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Post-Chernobyl ^{137}Cs in the atmosphere of Thessaloniki: a consequence of the financial crisis in Greece

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ABSTRACT

The background radiation level of ^{137}Cs at the urban atmosphere of Thessaloniki has been increased during the recent decade only due to the Fukushima accident fallout. Since then, no other signal of ^{137}Cs was observed until the winter period of 2013, when slightly elevated ^{137}Cs concentrations were measured. The ^{137}Cs signals observed were up to 12 μBq m^{-3}, mainly during holidays and weekends followed by lower or even non-detectable activities in the next working days. Those episodes are attributed to the increase of biomass products combustion for residential heating as this year the tax of oil for heating was drastically raised as a consequence of the financial crisis. A preliminary survey of various wood products as well as of bottom ashes from different domestic burning devices is presented. ^{137}Cs concentrations up to 11 Bq kg^{-1} were measured in wood products and up to 500 Bq kg^{-1} in ash samples.

1. Introduction

The radioactivity in the atmosphere of Thessaloniki, Greece, is continuously measured during last decades after the Chernobyl accident as part of the environmental radioactivity monitoring program carried out by the Atomic and Nuclear Physics Laboratory of Aristotle University of Thessaloniki. Naturally occurring radionuclides like radon and thoron progeny as well as ^{7}Be are normally present in the urban air, while ^{137}Cs concentration was usually not detectable with an exemption during the period of the Fukushima accident when ^{134}Cs was also present (Manolopoulou et al., 2012). The relatively long lived ^{137}Cs due to Chernobyl accident was appearing in the surface air of Europe at trace levels below 1 μBq m^{-3} (AMAP, 2010; Masson et al., 2010), while ^{134}Cs had not been measured in the atmosphere since the middle of the 1990s due to its fairly short half-life.

Slightly elevated ^{137}Cs concentrations were measured during the first months of 2013, which were following a pattern: higher concentrations were measured during weekends and holidays, followed by lower or non-detectable activities in the subsequent working days. An analogous behavior has been reported regarding the PM_{10} (PM_{10}: Particulate Matter, diameter less than 10 microns) and $PM_{2.5}$ concentrations measured in the atmosphere at the center of the city for the same time span. An increase of 13% and 25% was observed in the concentrations of PM_{10} and $PM_{2.5}$ respectively, compared with the corresponding values before the financial crisis and it was attributed to the biomass combustion for domestic heating mainly (Petrakakis et al., 2013).

The recent financial crisis, raised in Greece after 2009, among the other consequences, has led to the transition of residents in biomass combustion as the taxation of the oil for domestic heating was significantly increased, according to the financial recommendations, leading to a rise of 30% of its price. According to the Hellenic Statistical Authority the oil consumption between October to February of 2011–12 and 2012–13 was decreased by 68.7%. Additionally, the gas consumption in the urban area of Thessaloniki has been reduced by about 27%. Thus, the customary fossil fuels have been replaced by biomass products due to the lower taxation.

Wood can be used in raw form or in processed form like pellets, briquettes and chips. Combustion of wood and other bio-fuels is one of the main supplies of particulate matter, organic compounds and aerosol formations in urban areas during winter. The pollutants production is a function of various factors such as the type and quality of the biomass products and the burning appliance, while the pollutants accumulation in the atmosphere is influenced by the meteorological conditions and the topography of the area (Johansson et al., 2004; Caseiro et al., 2009; Chrysikou and Samara, 2009; Gonçalves et al., 2010; Holden et al., 2011; Vu et al., 2012). Among the pollutants released in the atmosphere due to biomass

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combustion is the long-lived radioactive isotope ^{137}Cs (Bourcier et al., 2010). The specific radionuclide has been inserted in the atmosphere due mainly to Chernobyl accident in 1986 and since then the forest ecosystem in Europe has been contaminated (Bunzl and Kracke, 1988; Ronneau et al., 1991; Ravila and Holm, 1996; Fogh and Andersson, 2001; Clouvas et al., 2007; Zhiyanski et al., 2010). The contaminated biomass is not dangerous itself; however, there are some health impacts especially occurred due to the inhalation of the smoke and fine ash aerosols produced during the burning process as well as due to the ash usage as fertilizer for the soil (Bølling et al., 2009; Ladygienė et al., 2010). In addition, they degraded the air quality of the urban Thessaloniki area, due to large quantities of gaseous air pollutants and particles emitted (Petrakakis et al., 2013).

A preliminary survey of various kinds of wood and pellet samples as well as of bottom ashes from different type of domestic burning devices has been implemented, in addition to the atmospheric radioactivity monitoring program. Biomass combustions occurring in highly populated cities are of particular concern since they add to the impacts caused by high urban pollution levels. Further elaboration on the processes taking place during similar usage in urban centers could make use of the results presented in this paper.

2. Experimental methods and materials

The air sampling was carried out using a Staplex type TFIA-2 high volume air sampler operating at a regulated air flow rate ranging from 28 to 32 L s^{-1} with a 8″ × 10″ glass fiber filter type TFAGF810. With this design the collection efficiency is 99.98% for particle size 0.3 μm and over. Air sampling duration was about 24 h and the air sample volume was around 2600 m^3. The location of the air sampling was 50 m above sea level at the roof of the Faculty of Sciences located at the center of the city (Fig. 1). The Thessaloniki municipality area is densely populated (16,703 residents/km^2 according to the census of 2011) whilst the majority of the apartment buildings were built during '60s and '70s with central heating systems, whereas the older apartment buildings although they have chimneys for stove installation do not provide storage areas for wood. Thus this area is not expected to contribute significantly to the atmospheric pollution because of biomass combustion. The only area near the center of the city where open fireplaces are widespread is the historic "old city" (see Fig. 1), where inhabits only 4% of the total permanent population of the urban area of Thessaloniki (about 800,000). The other region with newer buildings which typically are equipped with open fireplaces is the neighboring area around the municipality of Thessaloniki with 59% of the total permanent population (see Fig. 1).

The wood samples examined during this study were from two areas of Northern Greece Hmathia and Chalkidiki, which are the main local sources of biomass products (see Fig. 2). Three types of wood have been measured: oak, beech and plane. Imported wood and pellet samples produced in Serbia, Bulgaria and Romania were also measured. The samples were oven-dried at 40 °C to constant weight. The bottom ash samples were collected from different type of domestic burning devices like fireplaces as well as wood- and pellet-stoves.

All samples were measured for radioactivity using a high purity, low background Ge detector with resolution 1.8 keV at 1.33 MeV and relative efficiency 42%. The air filters were measured using the 'standard filter' geometry (Ø 6 cm) for 4–5 × 10^5 s to achieve a determination limit of 2 μBq m^{-3} with 50% uncertainty (Gilmore and Hemingway, 1995). Marinelli geometry (1 L) was used for ash and crashed pellet samples while the wood samples were measured in raw form. The γ-spectrometric system was calibrated for filter, pellets and ash samples using standard reference sources and IAEA samples of worldwide proficiency tests in which the laboratory takes part for more than 15 years. The wood samples were measured without bark and the counting efficiency was determined individually for each sample using GEANT4 (Agostinelli et al., 2003) code simulations. The overall uncertainty of the measurements ranged between 20 and 40% for the filter geometry, < 5% for the ash samples and 10–40% for woods and pellets.

3. Results and discussions

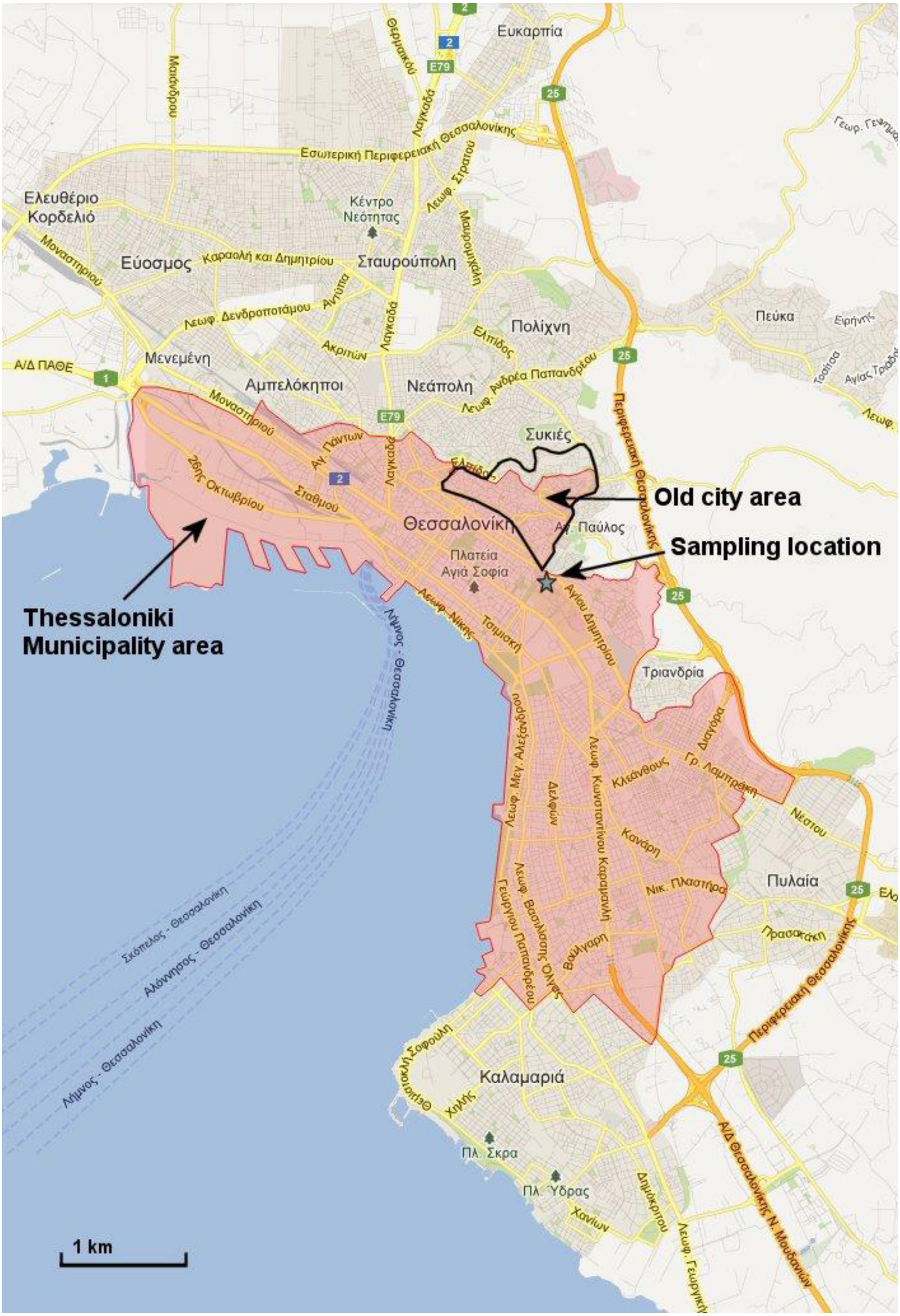
3.1. ^{137}Cs Concentrations in surface air

During the recent decades (2000–2013), the background radiation level of ^{137}Cs at the urban atmosphere of Thessaloniki was lower than the determination limit of the counting systems used, i.e. <4 μBq m^{-3} for counting time of about 2 × 10^5 s with an exemption during the Fukushima accident fallout. The activity concentrations of aerosol-bound ^{137}Cs in air from March 24, 2011 through April 25, 2011 ranged from <5 up to 145 μBq m^{-3} accompanied by ^{134}Cs in similar concentrations (Manolopoulou et al., 2012). Since then, no other signal of ^{137}Cs was observed until the winter period of 2012–2013.

The ^{137}Cs concentrations measured during January–March 2013 in the urban atmosphere of Thessaloniki are presented in Fig. 3. The determination limit for these measurements was 2 μBq m^{-3} for counting time of about 5 × 10^5 s. In the same figure hourly precipitation rate and average night-time (19:00–07:00) temperature and wind speed are presented. The night time averages were selected instead of the 24 h averages, as they are considered more indicative for the operation of domestic heaters. The higher values, above 5 μBq m^{-3}, were measured during weekends and holidays (marked with vertical lines in Fig. 3) when the inhabitants spend more time indoors with the domestic wood-heating devices working almost all day long. It must be noted that during the nights of this winter it was the first time that the smell of burning wood was present in the atmosphere of the central city. Thus, it is expected that the night time concentrations would be higher than the average of the 24 h concentrations that was measured and presented in Fig. 3. The following working days the signal of radioactivity was smaller or even removed since the people were using heating devices for shorter periods.

From Fig. 3 it comes out that the measured concentration of ^{137}Cs in the atmosphere was influenced by the meteorological conditions of the time period studied, i.e. by the temperature and wind velocity. While the low temperature triggers the increase of biomass products consumption for heating purposes, its combination with low wind velocity favors the accumulation of aerosol-bound ^{137}Cs in the atmosphere. That could be the reason of the higher concentrations measured during 19 and 27 of January and 2, 9 and 10 of February (see Fig. 3) when both wind velocity and temperature were the lowest. On the contrary, during 12 and 26 of January and 16 of February lower measurements were recorded probably due to the higher wind velocity for these time periods.

In Fig. 3 with a diamond shape is presented the concentration of ^{137}Cs in the atmosphere of Kardia, a suburb of Thessaloniki (20 km distance, see Fig. 2) with a population of about 3400 people. The higher value, 12.1 (±16%) μBq m^{-3}, that was measured at Kardia area may be attributed either to the proximity of the emission sources, as this is a newly built-up area where open fireplaces are available to the majority of the houses, or to the topography of the location which can be resembled as a closed urban valley where air masses can remain stagnant. The concentration of ^{137}Cs measured during the same day at the standard sampling location was 7.4 (±18%) μBq m^{-3} The standard sampling location is at the center of the city at a relatively open landscape while the position of the emission sources can be at the "old city", about 1–1.5 km away from the sampling site, as well as at the areas around the municipality of Thessaloniki, around 2–7 km away (see Fig. 1). A more detailed


Old city area
Sampling location
Thessaloniki
Municipality area
1 km
Ευκαρπία
Ελευθέριο
Κορδελιό
Εύοσμος
Σταυρούπολη
Κέντρο
Νεότητας
Πολίχνη
Πεύκα
Μενεμένη
Αμπελόκηποι
Νεάπολη
Συκιές
Θεσσαλονίκη
Αγ. Παύλος
Πλατεία
Αγιά Σοφία
Τριανδρία
Πυλαία
Καλαμαριά
Πλ. Σκρα
Πλ. Ύδρας
Α/Δ ΠΑΘΕ
Εσωτερική Περιφερειακή Θεσσαλονίκης
Περιφερειακή Θεσσαλονίκης
Α/Δ Θεσσαλονίκης Ν. Μουδανιών
Μοναστηρίου
26ης Οκτωβρίου
Σταθμού
Λαγκαδά
Λεωφ. Νίκης
Τσιμισκή
Αγίου Δημητρίου
Λεωφ. Μεγ. Αλεξάνδρου
Λεωφ. Κωνσταντίνου Καραμανλή
Κλεάνθους
Γρ. Λαμπράκη
Νέστου
Κανάρη
Νικ. Πλαστήρα
Βούλγαρη
Λεωφ. Βασιλίσσης Όλγας
Γεωργίου Παπανδρέου
Σκόπελος - Θεσσαλονίκη
Αλόννησος - Θεσσαλονίκη
Λήμνος - Θεσσαλονίκη
Χανίων
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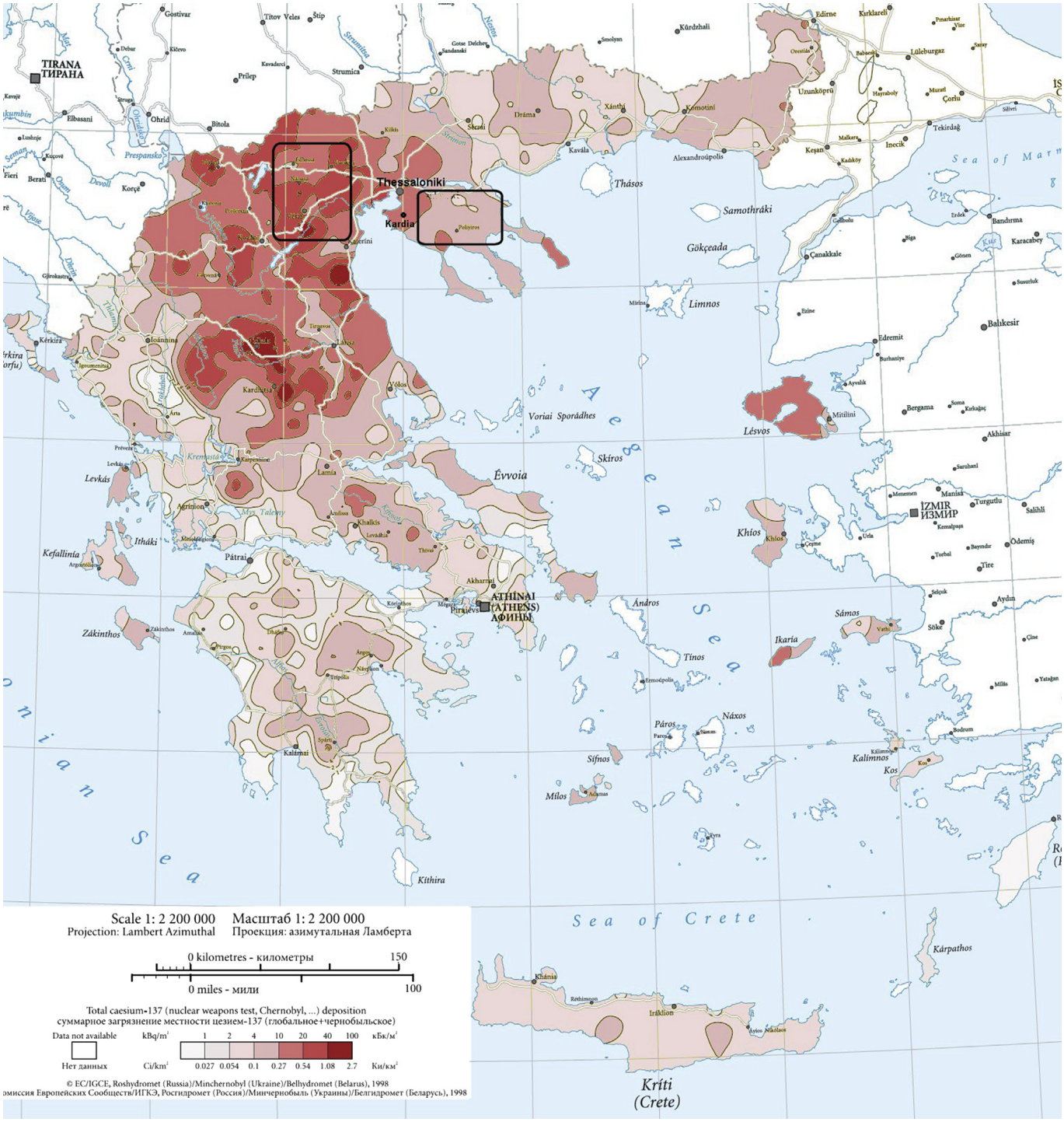

Fig. 2. Total ^{137}Cs deposition in North Greece (De Cort et al., 1998). The origin of the wood and ash samples is the marked areas in the rectangular.

study of the influence of meteorological conditions as well as the topography of the urban area is necessary to clarify the issue.

3.2. ^{137}Cs concentrations in wood and bottom ash

A preliminary survey of various kind of wood and pellet samples as well as of bottom ashes from different type of domestic burning devices has been implemented in order to examine the contribution of biomass products combustion to the increase of ^{137}Cs measured in the urban atmosphere of Thessaloniki. The ^{137}Cs concentrations of the ash samples produced by imported woods from the Balkan area ranged from 31 up to 64 Bq kg^{-1}, and from imported pellets was 174 Bq kg^{-1} according to the results presented in Table 1. The concentration of ^{137}Cs in the ash produced from local

Fig. 1. The urban area of Thessaloniki is presented. The oldest part, corresponding to the Thessaloniki municipality area, is indicated. Air sampling location is indicated with a star, while the nearby small marked area is the historic "Old city" where open fireplaces are widespread. (Map data ©2013 Google).

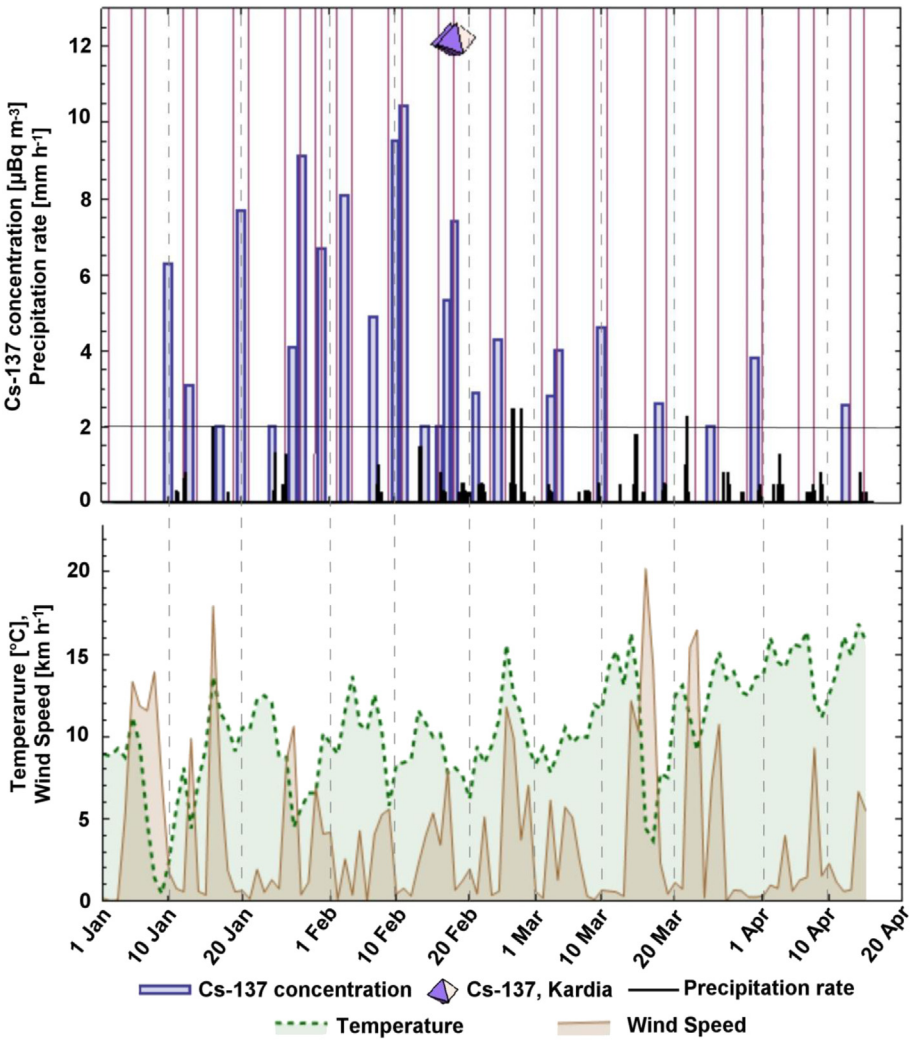

Fig. 3. Time variation of ^{137}Cs concentrations measured in the atmosphere of Thessaloniki during the winter of 2013 together with hourly data of precipitation rate. The determination limit for ^{137}Cs concentrations in the atmosphere is 2 μBq m^{-3} (horizontal line). The vertical lines delimit the weekends and holidays periods. In the lower part the average night-time (19:00–07:00) temperature and wind speed.

origin wood combustion ranged from 25 to 500 Bq kg^{-1}, which are higher than ashes produced from imported wood (see Table 1) probably due to the higher Chernobyl fallout deposition in Northern Greece than in the forest area of neighboring countries. It must be noticed that ^{137}Cs presence is attributed mainly to the fallout of the Chernobyl accident (see Fig. 2) because ^{134}Cs ($T_{1/2} = 2.07$ y) has not been measured in the atmosphere, in the wood or ash samples, although its concentrations were similar to that of ^{137}Cs during the Fukushima accident. The ashes from household fireplaces are usually used as fertilizers in the domestic gardens. So far, the maximum measured ^{137}Cs concentration in ashes is half the exemption level (1000 Bq kg^{-1}) defined for ^{137}Cs in Hellenic Gazette B216/6-3-2001.

Oak trees samples contained the lowest ^{137}Cs concentrations measured, lower than 1.3 Bq kg^{-1}, while the concentrations in beech and plane trees ranged up to 11.0 Bq kg^{-1} (Table 2). This could be attributed to the different growing location of each type in the forest ecosystem but the number of samples is small (9 in total) to draw a definite conclusion. The activities presented in Table 2 are in good agreement with previously reported data during the last years by other researchers reported for the same areas and biomass products (Clouvas et al., 2007; Zhiyanski et al., 2010; Ladygienė et al., 2010; Desideri et al., 2012).

The activities measured in the ash samples were enriched in respect to the activities of the burned biomass products due to the

Table 1
Activity concentrations measured in ash samples (Bq kg^{-1}).

Ash samples produced from	^{137}Cs	^{40}K
Wood imported from Bulgaria-Romania [7][a]	31–64	3280–4370
Pellets imported from Serbia [1]	174	3082
Local origin wood [16]	25–499	1592–7060

[a] The values in the brackets refer to the number of samples.

Table 2
Activity concentrations measured in biomass products (Bq kg^{-1}).

	^{137}Cs	^{40}K	^{137}Cs
Oak trees [4][a]	<0.05–1.3	12–37	0.8–1.1 Zhiyanski et al., 2010
Beech trees [4]	<0.05–11.0	34–35	1–12 Clouvas et al., 2007
Plane tree [1]	8.2	23	
Pellets imported from Serbia [1]	5.2	36	

[a] The values in the brackets refer to the number of samples.

Table 3
"Wood-to-Ash" enrichment factor estimated using ^{40}K concentrations.

Type of appliance	Wood (Bq kg^{-1})	Ash (Bq kg^{-1})	Ash/Wood
Fireplace [5][a]	12–34	2602–3322	77–209
Wood-stove [2]	35–37	3928–4096	107–118
Pellet-stove [1]	36	3082	85

[a] The values in the brackets refer to the number of samples.

removal of organic compounds during combustion. To estimate the *"wood-to-ash"* enrichment factor the ^{40}K concentrations were used, assuming that K is not volatile like Cs and thus the potassium present in the raw material remains in the ash produced during the combustion (Hedvall and Erlandsson, 1992). The reciprocal of this factor can give the percentage of ash produced by each type of wood and/or type of heater. For this assessment the people who provided the wood and ash samples were asked to clean the fireplaces and burn only the specific type of wood that was measured. The wood-to-ash enrichment factor ranged from 77 up to 209 for open fireplaces, 107 up to 118 for wood-stoves and 85 for the pellet-stove, according to the results presented in Table 3. The enrichment factors estimated in the present study were in the range reported in the literature for wood chips (86–370) (Ladygiené et al., 2010).

The wood-to-ash enrichment factors could be used to estimate the mass balance of ^{137}Cs in biomass combustion. However, for biofuel power plants, it has been reported that it is questionable if the specific procedure can be utilized to assess the amount of ^{137}Cs released in the atmosphere especially because of grab sampling (Hedvall et al., 1996). Other researchers reached to the same conclusion even though they measured integrated samples over time in a Lithuanian factory (Ladygiené et al., 2010). So far, to the knowledge of the authors, similar calculations were not reported for domestic heating devices. Using the data presented in Tables 1–3 mass balance calculations were performed indicating that 41% up to 79%, of ^{137}Cs contained in the biomass is not accounted for in the ash produced. These percentages are reaching quite high values that are comparable only with open field fire data for Cesium (Amiro et al., 1996). The above findings indicate the necessity of more extensive studies to assess the emissions of ^{137}Cs in the atmosphere from domestic heaters.

4. Conclusions

Signals of ^{137}Cs have been observed during the winter of 2013 in the urban atmosphere of Thessaloniki, which were up to one order of magnitude higher than the background measurements. The ^{137}Cs concentrations measured, up to 12 μBq m^{-3}, are attributed to the biomass combustion for residential heating thus releasing into the atmosphere a part of ^{137}Cs contained in the wood products. Systematically higher values appeared during weekends followed by lower or not detectable values during the next working days. The signals were influenced by meteorological conditions and the topography of the area. Although these concentrations do not pose a hazard for the residents of the city, samplings should be performed locally in the areas where biomass is used as main or additional fuel for residential heating. The signals of ^{137}Cs in the atmosphere that were observed for the first time during the first months of 2013 resulted from the increased use of biomass products combustion for residential heating as a consequence of the rising prices of fossil fuels.

According to the preliminary results of this study the wood samples from oak trees contained the lowest ^{137}Cs concentrations measured, <1.3 Bq kg^{-1}, while the concentrations in wood from beech and plane trees ranged up to 11.0 Bq kg^{-1}. The ^{137}Cs concentrations measured in the ashes were up to 500 Bq kg^{-1}. The wood-to-ash enrichment factor ranged from 77 up to 209 for various type of heating devices, according to the results obtained. The release of ^{137}Cs estimated from 41% up to 79% of the wood concentrations corresponds to activities ranged from 0.8 up to 6.5 Bq of ^{137}Cs per kg of wood burned. Although biomass combustion when wood is used for domestic heating is a dominant source of increased ^{137}Cs concentrations in urban environments, a more extensive study is necessary in order to validate the preliminary results obtained.

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