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Inventory of Commercial Cooking Activities and Emissions in a Typical Urban Area in Greece

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Abstract: The pollutants emitted during meal preparation in restaurants deteriorate the air quality. Thus, it is an environmental issue that needs to be addressed, especially in areas where these activities are densely located. The purpose of this study is to examine the impact on air quality from commercial cooking activities by performing a qualitative and quantitative analysis of the related parameters. The area of interest is located in the southeastern Mediterranean (Greater Athens area in Greece). Due to the lack of the necessary activity information, a survey was conducted. Emissions from the fuel burnt during the cooking procedures were calculated and it was found that, overall, 940.1 tonnes are attributed to commercial cooking activities annually (generated by classical pollutants, heavy metals, particulates and polycyclic aromatic hydrocarbon emissions). Comparing the contribution of different sources to the pollutants emitted, it was found that commercial cooking is responsible for about 0.6%, 0.8% and 1.0% of the total CO, NOx and PM10 values. Cooking organic aerosol (COA) and volatile organic compound (VOC) emissions from grilled meat were also calculated, accounting for 724.9 tonnes and 37.1 tonnes, respectively. Monthly, daily and hourly profiles of the cooking activities were developed and emissions were spatially disaggregated, indicating the city center as the area with higher values. Numerical simulations were performed with the WRF/CAMx modeling system and the results revealed a contribution of about 6% to the total PM10 concentrations in the urban center, where the majority of restaurants are located.

Keywords: commercial cooking; emissions; Greece; particulate matter; temporal factors; CAMx

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1. Introduction

Commercial cooking refers to all of the activities related to professional food preparation for other people that will compensate for the received service. In Greece, cooking in restaurants, cafeterias, canteens and taverns are included in this category. The above-mentioned services are either located in specific areas where comprehensive entertainment services are provided, such as nightclubs, theaters, cinemas, in touristic areas or established in each neighborhood, mostly as local cuisine restaurants. There is also a small part made up of mobile cooking services, located mostly on the main urban roads of the city center.

The pollutants emitted during meal preparation deteriorate the indoor air quality and pose potential risks to human health, since they are usually emitted relatively close to breathing distances [1–3]. Moreover, the cooking fumes that are created affects the air quality in the surrounding area [4], especially in areas where these activities are densely located [5–8]. However, air pollution control policies and scientific studies have focused on the control of the transport, industrial and household sector emissions, while much less attention has been paid to commercial cooking emissions. Thus, it is an environmental issue that needs to be addressed in urban areas.

The chemical profile of the pollutants emitted from cooking is highly dependent on the cooking style, defined by the appliance and fuel used for the food preparation [9–12]. The fuel and energy types used are mainly coal, liquid petroleum gas (LPG), electricity and natural gas. With the exception of electricity, the rest of the fuels are important sources of CO, particulates and VOC emissions [13–16].

The energy required for food preparation (both in restaurants and households) is a significant factor that needs to be studied in depth, in order to define the parameters (e.g., fuel, type of appliance) that could possibly reduce cooking appliance consumption [17] Thus, many researchers have focused on testing different cooking appliances in terms of cooking time, as well as the energy efficiency of the process, which stands for the ratio of the actual energy consumption required to cook the sample to the actual energy consumption of the appliance [18–21].

Apart from the fuel burnt, measurements have been carried out in order to provide a better understanding of the chemical composition, as well as the size of particles generated due to the cooking of meat [22–24]. Particulate emissions measured from meat charbroiling were found to consist mainly of organics (>99%) and much less of inorganics (0.5%) and BC (0.3%) [25]. The chemical characteristics of the fine particulate matter (PM2.5) emitted at three restaurants in Shanghai indicated that organic matter (OM) was the predominant contributor to cooking-related PM2.5 mass, with a proportion of 69.1–77.1% [26].

Polycyclic aromatic hydrocarbons (PAHs) should also be highlighted as an important group of pollutants formed mainly by the incomplete combustion of fuels. as well as by organic compounds such as grilled meat [27–30]. The authors in [31] found that PAH emissions from cooking activities, including both restaurants and home kitchens, were slightly lower than those from traffic sources in a representative city of Taiwan. The carbonyl composition of the cooking fumes was examined by [6] and it was found that formaldehyde and acrolein were the predominant species.

The literature review carried out by [32] for cooking activities (both commercial and residential) worldwide revealed that researchers, mainly in Asian countries and the United States, have focused on this source. Very few efforts have been made worldwide to compile an emissions inventory of commercial cooking activities, either on a national scale or at a regional level. The only two research studies available are [33,34], in which the research was based on surveys and official data. However, in European emissions inventories, cooking organic aerosol (COA), as well as other pollutants, is not usually included. Studies in the UK [35], Spain [36] and France [37] were performed via positive matrix factorization (PMF) analyses of aerosol mass spectrometer (AMS) measurements.

Concerning Greece, no official or other emissions inventory regarding pollutants emitted from commercial cooking activities exists, to the best of our knowledge. Emissions from the tertiary sector are usually included only for water and space heating. Information is available on the profile of emissions from restaurants from measurements only, and for specific species. Separate measurement campaigns at various periods, with different instrumentations and techniques, were conducted in the past, providing evidence that it is an important source that needs to be further studied and addressed in the local emissions inventories. More specifically, measurements of PM1 (HR-ToF-AMS), PM2.5 (24 h sampling with Teflon filters), BC (with a multi-angle absorption photometer in Patras and an aethalometer in Athens), as well as other pollutants, conducted by [38] in two Greek cities (Athens and Patras) from 8 to 26 July 2012, revealed that the contribution of cooking meat to the organic aerosol (OA) ranges from 15% (in Patras) to 17% (in Athens). During the wintertime, campaigns were conducted in 2012 and 2013, at the above cities, with the use of a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS). The PMF source apportionment algorithm was complementarily applied to the corresponding datasets for the estimation of the contributions of the different OA sources. Results revealed that organic aerosols attributed to cooking activities reached a 75% contribution to the total values during the noon hours [39]. In accordance with the above studies, ref. [40] found that the contribution of COA to the total organic fraction, during three campaigns in Athens (18 December 2013–21 February 2014, 23 December 2015–17 February 2016 and 26 July 2016–31 July 2017), by using an aerosol chemical speciation monitor, was about 10%, while the respective percentage was also 10% in summer. COA emissions were estimated by [41] for a middle sized city in Greece (Patras) at about 0.6 g/day per person, which corresponds to 54.8 tonnes annually. It should also be mentioned that, to the best knowledge of the authors, it is the first time that coefficients representing the operation of commercial kitchens in Attica are produced. Previous diurnal profiles [40] were based on measurements, so the peak values were recorded later than the actual beginning of the kitchens' operation. However, emissions from cooking activities consist not only of COA and particles in general (already measured and identified), but also of pollutants such as CO, NOx, NMVOCs and PAHs, which have been proven to significantly affect the air quality in Athens and have already been identified by multiple researchers as leading to O_3 and particulate pollution episodes [42–45]. Consequently, it is of great importance to include them in an emissions inventory.

Moreover previous studies have not analyzed and quantified the relationship between emissions and parameters such as the fuel, cooking appliance, cooking method and spatial distribution of emissions in a highly populated area, and have not clarified the contribution of this source to the local air quality. The authors in [32], while performing a literature review on cooking activities, realized that "an optimal cooking process, with emissions and related impacts, has not been developed widely". As mentioned above studies have been conducted based mainly on measurements either for a limited time period or for selected pollutants. Commercial cooking emissions are not usually included in the national emissions inventory, thus an important source for some areas is missing.

Within that frame, the purpose of this work is to study the impact on air quality from commercial cooking activities by performing a qualitative and quantitative analysis of the related parameters. The following approach included the below steps:

(i) Collection of data. The necessary activity information was gathered by the conduction of a survey of the restaurants located in the region of interest. The main purpose was to (a) record the activity profiles in terms of the kitchen's operation, (b) define the cooking equipment, as well as the fuel used by each appliance and (c) find the most preferable raw materials and the respective consumption.

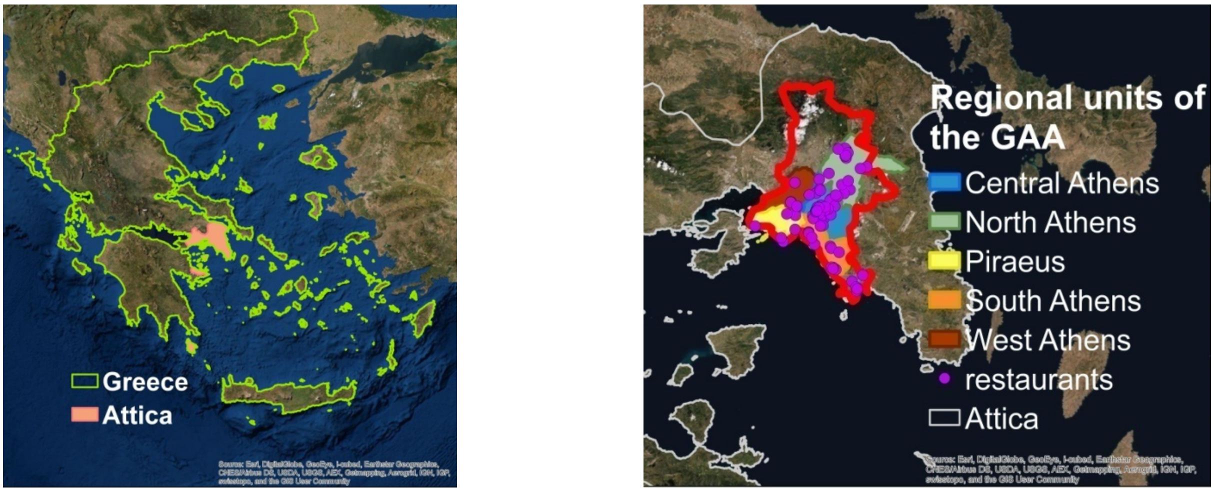
(ii) Calculation of emissions. The energy consumption during cooking was calculated based on the survey findings and official data. Hence, the respective annual emissions produced by the fuel combustion, as well as the meat cooked, were calculated. Finally, temporal coefficients indicative of the local activities were produced for the disaggregation of the annual emissions to monthly, daily and hourly values. The FEI-GREGAA emissions inventory was updated accordingly [46].

(iii) Dispersion of pollutants. On–off numerical simulations were performed to assess the impact on local air quality, and model results were compared with data from existing measurements.

2. Materials and Methods

2.1. The Commercial Cooking Activity in the Greater Athens Area

The Greater Athens area (GAA) is a densely populated area (approx. 3.2 million people live in 415 km^2), subdivided into five regional units (Figure 1). According to the Hellenic Statistical Authority (ELSTAT), in 2017, there were 7383 restaurants (approximately 24 restaurants per 104 people) and mobile food service activities in the GAA, spatially distributed in each regional unit as shown in Table 1. The majority of them (39.2%) are allocated at the Central Athens unit since this area is the most populated (about 1 million people), and it is the center of entertainment for the locals, as well as the majority of touristic attractions being located there. The North Athens, South Athens and Piraeus units follow, having 17.1%, 16.1% and 15.1% participation in the total number of services. In general, Athens is among the capitals where nightlife is important, since people love to socialize. Commercial cooking activities are an important sector for the local community. As presented in Table 1, the total annual turnover was 1,266,274 thousands of Euros in 2017 while 68,559 employees work in these services, with more than one third of them being in Central Athens.



(**a**) (**b**)

Figure 1. (**a**) Map of Greece and the Attica region and (**b**) the restaurants participating in the survey, spatially allocated in the regional units of Attica.

Table 1. Statistical data for the commercial cooking activities in the GAA for the year 2017 (Source: Hellenic Statistical Authority).

Regional Unit	Restaurants and Mobile Cooking Services (Number of Legal Companies)	Annual Turnover (in Thousands of Euros)	Employees (Number of People)
Central Athens	2892	488,018	26,313
North Athens	1263	296,642	14,089
West Athens	920	101,699	7204
South Athens	1190	257,925	13,087
Piraeus	1118	121,991	7866
Total	***7383***	***1,266,274***	***68,559***

2.2. Emissions Calculation

Emissions from commercial cooking were calculated for the year 2017, for the GAA, following the Tier 2 methodology proposed by the EMEP/EEA (European Monitoring and Evaluation Programme/European Environmental Agency) emission inventory guidebook 2019 [47] for small combustion, which is based on the appliance technology. The calculation of the annual emissions was based on the equation

$$E_i = \sum_{j,k} EF_{i,j,k} \times A_{j,k} \quad (1)$$

where

E_i is the annual emission value of pollutant i,
$EF_{i,j,k}$ is the emission factor for pollutant i, source category j and fuel k,
$A_{j,k}$ is the annual consumption of fuel type k and appliance j.

Calculated pollutants are SO_2, NOX, CO, NMVOC, SOx, particulate matter (PM10 and PM2.5), black carbon (BC), heavy metals (Pb, Cd, Hg, As, Cr, Cu, Ni, Se and Zn), polycyclic aromatic hydrocarbons (PAHs), polychlorinated biphenyls (PCBs), polychlorinated dibenzo-dioxins and furans (PCDD/F), and hexachlorobenzene (HCB). The types of fuels and energy used are natural gas, liquid petroleum gas (LPG), coal, electricity and, less commonly, wood. However, electricity was not included in the calculation process since the pollutants for the production of this type of energy are considered to be emitted close to the public electricity industries. The emission factors (EFs) are presented in Appendix A and they are the ones proposed by the EMEP/EEA emission inventory guidebook for medium-sized gas-fired boilers (Table A1) and manual boilers burning coal fuels (Table A2). The same EFs were used for coal and wood consumption, since the latter one was very limited. It is obvious that much higher values are attributed to the combustion of coal. In particular, the EFs for CO and particulates are 62 and 300 times higher for coal than gas, while for NOx the relevant ratio is only 2.74.

2.3. Energy Consumption Data

2.3.1. Conduction of a Survey

One of the major difficulties encountered in the present study was the lack of data on fuel consumption (e.g., coal, electricity, LPG) during cooking, raw material consumption (e.g., meat, poultry and fish) and equipment type (e.g., grill, pan). From the Odyssee–Mure project (https://www.odyssee-mure.eu/, accessed on 20 October 2020), fuel consumption data for the tertiary sector were collected, which includes hotels, restaurants and cafes, hospitals, education, offices, and trade. Electricity consumption is superior to other fuels with a percentage ranging from 73.3% (in 2006) to 84.4% (in 2014). Oil consumption follows until 2013, while for the rest of the period either gas or biomass has second place, depending on the year. There is a decrease in values for all types of fuel during the period 2010–2014 (the total fuel consumption was 81.71 PJ in 2010 and 71.65 PJ), with the exception of electricity consumption in the year 2012, while values increased onwards (the total fuel consumption was 91.74 PJ). The abovementioned values include several activities such as cooking, water and space heating.

In order to find the amount corresponding to commercial cooking, it was necessary to conduct a survey through a questionnaire in selected facilities. The questionnaire was addressed to the employees and/or owners of selected restaurants and barbecues, spatially covering the regional units of Attica, where most of the population is concentrated. This area is characterized as the Greater Athens area (GAA) and includes the four units of Athens (North, West, South and East) as well as Piraeus. The rest of the regional units of Attica were not included in the present research, since the main purpose was to estimate the pollutants affecting the air quality within the city level. The aim was to collect data in order to estimate the total energy consumption, the type of fuel used, the annual emissions and the temporal distribution of the emissions, on an hourly scale. For this reason, the parameters recorded were:

- The duration of operation of the kitchen (per month, per day and the daily schedule), in order to record the duration of the emitted pollutants and particles,
- The type and amount of meat and fish used on a daily, weekly or monthly basis,
- The type of equipment and fuel used.

Prior to deployment, the questionnaire was addressed to three pilot owners, the main comment of whom was to limit the duration to a maximum of ten minutes. As a result the final questionnaire was developed (see Appendix B) comprised of eight questions (three open and five closed). The survey was conducted via telephone calls (due to COVID-19 restrictions) to almost 500 facilities which met the following criteria: (a) they were located at different regional units; (b) their cuisine was Greek, Italian, European, American or Asian and (c) they were of different size. Fast food chains were not included in the survey. Finally, 90 valid responses were received, spatially allocated as shown in Figure 1. The main drawback encountered during the questionnaire filling procedure was the lack of time from the restaurant staff. Apparently, this occurred due to the health crisis, since restaurants had been working overtime to fulfill the sheer number of customers who emerged after the end of the lockdown period. Moreover, almost all the responders could not provide the amount of raw material (question 8). The brand name and the coordinates of each facility were collected as well.

2.3.2. Calculation of the Energy Consumed

For the calculation of the energy consumption per fuel type, enquiries were conducted with the main propane suppliers (EKO, GAS Express 2020) in the GAA in the first instance, in order to estimate the weekly or the monthly consumption. It was found that the mean weekly consumption of propane by a typical restaurant in this area is equal to 125 kg. Secondly, based on the energy content of propane (ETB 2020) it was estimated that the mean annual LPG consumption per restaurant is about 88,400 KWh, including both cooking and heating demands. However, the mean annual energy consumption for space heating per restaurant in Greece has been estimated by [48] to about 250 KWh/m^2/year. Moreover, the mean area of restaurants in Greece is about 120 m^2 [49], which was also verified through calculations using satellite data (Google Earth 2020) for selected restaurants in the GAA. As a result, the annual energy consumption for space heating per restaurant is about 30,000 KWh. Using the conversion coefficient for propane, it was concluded that the mean annual energy consumption for space heating, when totally covered by propane, is 28,571 KWh. Therefore, the minimum annual amount of LPG consumption for cooking was found to be 59,829 KWh (88,400–28,571). Whereas, the possible maximum amount is 88,400 KWh, considering that propane does not contribute to space heating and, thus, the average value is 74,114 KWh (having a maximum of 88,400 KWh and minimum of 59,829 KWh). Since half of the restaurants use LPG for cooking (45 out of 90 responses), it was assumed that, for the whole GAA, approximately 3692 facilities use LPG. Thus, it can be estimated that the annual LPG consumption for commercial cooking services in the GAA is equal to 274 GWh.

In addition, according to the responses received, it was estimated that the contribution of propane to the energy demands for cooking at restaurants is about 33.2% of the total energy used, by all types of fuels which was found to be equal to 824 GWh. By using this value, the data in Table 2 were derived. The final energy consumption used in the calculation of emissions was 0.68 PJ for natural gas, 0.98PJ for LPG and 0.27 PJ for coal. These values stand for 10.8%, 19.1% and 2.5% of the national energy consumption for the tertiary sector, respectively.

Table 2. Energy consumption from commercial cooking activities in the GAA, as produced by the present study.

	Energy (GWh)
Electricity	285
Coal	76
Natural Gas	189
LPG	274
Total	**824**

2.4. Spatial and Temporal Allocation of Emissions

In order to perform in depth study regarding the impact of emissions on the local air quality, it is necessary to spatially disaggregate the values calculated for the GAA on a finer scale and in gridded form, especially for applications with chemical transport models. Thus the annual emissions were allocated at the five regional units of the GAA, based on the number of restaurants and mobile cooking services presented at Table 1. Afterwards, the emissions of each regional unit were mapped on the GEOSTAT grid map of 1 × 1 km cells (source: https://ec.europa.eu/eurostat/web/gisco/geodata/reference-data/population-distribution-demography/geostat, accessed on 15 June 2021), by using the number of restaurants allocated in each cell (source: OpenStreetMap data retrieved on 1 December 2021 by http://download.geofabrik.de/europe/). The methodology that followed is described by Figure 2: First step (grey arrow): GAA annual total CO emissions to emissions per regional unit, proxy value: number of restaurants per regional unit (Table 1); Second step (light green arrow): regional emissions to gridded emissions, proxy values: GEOSTAT grid and shapefile with the location of the restaurants (brow arrows).For the temporal disaggregation of emissions at monthly, daily and hourly values, temporal coefficients were produced by the monthly, daily and hourly profiles of the restaurants' operation described in Section 3.

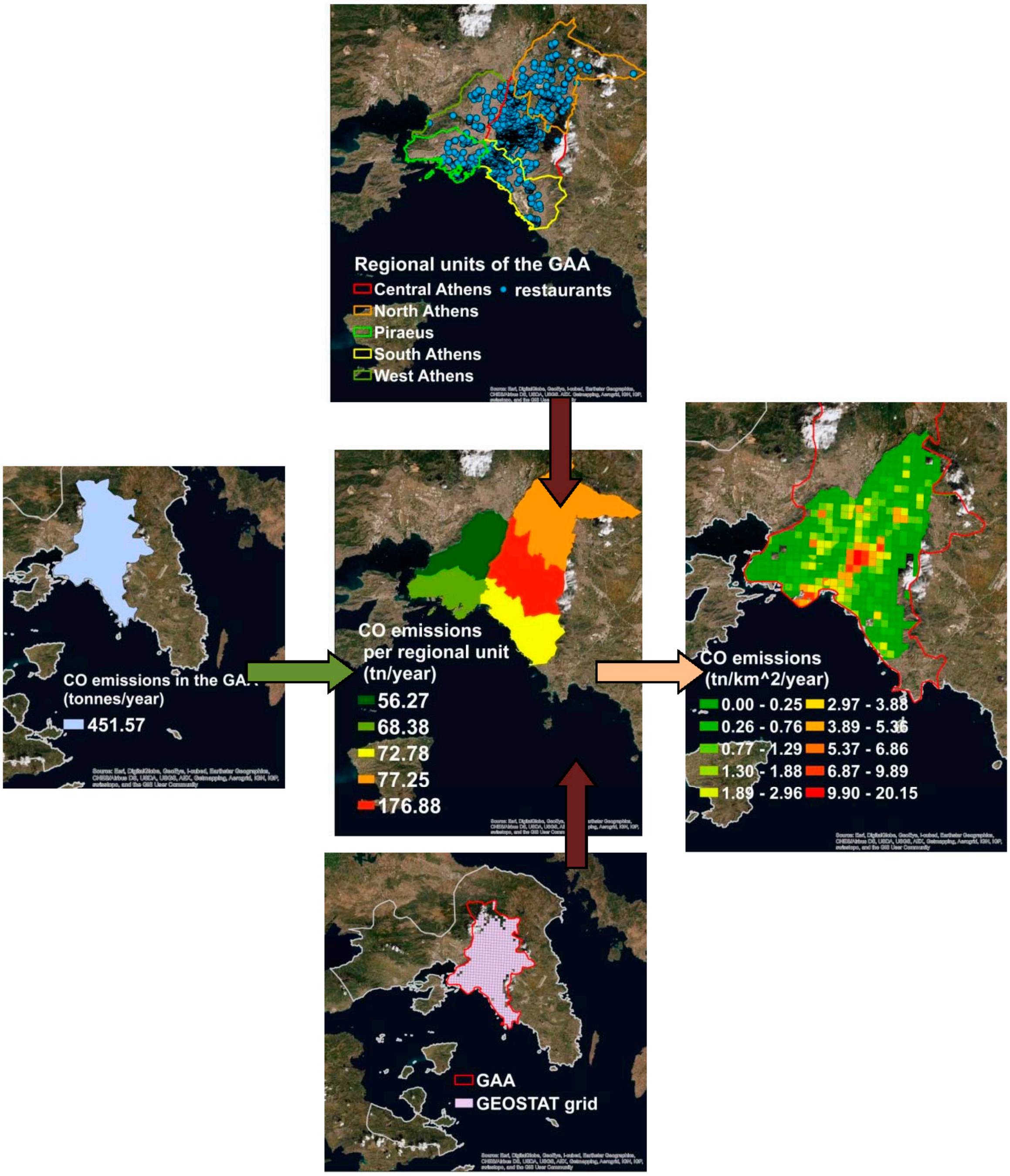


Figure 2. Methodology for the spatial allocation of emissions for the GAA to gridded values (1×1 km^2).

2.5. Numerical Simulations

In order to study the impact from commercial cooking activities on the air quality of the GAA, a source apportionment approach was followed. The simulations were performed with the aid of the comprehensive air quality model with extensions CAMx (www.camx.com, accessed on 10 April 2020). CAMx is a widely used Eulerian photochemical model that simulates the emission, dispersion, chemical reaction and removal of pollutants in the troposphere. For the present study a grid of 2×2 km^2, covering Greece and part of the surrounding countries, was used. The simulation period lasted 4 days (including a two-day spin up period in an attempt to minimize the effects from the initial and boundary conditions on the computed results), from 00.00 UTC on 13 March 2022 to 00.00 UTC on

17 March 2022. The necessary meteorological parameters for CAMx were produced by the model WRF. Concerning emissions, hourly gridded emissions from the FEI-GREGAA database were used for Greece [46] and gridded emissions from the WebDad, which is the emission database of EMEP (co-operative programme for monitoring and evaluation of long range transmission of air pollutants in Europe, https://www.ceip.at/webdab-emission-database/emissions-as-used-in-emep-models, accessed on 10 December 2021), for the rest of the countries. Two different case studies for the emissions profile were used:

First case study: All emission sources were included (**all_sources**)

Second case study: Only commercial cooking emissions were used as input (**cook_only**)

For the evaluation of the model results, a comparison of the hourly gridded concentrations with measurements from the national AQ network was performed, for the "*all_sources*" case study, as well as statistical analysis by using statistical parameters such as the correlation coefficient (r), mean bias (MB), root mean square error (RMSE), normalized mean error (NME), normalized mean bias (NMB), mean observation (MO)value and mean simulation (MS)value. The calculation method for each statistical parameter is presented in Appendix C.

3. Results and Discussion

3.1. Survey Results

Due to the fact that most facilities use multiple types of fuel, raw material and equipment, multiple answers were given to the questionnaire. The responses received, in terms of cooking fuel and energy, are presented in Table 3. It is obvious that a great proportion of facilities have LPG, either as the only fuel (21% of responses) or supplementary to other fuels (29%). Electricity is used by the majority of restaurants, while only 9% of them depend totally on it. Only one owner (of a Greek barbeque) mentioned that they use only coal for all cooking procedures. Results were further analyzed by each type of fuel separately. Therefore, 153, 308 and 211 responses were collected regarding the type of fuel, raw material and equipment, respectively (Figure 3). Concerning cooking fuel, electricity (54 responses) and LPG (45 responses) prevail, while natural gas (33 responses) and coal (21 responses) follow. It should be mentioned that the use of wood is included in the category of coal. The natural gas network is not available in many areas in Attica so the responses received are mainly from restaurants located in Central (16 responses), North (6 responses) and South (8 responses) Athens. Electricity is widely used, mostly as a secondary energy source, since many appliances are compatible with electricity supplies. As show in Table 4, it is equally used with all cooking appliances (87% for pan/wok/fryer and grill, and 81% for oven). The use of coal is a trend advertised by the owners since it provides a unique flavoring to food. Concerning LPG, many commercial kitchens prefer it for cooking since it provides heat faster and provides control over temperature better than other methods, which is important for the preparation of meals, thus, it received half of the responses. Moreover, it is easy to use and provides independence with possible electricity blackouts. LPG bottles can also be used for space heating, so the restaurant owners can probably save money from ordering greater LPG supplies.

Table 3. Responses received to the question about the type of fuel and energy used for the cooking demands of the restaurants.

Fuel/Energy	Responses	Frequency (%)
Only electricity	8	9%
Electricity & coal	6	7%
Only coal	1	1%
Only LPG	19	21%
LPG & Electricity	15	17%
LPG & Coal	2	2%
LPG, Coal & Electricity	6	7%
LPG, Coal, Natural Gas & Electricity	3	3%
Only Natural Gas	11	12%
Natural Gas & Electricity	16	18%
Natural Gas, Coal & Electricity	2	2%
Natural Gas & Coal	1	1%
Total	**90**	**100%**

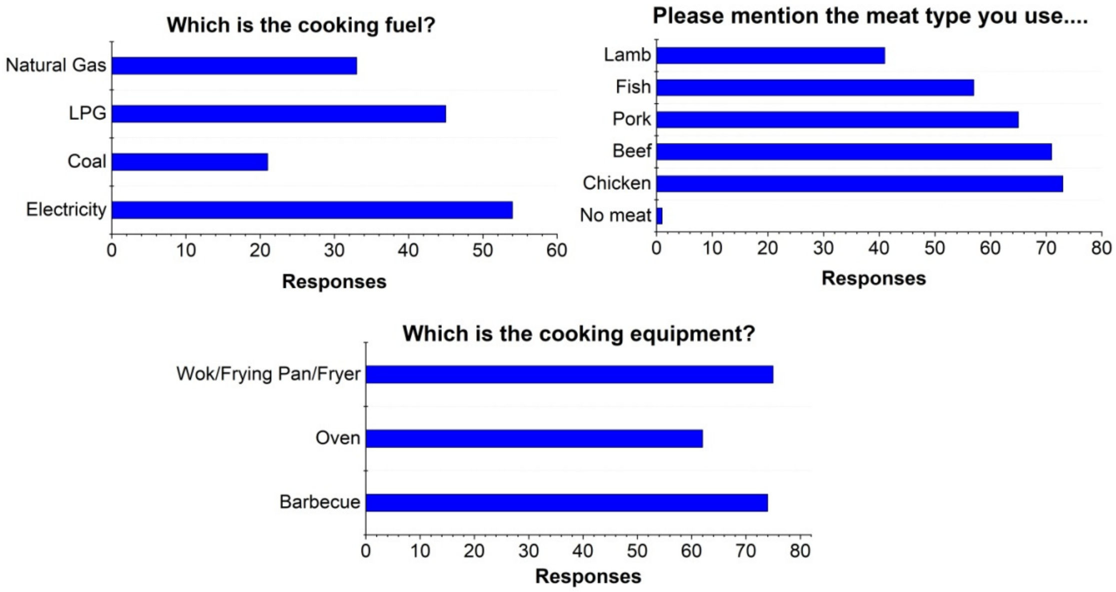


Figure 3. Survey responses concerning the type of fuel, meat and equipment used.

Table 4. Equipment type using natural gas, coal, LPG and electricity.

Equipment	Responses	Frequency (%)	Responses	Frequency (%)
	Natural gas		*Coal*	
Grill	38	84%	18	86%
Oven	31	69%	13	62%
Pan/Wok/Fryer	35	78%	2	10%
	LPG		*Electricity*	
Grill	26	79%	47	87%
Oven	26	79%	44	81%
Pan/Wok/Fryer	30	91%	47	87%

In terms of the equipment, the following three types are almost equally used by each facility (without counting the frequency of their usage): wok/frying pan/fryer (75 responses), grill (74 responses) and oven (62 responses) (83%, 82% and 69%, respectively) (Figure 3, bottom).

The most popular types of meat were found to be chicken and beef, and, secondarily, pork and fish, representing the 81%, 79%, 72% and 63% of the total number of restaurants. It is true that there are many dishes in the Greek cuisine made with chicken and beef. Moreover, dishes that include chicken are usually cheaper so more preferable by customers.

Table 4 presents the responses, as well as the frequency (for the total number of restaurants that use this type of energy), regarding the type of equipment by fuel category. As mentioned before, electricity is used equally by all types of equipment (grill, oven and pan). Coal/wood is mainly related to a grill (86%) and oven (62%) since some Italian restaurants have a wood-fired oven for baking pizzas. The use of natural gas is mainly associated with a grill (84%), while LPG was mainly associated with apan/wok/fryer (91%). It is usually stated by chefs, that gas cooktops are flexible to use since they provide heat across the entire pan, so they don't have to worry about having a flat bottomed pan to cook.

3.2. Activity Profiles

The operation of restaurants on a monthly, daily and hourly scale is shown in Figure 4. Almost all the facilities (>98%) are in full operation from October to April. A small proportion of restaurants (4–9%) in Attica remain closed from the beginning of the touristic season (May) until September, probably because the owners have another restaurant close to the coastal zone of Attica or in a touristic area. In August in particular, 9% of the restaurants participating in the survey, mostly the family businesses, are closed due to summer vacations for the owners and staff. This is the case for many other activities in Attica. The daily profile revealed that all the restaurants are open from Wednesday to Saturday, while most of them are open on Tuesday (96%). A proportion of the local restaurants are closed on Sunday (10%) because Athenians prefer to go for a walk far from the municipality that they live in. On Monday, the restaurants have a low stream of customers. As a result, 11% of owners mentioned that they prefer to provide a day off for their staff. The hourly operation of the commercial kitchens, and not the restaurants, is presented in Figure 4, bottom. This is because many cooking preparations begin before the opening of the restaurants to the public. It is obvious that very few commercial kitchens start in the morning (9:00–11:00 LT), while in almost half of them (41 responses), the cooking preparations begin at 12:00 LT, indicating that these restaurants are open for both lunch and dinner. From 13:00 LT to 17:00 LT the mean number of commercial kitchens that operate is 61, while almost all kitchens (>90%) are open from 19:00 LT to 22:00 LT, representing the peak period that customers in Greece prefer to have dinner. A total of 75% of the restaurants that serve grilled dishes have a daily operation time of about 12 h. This is consistent with the study of [40], for Athens. COA concentrations presented a 50% increase of the daily average during the lunchtime hours (12:00–15:00 LT) and a large peak at night (at about 22:00 LT). A similar diurnal profile was also recorded at the COA measurements conducted by [39]. A separation was also made in the present study for weekdays and weekends (questions 3 and 4), revealing that more facilities serve lunch on weekends in comparison to weekdays. The temporal coefficients produced by the responses to the survey are included in Tables 5–8, following the monthly, daily and hourly profiles described previously.

Table 5. Monthly coefficients representing the operation of restaurants in Attica.

Month	Coefficient	Month	Coefficient
January	1.033	July	0.963
February	1.021	August	0.940
March	1.010	September	0.986
April	1.010	October	1.01
May	0.986	November	1.033
June	0.975	December	1.033

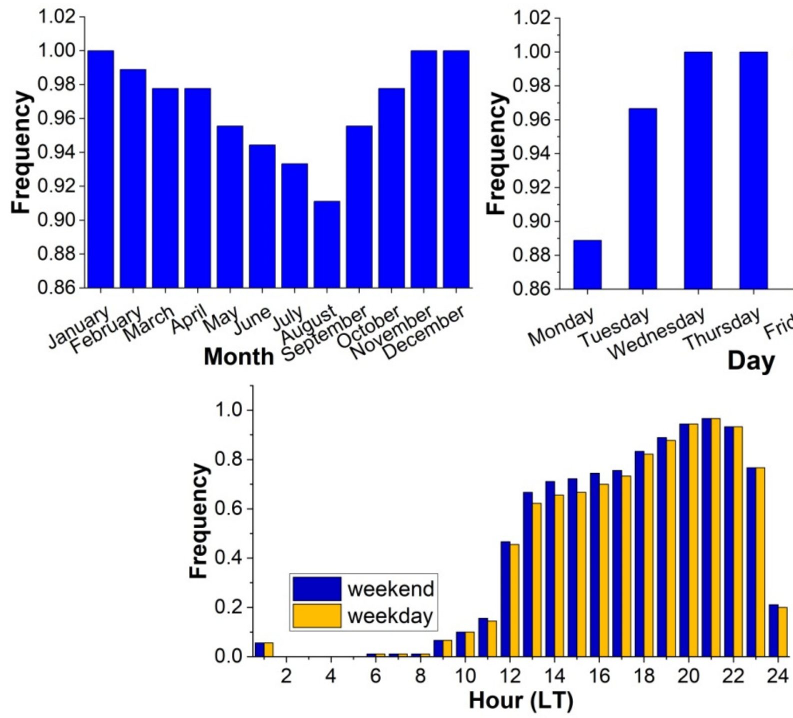


Figure 4. The monthly, daily and hourly profiles of the restaurants' operation.

Table 6. Daily coefficients representing the operation of restaurants in Attica.

Day	Monday	Tuesday	Wednesday	Thursday	Friday	Saturday	Sunday
coefficient	0.917	1.000	1.035	1.035	1.035	1.035	0.943

Table 7. Hourly coefficients representing the operation of commercial kitchens in Attica during weekdays (in Greece, local time = UTC + 2 h in winter and UTC + 3 h in summer).

Time (LT)	Coefficient	Time (LT)	Coefficient
1	0.137	13	1.532
2	0.000	14	1.615
3	0.000	15	1.642
4	0.000	16	1.724
5	0.000	17	1.806
6	0.027	18	2.025
7	0.027	19	2.162
8	0.027	20	2.326
9	0.164	21	2.381
10	0.246	22	2.299
11	0.357	23	1.888
12	1.122	24	0.493

Table 8. Hourly coefficients representing the operation of commercial kitchens in Attica during weekends (in Greece, local time = UTC + 2 h in winter and UTC + 3 h in summer).

Time (LT)	Coefficient	Time (LT)	Coefficient
1	0.132	13	1.596
2	0.000	14	1.703
3	0.000	15	1.729
4	0.000	16	1.783
5	0.000	17	1.809
6	0.027	18	1.996
7	0.027	19	2.129
8	0.027	20	2.262
9	0.160	21	2.315
10	0.239	22	2.235
11	0.372	23	1.835
12	1.118	24	0.506

3.3. Emissions

The annual emissions from commercial cooking in the GAA, calculated in the present study, are shown in Figure 5. Overall, 940.1 tonnes are attributed to commercial cooking activities. Values represent the species emitted only by the fuel combustion, and they are independent of the raw material used for the production of the dishes. From Figure 5, left, it is obvious that CO emissions prevail (451.6 tonnes) and they define the total value by having a 48.0% percentage contribution. NOx (176.6 tonnes) and SOx (125.8 tonnes) follow, while PM10 and PM2.5 accounted for about 4.2% (39.2 tonnes) and 3.9% (36.4 tonnes), respectively. The in-depth study of the different fuel contributions revealed that the abovementioned values are mostly due to coal combustion. More specifically, the percentage contribution of pollutants emitted at restaurants using coal to produce grilled dishes, to the total value of each pollutant, ranged from 91.1% (for CO) to 98.1% (for particulates). However, this was not the case for NOx, for which the percentage contribution to the total value was 68.9% from gas and 31.1% from coal.

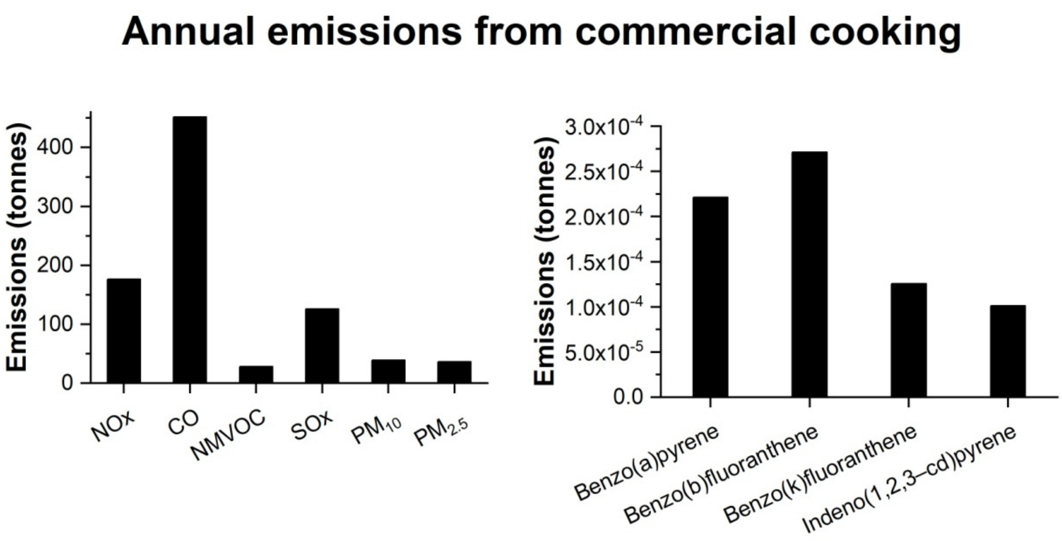


Figure 5. Emissions from the fuel combustion during commercial cooking activities in the Greater Athens area.

From the group of PAHs, benzo(b)fluoranthene had the highest value (0.03 tonnes, 37.9% of the total PAHs value) followed by benzo(a)pyrene (0.02 tonnes), while much lower was the percentage contribution of benzo(k)fluoranthene and indeno(1,2,3–cd)pyrene to the total PAHs emissions (17.2% and 13.8%, respectively). These emissions are almost totally due to coal and wood burning. Despite the effort to collect data following a bottom up approach, uncertainties in emissions calculation are unavoidable and in the present study they mainly concern the estimation of fuel consumption. Moreover, according to the methodology proposed by [47], uncertainties exist on the EFs used for the estimation of emissions. Finally, when conducting a survey, the sample size is always expected to be bigger in order to increase the representativeness of the results.

Comparison with other anthropogenic emission sources revealed that commercial cooking contribution, to the annual CO, NOx and PM10 total values for the GAA, is 0.6%, 0.8% and 1.0% (Figure 6, Fameli and Assimakopoulos 2016). It should be mentioned that, concerning navigation only, the emissions attributed to the urban cells close to the port of Piraeus were used. This is in agreement with [34], who presented that the commercial cooking contribution to the national PM2.5 emissions is 1% for the USA.

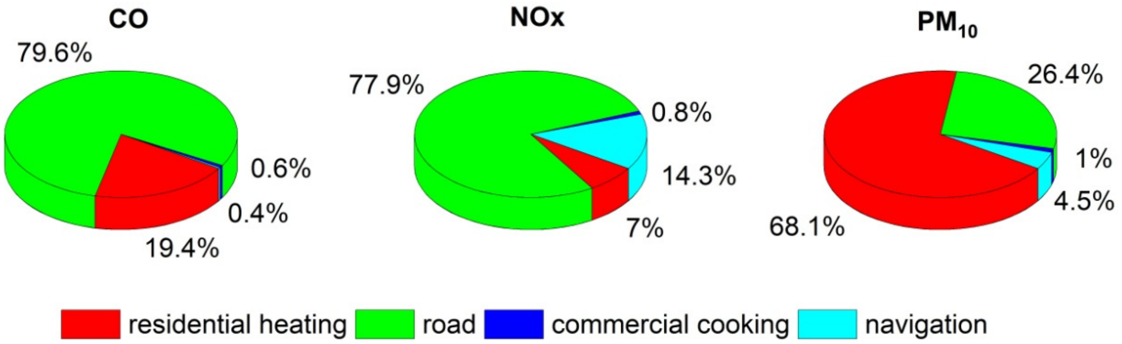


Figure 6. Percentage contribution of different sources (residential heating, road transport, commercial cooking and navigation) to the total emissions at the GAA.

NOx and PM2.5 annual emissions per regional unit in gridded form are presented in Figure 7. Higher values are attributed to the regional unit of Central Athens (69.1 tonnes for NOx and 14.2 tonnes for PM2.5) due to the fact that the majority of restaurants are located there. Many grilled restaurants also exist in the regional unit of North Athens. The maximum annual NOx and PM2.5 emissions are about 7.9 tonnes/km^2 and 1.6 tonnes/km^2, respectively, while lower than 1.5 tonnes/km^2 for NOx and 0.3 tonnes/km^2 for PM2.5 are attributed to the cells located in the regional unit of South Athens. It should be mentioned that the NOx and PM2.5 maximum gridded emissions from the other main sources in Central Athens are about 134.7 tonnes/km^2 and 6.1 tonnes/km^2, respectively, for road transport, and 11.4 tonnes/km^2 and 17.5 tonnes/km^2, respectively, for residential heating.

An effort to calculate emissions from grilled meat was also made based on the methodology proposed by [41]. By considering an average meat consumption of 28.4 kg per person in Greece [41] and taking into account (i) the population of the GAA (about 3.2 millions, source: Hellenic Statistical Authority, 2011), (ii) the emission factors (EFs) proposed by [25] for VOCs and (iii) the mean EFs for COA proposed by [41], the annual total emissions were produced for the GAA (Table 9). Bearing in mind that the total emissions from the fuel burnt are 940.1 tonnes, it is obvious that COA emissions (724.9 tonnes) are not negligible, and surely they define the air quality of the area where restaurants and related activities are located. COA emissions for January at 21:00LT are spatially distributed in cells of 1 km^2 in Figure 8b. Maximum values are reported in the city center (>4.5 kg/km^2). The diurnal profile of COA emissions for winter at a selected cell in Central Athens (Figure 8a, total daily COA emission = 2.7 kg/km^2) displays a first peak (0.17 kg/km^2 at 13:00 LT) during lunchtime (13:00–15:00 LT) while the largest hump appears at night from 20:00–22:00 LT (0.27 kg/km^2 at 21:00 LT). Similar diurnal profiles, but with a small delay (of approximately a couple of hours), were found by [40] for Athens. This is because the temporal profiles proposed in the present study refer to the preparation of the meals and thus represent, as realistically as possible, the emissions from the operation of the kitchen, while the diurnal profiles reported by [40] were produced by the pollutants measured in the atmosphere, and thus, maximum concentrations are reported delayed.

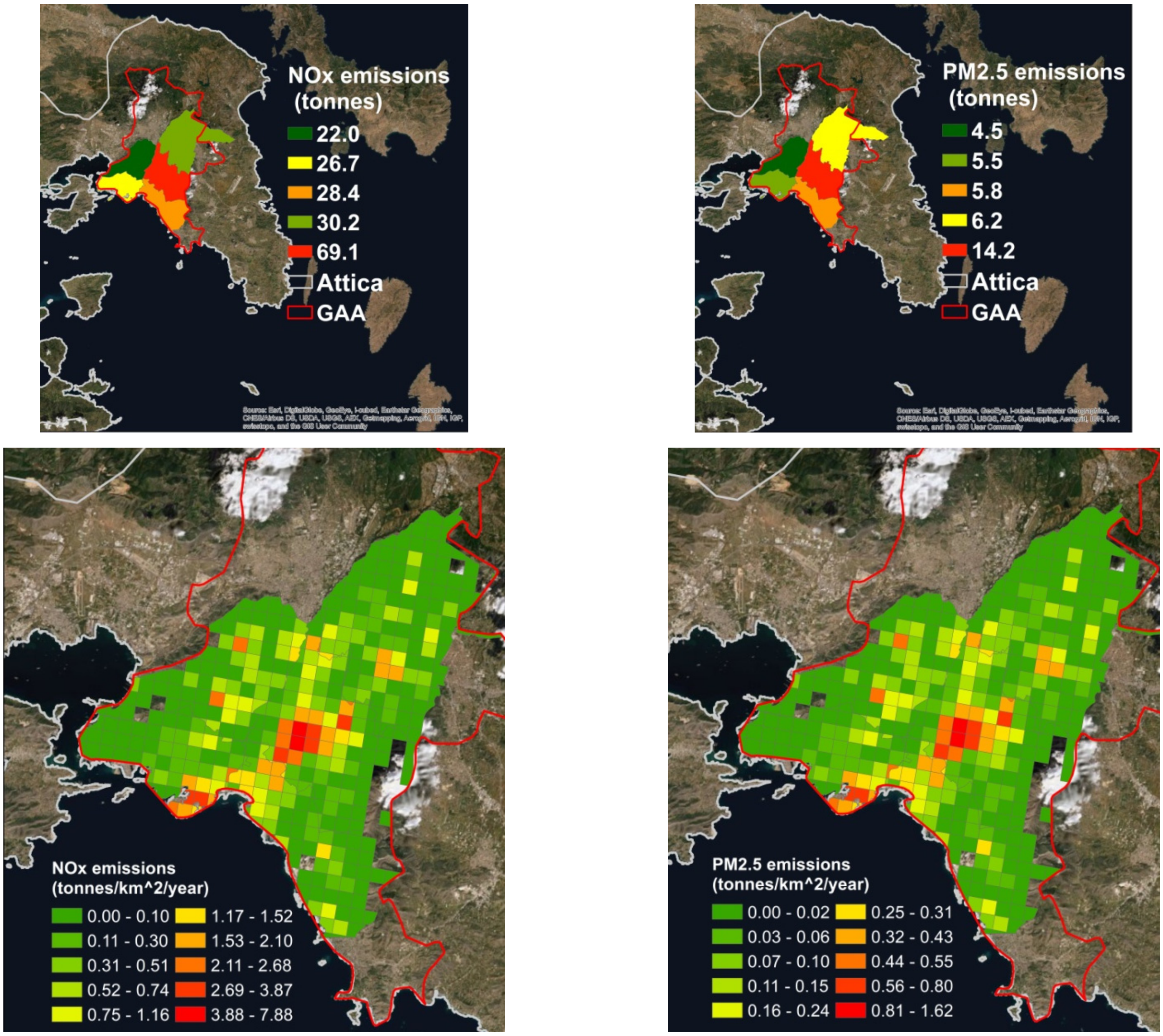
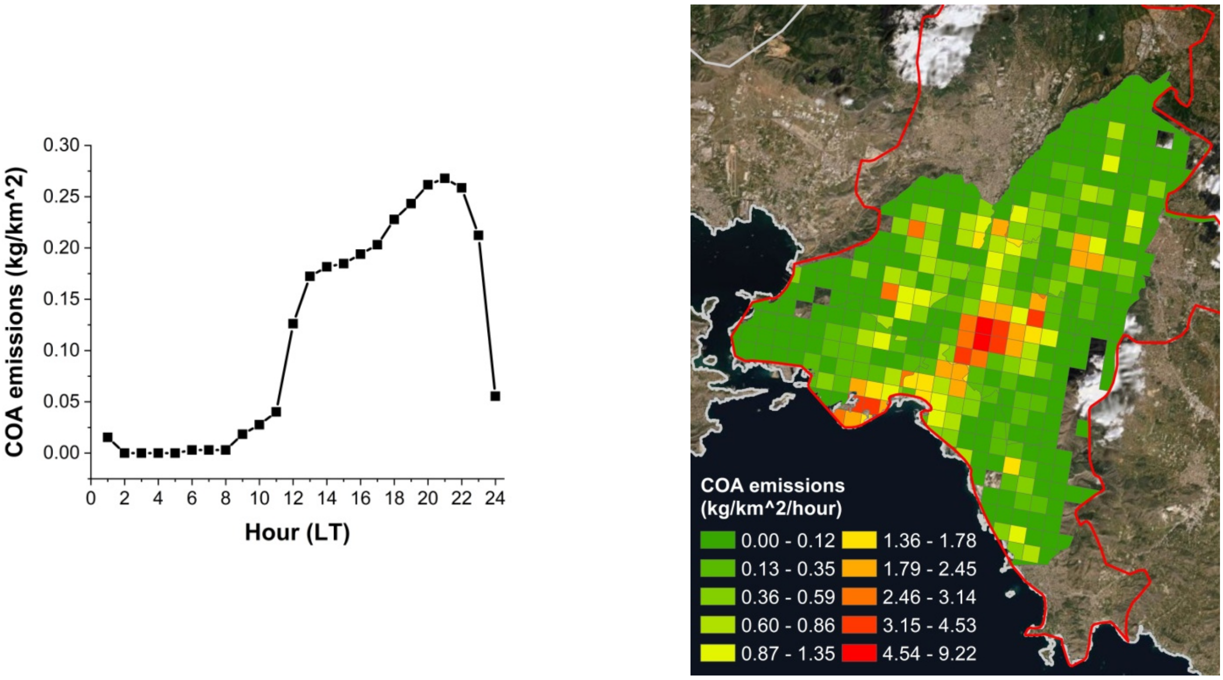


Figure 7. NOx and PM2.5 emissions per regional unit (**top**) and gridded emissions (**bottom**).

Table 9. Annual emissions produced by the grilled meat at the GAA.

Pollutant	Emissions (tn)
VOCs	
Acetonitrile	0.9
Acetone	2.7
Isoprene	4.5
Benzene	8.2
Toluene	8.2
Xylenes	9.1
Monoterpenes	3.6
COA	724.9



(**a**) (**b**)

Figure 8. (**a**) The diurnal profile of COA gridded emissions (in kg/km^2, for a Friday in January) for a selected cell in Central Athens and (**b**) the spatial distribution of COA emissions in January at 21:00 LT.

3.4. Numerical Approach

Figure 9 shows the distribution of the hourly PM10 ground concentrations, as predicted by CAMx for both cases (*all_sources* and *only_cook*), at 20:00 LT and 22:00 LT, when the commercial cooking emissions reached their peak value. During the simulated period low winds prevailed so the pollutants were mainly affected by the local emissions. The highest value was observed in the urban center, where the majority of restaurants are located, as well as other anthropogenic activities (22 ug/m^3 at 22:00 LT when emissions from all sources were used in the simulation; Figure 9, top). The maximum contribution of commercial cooking to the total PM10 concentrations was about 6% (1.2 ug/m^3), indicating that it is an important source that needs to be addressed in local emissions inventories. The cooking fumes that were created at approximately18:00 LT, with concentrations ranging from 0.15 ug/m^3 to 0.6 ug/m^3, increased and had the maximum spatial coverage in the evening at 22:00 LT (Figure 9, bottom).

The calculated statistical parameters for the evaluation of model results are presented in Table 10.

Table 10. Statistical parameters for the evaluation of model concentrations for 16 March 2022.

	O_3, all_Sources			PM10, all_Sources		
	Port	*South Athens*	*Center*	*Port*	*South Athens*	*Center*
MO	32.96	47.50	36.00	60.88	46.83	64.92
MS	34.53	50.14	36.05	15.94	9.20	13.49
MB	1.57	2.64	0.05	−44.94	−37.63	−51.43
NMB	0.05	0.06	0.00	−0.74	−0.80	−0.79
NME	0.36	0.25	0.37	0.74	0.80	0.79
RMSE	16.58	15.46	18.08	51.04	42.06	56.81
r	0.84	0.94	0.86	0.59	0.59	0.70

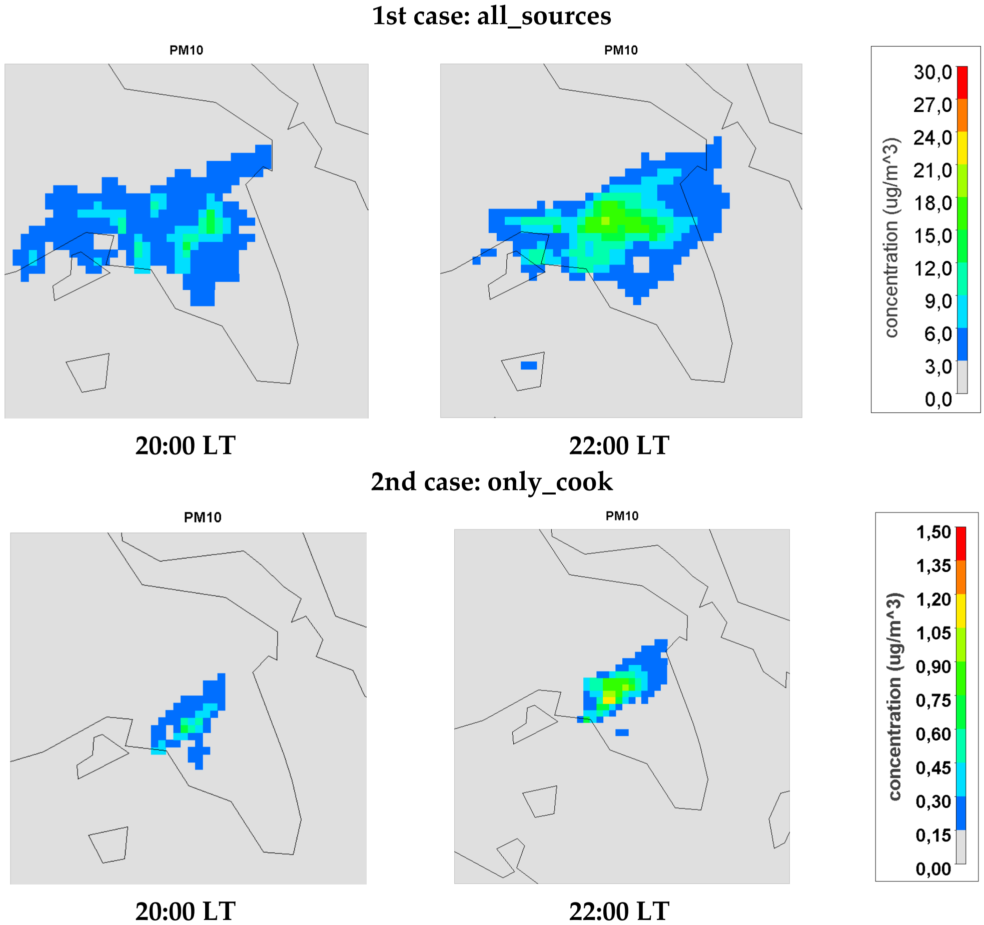


Figure 9. PM10 concentration dispersion at 20:00 LT and 22:00 LT at the GAA, having all emission sources (**top**) and only commercial cooking emissions (**bottom**) as input at the numerical simulations.

Model reproduced satisfactory O_3 concentrations within the urban area as the MS values are very close to the measured (MO) ones, and the correlation coefficient ranged from 0.94 in South Athens, to 0.84 at the port of Piraeus. Model performance was quite good for the particulates, since the model results were lower for the measurements for all three sites.

4. Conclusions

The cooking fumes that are created during meal preparation in restaurants deteriorates air quality in the surrounding area, especially in areas where these activities are densely located. Very few efforts have been made worldwide to compile emissions inventories of commercial cooking activities, either on a national scale or at a regional level, mainly due to the fact that the provision of the information needed in order to perform the relevant study is not easy and existing data are scarce. Thus, the purpose of the current work was to present the methodology for the development of an inventory of the commercial cooking sector, in the region of Attica. For this reason, a survey was conducted of the restaurants located in the Greater Athens area, in order to gather as much information as possible.

Among the main conclusions are:

- The chemical profile of the pollutants emitted from cooking is highly dependent on the cooking style, defined by the appliance and the fuel used for the food preparation.
- Electricity and LPG were the most preferable fuels, while natural gas and coal followed.
- The use of coal (and wood) for the production of grilled dishes is responsible for 98.1% of the total emitted particulates (PM10 and PM2.5). NOx emissions mostly originated from gas (68.9%), and, secondarily, from coal (31.1%).

- The hourly operation of the kitchen, and not the restaurant, was recorded by the survey since many cooking preparations begin before the opening of the restaurants to the public.
- Temporal coefficients were produced from the responses to the survey for the disaggregation of the annual emissions, to monthly, daily and hourly values.
- Very few commercial kitchens start in the morning (9:00–11:00 LT), while almost half of them begin cooking preparations at 12:00 LT. From 19:00 LT to 22:00 LT, almost all kitchens (>90%) are open, representing the peak period that customers in Greece prefer to have dinner.
- COA emissions from grilled meat were also calculated, accounting for about 724.9 tonnes, while PM10 emissions from the fuel burnt is responsible for only 39.2 tonnes, indicating that the raw material also plays an important role.
- The total PM10 emissions from commercial cooking accounts for 1% of total emissions at the GAA, as compared to traffic, residential heating and navigation.
- The most important pollutants are COA, CO and NOx.
- Spatial and temporal disaggregation of pollutants can be used for exposure studies, as well as for applications with chemical transfer models (CTMs).
- The maximum contribution of commercial cooking to the total PM10 concentrations was about 6% in the urban center, indicating that it is an important source that needs to be addressed in local emissions inventories.

Author Contributions: Conceptualization, K.-M.F., A.K. and V.D.A.; methodology, K.-M.F., A.K. and V.D.A.; validation, K.-M.F. and A.K.; data analysis, K.-M.F., A.K. and V.D.A.; writing—original draft preparation, K.-M.F., A.K. and V.D.A.; visualization, K.-M.F.; supervision, K.-M.F.; project administration, K.-M.F. All authors have read and agreed to the published version of the manuscript.

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Appendix A. Factors Used for the Calculation of Emissions

Table A1. Emission factors for medium-sized (>50 KWth to ≤1 MWth) boilers burning natural gas (source: EMEP/EEA emission inventory guidebook). *KWth and MWth stands for Kilowatt Thermal and Megawatt Thermal, respectively.*

Pollutant	EF	Unit
NOx	73	g/GJ
CO	24	g/GJ
NMVOC	0.36	g/GJ
SOx	1.4	g/GJ
PM10	0.45	g/GJ
PM2.5	0.45	g/GJ
BC	5.4	% of PM2.5
Pb	0.0015	mg/GJ
Cd	0.00025	mg/GJ
Hg	0.1	mg/GJ
As	0.12	mg/GJ
Cr	0.00076	mg/GJ
Cu	0.000076	mg/GJ
Ni	0.00051	mg/GJ
Se	0.011	mg/GJ

Table A1. *Cont.*

Pollutant	EF	Unit
Zn	0.0015	mg/GJ
PCDD/F	0.5	ng I-TEQ/GJ
Benzo(a)pyrene	0.56	µg/GJ
Benzo(b)fluoranthene	0.84	µg/GJ
Benzo(k)fluoranthene	0.84	µg/GJ
Indeno(1,2,3-cd)pyrene	0.84	µg/GJ

Table A2. Emission factors for medium-sized (>50 KWth to $\leq$1 MWth) manual boilers burning coal fuels (source: EMEP/EEA emission inventory guidebook). *KWth stands for Kilowatt Thermal and Megawatt Thermal, respectively*.

Pollutant	EF	Unit
NOx	200	g/GJ
CO	1500	g/GJ
NMVOC	100	g/GJ
Sox	450	g/GJ
PM10	140	g/GJ
PM2.5	130	g/GJ
BC	6.4	% of PM2.5
Pb	150	mg/GJ
Cd	2	mg/GJ
Hg	6	mg/GJ
As	4	mg/GJ
Cr	10	mg/GJ
Cu	15	mg/GJ
Ni	20	mg/GJ
Se	2	mg/GJ
Zn	200	mg/GJ
PCDD/F	200	ng I-TEQ/GJ
Benzo(a)pyrene	90	mg/GJ
Benzo(b)fluoranthene	110	mg/GJ
Benzo(k)fluoranthene	50	mg/GJ
Indeno(1,2,3-cd)pyrene	40	mg/GJ

Appendix B. Questionnaire about Commercial Cooking Activities

Question 1: **Please select the month(s) that the restaurant is closed.**

January	July
February	August
March	September
April	October
May	November
June	December

Question 2: **Please select the day(s) that the restaurant is closed.**

Monday	Tuesday	Thursday	Wednesday	Thursday	Friday	Saturday	Sunday

Question 3: Please mention the period that the kitchen is open in weekdays (open question).

Question 4: Please mention the period that the kitchen is open in weekends (open question).

Question 5: Please select the cooking equipment that is used for the cooking demands of your restaurant. You can make more than one selections.
(a) grill
(b) oven
(c) pan/wok/fryer

Question 6: Please mention the fuel(s) that you use for each cooking equipment. You can make more than one selections.

Equipment	Natural Gas	LPG
Grill		
Oven		
Pan/Wok/Fryer		
	Coal/Wood	Electricity
Grill		
Oven		
Pan/Wok/Fryer		

Question 7: Please select which of the below type(s) of raw material you use.
(a) lamp (d) beef
(b) fish (e) chicken
(c) pork (f) no meat

Question 8: Please mention the amount of each raw material that you use in a daily, weekly, or monthly scale (open question).
(a) lamp (d) beef
(b) fish (e) chicken
(c) pork

Appendix C. The Statistical Parameters Used for the Evaluation of Model Results

For the evaluation of the model results, a comparison of the hourly model concentrations (simulation values—S) with measurements from the national air quality network (observation values—O) was performed following the methodology proposed by [50].

Correlation Coefficient (r)

$$r = \frac{\sum_{i=1}^{N}\left(O_i - \overline{O_i}\right)\left(S_i - \overline{S_i}\right)}{\sqrt{\sum_{i=1}^{N}\left(O_i - \overline{O_i}\right)^2 \sum_{i=1}^{N}\left(S_i - \overline{S_i}\right)^2}} \quad \text{(A1)}$$

It is a measure of the relationship between the observation value (O) and simulation value (S). When r is close to 1 (±1) the relationship between the above values is strong, while, when r is close to 0, the relationship is weak.

Mean Bias (MB)

$$MB = \frac{1}{N}\sum_{i=1}^{N}(S_i - O_i) \quad \text{(A2)}$$

It provides a measure of the difference between the observation value and simulation value. When MB > 0, the model has overestimated the expected concentration, while MB < 0 means that that the model has underestimated the expected concentration.

Root Mean Square Error (RMSE)

$$RMSE = \sqrt{\frac{1}{N}\sum_{i=1}^{N}(S_i - O_i)^2} \quad \text{(A3)}$$

It represents the standard deviation of the differences between observations (O) and model results (S). RMSE takes values ≥ 0, with zero being the most satisfactory value in terms of model performance.

Normalized Mean Error (NME)

$$\mathrm{NME} = \frac{\sum_{i=1}^{N} |S_i - O_i|}{\sum_{i=1}^{N} O_i} \tag{A4}$$

Normalized Mean Bias (NMB)

$$\mathrm{NMB} = \frac{\sum_{i=1}^{N} (S_i - O_i)}{\sum_{i=1}^{N} O_i} \tag{A5}$$

Mean Normalized Biass Error (MNBE)

$$\mathrm{MNBE} = \frac{1}{N} \sum_{i=1}^{N} \frac{S_i - O_i}{O_i} \tag{A6}$$

Mean Normalized Gross Error (MNGE)

$$\mathrm{MNGE} = \frac{1}{N} \sum_{i=1}^{N} \frac{|S_i - O_i|}{O_i} \tag{A7}$$

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