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Activity concentration of ^{137}Cs in undisturbed attic dust collected from Salgótarján and Ózd (northern Hungary)

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ABSTRACT

Due to the Chernobyl nuclear power plant accident, contaminated air masses, containing ^{137}Cs, were widely propagated across all of Europe. Cesium-137 is easily adsorbed on aerosol particles as it returns to the lithosphere/pedosphere/via wet and dry deposition in the form of a radioactive fallout component. Following the nuclear accident, primary attention was paid to agricultural areas and less to urban environments. Our ^{137}Cs activity study using undisturbed attic dust samples has been carried out from two residential areas (city of Salgótarján and Ózd) in northern Hungary, approx. 1000 km away from Chernobyl. A total of 61 attic dust samples were collected in 2016 and 2018 from houses (>30 years) functioning as family house, kindergarten, blockhouse and church. Activity concentration of ^{137}Cs was determined for 1–2 g homogenized (<125 μm) attic dust samples in a low background iron chamber with a well-type HPGe detector. The mean ^{137}Cs activity concentrations in attic dust samples are 88.5 ± 5.1 Bq kg^{-1} and 87.8 ± 4.5 Bq kg^{-1} in Salgótarján and Ózd, respectively. The dependence between ^{137}Cs activities and the age of the houses was found to be significant (p=*0.02*), which could be explained by Chernobyl nuclear accident-causing elevated activity concentrations in location built prior to the accident. Three outliers in Ózd (>223 Bq kg^{-1}), are probably related to the first rainfall event after the Chernobyl accident. Isotopic landscapes (isoscapes) of ^{137}Cs were derived for both cities by means of kriging interpolation. In Salgótarján the ^{137}Cs activity concentrations were higher than in Ózd which might have been due to redistribution loadings and local topographical features. We concluded that components of attic dust are highly useful indicators of home exposure to pollution events and remain detectable after several decades.

1. Introduction

Cesium-137 is a principal artificial radionuclide introduced into the environment through atmospheric nuclear weapon tests from the middle of the 1940s to the 1980s and from major nuclear accidents (Chernobyl, 1986 and Fukushima, 2011). As a result of the Chernobyl nuclear accident, the largest accident, up to 85 PBq of ^{137}Cs activities were released into the atmosphere (UNSCEAR, 2000), notably over vast areas of Europe. The initial ^{137}Cs deposition rate was strongly dependent on meteorological conditions during the first days after the event (1986). A high level of radionuclide fallout deposited mostly over Belarus, North Ukraine, and the European part of Russia (De Cort et al., 1998). Due to the changes of wind direction, the contaminated air masses were initially transported to west and north-west reaching Poland and the Scandinavian countries, but in the following days the wind patterns changed substantially (IAEA, 2006). Therefore, deposition rates from

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the radioactive cloud were not constant during its dispersal, especially considering wet deposition associated with rainfalls at that time (De Cort et al., 1998; Beresford et al., 2016).

In the absence of washout effects, ^{137}Cs attached to airborne particles were transported to large distances and depleted by dry deposition all over the European continent (Balonov et al., 1996; De Cort et al., 1998). In general, both dry and wet depositions are affected by local surface properties, such as aerodynamic roughness determined by local surface topography and land cover (Van der Perk et al., 2002). The ^{137}Cs adhered to aerosol and dust particles were also capable of entering houses (e.g., through open windows, vents and cracks) in residential areas and accumulated inside, and remained undisturbed in areas that are not easily accessible for regular cleaning like attics (Ilacqua et al., 2003). In addition, deposition processes and activities within urban territories contributed to radionuclide redistribution altering atmospheric deposition, particularly over long time periods, i.e., decades.

Numerous studies have examined attic dust samples for determining the levels of toxicants including metal/loid/ trace elements (Cizdziel and Hodge, 2000; Davis and Gulson, 2005; Šajn, 2005; Gosar et al., 2006; Balabanova et al., 2011, 2017; Völgyesi et al., 2014), or polycyclic aromatic hydrocarbon (Coronas et al., 2013; Wheeler et al., 2020). The first paper on ^{137}Cs and ^{239}Pu activity concentrations measured from attic dust was published by Cizdziel et al. (1998), analyzing samples from the surroundings of the Nevada nuclear test site (USA). Subsequently, Ilacqua et al. (2003) concluded that global fallout from nuclear weapon testing was the source of ^{137}Cs deposition in 201 New Jersey homes.

Airborne particles infiltrate residences by diffusion and advection through active and passive ventilation portals and eventually they are deposited. Since attic dusts are less subjected to environmental degradations (e.g., sunlight effects, decomposition by microbial influence, chemical weathering, ventilation, precipitation), they can be preserved in long-terms (Coronas et al., 2013). As such, attic dust is resulted by the long-term dynamic accumulation of material influenced for many years by the natural movement of air, penetration of the dust indoors (resupply), and settling on solid surfaces (Cizdziel and Hodge, 2000). The undisturbed "archive" deposited dust particles can be used to examine regularly the activity of radionuclides following various fallout events and nuclear power plant emissions (Lioy et al., 2002).

After the Chernobyl accident, several European research groups started to study the fate of ^{137}Cs radionuclides in the environment (De Cort et al., 1998). The total activity of ^{137}Cs deposited in Hungary accounted to Chernobyl was 0.15 PBq (De Cort et al., 1998), which is a comparatively low value. As for the geographic distribution, slightly higher ^{137}Cs activities were found in the western parts of Hungary than the other areas based on the 86 field (in situ) gamma-spectrometry measurements (De Cort et al., 1998). Similar findings were reported by Szerbin et al. (1999) for 19 additional measurements. The ^{137}Cs activity concentrations of the undisturbed upper 30 cm of soils in Pest County (Central Hungary) ranged between 0.5 and 61.1 ± 2.2 Bq kg^{-1} (Szabó et al., 2012). As reported by De Cort et al. (1998), airborne gamma radiation surveys were performed between 1963 and 1973 for the European area just prior to the Chernobyl accident, and the estimated total activity of ^{137}Cs was 20 PBq, and the activity concentration

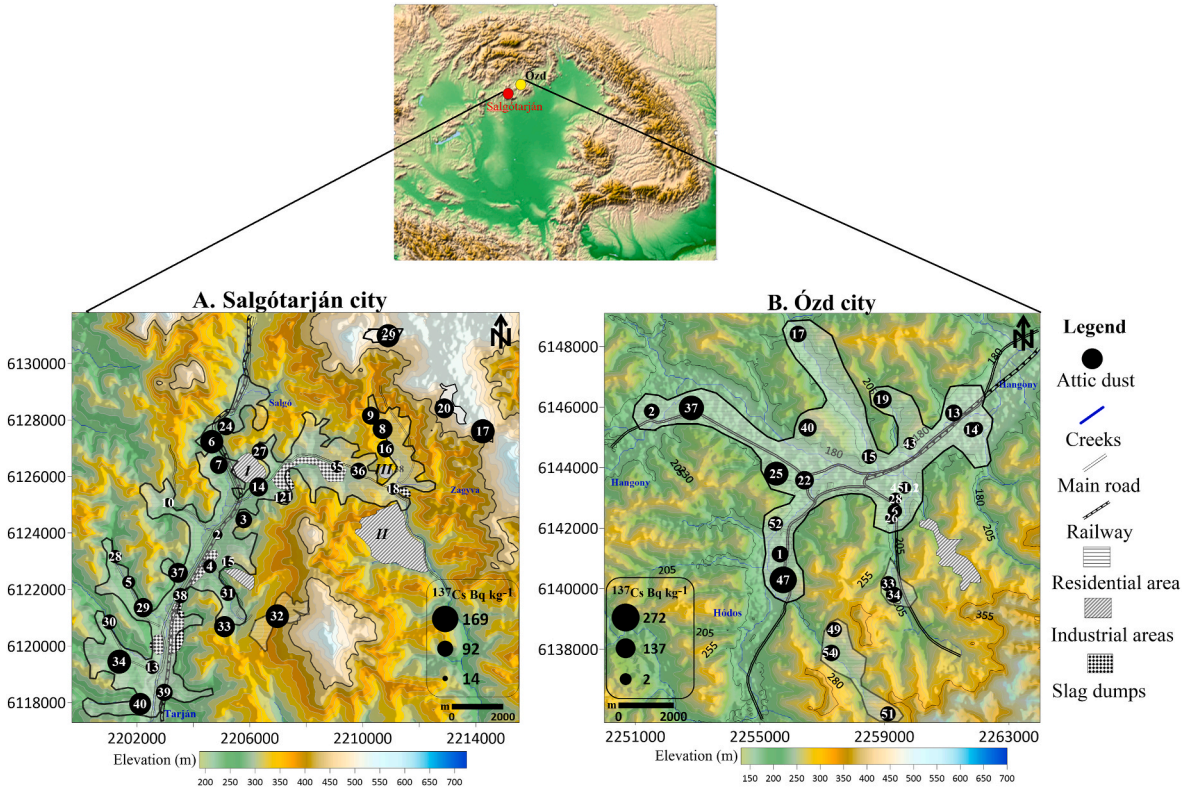

Fig. 1. Maps of the studied urban areas. A. Salgótarján study area with the attic dust sampling sites (n = 36) and location of a coal-fired power plant (Vízválasztó) and slag hills (I – Kucord Hill; II – Inászó; III – Coal ash dump). B. Ózd study area with the attic dust sampling sites (n = 25). The radii of the circles are proportional to the ^{137}Cs activity concentration values (Bq kg^{-1}) at each measurement point. In both cases, the topographic shaded relief model with elevation contour lines are overlaid. Map projections are in EPSG:3857, WGS 84 (Mercator system).

in Hungary it was accounted 2.5–3.0 kBq m^2.

Due to the long half-life ($T_{1/2} = 30.17$ years), as a gamma emitter, its emissions deeply penetrates into biological materials and it can trigger off extensive chemical and genetic damages (Brisban and Dallas, 2008). In addition, ^{137}Cs possesses characteristics enhance its importance as a contributor to the radiation exposure (Ashraf et al., 2014). Therefore, it is worth to study whether attic dust is a concern to public or not. Since the period of fallouts following the Chernobyl disaster, primary attentions were paid on radioactive effects in natural and agricultural ecosystems in Europe, with a much less attention to urban environment.

Our principal goal is to investigate ^{137}Cs accumulation in attic dust samples was performed in Salgótarján and Ózd cities (North Hungary). The main aims of the study were to:

- Determine the ^{137}Cs activity concentrations over the residential areas,
- Facilitate a better understanding of the pathways of ^{137}Cs contamination in the urban environments,
- Clarify whether dust composition can serve as a proxy for historical pollution events,
- To obtain spatial distribution of ^{137}Cs accumulations by means of geostatistical tools.

Our study offers a new perspective and strengthen approaches to studies on attic dust in the future.

2. Material and methods

2.1. Studied urban areas

2.1.1. Salgótarján city

Salgótarján (*48.0853°N, 19.7868°E)* is located at ~220–500 m above sea level in the northern part of Nógrád county, Hungary, with a total area of 103 km^2 and population of ~35,000. The area of the northeastern part of Hungary has diverse hilly and mountainous orography (Fig. 1A). The main geological formations of the Salgótarján study area are Neogene volcanic (effusive and pyroclastic) and different sedimentary (sandstones, marl and brown coal) rocks (Kercsmar et al., 2010). The dominating soil type around the urban area is brown forest soil. The dominant wind direction is north-western. The annual average temperature is around 8–9 °C, and the average annual precipitation is 500–550 mm (Bihari et al., 2018). Salgótarján was a major industrial centre between the 19th and the end of the 20th century. Its significant economic role was based on the local brown coal energy source. For about 150 years, the industrial production of iron and glass wastes (steel and glass slag) and from coal burning (fly-ash, slag, and sludge) have accumulated without any regulation in the city area and outskirts such as Kucord Hill, Pintértelep, Inászó and Vízválasztó (Fig. 1A).

2.1.2. Ózd city

Ózd (*48.2241°N, 20.2889°E)* is located 40 km northeast of Salgótarján, at 150–200 m above sea level in Borsod-Abaúj-Zemplén county, Hungary, with a total area of 92 km^2 and population ~35,000 (Fig. 1B). The main geological formations of the Ózd study area Neogene sedimentary (sandstones, marl and brown coal) and pyroclastic rocks (Kercsmar et al., 2010). Brown forest soil is dominant around residentials. The prevailing wind direction is northern. The region has 8–9 °C average annual temperature and 700–800 mm average annual precipitation (Bihari et al., 2018). The region also played a significant role for iron and steel works between 1835 and 1990 operated in the city centre, whereas coal mines opened around Ózd producing coal for iron and steel factories (Fig. 1B). Currently, a new steel factory is in operation at the northeastern part of Ózd. For about 150 years, the industrial production of iron and steel wastes (smelter slag) have accumulated without any regulation in the city area and outskirts such as slag dumps (Fig. 1B). As a consequence of the collapse of the old political and economic system, the industrial structure, in both cities, has been restructured in the early 1990s, coal mines and heavy industry have been abandoned, but large amounts of industrial waste have been remained in the whole area.

2.2. Sampling strategy

The collection of attic dust samples followed the Euro-Geo-Surveys international urban geochemical sampling protocol (Demetriades and Birke, 2015). Although this protocol has been used as a norm for urban geochemical sampling, we adapted some changes in this protocol. The methodology is designed for sampling inorganic stable chemical elements and organic chemical compounds, but we studied a radioactive nuclide, ^{137}Cs. Also, previous experiences were taken into account (Cizdziel et al., 1998; Ilacqua et al., 2003; Šajn, 2005; Völgyesi et al., 2014). The sampling strategy followed a grid-based stratified sampling design, where the total area was covered by 1×1 km^2 grid cells.

From each grid cell one sample was collected. Altogether 36 and 25 buildings were sampled in Salgótarján and Ózd, respectively. A 16% duplicate sampling was applied to ensure the reproducibility of the sampling method. Relative differences between the ^{137}Cs activity concentrations of the duplicates were less than 10% as measured by the root mean square error, which is acceptable for these samples. Sampling was carried out in 2016 for Salgótarján and in 2018 for Ózd after the required local authorizations permissions were obtained.

2.3. Sampling material: attic dust

Attic dust was collected from 36 buildings in the residential area of Salgótarján, 27 taken from family houses, 4 from churches, 3 from kindergartens and 2 from blockhouses. These buildings were built between 1890 and 1989 (Table 1). From Ózd, 25 attic dust samples were collected, 11 from family houses, 5 from churches, and 1 from a kindergarten and 8 from blockhouses (Table 2). These buildings were constructed between 1790 and 1971. Attic dust samples were collected far from the attic entrance and at the best possible point of the ceiling to minimize possible disturbing effects of resident activities (Völgyesi et al., 2014). Attic floor was not sampled and organic materials, such as insects, feces, etc., were eliminated during the sampling. Two to 20 g of dusts, composed of 3–5 sub-samples, depending on attic condition were collected into polyethylene bags, using disposable fine brushes and plastic gloves. To avoid cross-contamination, new fine brush and plastic gloves were used to collect each sample, including duplicate samples. Notably, in one building within the territory of the former iron factory (in Ózd) we observed a two-layered attic dust accumulation, where we did a layered sample collection based on different appearance in color (Fig. S1), upper layer (OZD_AD45UL) and bottom layer (OZD_AD45LL; Fig. S1). Accordingly, both layers were collected using different brushes and gloves. During the sampling process, photographic documentation, geographic coordinates, and building characteristics such as the year of built and major reconstruction, roof material, function of the building (e.g., family house, church), construction material, slope of the ground below the building and elevation (construction height was added) are recorded (Tables 1 and 2). Residents were interviewed about the development history and renovations to ensure undisturbed samples.

Table 1
The ^{137}Cs activity concentration (Bq kg^{-1}) of the attic dust (AD) samples and the characteristics of the sampling locations in Salgótarján urban area (STN). The projection are coordinates in EPSG:3857, WGS 84 (Mercator system).

Sample ID	Coordinates of the sampling locations (X, Y), in meter		Year of built (renovation)	Function of the building	Roof material	Construction material	Slope[b] (°)	Terrain aspect (°)	Elevation (m) a. s. l.[a]	^{137}Cs activity concentration (Bq kg^{-1})	Mean values, per building
STN AD1	2205477	6124930	1936	Church	Tin	Mixed with stone, bricks and wood	4.5	316	310	15.9 ± 1.1	59.5
STN AD2	2204815	6123910	1914	Church	Tile	Bricks	3.6	278	324	13.0 ± 1.3	
STN AD25	2210931	6130914	1930	Church	Tin	Bricks	1.7	265	492	159.6 ± 8.4	
STN AD28	2201138	6123143	1934 (2013)	Church	Slate (red)	Wood and bricks	1.8	209	275	51.0 ± 3.3	
STN AD3	2205749	6124438	1957	Family house	Tile	Bricks	2.3	186	309	91.7 ± 6.0	94.2
STN AD6	2204602	6127185	1960	Family house	Tile	Blocks	2.2	276	267	162.3 ± 8.8	
STN AD7	2204855	6126365	1944	Family house	Tile	Blocks (slag and stone included)	7.9	92	273	104.1 ± 6.2	
STN AD9	2210308	6128102	1961	Family house	Slate	Adobe	3.8	77	349	101.5 ± 5.6	
STN AD10	2203028	6125036	1980	Family house	Slate (red)	Blocks	1.5	264	287	5.5 ± 0.9	
STN AD12	2207059	6125216	1890	Family house	Slate	Mixed with wood, cement and bricks	9.4	17	307	35.4 ± 2.5	
STN AD13	2202467	6119246	1965	Family house	Slate	Bricks	1.1	35	233	54.8 ± 3.4	
STN AD16	2210836	6126935	1970	Family house	Slate	Blocks	1.3	120	337	93.9 ± 5.3	
STN AD17	2214330	6127551	1922	Family house	Tile	Wood	8.3	182	495	169.8 ± 8.8	
STN AD18	2211109	6125522	1970	Family house	Slate	Bricks	3.5	279	314	35.7 ± 2.2	
STN AD20	2212946	6128359	1960	Family house	Slate and tile	Adobe	7.8	226	507	123.4 ± 7.6	
STN AD21	2207291	6125248	1916	Family house	Tile	Bricks	6.5	341	305	41.8 ± 2.6	
STN AD24	2205113	6127722	1970	Family house	Slate	Blocks	1.0	205	276	100.5 ± 5.6	
STN AD26	2210963	6131022	1920	Family house	Tin	Bricks	3.0	261	496	41.3 ± 2.2	
STN AD27	2206333	6126840	1940	Family house	Tile	Adobe	2.5	90	331	90.3 ± 5.2	
STN AD29	2202141	6121341	1936	Family house	Tile	Bricks	1.4	203	245	117.8 ± 6.6	
STN AD30	2200930	6120839	1950	Family house	Slate	Wood and bricks	4.5	80	253	70.7 ± 4.5	
STN AD31	2205184	6121852	1910	Family house	Slate (wood inside)	Blocks	2.8	56	267	77.1 ± 5.1	
STN AD32	2206954	6121042	1900	Family house	Tile	Bricks	5.6	284	412	165.8 ± 8.9	
STN AD33	2205060	6120675	1955	Family house	Tile	Bricks	6.2	74	299	137.8 ± 7.9	
STN AD34	2201280	6119441	1936	Family house	Tile	Adobe	7.8	303	261	169.3 ± 9.4	
STN AD35	2209100	6126310	1946	Family house	Slate (red)	Bricks	5.3	213	306	41.6 ± 2.5	
STN AD36	2209873	6126154	1940	Family house	Tile	Bricks and blocks	2.8	254	306	87.3 ± 5.0	
STN AD37	2203389	6122568	1961	Family house	Tile	Bricks	3.9	66	253	124.7 ± 6.5	
STN AD38	2203483	6121767	1880 (1984)	Family house	Tile and wood	Bricks	3.9	284	245	73.9 ± 4.3	
STN AD39	2202896	6118359	1910	Family house	Tile	Adobe	4.9	301	237	93.6 ± 5.0	
STN AD40	2202031	6117952	1964 (1991)	Family house	Tile	Blocks	2.7	71	230	149.0 ± 7.7	
STN AD15	2205184	6122957	1965 (2006)	Blockhouse	Tile (iron structured)	Bricks	6.8	212	308	36.7 ± 2.7	75.5
STN AD14	2206280	6125594	1950	Blockhouse	Slate	Concrete and wood	3.0	177	275	115.8 ± 6.7	
STN AD5	2201621	6122238	1990	Kindergarten	Tile	Tile	1.0	176	254	49.6 ± 3.1	77.3
STN AD4	2204534	6122799	1980	Kindergarten	Slate and tile	Cement and bricks	1.2	78	270	57.8 ± 3.1	
STN AD8	2210734	6127634	1960	Kindergarten	Slate	Wood	5.0	103	351	126.5 ± 7.1	

[a] The height of the building (m) is added.
[b] Slope of the ground below the building.

Table 2
The ^{137}Cs activity concentration (Bq kg^{-1}) of the attic dust (AD) samples and the characteristics of the sampling locations in Ózd urban area (OZD). The projection are coordinates in EPSG:3857, WGS 84 (Mercator system).

Sample ID	Coordinates of the sampling locations (X, Y), in meter		Year of built	Function of the building	Roof material	Construction material	Slope[b](°)	Terrain aspect (°)	Elevation (m) a. s. l.[a]	^{137}Cs activity concentration (Bq kg^{-1})	Mean values per building
OZD AD1	2255695	6141110	1897	Priest house	Tin	Bricks	7.2	297	179	93.3 ± 4.7	
OZD AD2	2251528	6145835	1950	Priest house	Slate	Bricks	1.1	97	180	74.1 ± 3.8	
OZD AD6	2259419	6142538	1903	Church	Slate	Bricks	1.1	163	192	51.3 ± 2.7	
OZD AD8	2262064	6145264	1790	Priest house	Tile	Bricks	5.6	141	179	39.7 ± 2.0	
OZD AD54	2257283	6137854	1945	Church	Slate	Cements	0.9	173	215	38.4 ± 2.6	
OZD AD17	2256280	6148391	1903	Family house	Tile	Bricks	1.6	235	174	95.1 ± 5.0	111.7
OZD AD19	2259019	6146228	1957	Family house	Tile	Bricks and concrete	2.0	158	186	120.9 ± 5.9	
OZD AD33	2259208	6140130	1950	Family house	Slate	Bricks	3.8	47	197	74.9 ± 3.8	
OZD AD34	2259394	6139765	1930	Family house	Tile	Bricks	7.2	54	205	85.5 ± 4.3	
OZD AD37	2252814	6145944	1970	Family house	Tile	Bricks	2.8	260	197	238.6 ± 11.5	
OZD AD40	2256587	6145293	1971	Family house	Tile	Bricks	2.5	199	197	116.7 ± 5.7	
OZD AD43	2259908	6144774	1940	Family house	Tile	Bricks	4.0	117	165	25.5 ± 1.6	
OZD AD47	2255796	6140258	1978	Family house	Slate	Bricks	5.5	262	198	272.8 ± 13.0	
OZD AD49	2257449	6138615	1978	Family house	Tile	Bricks	5.1	146	266	71.4 ± 3.8	
OZD AD51	2259197	6135827	1920	Family house	Tile	Bricks	3.7	214	212	71.0 ± 3.9	
OZD AD52	2255547	6142116	1940	Family house	Tile	Bricks	7.2	127	180	62.1 ± 3.4	
OZDAD45UL	2259746	6143306	1878	Blockhouse	Tin	Bricks	3.2	333	187	1.9 ± 1.0[c]	25.7
OZD AD45LL	2259746	6143306	1878	Blockhouse	Tin	Bricks	3.2	333	187	16.0 ± 1.4	
OZD AD13	2261332	6145778	1920	Blockhouse	Tile	Bricks and adobe	2.3	106	165	100.8 ± 5.2	
OZD AD14	2261910	6145226	1874	Blockhouse	Tile	Adobe	3.1	106	170	72.0 ± 3.6	
OZD AD15	2258591	6144336	1957	Blockhouse	Tile	Concrete	3.1	228	135	62.8 ± 3.3	
OZD AD22	2256488	6143552	1951	Blockhouse	Tile	Bricks	1.2	183	187	129.1 ± 6.5	
OZD AD25	2255570	6143772	1954	Blockhouse	Tile	Bricks	4.0	270	203	223.4 ± 6.5	
OZD AD26	2259310	6142306	1926	Blockhouse	Slate	Bricks	3.4	193	184	21.5 ± 1.4	
OZD AD28	2259444	6142935	1945	Blockhouse	Slate	Bricks	1.5	227	184	28.3 ± 1.5	
OZD AD20	2257365	6137842	1890	Kindergarten	Wood and tile	Bricks	3.1	265	202	93.3 ± 4.8	93.3

[a] - The height of the building (m) is added.
[b] Slope of the ground below the building.
[c] Limit of Detection (LOD) is 1.3 Bq kg^{-1}.

2.4. Sample preparation

The attic dust samples were sieved through 125 μm to remove residuals of insects, plants, feces, wood, debris of roof, etc. After aliquot homogenized subsamples were selected for gamma spectrometry measurements, 1–2 g of samples were stored in plastic vials (test tube). Nikon stereo microscope installed in Lithosphere Fluid Research Lab, Eötvös Loránd University, Budapest, was used to observe morphological features of grains which are commonly occur in our samples. Two types of dominated grains, geogenic quartz and anthropogenic slag, were identified.

2.5. Gamma spectrometry measurement

Activity concentration of ^{137}Cs in attic dust samples was measured with a well-type HPGe detector (Type: Canberra GCW 6023, relative efficiency is 60%, resolution is 1.5 keV at 122 keV and 2.3 keV at 1332.5 keV, cryostat well diameter is 16 mm, well depth is 40 mm) in a low-background iron chamber (background is 1.1 cps over the energy range of 7 keV–3.3 MeV) at the laboratory of Nuclear Security Department, Centre for Energy Research. The detector efficiency is 0.22 at 661.66 keV (^{137}Cs gamma line). The well-type detector is ideal for small amounts of environmental samples as it combines both low background and high detection efficiency (Laborie et al., 2000). Attic dust samples stored in plastic vials were placed in the well of the detector. The measurements were performed over different test times (from 200,000 to 600,000 s) to obtain sufficient statistics (error of the net area error is less than 5%). Background ^{137}Cs activity concentration was measured by putting an empty plastic vial into the well. Spectrum analysis was carried out using Fitz-peaks (Version 3.71) gamma analysis software. The ^{137}Cs activity concentration was decay corrected in the sampling year 2016 (Salgótarján) and 2018 (Ózd).

2.6. Meteorological data evaluation

Meteorological data evaluations were based on the ERA5 reanalysis records of the European Centre for Medium-Range Weather Forecasts (ECMWF)[1]. The temporal period ranged from 1 January 1979 to 1 July 2018. The data sets cover the north-eastern (NE) part of Hungary (47.0°N - 49.0°N, 19.0°E − 21.0°E) with a spatial resolution of about 8 × 8 km (geographic grid 0.125° × 0.125°) and a time resolution of 3 h. All computations were performed using Jupyter in Python 3.7 with standard modules.

2.7. Statistical analyses

2.7.1. Regression analysis

Regression analysis is a common statistical procedure used to determine the relationship between an independent- and one or more dependent variable(s). Ordinary least squares (OLS; Goldberger, 1964) and robust regression were considered to be applied to the data. Former is known to be sensitive to statistical outliers (Yu and Yao, 2017), whereas latter is capable of providing more reliable results by limiting the weight of outlier based on comparative studies (R.Wilcox and Keselman, 2012; Yu and Yao, 2017; Kalina and Tichavský, 2020). Therefore, robust MM (maximum likelihood method) estimation was applied when assessing the house ages vs. ^{137}Cs activity concentration with rlm function of the MASS package in R. However, when exploring the altitude effect, OLS regression was sufficient due to the lack of outliers.

2.7.2. Geostatistical analysis

In the first step, samples were grouped by the date when the house was built. Only those samples were considered which originated from houses constructed prior to 1945 (Tables 1 and 2), thus the attic dust are assumed to archive all the anthropogenic activities that might have caused ^{137}Cs fallout. As a result, 18 and 17 attic dusts samples were taken into account from Salgótarján and Ózd cities, respectively. The elevation (m, above sea level; a.s.l.) of the houses was extracted from a digital elevation model with a 30 * 30 m grid-size, corrected by the heights of the attics (Tables 1 and 2). The corrected elevation is referred as ELE* hereinafter.

The spatial autocorrelation of ^{137}Cs activity concentration was determined with the basic function of geostatistics the semivariogram, which was then used as the weighting function in kriging (Cressie, 1990) used to interpolate the isotopic landscapes (isoscapes) of ^{137}Cs activity concentration values. The empirical semivariograms were obtained with the Matheron and Marie (1965) algorithm (Eq. 1):

$$\gamma(h) = \frac{1}{2N(h)} \sum_{i=1}^{N(h)} [Z(x_i) - Z(x_{i+h})]^2 \quad \text{Eq. 1}$$

where $\gamma(h)$ represents the semivariance, $N(h)$ indicates the number of sample point pairs with a distance h, $Z(x_i)$ indicates the measured value at observation site i, and $Z(x_i + h)$ is the measured value at observation site $i + h$.

Next, theoretical semivariograms were fitted to the empirical ones using least squares fitting. Kriging is one of the best interpolation algorithms in providing prediction for radionuclide fallout (e.g., Mabit and Bernard, 2007). One of the most important characteristics of the semivariogram is its spatial range where the samples become uncorrelated (Chilés and Delfiner, 2012). In fact, the obtained range is usually attributed to environmental processes that act on the same scale (e.g., Hatvani et al., 2017, 2020) Note that reported ranges in the study are planar distances in km (EPSG: 3857) and the fit statistics are reported along with the semivariograms.

The interpolation of the ^{137}Cs values was done with ordinary point kriging (Cressie, 1990). As a preprocessing step, semivariogram clouds were utilized to search for outlying values, thus discarding samples were STN_AD26 and OZD_AD54. All data processing and calculations, geostatistical data interpretation and visualization have been performed using Golden Software Surfer 15 and GS +10.

3. Results

3.1. The ^{137}Cs activity concentration in Salgótarján city

The ^{137}Cs activity concentration of 36 attic dust samples from Salgótarján together with characteristics of the sampling locations are shown in Table 1. The obtained ^{137}Cs activity concentrations of attic dust samples ranged from 5.5 ± 0.9 Bq kg^{-1} to 169.8 ± 8.8 Bq kg^{-1} with arithmetic mean of 88.5 ± 5.1 Bq kg^{-1}, and standard deviation of 48.0 Bq kg^{-1}. The ^{137}Cs activity concentrations, according to the function of the buildings, ranged in family houses ($n = 27$) from 5.5 ± 0.9 to 169.8 ± 8.8 Bq kg^{-1} built between 1880 and 1990 without renovations, in churches ($n = 4$) from 13.0 ± 1.3 to 159.6 ± 8.4 Bq kg^{-1} built between 1914 and 1936 (STN_AD28 ceiling tiles were reconstructed in 2013, see Table 1), in kindergartens ($n = 3$) from 49.6 ± 3.1 to 126.5 ± 7.1 Bq kg^{-1} built between 1960 and 1990, without renovations and in blockhouses ($n = 2$) from 36.7 ± 2.7 and 115.8 ± 6.7 Bq kg^{-1}, built between 1950 and 1965 (STN_AD15 attic area fully renovated in 2006, see Table 1). Considering roof type of the sampled houses, most roofs are tiles ($n = 19$) and slate ($n = 13$) with a small number of combined slate/tile ($n = 2$) and tin roofs ($n = 2$), see Table 1.

3.2. The ^{137}Cs activity concentration in Ózd city

The ^{137}Cs activity concentration of 25 attic dust samples from Ózd and the characteristics of the sampling locations are shown in Table 2.

[1] https://www.ecmwf.int/

The ^{137}Cs activity concentration in attic dust samples from Ózd ranged from 1.9 ± 1 Bq kg^{-1} to 272.8 ± 13.0 Bq kg^{-1}, with an arithmetic mean of 87.8 ± 4.5 Bq kg^{-1} and standard deviation 65.7 Bq kg^{-1}. Regarding to the different functions of the buildings, ^{137}Cs activity concentrations range from 25.5 ± 1.6 Bq kg^{1} to 272.8 ± 13.0 Bq kg^{-1}in family houses (n = 11), (built in 1903–1971), without renovations; from 38.4 ± 2.6 Bq kg^{-1} to 93.3 ± 4.7 Bq kg^{-1}in churches (n = 5), (built in 1790–1950), without renovations; from 93.3 ± 4.8 Bq kg^{-1} in a kindergarten (n = 1), (built in 1890), without renovations and from 1.9 ± 1.0 Bq kg^{-1} to 223.4 ± 6.5 Bq kg^{-1}in blockhouses (n = 9), (built in 1874–1954), without no renovations.

Three outlying values were found: OZD_AD25 (blockhouse) 223.4 ± 6.5 Bq kg^{-1}, OZD_AD37 (family house) 238.6 ± 11.5 Bq kg^{-1} and OZD_AD47 (family house) 272.8 ± 13.0 Bq kg^{-1} from the southern and western parts of the urban area (Fig. 1 and Table 2). In Ózd, the majority of the attic roof was tile (n = 16) following by slate (n = 7) and tin (n = 3), see Table 2. At two-layered attic dust sampling site (blockhouse) (Fig. S1). The upper layer (OZD_AD45UL) contains lower ^{137}Cs activity concentration 1.9 ± 1.0 Bq kg^{1} than the bottom layer (OZD_AD45LL) 16.0 ± 1.4 Bq kg^{-1} (Fig. S1, Table 2).

4. Discussion

4.1. Comparison (world-wide samples from different environments)

The ^{137}Cs activity concentrations (Bq kg^{-1}) found in Salgótarján and Ózd attic dusts are compared with those measured in the USA (Table 3). Unfortunately, there is no information about ^{137}Cs in attic dust in the other continents. Therefore, our results can be compared with only these two studies (Cizdziel et al., 1998; Ilacqua et al., 2003). The arithmetic average of ^{137}Cs activity levels in attic dust (STN 88.5 ± 5.1 Bq kg^{-1} and OZD 87.8 ± 4.5 Bq kg^{-1}) are comparable with previous findings in Dover and New Jersey (Cizdziel et al., 1998) with arithmetic mean of 42 ± 28 Bq kg^{-1}. Arithmetic mean values are the same (within errors) as at the Nevada Test sites (NTS) including areas in Utah (Ilacqua et al., 2003), 75 ± 53 Bq kg^{-1}. However, in Europe the total ^{137}Cs activity from the Chernobyl accident was estimated around 85 PBq, whereas the global mean fallout was considerable lower at around 20 PBq (De Cort et al., 1998). These data clearly suggest that most of ^{137}Cs contribution to Europe has originated from the Chernobyl accident.

Since measured ^{137}Cs activity concentrations in attic dust samples are globally scarce, present results are also compared to soil samples from Hungary and surrounded countries (Table 3). The studied soils from central Hungary (Szabó et al., 2012) and northern part of Nógrád and Borsod-Abaúj-Zemplén counties Hungary (Szerbin et al., 1999) have lower ^{137}Cs activities (0–61.1 Bq kg^{-1}; 1–47 and 2–60 Bq kg^{-1}, respectively) than the attic dust (Table 3). A possible explanation is that the uppermost layer of soils can be disturbed by anthropogenic activities (building and road constructions, greening the urban environment with soil changes, etc.) or by natural physical and chemical weathering processes, thus soil cannot preserve the past atmospheric ^{137}Cs pollution such efficiently as attic dust.

Comparing our attic dust ^{137}Cs results with that of soils from some European countries such as Spain (Navas et al., 2011); Macedonia (Dimovska et al., 2011); Greece (Vosniakos, 2012); Lithuania (Lukšienė et al., 2015); Serbia (Milenkovic et al., 2015; Vukašinović et al., 2018), Montenegro (Antovic et al., 2012), we obtained higher average values (Table 3). However, in Austria and Romania the studied soils show elevated ^{137}Cs activity (over >200 kBq m^{2} and from 6.6 to 2316.1 Bq kg^{-1}, respectively) (Bossew et al., 2001; Begy et al., 2017) than the Hungarian attic dusts, indicating a positive correlation with high annual rainfall at mountainous (i.e., Alps, Carpathians, Dinarides) areas as Begy et al. (2017) suggested.

Although measured records of ^{137}Cs activity concentration in urban areas, particularly in attic dust, are globally scarce, we assume that ^{137}Cs records in attic dust might support an approximate estimate of atmospheric deposition. It is more than interesting that a reported ^{137}Cs arithmetic mean of urban puddle sediment from Ekaterinburg (Ural region, Russia) is 80 Bqkg^{-1} (Seleznev et al., 2010). These authors proved that puddle sediment apparently traps ^{137}Cs total accumulation of activity (Table 3). This average ^{137}Cs activity value is consistent with

Table 3
Comparison of ^{137}Cs activity concentration (Bq kg^{-1}) in different environmental samples (attic dust, urban soil, sediment and moss) of different countries deposited after Chernobyl nuclear power plant accident.

Country	Type of samples (Particle size, mm)	^{137}Cs activity concentration (Bq kg^{-1}), average (range)	References
Attic dust samples			
Salgótarján city, Hungary	Attic dust n = 36 (undisturbed); <0.125 mm	88.5 ± 5.1 Bq kg^{-1} (5.5–169.9 Bq kg^{-1})	*Present study*
Ózd city, Hungary	Attic dust n = 25 (undisturbed); <0.125 mm	87.8 ± 4.5 Bq kg^{-1} (16.0–272.9 Bq kg^{-1})	*Present study*
Nevada and Utah, USA	Attic dust n = 14 (undisturbed); <2.36 mm	42 ± 28 Bq kg^{-1} (6–105 Bq kg^{-1})	Cizdziel et al. (1998)
New Jersey, USA	Attic dust n = 201 (undisturbed); NA	75 ± 53 Bq kg^{-1} (14–286 Bq kg^{-1})	Ilacqua et al. (2003)
Soil samples			
Pest county, Central Hungary	Soil samples, n = 45 (0–30 cm, undisturbed); <2 mm	9.5 Bq kg^{-1} (0–61.1 Bq kg^{-1})	Szabó et al. (2012)
19 Hungarian counties, across the whole Hungary	Soil samples, n = 19 (0–20 cm, disturbed, uncultivated); <1.25 mm	1-47 Bq kg^{-1} (Nógrád county); 2–60 Bq kg-1 (Borsod-Abaúj-Zemplén county)	Szerbin et al. (1999)
Across the other countries	**Soil and sediments samples**		
Austria	Soil samples n = 2115 (measurements); NA	18.7 kBq/m^{2} (>200 kBq/m^{2})	Bossew et al. (2001)
Montenegro	Soil samples n = 24 (0–5; 5–10; 10–15 cm, uncultivated); <2 mm	65.3 Bq kg^{-1} (1.82–413 Bqkg^{-1}) 46 Bq kg^{-1} (1.9–141 Bq kg^{-1}) 36 Bq kg^{-1} (2–112 Bq kg^{-1})	*Antovic* et al. *(2012)*
Belgrade capital city, Serbia	Urban soils n = 22 (0–20 cm); <2 mm	17 (0.7–35 Bq kg^{-1})	Vukašinović et al. (2018)
Kragujevac city, Serbia	Soil samples n = 30 (0–10 cm, undisturbed); <2 mm	40.2 Bq kg^{-1} (0.5–90.5 Bq kg^{-1})	*Milenkovic* et al. *(2015)*
Kavadarci city, Macedonia	Urban soil samples n = 45 (0–5 cm); <2 mm	41.5 Bq kg^{-1} (4–220 Bq kg^{-1})	*Dimovska* et al. *(2011)*
Lithuanian soil	Open meadow soil n = 24 (0–10 cm, undisturbed); <0.2 mm	5.04 Bq kg^{-1} (2–16.6 Bq kg^{-1})	*Lukšienė* et al. *(2015)*
Transylvania region (Transylvanian Plateau and the Western Plain and Hills)	Soil samples (undisturbed) n = 153 (0–20 cm); <0.1 mm	177.3 Bq kg^{-1}(6.6–2316.1 Bq kg^{-1})	Begy et al. (2017)
Catchment in the Pyrenees, Spain	Soil samples (0–20 cm); <2 mm	30.9 Bq kg^{-1} (4.4–64.7 Bq kg^{-1})	Navas et al. (2011)
All over Greece	Soil samples, n = 780 (0–5 cm, undisturbed); <2 mm	23.1–51.1 Bq kg^{-1}	Vosniakos (2012)
Ekaterinburg city, Ural region, Russia	Puddle sediment (upper 0–5 cm); NA	80 Bq kg^{-1} (0–540 Bq kg^{-1})	Seleznev et al. (2010)

our estimated ^{137}Cs averages in Salgótarján (88.5 ± 5.1 Bq kg^{-1}) and Ózd (87.8 ± 4.5 Bq kg^{-1}) urban areas (Table 3) suggesting also a long-term accumulation of ^{137}Cs in attic dust.

The layered attic dust sample from Ózd shows higher ^{137}Cs activities in bottom layer (OZD_AD45LL; 16.0 ± 1.4 Bq kg^{-1}) than in the upper layer (OZD_AD45UL; 1.9 ± 1.0 Bq kg^{-1}) (Table 2; Fig. S1). The sampled building was the repair hall of the former steel factory more than a century, which activity is assumed to provide the bottom layer. Later it was operating as a small horsebox iron factory during the past ~25 years. This activity also produced and released dust to form the upper layer, which show lower variability in heavy metal and metalloids than the lower layer (Salazar et al., 2021). This observation might indicate that effects of heavy metals/loids to decrease concentrations of ^{137}Cs, which is supported by Outola et al. (2003) in a Finnish heavy metal industrial area.

4.2. Age of house versus ^{137}Cs activity concentrations

Activity concentrations of ^{137}Cs in all studied attic dust samples from Salgótarján and Ózd seem to gradually decrease towards the older buildings (Fig. 2). The robust regression (MM-estimator, Kalina and Tichavský., 2020) between ^{137}Cs and age of buildings is statistically significant: ($r^2 = 0.05$, $p = 0.025$). In houses, built after 1950 (beginning of nuclear test), approx. 50% of total attic dust samples have wide range (from 272.8 ± 13.0 to 5.5 ± 0.9 Bq kg $^{-1}$) and high ^{137}Cs average activity concentrations (108 Bq kg $^{-1}$). Whereas attic dust, collected in houses constructed before 1950, show narrower range (from 169.8 ± 8.8 to 1.9 ± 1.0 Bq kg^{1}) and lower average activity concentration (71.9 Bq kg $^{-1}$; Tables 1 and 2, Fig. 2). Previous study in New Jersey (Ilacqua et al., 2003) has found elevated mean ^{137}Cs activity concentrations in older houses, built between 1955 and 1965, compared with younger constructions illustrated in Fig. 2. Our finding (i.e., higher ^{137}Cs activities found houses built after 1950) is consistent with results of Ilacqua et al. (2003) (Fig. 2). However, pattern of the three outlier samples, with the highest ^{137}Cs values from Ózd (OZD_AD47, 37 and 25), shows a clear opposite trend: most elevated values derive from younger buildings (Fig. 2). This cannot be explained easily, some unknown mechanism(s) resulting in ^{137}Cs enrichment seem to be necessary or perfect isolations might be occurred.

The remaining ~50% of total attic dust samples collected from houses built before 1950 show a slightly decreasing tendency in activity concentration with increasing age of houses (Tables 1 and 2, Fig. 2). This trend indicates that ^{137}Cs activity concentration drops back to background values as 0 if there is no additional source to create overprint it. However, the elevated ^{137}Cs levels from four Salgótarján houses (STN_AD17, 25, 32 and 34) are noticeably higher than the tendency line. This variability in our data could be explained by further accumulation process(es). Three sampling sites (STN_AD17, 25, 32) of these four are situated at the highest elevation of the study area (>410 m; Fig. 1A and Table 1), therefore probably local topography played significant role in accumulation. On the other hand, all of the other houses with average values below 59.2 Bq kg^{-1} can be explained by regular cleaning of attic areas, complete renovations as houses getting older, or an almost airtight isolation. ^{137}Cs activities in houses built before ~1950 is assumed to have a long-term initial deposition in both cities which will be readdressed in the geostatistical analysis.

The two-layered attic dust sampling site (OZD_AD45, former repair hall as discussed above) was built far before 1950 and show also very low ^{137}Cs activity value in the bottom layer (OZD_AD45LL, 16.0 ± 1.4 Bq kg^{-1}; Table 2, Fig. 2). The upper dust layer accumulated during the past ~25 years and it also exhibits the lowest activity (OZD_AD45UL, 1.9 ± 1.0 Bq kg^{1}). It cannot be excluded that the upper layer at its bottom contains certain quantity from older lower layer by reaccumulations and source of ^{137}Cs was not available in the local environment.

4.3. Dust deposition processes

Dry and wet deposition processes are important pathways of the accumulation of radioactive pollutants on the surface (De Cort et al., 1998). To get a hint of the importance of meteorological conditions, we evaluated surface wind direction (Fig. S2), wind speed and precipitation records from the ERA reanalysis from 1979 to 2017 (see Fig. S2). The prevailing wind direction was North to South throughout the Salgótarján and Ózd study areas during the last 4 decades, while wind speed did not exceed 2.6 m s^{-1} in Salgótarján and 2.5 m s^{-1} in Ózd urban areas. However, within the analyzed area (spatial resolution of 0.125° × 0.125°) including both cities, wind speed varied between 0.002 and 14.77 m s^{-1} (Fig. S2).

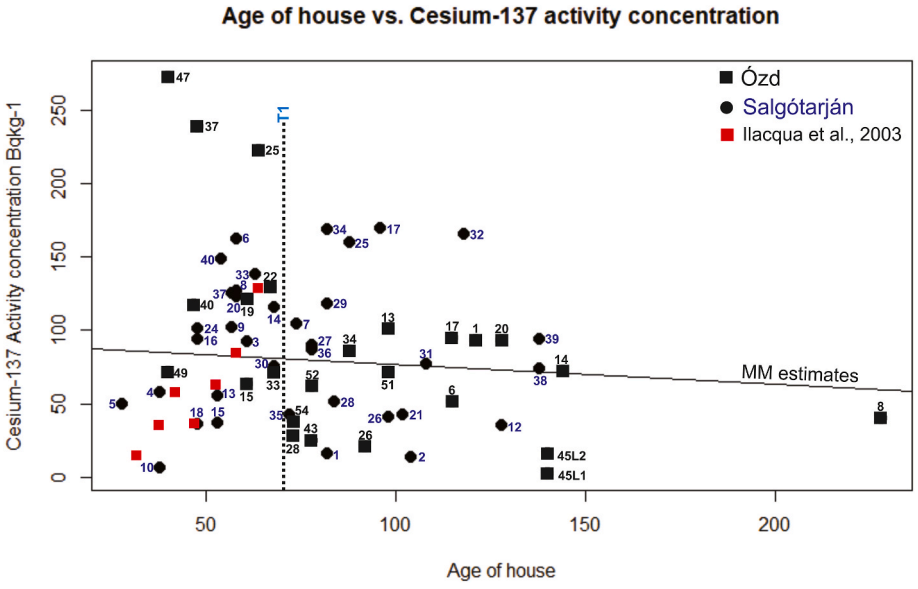

Fig. 2. Robust relationship (MM estimates) between ^{137}Cs activity concentration of labelled attic dusts and age of houses in both studied cities (Salgótarján and Ózd) (Tables 1 and 2). Vertical dotted and solid lines define time marks (T1 = 1950). The red square plot displayed ^{137}Cs activity concentration in attic dust from Dover, New Jersey (Ilacqua et al., 2003). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

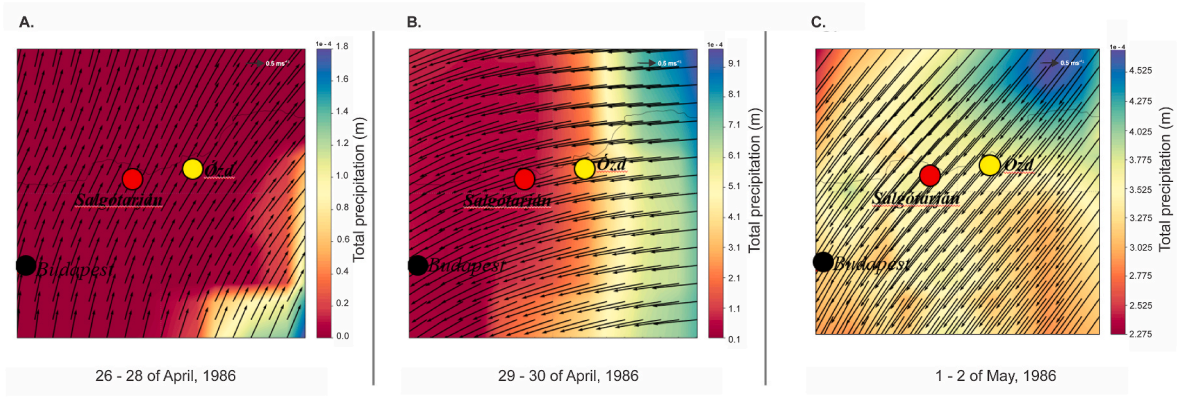

Fig. 3. Seven days interval total precipitation (m) versus wind direction (↑), from 26th of April 1986, 12:00 (CET). Total precipitation and wind direction of time intervals A. 26–28th of April 1986; B. 29–30th of April 1986; C. 1–2 of May 1986.

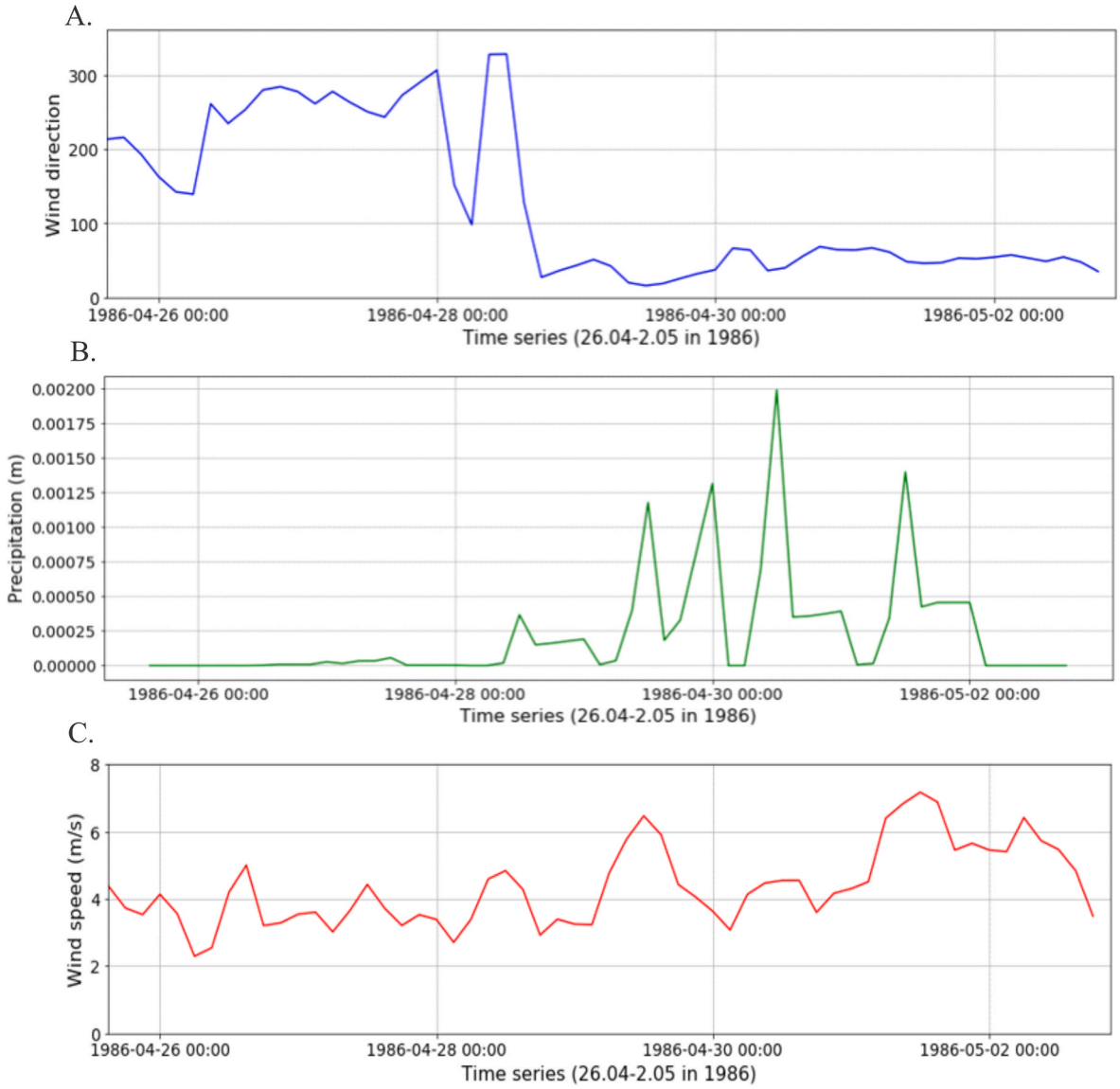

Fig. 4. Meteorological data during the Chernobyl accident period (26th of April 1986 to 2nd of May) at the study areas. A. Wind direction (blue); B. precipitation (unit: m/1000; green); C. wind speed (unit: m/s; red) are shown. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

After the Chernobyl accident (1986), ^{137}Cs contaminated particles distributed non-uniformly and the dispersal was controlled primarily by winds and precipitations. The major part of the initial emission dispersed at relatively high altitudes (Brandt et al., 2002), and the initial transport of the radioactive species was directed towards north-west up to Sweden and Finland. In the following few days, the contaminations were distributed over most of Europe with major exposures in southern, eastern and central Europe (De Cort et al., 1998).

Our analyses (Fig. 3A and Fig. 4A) illustrate that during a two-day interval (26th - 28th of April, 1986), surface wind direction was from south to north and turned to northeast, while no precipitation occurred. However, from 29th to 30th of April (Figs. 3B and 4B) the wind direction changed from east to west, meanwhile precipitation appeared. It is clearly demonstrated that Ózd urban environment received the first rainfall during this day (Fig. 3B and 4B). Since wet deposition is known as an important pathway of the fallout of radioactive plum (e.g., De Cort et al. (1998); Masson et al. (2011), the earlier precipitation might explain the appearance of higher ^{137}Cs activity concentrations in Ózd than Salgótarján (Tables 1 and 2). Our consequence is in agreement with a conclusion of a comprehensive European study by De Cort et al. (1998), in which high level concentrations were found to be correlated with wet deposition. From the day of 1st of May, higher level (1.5 km above ground) wind direction shifted to south-westerly in the Central-Eastern European region (De Cort et al., 1998). This is also compatible with our results which indicate increased wind speed (Figs. 3C and 4C).

Atmospheric deposition of pollutants onto rough surfaces, such as an urban environment, depends on various factors like elevation, local variability of topography, local slope, season, building heights and arrangements, etc. The latter can enhance the speed of wind in the wind tunnels between buildings (Buccolieri and Sandberg, 2008), furthermore the quasi-laminar sublayer can be affected by local features triggering off turbulence or increasing its intensity (Giardina et al., 2019). Obvious differences of dust deposition are observed between the two studied urban areas (Fig. 1A and B). In the highly hilly region at Salgótarján (up to ~700 m; Fig. 1A), the pattern of dust redistribution can be different from Ózd where the elevation is only up to ~200 m (Fig. 1B). In addition, different landscapes reflect the unequal deposition of a ^{137}Cs in attic dust (Fig. 2). Dry deposition process is recognized as an important pathway among the various removal processes of radioactive pollutants in the atmosphere (Giardina et al., 2019) for over a time.

4.4. Slope exposure

The slope exposure can be a significant factor in studying surface contaminations or depositions depending on the movement of air-masses (e.g., Begy et al., 2017). Considering this factor, our attic dust sampling sites were classified into 8 classes of ordinal and cardinal regions (N, NE, E, SE, S, SW, W and NW) determining slope exposure of each sampling points in keeping with terrain modelling (Moore et al., 1991). One can observe an increasing tendency from north (38.6 Bq kg $^{-1}$) to west (120.9 Bq kg $^{-1}$) for mean activity levels of these classes of samples (Fig. 5, Tables 1 and 2), using the whole data set. The largest average ^{137}Cs levels are observed at W (120.9 Bq kg $^{-1}$) slope exposure, including 13 samples (4 samples from Ózd and 9 samples from Salgótarján; Fig. 5. Notably the three Ózd outlier values (Fig. 2, Table 2) are located on this slope. In agreement with the present results, Szerbin et al. (1999) also found that W and S sloping parts of Hungary received the highest ^{137}Cs concentrations after the Chernobyl accident. Furthermore,

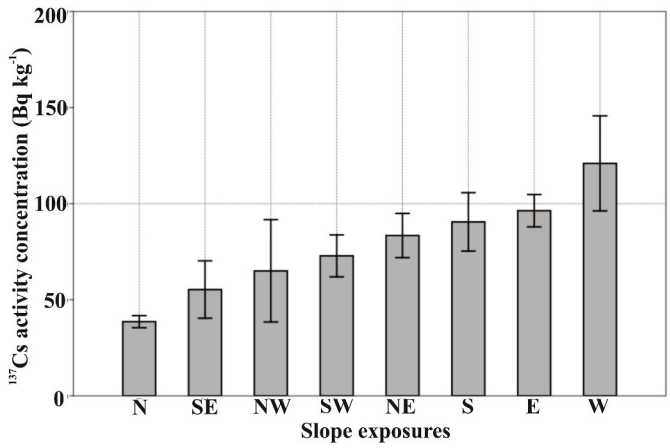

Fig. 5. Bar chart of slope exposures of attic dust sampling points (total number of samples n = 61; 36 from Salgótarján and 25 from Ózd), Slope exposure intention adopted in the work by Begy et al. (2017). Error bar illustrates standard deviations of the results.

it has also mentioned by Bossew et al. (2001) that in SE Austria the W directed stretches of Hungary turned out to be a strong fallout zone, which deposition vast influence on this slope.

Since dusts started to accumulate when a house was built on each slope sites, it remained undisturbed, but influenced by inertial and turbulence impact phenomena as well as human activity.

4.5. Isoscapes of ^{137}Cs activity concentrations in northern Hungary

In general, it is accepted that noticeable higher activities tend to be directly proportional to the sampling altitude. This altitude effect was observed in Transylvania after the Chernobyl (Begy et al. (2017)) and in Europe after the Fukushima accidents (Masson et al. (2016)). To explore whether ELE* has a significant role in the observed ^{137}Cs activity values in Salgótarján and Ózd, OLS regression analysis was applied with elevation as the predictor and ^{137}Cs activity as the dependent variable. For ^{137}Cs concentration activity values, the linear relationship with ELE* was proven insignificant at $\alpha = 0.05$ ($r^2 = 0.09$, F (1, 17) = 1.58, $p = 0.22$) and ($r^2 = 0.07$, F (1, 16) = 1.22, $p = 0.28$), respectively. The reason behind the insignificant relationship between the ^{137}Cs activity concentration and ELE* is probably due to the relatively small elevation range, 258 m and 40 m for Salgótarján and Ózd, respectively, since after the Fukushima accident for example the altitude effect was determined from samples spanning >3000 m in Europe (Masson et al., 2016).

Isotropic spherical semivariogram models were fitted to the empirical semivariograms of Salgótarján (Fig. 6A–A1) and Ózd (Fig. 6B–B.1). Since the semivariograms indicate a meaningful spatial autocorrelation structure (see e.g Chilés and Delfiner, 2012: Sect. 2.2 therein) of the ^{137}Cs activity samples the fitted theoretical semivariograms were applicable for kriging to obtain the ^{137}Cs isoscapes of the two cities (Fig. 7).

The elevated ^{137}Cs activity concentrations in attic dust over the exterior part of urban areas in Salgótarján (Fig. 7A) and Ózd (Fig. 7B) show an increasing level of ^{137}Cs similarly to the central part of cities. The lowest activity concentrations, found from valleys where former industrial areas located, might decrease of ^{137}Cs levels in both studied area, in an agreement with Outola et al. (2003). Because of less environmental degradations in attic area, attic deposition probably contains

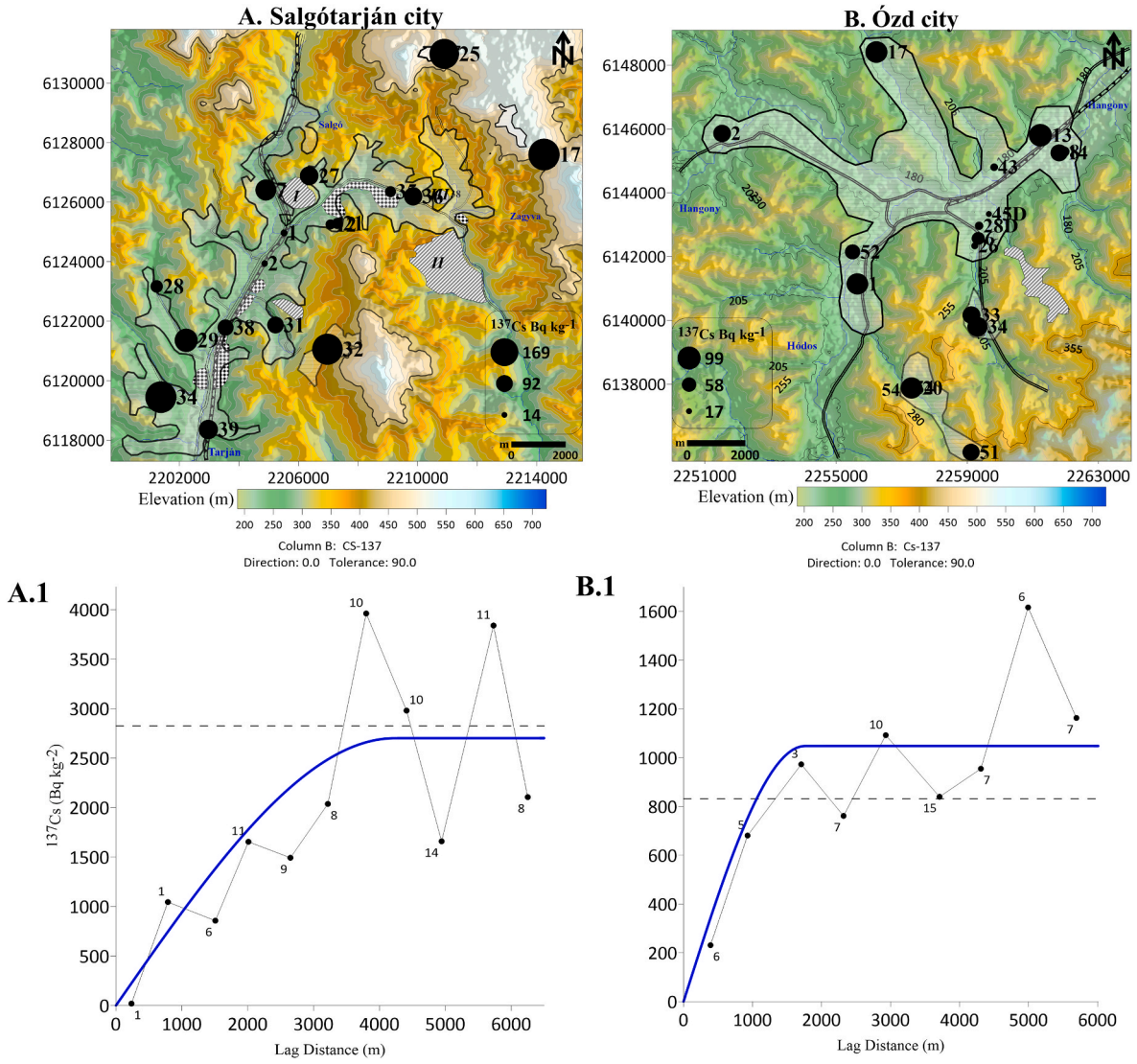

Fig. 6. Fitted topography of studied urban area maps for the kriging methodology. The diameters of filled circles are proportional to ^{137}Cs activities (A and B), with labels; blue lines: creeks (map projection: EPSG:3857, WGS 84/Mercator projection (m)). The empirical (black dotted line) and fitted spherical semivariogram (blue line) derived from ^{137}Cs activity values for Salgótarján (A.1) and Ózd (B.1). For Salgótarján, the parameters of the spherical semivariogram model are $C_0 = 1$; C_0+C = 2672; a = 4.2 km; $r^2 = 0.72$ (A.1); for Ózd, $C_0 = 1$; C_0+C = 1048; a = 1.7 km; $r^2 = 0.51$ (B.1). Numbers next to values of the empirical semivariogram (black numbered dots) represent the data pairs within a given distance; bin width was 590.9 m for Salgótarján and 610.0 m for Ózd. Max lag distances were 6500 m and 6010 m for Salgótarján and Ózd, respectively. The dashed line indicates the variance. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

rich heavy metal/loids dust particles which is not a ^{137}Cs preferred place. Whereas, ^{137}Cs is adsorbed reversibly and/or irreversibly onto the clay minerals, particularly onto illite (Lee et al., 2017).

Distribution of atmospherically derived radionuclide ^{137}Cs depends on various environmental properties and exhibits complex spatial variability (Van der Perk et al., 2002; Navas et al., 2011). Apparently, Salgótarján received larger amount of ^{137}Cs than Ózd (Fig. 7A–B), where regular cleaning and renovation of attic areas might have caused a depletion of ^{137}Cs. Based on geostatistical reason, attic dust STN_AD26 and OZD_AD54 were left out of the analysis due to their unacceptably high variance compared with their neighboring sites (Fig. 7A–B, marked in white circles). Several other factors, such as attic height changes, changing ventilation circumstances and renovations assumed to be reasons for distracting. Our constructed ^{137}Cs spatial distribution patterns for the two residential areas are based on higher sampling density than previous assessments in Hungary (De Cort et al., 1998; Szerbin et al., 1999; Szabó et al., 2012).

Overall, the long-term preserved dusts without severe disturbances could result in higher ^{137}Cs activities in attic areas. The studied regions had intensive heavy-industrial history in the past centuries, where dust accumulated from local anthropogenic activities as industrial (commonly iron and steel work processes, coal mining, transportation, and in case of Salgótarján coal-fired power plant) and mixed natural sources and with atmosphere derived dust particles. All of these resulted in a large variability of dusts. The activity concentrations of ^{137}Cs measured in different age of buildings (Fig. 2, Tables 1 and 2) provide a better basis for further considerations than previous studies (Cizdziel et al., 1999; Ilacqua et al., 2003). Opportunity of study layered attic dust samples in Ózd from the former steel factory (Fig. S1, Table 2) sheds light on the significance of ^{137}Cs investigation in urban area. All these findings confirm that attic dust apparently traps ^{137}Cs for decades, which remain detectable. The presented research is one of the first steps towards evaluation of interdisciplinary to this field.

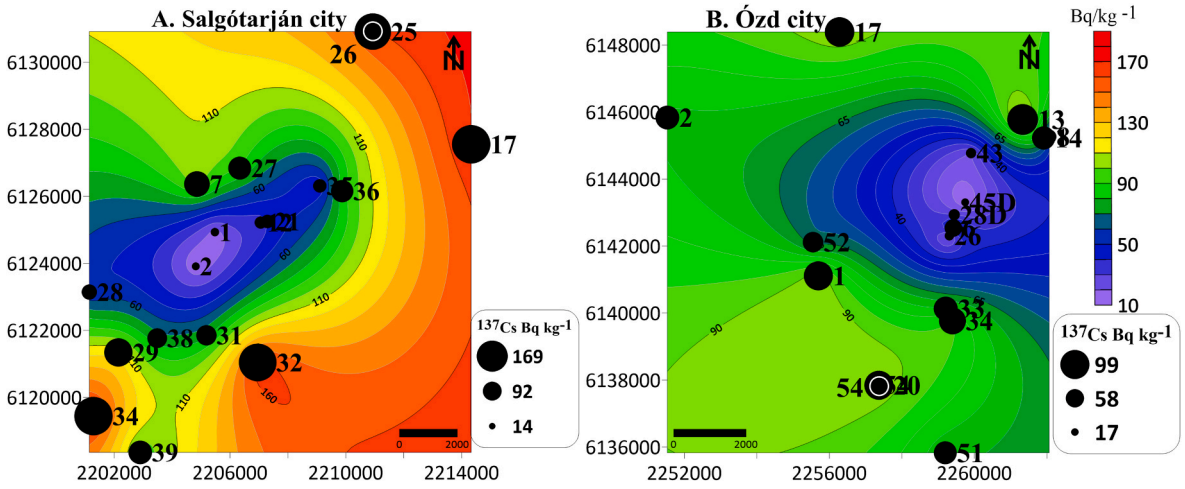

Fig. 7. The kriged contour map (isoscape) (Map projection: EPSG:3857, WGS 84/Mercator projection (m)) of the ^{137}Cs activity from the studied urban environments (A) Salgótarján ($n = 17$) and (B) Ózd ($n = 16$). Grid resolution: 0.9 × 1 km for Salgótarján and 0.8 × 1 km for Ózd. Interpolation was done with ordinary point kriging using the spherical semivariogram models. (Note samples of Salgótarjan AD26 and Ózd AD54 was excluded in the interpolated contour map, showing in white circles).

5. Concluding remarks

Our hypothesis that ^{137}Cs resides in attic dust within residential area is proven based on our study on a total of 61 samples. The average values of activity concentration in both studied cities are highly similar: 88.5 ± 5.1 Bq kg^{-1} for Salgótarján and 87.8 ± 4.5 Bq kg^{-1} for Ózd. The highest and lowest values (272 and 1.9 Bq kg^{-1}, separately) were measured in Ózd. The activity values of ^{137}Cs in attic dust samples are commonly higher than those of obtained in soils from Hungary, other countries. This finding indicates that attic dust, accumulating in closed and safe area, can more efficiently preserve fingerprints of the past atmospheric ^{137}Cs pollution compared to soil (i.e., urban, street, agricultural, forest ones, etc.) occurring in open environments.

Variation of ^{137}Cs activity concentration in function of year of building construction gradually decrease towards older buildings form the three extremely high values (223.4; 238.6 and 272.8 Bq kg^{-1}) measured in Ózd houses built between 1954 and 1978. This trend indicates that ^{137}Cs activity concentration reaches back to background values as 0 if there is no additional source to create overprint it. In contrast, houses built after the beginning of nuclear test shows positive correlation between ^{137}Cs activity concentration and year of building construction suggesting possibility of redistribution of attic dust.

Study of layered attic dust samples in Ózd from the former steel factory sheds light on the significance of ^{137}Cs investigation in residential area: the older layered dust, accumulated during operation of the former steel factory, shows one order higher activity concentration compared to the younger one, accumulated during the past ~25 years after steel factory was shut down and source of ^{137}Cs was not available in the local environment.

Meteorological simulations showed that Ózd city had earlier contaminated precipitation from the radioactive plum of the Chernobyl NPP accident than Salgótarján, which can explain the appearance of outlier values of ^{137}Cs activity concentrations in three Ózd houses.

In both cities, houses built on the westerly orientated slopes are the mostly loaded by ^{137}Cs, showing elevated activity concentration according to terrain aspects modelling.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvrad.2022.106950.

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