimation of pollution levels (Taghavi et al., 2005).

In Greece, particularly in the two largest urban agglomerations of Athens and Thessaloniki, restriction policies implemented in the last years has reduced gaseous pollutant levels below the ambient EU thresholds with a few exceptions, depending on location and time period. On the contrary, PM_{10} continues to present extensive exceedences in both Thessaloniki and Athens as well as in other areas of the country e.g. where the major power plant complexes are located (Tzimou-Tsitouridou, 2004). Twenty four hour averaged PM_{10} concentrations at four monitoring stations distributed through the greater Athens area reveal a substantially higher number of days, than the allowed frequency of 35 days per year, for which the concentrations exceeded the 50 $\mu g\ m^{-3}$ limit (HMEPPPW, 2006). Moreover, in the aforementioned stations, the annual mean concentrations are well above the threshold goal limit of 40 $\mu g\ m^{-3}$. The situation in the city of Thessaloniki is equally bad where at an urban traffic monitoring station the 24 h concentration threshold is exceeded almost every day of the year (District of central Macedonia, 2006) ranking the city among the most polluted in Europe regarding PM_{10}. Taking into account the scientific issues associating ambient concentrations of particulate matter with adverse respiratory health effects and mortality (Pope and Dockery, 2006), there is a growing concern for a detailed PM_{10} emission inventory for Greece in order to study air pollution, introduce effective abatement measures and thus fulfill the environmental goals.

To date, there have been only a limited number of studies regarding the compilation of PM_{10} emission inventories in Greece while no published PM_{10} quantification study exists for Greece with high spatial and temporal resolution (Economopoulou and Economopoulos, 2002; Tsilingiridis et al., 2002; Aleksandropoulou and Lazaridis, 2004; Giannouli et al., 2006; Poupkou et al., 2007; Markakis et al., 2009). These studies present one or more of the following limitations: they are either limited to urban scale, lack all anthropogenic sources or they lack the spatial and/or temporal processing. In addition, there is hardly any information concerning the quantification of the non-exhaust PM_{10} emissions (e.g. tire-wear, break-lining and road abrasion) in Greece. For the aforementioned reasons, these studies cannot provide a PM model ready emission inventory (gridded, temporally analyzed, and chemical speciated) that can be used for modelling studies in Greece.

To fill the gap, this paper presents the development of a PM_{10} emission inventory for Greece, with a high spatial and temporal resolution. The best available activity and statistical information were used to either quantify emissions or pre-defined gridded or point source emissions are combined with geographical information (Dai and Rocke, 2000) consisting of high resolution maps (cruising lines of ships, road network, population as well as the 100 m resolution land cover maps of CORINE- Coordination of Information on the Environment) and scripted into GIS software aiming to develop an emission processor which can easily produce and/or update annual/weekly/hourly gridded emission fields for five PM chemical compounds.

2. General Description of the Emission Inventory

The PM_{10} emission inventory presented in this paper is spatially allocated in a 10 km resolution grid over Greece, compiled for the reference year 2003 (Figure 1). The domain was determined in Lambert conformal conic (LCC) coordinate system. Emissions considered here are those from: energy sector (power plants), industrial sector, non–industrial combustion plants, coal extraction, road transport (exhaust and non–exhaust emissions), other mobile sources and machinery and agriculture. The emissions of the non–industrial combustion plants are considered to originate from heat production/heating operation (residential, commercial and institutional) (Klimont et al., 2002). Therefore, this sector will be referred as heat production. In addition, only anthropogenic emissions have been considered in the present study.

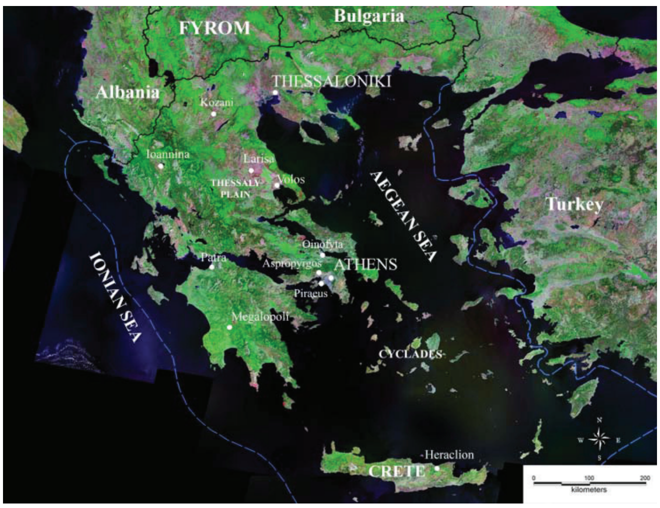


Figure 1. *Landsat overview of the study area.*

The presented inventory includes three different types of sources that also represent the quality level of the derived emissions:

(1) Real activity data which are used to quantify the emissions from the road transport sector, the other mobile sources and machinery sector (except international shipping), and the lignite mining operations. The dataset for the transport sectors represents the best available activity data in Greece and an additional effort was made to update the dataset. The data employed for the quantification of the transport sectors are assembled during the "Environmental Inventory System for Transport" (EIST) (Symeonidis et al., 2003) and for the mining operations data were provided by the mining company.

(2) Emission databases derived from national sources. The power plants and the industrial sector are covered from official databases. The datasets consist of the annual 2004 emissions for all power plants, provided by the Greek Public Power Corporation and for the industrial sector the annual emission data originate from the 2001 National industrial registry (HMEPPPW, 2001) covering more than 900 small, medium and large industrial units in Greece. The emissions of a small number of industrial units of the HMEPPPW (2001) database were replaced by the updated emissions for the year 2004, reported in the European Pollutants Emission Register (EC, 2000).

(3) Emissions derived from pre-compiled spatially disaggregated emission inventories. To fill for the remaining sectors (heat production, waste treatment and disposal, international shipping and agriculture) the annual 2003 emission data of the EMEP (European Monitoring and Evaluation Programme) database were used (Vestreng et al., 2006) (expert estimates retrieved from http://www.ceip.at/emission-data-webdab/emissions-used-in-emep-models/).

The inventory for each source sector is compiled in four steps. In the first step, monthly or annual emission data are either calculated or derived from existing databases. To achieve this, the code uses different tables containing the activity data or the statistical data as well as tables containing the parameters used for the quantification (emission factors).The second and third steps introduce the spatial disaggregation and temporal variation (monthly, weekly, and diurnal profiles) of emissions. To provide spatially resolved emissions the code uses geographical maps (i.e., population density, landuses) in combination with source specific spatial indicators which link an emission source with its geographical information. The high resolution landuses of CORINE (EEA, 2000) provided in raster format is used to spatially disaggregate a number of emission sources. Table 1 summarizes the main characteristics of the inventory including the monthly temporal and the spatial indicators implemented as well as the input data used to compile the emission inventory. All weekly profiles were compiled specifically for Greece within the scope of the EUROTRAC/GENEMIS project (Friedrich, 1997). The latter is also the reference source for all diurnal variation profiles with the exception of the power generation sector and the industrial sector taken from a local study (Tsilingiridis et al., 2002), the diurnal profiles for the road transport sectors' bottom–up emissions which derive from the input activity data and heat production diurnal profile for the large urban centres of Athens and Thessaloniki. Furthermore, splitting of PM emissions is needed for the characterisation of their chemical composition. Since specific chemical profiles which represent the technology used in Greece are lacking the chemical splitting was performed using source specific profiles derived from CARB (2007). PM_{10} emissions were chemically speciated in organic and elemental carbon, nitrates, sulphates and other particles.

For the aforementioned steps to be completed an emission processor was developed. The processor is written in Mapbasic 9.0 programming language and integrated into Mapinfo 9.0 GIS software. The advantage of integrating computer code with GIS is the ability to directly link the emission sources to their geographical activity level, relatively easy (few simple SQL commands). The processor was structured to produce spatially, temporally and chemically disaggregated PM_{10} emissions over a gridded domain starting from yearly or monthly emissions either calculated or derived from the pre-existing databases with the use of source relevant spatiotemporal indicators.

Table 1. *Input data used in the inventory and their geographical activity level*

Emission source sector (spatial analysis)	Input data	Monthly profile applied[a]	Spatial Allocation
Power plants (point)	Annual emissions and stack parameters for 64 individual stacks, part of 15 thermoelectric power generation units.	IPI[b]	Geographical location
Industry[c] (point)	Annual emissions for the industrial units of Greece. (HMEPPPW, 2001).	IPI per industry type	Geographical location
Bottom-up road transport emissions[d] (line)	Traffic loads provided for 700 segments of the national road network of Greece and 1 700 segments of the road network of Thessaloniki, segment length	EIST/GENEMIS[f]	Road segments
Top-down road transport emissions[d] (area)	Mileage driven per vehicle type and engine technology[e], vehicle fleet per vehicle type and engine technology	EIST/GENEMIS[f]	Bottom-up information/road length (section 3.3)
Passenger/ferries[d] (line)	Emission factor (compiled from engine output power and number of engines per vessel), vessels fleet per route/month, route distance, vessel cruising speed	Monthly activity per cruising route	Sea routes
Recreation vessels[d] (area)	Vessels number per prefecture, typical travel distance and speed, fleet gross tonnage	GENEMIS	Prefecture sea area
Fishing boats[d] (area)	Vessels fleet per prefecture, working time, fleet gross tonnage	GENEMIS	Prefecture sea area[h]
Off-road agriculture and industrial machinery[d] (area)	Vehicles per prefecture, working hours, engine power, load factor, emission factor	Monthly activity per vehicle type	Agriculture areas/ Population
Railway transport[d] (line)	Routes per line/month, working hours, engine power, load factor	Monthly activity per route	Railway network
Aviation (airport)	LTO cycles per airport[i], aircraft type	Monthly activity per airport	Geographical location
Coal extraction (mining areas)	Extraction capacity in the major coal fields of Greece	Monthly activity per coal field	Mining areas
1. Central heating 2. Waste treatment and disposal 3. International cargo shipping 4. Agriculture	EMEP database for the year 2003 (provided in 50 km resolution grid).	1. See section 3.2 2. GENEMIS 3. Port activity[j] 4. GENEMIS	1. See section 3.2 2. Population density 3. Area proportion 4. CORINE land cover

[a] The GENEMIS project (Friedrich, 1997) is the reference source for all diurnal and weekly profiles with the exception of the diurnal profile for the power plants and the industrial sector taken from Tsilingiridis et al. (2000) and the diurnal profiles for the road transport bottom-up emissions which derive from the input activity data.

[b] For industrial facilities the industrial production indicator (IPI), provided by the Greek statistical service, was used as a monthly surrogate. The IPI of a product is linked with the emitted quantities of pollutants relevant to that facility (Pham et al., 2008).

[c] The industrial sector units were classified in 7 categories: food, metal, ceramics, lime, cement, chemical, other.

[d] Data derived from EIST (Symeonidis et al., 2003).

[e] Calculated from fuel consumption statistics (Symeonidis et al., 2003).

[f] For the network of Greece in which no traffic counts were available the annual and diurnal profiles as in GENEMIS. For the road network in which traffic counts were measured the diurnal variation was based on the traffic counts and the annual variation as in GENEMIS. The traffic counts provided the diurnal variation for two large urban centers and the annual variation of emissions as in GENEMIS for Athens and as in Tsilingiridis et al. (2002) for Thessaloniki.

[g] The fuel consumption per vessel is calculated from engine power data.

[h] Except for Thessaloniki where the fishing areas were derived from Tsilingiridis et al. (2002).

[i] In the EIST database the old Athens airport (Elliniko) was included. In our input database the LTOs and the location of the currently operational Venizelou international airport is included.

[j] Activity data for the activity of cargo ships in major ports of the country were available from the EIST database (Symeonidis et al., 2003).

3. Methodology

3.1. Power plants and industries

The emissions for all power plants in the country were provided by the Greek Public Power Corporation. For a number of power units located in the area of Kozani at the north which hosts the largest and most polluting units of the country (75% of the national totals) the emissions are measured on continue basis in the stacks. The database contains 15 individual units (stacks) grouped in 5 large power plants. These are provided as monthly emissions rates. For the rest of the power plants of the country, the emissions were derived either from circumstantial stack measurements or calculations performed by the company which are based on the equipped boiler technology and various operational characteristics. Details on these calculations were not known.

The emissions of the industrial sector is the outcome of a large project of the ministry of the environment (HMEPPPW, 2001) which has commissioned five companies aiming to develop a complete emission database containing all the major industrial units of the country, categorized according to type of manufactured product, type of pollution (water, air) and geographical location. The project followed two phases. During the first phase an emission factor database was developed for the first time in order to reflect the technological status of the Greek industry e.g. the industry types, specific processes, fuels used and abatement measures implemented. For this, a thorough survey of the international literature (WMO, 1993; EPA, 1999; EPA, 2000; EMEP/CORINAIR, 2001) was carried out. In addition for several industrial processes like oil press units, oil refineries, various chemicals, ceramic industry, cement and lime production industries, aluminium production and lead foundries, the emissions factors were compiled from field measurements. An inter–comparison between the compiled emission factors and those derived from the bibliographical sources was made in order to select the appropriate emissions factor for the Greek case. Finally, an uncertainty analysis was done for each of the industry types and processes contained in the database. The second phase involved the calculation of emissions. The latter was done from direct measurements in some cases or quantified from the assembled activity data which derived from local authorities, ministries or the industries themselves either through the use of questionnaires or after personal communication with the representatives of the industries. The database includes more than 900 individual units in the country.

3.2. Heat production

The gridded annual 2003 emission data of the EMEP database were spatially disaggregated on the 10 km resolution grid over Greece on the basis of geographically resolved population statistics. The downscaling of emissions within the large urban agglomerations of Athens and Thessaloniki was performed based on building statistics (categorized as residential, commercial buildings and buildings for other uses). Regarding the temporal split of emission data, two different approaches were used for Greece and the two largest urban centres. For Greece, the monthly variation of emissions in each 10 km grid cell was produced based on climatic temperature data. More particularly, mean monthly temperature maps were created using data from the CRU global climate dataset (Brohan et al., 2006) interpolated in the 10 km grid. On the basis of those maps and the critical temperature of 14 °C, below which the boilers are considered operational in Greece, (Matzarakis and Balafoutis, 2004) the heating degree day indicator was calculated for each month from the difference between the mean monthly climatic temperature and the critical temperature (Poupkou et al., 2007) to represent the monthly proportion of annual emissions in each cell. Weekly and diurnal profiles from the GENEMIS project were used (Friedrich, 1997) for Greece. For Athens and Thessaloniki, the monthly variation of emissions was based on the consumption of diesel for heating, whereas the diurnal profile was based on typical working hour profiles for each building type (HMEPPPW, 2001).

3.3. Road transport

The quantification of the road transport sector emissions was completed using both the macroscale and the microscale approaches (EMEP/CORINAIR, 2001). The microscale approach (bottom–up) is based on activity data e.g. traffic loads which are measured under real driving conditions in specific roads of the network under investigation. This study uses data which were assembled in the year 1998 in the framework of the EIST project and consisted of hourly traffic loads and vehicle speeds for 7 vehicle categories (passenger cars, taxis, light duty vehicles, heavy duty vehicles, urban buses, coaches and two-wheeled vehicles) (Symeonidis et al., 2003). The database contains about 700 road segments of the national road network of Greece, 1 001 road segments in Athens and 1 710 road segments in the city of Thessaloniki, all derived either by street measurements or as the output of sophisticated traffic circulation models. It has to be stated that, the EIST study as regards the road data was conducted in a typical weekday and does not provide other temporal information, thus in order to derive the annual emission amounts, the daily total bottom–up emissions were multiplied by 330 in order to take into account the reduction of the emitted quantities during the weekends (Symeonidis, 2002). The macroscale approach (top–down) is based on general statistics like vehicle numbers and composition as well as fuel consumption amounts. The aforementioned data (also for 1998) were assembled within the EIST project. The database contains the circulating fleet in Greece as well as in the two large urban centres, categorised by vehicle type, engine capacity and technology as well as the annual gasoline and diesel consumption.

The calculations for the exhaust traffic emissions (gasoline and diesel powered vehicles) were performed using the methodology of EMEP/CORINAIR (2001). To estimate spatially resolved emissions, a combined methodology of both top–down approach (area sources) and bottom–up approach (line sources) was employed. The national road network of Greece and a portion of the road networks of Athens and Thessaloniki were processed as line sources and their emissions were calculated on the basis of traffic loads, vehicle speeds and road lengths in each road segment (Table 1):

$$E_i = \Sigma_j (HTL \cdot L) \cdot e_{i,j} \qquad (1)$$

where E_i the calculated emissions (g), HTL the hourly traffic counts, L the road segment length (km) and $e_{i,j}$ the emission factor (g km^{-1}) per vehicle type i and engine technology j (except for the non–exhaust emission factors which are not engine technology specific).

The emissions factors used in Equation (1) were compiled based on the provided speeds in each segment and the equations presented in the CORINAIR emissions inventory guidebook for exhaust emissions (speed and vehicle type dependent) (EMEP/CORINAIR, 2001). The same formula as Equation (1) was also used for the calculation of tire-wear, break–lining and road abrasion emissions using the emission factors of the GAINS model (Klimont et al., 2002) that depend on the vehicle type.

For the calculation of top–down emissions the emission factors were compiled using the average speeds per driving behaviour (19 km h^{-1} for urban driving, 60 km h^{-1} for rural driving and 90 km h^{-1} for highway driving) provided by EMEP/CORINAIR (2001). The length of the segment in Equation (1) was replaced by the annual mileage driven for each vehicle type and engine technology (i.e., conventional, Euro I) that was also provided in the EIST database. The hourly traffic load was also replaced in Equation (1) by the national fleet also categorised in vehicle types and engine technologies (Table 1).

The input activity data (traffic loads, vehicle fleet) were developed within the framework of EIST for the reference year 1998 (Symeonidis et al., 2003) and are described in Table 1. An effort to update the traffic counts and vehicle fleet has been made. In order to do so, fleet growth factors and a retirement vehicle pattern (per vehicle type and engine technology) have been applied based on fleet data projections (Kyriakis et al., 1998). The traffic counts were updated using the fleet growth factors. In the urban centre of Thessaloniki and Athens, the traffic load projection was applied only on the outer part the city's road network, assuming that the centre has already reached saturation levels (Tsilingiridis et al., 2002). Mean vehicle speeds and diurnal temporal variation of traffic loads were considered unchanged.

The top–down approach results in annual emission estimations and the bottom–up in the emissions on specific road segments. The following methodology was implemented to map the area emissions. First, the area emissions in each driving mode were spatially allocated in Greece on the 10 km resolution grid. The line sources emissions (which are also categorised as urban, rural or highway roads) were also allocated in the same 10 km grid and subsequently they were extracted from the total area emissions for each driving mode in each cell. The residual was distributed to the remainder of the road network which is provided in a digital database according to the total length per grid cell and road type.

3.4. Other mobile sources and machinery

Calculations were performed for the following sub–categories using the CORINAIR simple or detailed methodologies, depending on the availability of detailed data: Agricultural and industrial off–road machinery, railway transport, passenger ships/ferries, fishing boats, recreation vessels and aviation. The input data employed for the calculations as well as the spatial disaggregation methods are presented in Table 1.

All calculated emissions for this category were based on activity information which was assembled in the framework of the EIST project (Table 1). The quantification of the railway emissions was based in engine output power of 16 different types of railcars and locomotives (in order to derive the emission factors for each type) and specific route information in a database which contains about 435 routes of the national railway network (in order to derive the travelled length) (Symeonidis et al., 2004). For the aviation, this study included only landing and takeoff (LTO) emissions. The calculations were based on fuel consumption information for specific aircraft types. The emissions of fishing activities were calculated based on the gross tonnage of the registered fleet in each prefecture of Greece which is used to derive the total fuel consumption. The majority of the fleet consists of small boats equipped with diesel engines (burning diesel of 0.05% sulphur content). The operating time was assumed to be 20 days per month (Symeonidis et al., 2004). The detailed methodology was implemented to quantify the emissions of passenger ships/ferries. The calculations were based on the travelled distance (provided by a GIS database) and cruising speeds (in knots) for 236 individual ships, operating in 103 lines in Greece. The aforementioned were combined with the emission factors derived from the total engine power (main and auxiliary engines) for each of the vessels considered (Symeonidis et al., 2004). For the passenger ships/ferries emissions for the hotelling and the manoeuvring phases were also quantified (assuming an operating time of 30 minutes for each of the 2 modes) (Symeonidis et al., 2004). For the quantification of recreation activity emissions, the main engine output power of the registered fleet in each prefecture of Greece were used to calculate the emission factors which were subsequently multiplied with the average cruising time (calculated from average movement speeds and travelling distances). For the larger vessels (sailing boats/workboats equipped with diesel or gasoline engines) an average trip length of 150 nautical miles and a speed of 20 knots were used and for the smaller boats (personal watercraft equipped with 2–stroke gasoline engines) an average trip length of 30 nautical miles and a speed of 30 knots were used (Symeonidis et al., 2004). Finally, international shipping emissions of the EMEP database were used and re-projected to the 10 km gridded domain.

3.5. Lignite extraction

The majority of the electricity production in Greece is based on lignite consumption. In the prefecture of Kozani at the north part of the country the largest lignite mines of Greece and one of the largest in Europe are located. Three major mines extract about 53 million tonnes of lignite each year. The other large mining site is located in the Peloponnese at the southern part of Greece. The amount of lignite extracted in these mines was combined with the emission factors of the TNO/CEPMEIP database (as kg of particles per tonne of lignite extracted) in order quantify the emissions of PM_{10}. The emission factor database is available at http://www.air.sk/tno/cepmeip/ and the production data are provided by the mining company for the year 2005. In addition, emissions were quantified for the fly ash amounts that are produced as a by-product of lignite combustion in the power units (12.5% of each tonne of lignite on average is fly ash) and are subsequently transferred to storage areas in the mines. An emission factor for fly ash was also extracted by the TNO/CEPMEIP database. Finally, the boundaries of the mines (mining and storage areas) are provided by the mining company as a GIS digital database.

4. Results and Discussion

4.1. Spatial distribution of emissions

Total emissions for Greece are estimated to be 182 219 metric tons for the base year 2003. Figure 2a depicts the annual emissions by source sector. The industrial sector represents a very high frac-

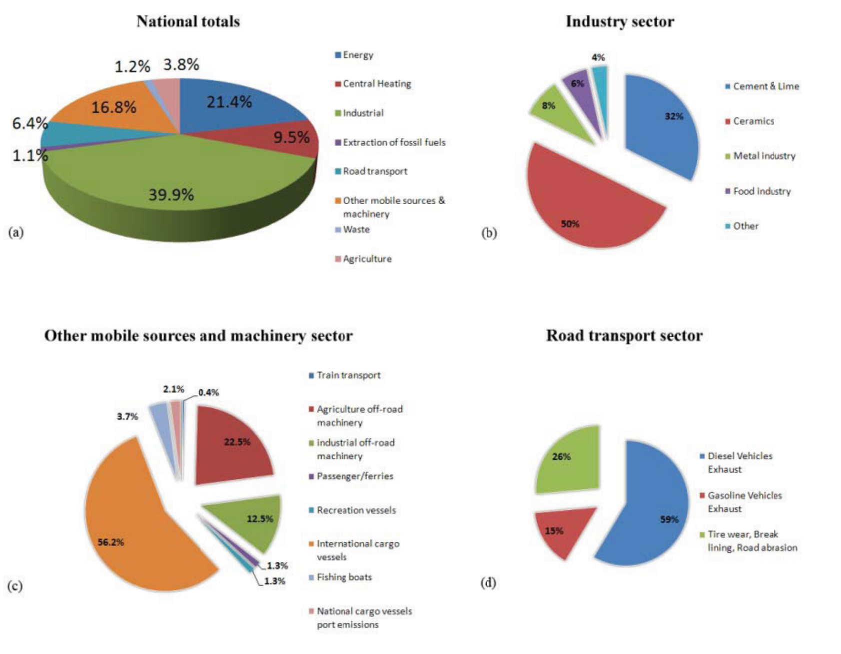


Figure 2. *Annual sectoral and sub-sectoral contribution to **(a)** national total **(b)** industrial **(c)** other mobile sources and machinery **(d)** road transport emissions.*

tion of the total emissions with the majority originating from ceramic products industries (36.3 kt yr^{-1}) and cement, lime production (23.5 kt yr^{-1}) (Figure 2b). Power plants are the second largest contributor to the total annual national emissions and their emissions display peak values in the two lignite–fired power plant complexes of Kozani (28.5 kt yr^{-1}) and Megalopoli (6.5 kt yr^{-1}). Practically all of the energy sector's emissions originate from the lignite–fired power plants due to the low heating value of the Greek lignite. Although 70% of the total electricity is produced by the lignite–fired power plants, they are responsible for the 96% of the annual power plant emissions. The industrial sector encumbers with emissions certain areas like the Aspropyrgos industrial complex outside the Athens basin (11 kt yr^{-1}), the industrial area of Volos (10.4 kt yr^{-1}) and Oinofyta (13.9 kt yr^{-1}).

The other mobile sources and machinery sector is also a considerable contributor, having almost 17% of the yearly emissions with the maritime sub–sector being the major emission source (Figure 2c). The national sea borders of Greece include major routes of international bunker vessels and their emissions peak at the southwest maritime area of Crete and at the Piraeus port (Figure 3a). Passenger ship emissions are higher over the Ionian Sea routes to Italy and the sea routes connecting Piraeus port to Crete and Cyclades. Important emission shares are also attributed to the diesel powered agriculture and industrial machinery (Figure 3b). Emission values are higher over the two large urban centres (dominated by construction machinery) as well as over major agricultural areas situated at the Thessaly plain (226 t yr^{-1}) and at the northern part of the country (Figure 3b).

Approximately 9.5% of the annual total PM_{10} emissions stem from heat production. Depending on the geographical location, the heating season may extend from October to April and emissions are higher during December, January and February. At the northern part of the country, where temperature values are low during a large period of the heating season, emissions during October and November can also be important. Emissions are higher over the urban agglomerations of Athens (32.5 kt yr^{-1}) and Thessaloniki (10.8 kt yr^{-1}), as well as over other major Greek cities like Patra (263 t yr^{-1}).

Traffic emissions are responsible for approximately 6.4% of the yearly nationwide PM_{10} totals, with diesel vehicles emissions dominating (Figure 2d). The main contributors of the diesel vehicles are the heavy duty vehicles (nearly 67%) and taxis (nearly 18%) due to the high emission factors of the former and the high mileage driven for the latter. Gasoline vehicles produce 15% of road traffic emissions, with two-wheeled vehicles representing 80% of that amount. The remaining gasoline vehicle emissions come from passenger cars. The latter is a result of the high share of conventional two–wheeled vehicles compared to the passenger vehicles equipped with EURO engines. Non–exhaust emissions account for 26% of the category with the majority to originate from tire wear (12.5 kt yr^{-1}) and road abrasion (13.1 kt yr^{-1}). The spatial distribution of exhaust emissions for the national network of Greece is depicted in Figure 4 which reveals that most polluting ones are the highways around the large urban agglomerations of Athens and Thessaloniki mainly due to high traffic loads of heavy duty vehicles. The spatialisation of the secondary (rural and urban) network exhaust emissions highlights the two largest urban centres and their surrounding areas. In Thessaloniki, the major road arteries are emphasized with high emission values mainly due to taxis and gasoline passenger cars. At the northwestern part of the city, which is closer to the industrial zone and where the main road entrances are located, heavy duty vehicles are the major producer of exhaust PM_{10} along with the tire wear, break lining and road abrasion emissions.

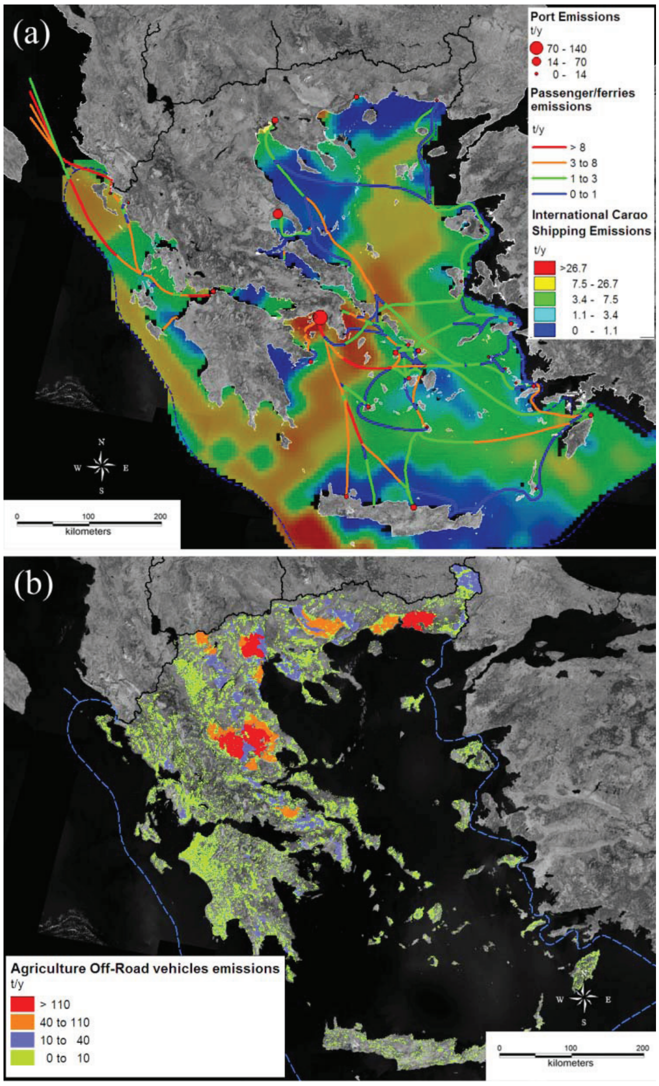


Figure 3. *Spatial distribution of annual emissions from* ***(a)*** *the power plants and the industrial sector,* ***(b)*** *the maritime sector allocated in the 100 m resolution CORINE landuses in Greece.*

Finally the spatialisation of the total emissions illustrated in Figure 5 indicates that the large urban agglomerations as well as major industrial zones (Oinofyta, Volos, Aspropyrgos, Kozani, and Megalopoli) are characterised by peak emissions. Medium sized urban areas also present high emission values with the sectoral contribution to reflect the influence of different anthropogenic activities. The emissions presented in Table 2 are the aggregated values of 9 grid cells of 10 km resolution defining a squared region centred in each small and medium sized city. Due to the significantly larger sizes of Athens and Thessaloniki urban centres, these emissions were derived from an area of 25 cells of 10 km size in order to take into consideration the greater areas of the cities. In Ioannina, heat production is the major PM_{10} producer given the low temperatures during the winter season. Larisa, a city in the heart of the agricultural area of the Thessaly plain, presents a strong agricultural emissions influence. In the city of Patra, industry and heat production are the main PM_{10} source sectors while in Volos and Heraclion, the industrial sector holds the majority. Athens and Thessaloniki show similar shares among the emission sources of which the industrial sector appears to hold the majority of emissions. It should be stated though that the industrial emissions in Thessaloniki are much closer to the city centre compared to Athens, in which the major industries are located several kilometres outside the city. It should be also stressed that the contribution of the maritime sector emissions presented in

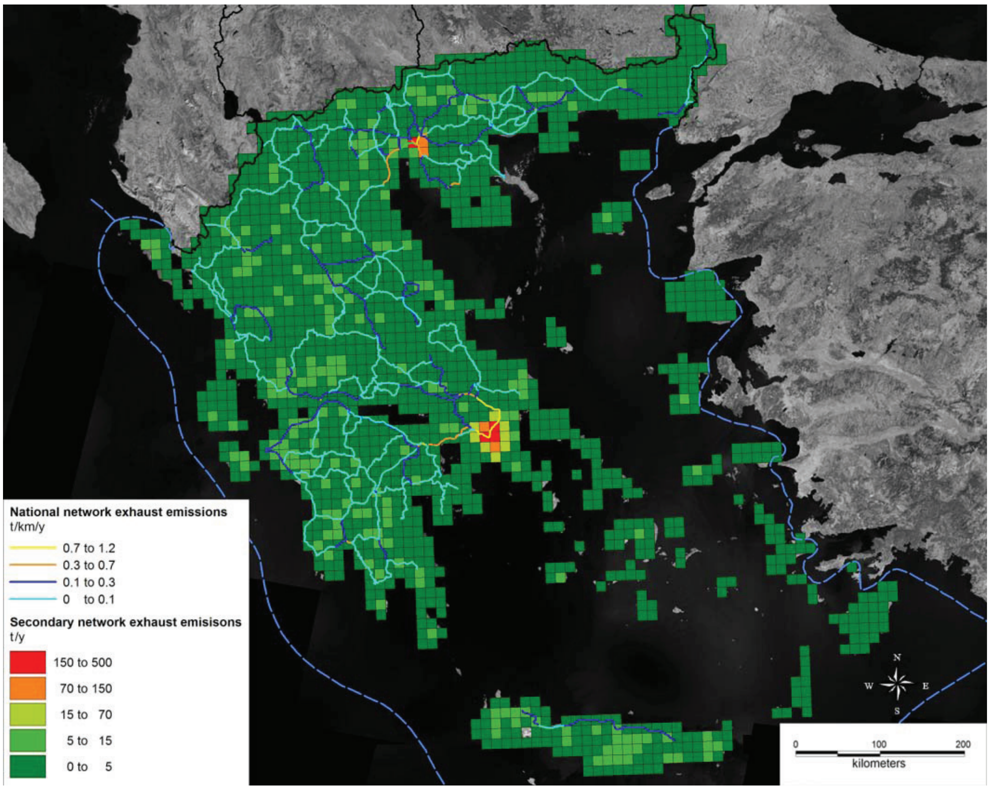


Figure 4. *Annual road transport exhaust emissions in the national and secondary road network of Greece.*

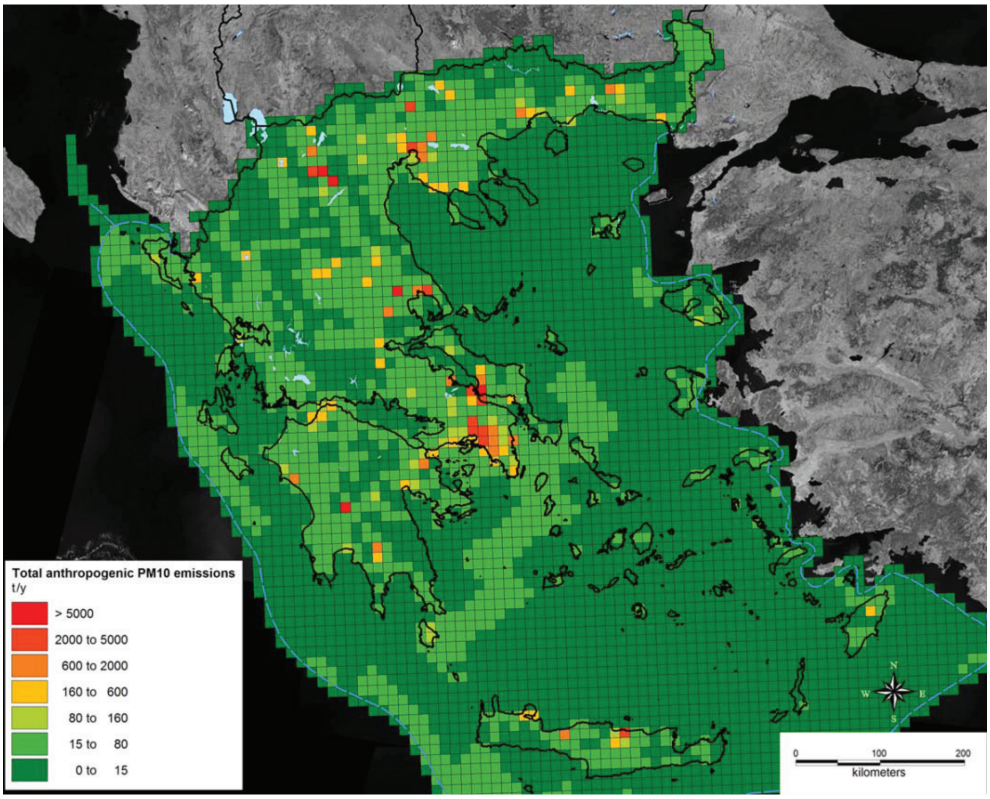


Figure 5. *Spatial distribution of the total PM_{10} anthropogenic emissions in Greece.*

Table 2 is probably underestimated in the port cities. The disaggregation method for international shipping scatters the 50 km EMEP cell emissions proportionally to the corresponding 10 km grid cells. A large share of these emissions is probably attributed to the port cells rather than the whole EMEP cell area.

***Table 2.** Sectoral contribution (%) to annual emission totals[a] for the large and medium sized Greek cities.*

City	Power plants	Industry	Central Heating	Road transport[a]	Maritime	Agriculture[b]	Industrial machinery	Waste	Total (in t y^{-1})
Ioannina	-	0.8	43.3	21.1	-	14.6	14.6	5.6	398
Larisa	-	10.3	33.2	10.5	-	32.9	8.5	4.6	742
Patra	-	55.3	20.3	5.6	6.7	3.6	5.3	3.2	1 296
Volos	-	90.8	3.9	1.1	1.4	1.3	0.9	0.6	5 509
Heraclion	5.8	75.1	8.9	2.5	1.5	3.1	1.8	1.3	3 052
Thessaloniki	-	58.4	17.3	12.4	1.5	3.9	3.7	2.8	7 188
Athens	-	57.2	18.0	13.0	1.9	0.8	5.5	3.6	20 596

[a] *Road transport includes exhaust emissions and non-exhaust emissions from tire wear, break lining and road abrasion.*
[b] *Agriculture includes off-road machinery exhaust emissions as well as other agricultural activities emissions that fall under the SNAP category.*

The majority of PM_{10} emissions (52%) are chemically characterized as other particles having inorganic origin. These particles are not combustion related but originate from the processes in the ceramics, lime and cement industry. Organic carbon is the second larger contributor to the national totals (22%), primarily emitted from the incomplete combustion processes in diesel oil–fired boilers for heat production (48%). The maritime sector, mainly the international cargo shipping is responsible for the majority of sulphate particles (37%) due to the high sulphur content of the fuels used. Elemental carbon major contributor is the off-road agricultural and industrial machinery (40%) as well as the industrial combustion (32%).

4.2. Temporal variation of emissions

Depicted in Figure 6 the power plant emissions display a small variation over the year with the maximum values during the summer months (July and August) when electricity demand is higher due to air-conditioning operations. Heat production emissions peak in January and February while industry and other mobile sources and machinery sector show summer maximum values in June for the former and July–August for the latter. In January and February, heat production emissions are comparable to industrial sector emissions highlighting the importance of the former during the heating period. The annual variation of total emissions is characterized by monthly emissions that are comparable for the months between January and March. Subsequently, emissions are decreased taking their minimum value in May. The increase of emissions during the summer period is attributed to the power plants and the other mobile sources and machinery sectors. There is a rising trend from September to December when emissions are maximum, mainly configured by the increase of the industrial and heat production emissions.

Inside Athens, the yearly emission variation of emissions presented in Figure 7a follows the operation of heat production with peaks in January (the coldest month of the year) and in November. In Thessaloniki (Figure 7a), the peaks in January and in March are a result of the heat production and the industrial sector respectively while the variation of emissions through the rest of the year is similar to Athens.

The weekly variation at the large urban centres (not shown) indicates a minor emission fluctuation during the weekdays, while emission values drop on weekends by 15% to 20% depending on the location and period. The diurnal variation of emissions depicted in Figure 7b for a typical weekday of January shows that in Athens and Thessaloniki, peak values are displayed in the morning hours (7:00 LT to 9:00 LT) due to the early morning enhanced anthropogenic activities (primarily road transport and heat production). The second peak in Athens is related to the return of people from their work and the late evening peak is attributed to the heat production operation. We have to state that heat production operates in specific hours and not in continue basis (HMEPPPW, 2001).

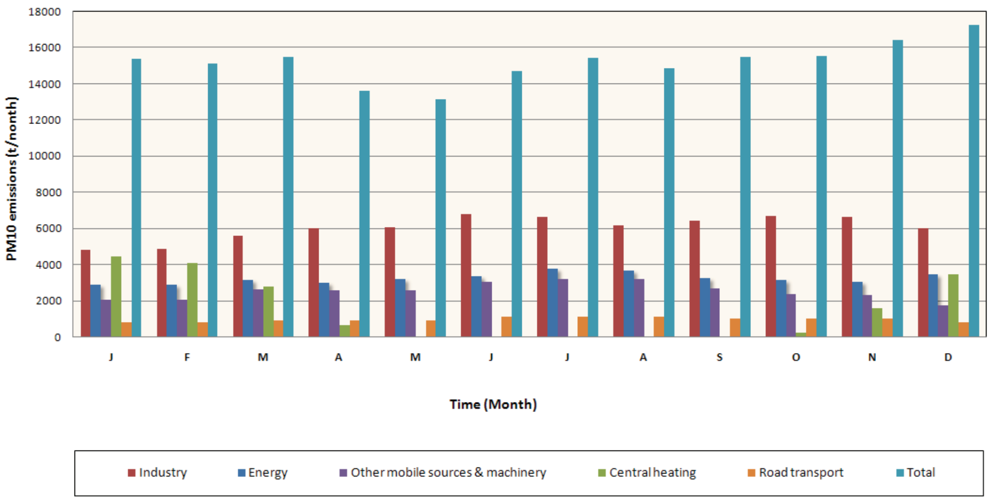


***Figure 6**. Annual total and sectoral emission variation.*

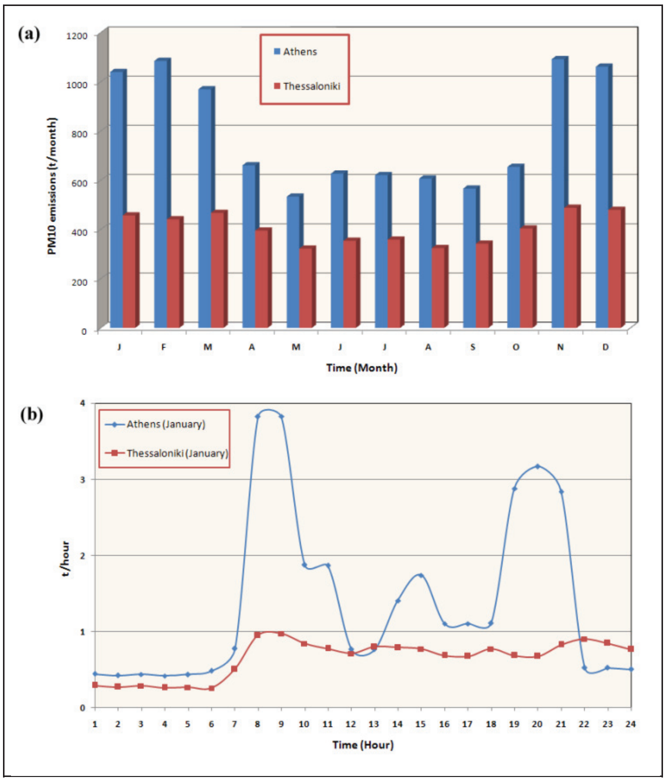


Figure 7. (a) *Annual and* ***(b)*** *diurnal variation of emissions at the urban centres of Athens and Thessaloniki.*

5. Evaluation and Data Limitations

In the current study, the best available input datasets were used for the calculation of the road transport and the other mobile sources and machinery sector emissions as well as for the major emitting sources like the power generation sector and the industrial sector. Nevertheless, limitations are present concerning the calculated estimates and the pre-existing emission data used. Regarding the industrial sector, the emission factors used to compile the emissions of the major contributing industrial types like ceramics, cement and lime industry present large uncertainties (HMEPPPW, 2001). More specifically for the production of ceramics and lime industry, the emission factors present an uncertainty of more than 100% while for cement production the emission factors for the four out of five individual processes are within 30% to 100% range, and less than 30% for the remaining three processes. Even so, the compiled emission factors are based on field studies and the uncertainties are related to the small number of conducted measurements. More measurements are needed to reduce the presented uncertainties. Also increased uncertainties are expected due to the spatial distribution of off–road construction machinery emissions having been calculated on prefectural level before being allocated according to the spatially resolved surrogate variables. This is due to the fact that registered machinery in a given prefecture does not necessarily operate in that prefecture. Furthermore the international shipping emissions are disaggregated to the 10 km resolution domain from a coarser grid and underestimation of emissions in the port grid cells is also introduced.

The annual sectoral estimations of this study were compared to the EMEP emission data. The results presented in Table 3 show that the emissions of this study are greater by a factor ranging from 1.6 (coal extraction) to 3.1 (industrial sector) compared to the emissions in the EMEP database. However, a comprehensive analysis of the EMEP emissions database (EMEP, 2006) indicates that there are considerable uncertainties regarding PM_{10} industrial emissions as well as underestimations of road transport and other mobile sources and machinery emissions for several European countries. But, most importantly it should be stated that the particulate matter emissions included in the EMEP database are a product of the expert panel of EMEP since Greece does not officially report particulate matter emissions to the organization. Consequently, the emissions of the EMEP database regarding PM for Greece are highly questionable and uncertain and the observed large discrepancies can be result of these uncertainties. The reason for which the emissions of the EMEP database were used to cover a number of source sectors was a necessity since no activity data were available to quantify these sources. It also has to be stated that the procedures of the expert panel which are adopted for the emission estimation are not known and thus further discussion regarding those differences is not possible. Finally, the road transport estimations of the present study regarding diesel vehicles emissions (6 720 t yr^{-1}) are in good agreement with a third party database (TRENDS database) that reported a value of 6 731 t yr^{-1} (Samaras et al., 2002).

Table 3. National sectoral emissions of this study and of the EMEP database (t yr^{-1}) for 2003[a].

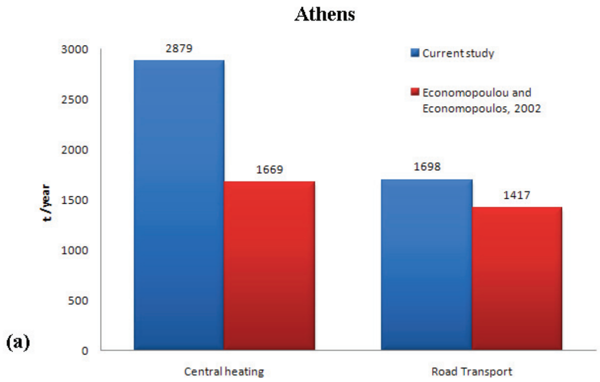
Source category	This study	EMEP
Power plants	38 907	17 100
Industry[b]	72 681	23 285
Road transport	6 720 (Diesel), 1 720 (Gasoline), 3 031 (tire, break, road abrasion)	6 378
Other mobile sources & machinery[c]	13 089	7 098
Coal extraction	3 100	1 998

[a] The activity data used for the quantification of coal extraction emissions are representative for 2005. The EMEP database emissions used for the comparison are also extracted for 2005.

[b] Includes combustion in the manufacturing industry and industrial processes emissions.

[c] Except international cargo shipping.

For the urban centres of Athens and Thessaloniki, a comparison with the only available studies regarding particulate matter emissions of Economopoulou and Economopoulos (2002) and Tsilingiridis et al. (2002) (Figures 8a and 8b) has been performed, respectively. Sectoral emissions of the present study are of the same order with the results of Economopoulou and Economopoulos (2002) and Tsilingiridis et al. (2002). It should be stressed though that the comparison is not straightforward taking into account the differences in the reference years and the particle sizes between the studies. In the study of Tsilingiridis et al. (2002) (emissions as TSP for the year 1995), it is not clear whether the industrial emissions, which is by far the most important emission source in the area, included the process and fuel combustion related emissions or fuel combustion emissions only. If process emissions are included then a considerable share of these would be above the size of 10 µm. The latter can potentially explain the observed difference in the estimated amounts of the studies. Heat production emissions of the present study for Athens (which stem from the EMEP database for the year 2003) are greater by 33% compared to Economopoulou and Economopoulos (2002) (which are based on the population census of 1991 and compiled for the year 1998). This can be attributed to the large increase of the population in the city, the emissions of the EMEP database which are used though are quite uncertain thus further explanation is not possible.



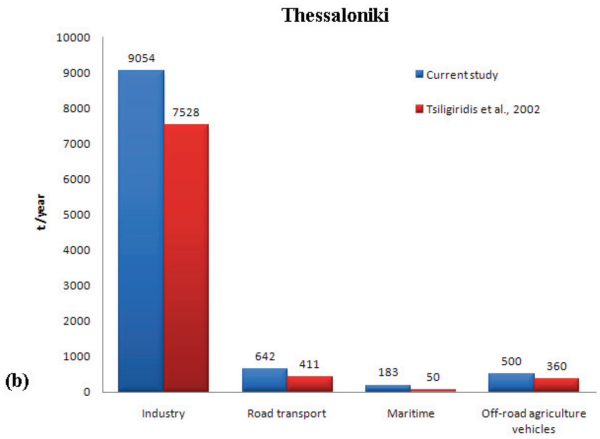


Figure 8. *Comparison of the present study's emission totals with previous studies for **(a)** Athens and **(b)** Thessaloniki.*

6. Conclusions

In this paper a structured methodology for the compilation of a spatially and temporarily resolved and chemically speciated anthropogenic PM_{10} emission inventory for Greece is presented. The inventory has high spatial (10 km) and temporal (hourly basis) resolution. Quantification of emissions was performed for the road transport, other mobile sources and machinery and lignite mining operations sectors following top–down and bottom–up methodologies. Existing emission data from national and European emission databases were also used. An emission processing kernel was developed in order to produce the spatial, temporal and chemical disaggregation of emissions based on source relevant spatio-temporal indicators.

In national scale, the main PM_{10} emission sources are the industrial, power generation sector and other mobile sources and machinery sectors. The majority of industrial emissions originates from ceramic and cement/lime production industries. Apart from the large urban agglomeration the major industrial areas of the country like Aspropyrgos, Volos, Oinofyta, Ptolemaida, and Megalopoli are emphasized with high emissions. The maritime areas of Greece allow intense shipping activities to take place that emit significant PM_{10} amounts especially from international shipping. The spatial distribution of PM_{10} totals shows that emissions peak over the major industrial zones of Greece and over the large urban agglomerations. Winter months contribute to the majority of annual emissions because of the increased anthropogenic activities.

In urban areas, the main emission sources are represented by different anthropogenic activities like industry, heat production, road transport and agriculture. In the greater areas of the large urban agglomerations of Athens and Thessaloniki, the industrial sector is the most important source of particles although the industrial areas are located outside the main urban structure. Inside the cities' main residential areas the road transport and heat production operation is the largest PM_{10} contributors. Especially heat production can be the major PM_{10} emission source in the urban centres on specific months during winter. In urban scale, road transport appears to be more significant than on a national level, especially if non-exhaust sources are accounted for (tire-wear, break-lining, road abrasion) due the accumulation of large number for vehicles in the cities which are also the reason of severe congestion phenomena.

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