Roughcast Analysis as a New Method of Environmental Research

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ABSTRACT: *This study aims to show the purpose of roughcast analysis as a new useful tool for research of the condition of the environment. Samples of roughcast were collected from buildings in 6 different cities in Europe (Cracow, Lublin, Warsaw, Lviv, Monchegorsk, and Murmansk) and their compositions were examined with the scanning electron microscope, ICP-OAS, and ASA methods. Also, a binocular magnifier was used. The samples were investigated for heavy metals presence. The result shows that despite the deposition of particles being bigger on the horizontal surface, the vertical layer of the roughcast accumulates significant amounts of impurities and heavy metals. The composition and content of metals differ accordingly to automotive traffic nowadays and in the past, the existence of road hubs, industrial districts, and historical manufacturers. Samples from Cracow show high content of non-ferrous metals and nickel, whereas the content of copper was significantly higher in Monchegorsk. In Lublin and Lviv, their agricultural past contributed to significantly lower heavy metal content Determination of heavy metals contents in roughcast can be used to evaluate anthropogenic impact in the cities through the years and to help to protect cities' populations from negative consequences of living in urban areas.*

KEYWORDS: *Roughcast surface analysis; Heavy metals pollution; Dust adsorption; Urban area; ICP-OAS.*

INTRODUCTION

Environmental analysis and research are crucial issues to determine factors that cause pollution in inhabited areas nowadays as they allow us to discover the anthropogenic impact of human behavior on the natural environment. These aspects through the years have become more and more important because of the multiple connections between

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the unpolluted environment and the quality of life and well-being of people. Many of the research issues focus on the impact of human behavior on i.e. freshwater quality, drinking water quality, air quality, or concentrations of heavy metals in plants.

Analysis of the environment can be conducted in an unchanged, wild, natural environment as well as in heavily transformed areas like city centers. Heavy metals contamination studies in the cities are conducted quite often, however, not many of them accentuate the problem of the place of occurrence and accumulation of heavy metals in the cities. Wahab with collaborators [1] raises the issue of the concentration of heavy metals (copper, lead, nickel, and zinc) in settled indoor dust collected directly from the room inside the house. The authors point to relevant connections between common sources of heavy metals in the dust (like traffic emissions, street dust, and industrial fumes) and their concentration in the examined indoor samples. The study aims at indoor air and dust quality but emphasizes the fact that heavy metals inside the houses can be derived directly from outdoor sources. *Dytłow* and *Górka-Kostrubiec* [2] as well as *Zgłobicki et al.* [3] using geochemical background data and spectrometry determine the heavy metals load in street dust in Warsaw and Lublin. The samples were collected in the vicinity of heavily used roads and the concentrations were defined via mass spectrometry. They show great anthropogenic impact on the heavy metals loads in examined swatches. Both of the studies stress that heavy metals occurrence in street dust raises the toxicity of the dust itself and leads to severe health problems such as i.e. kidney dysfunction.

The method that we would like to propose in this paper is based on an analysis of the exterior layer of roughcast covering the buildings. The presented method is pioneering in the field of determining the impact of heavy metals on human health -- analysis of the roughcast, in terms of long-term accumulation and exposure to heavy metals was never conducted before. This analysis is important, as the roughcast on the outer layer of the building has got persistent contact with every substance that appears in the atmospheric air like particular matter or toxic compounds from car fumes. The structure and texture of roughcast are prone to accumulate pollutants, likewise, adding more and more layers of the cast through the years should allow detection intervals of accumulations and their sources. In addition to this, roughcast analysis could be the indicator of the nuisance of different chemical compounds in the environment in urban areas. For instance, elements accumulated in exterior covers of buildings are washed off with precipitation and then they soak in the soil beneath the exposed surface. Thus, the roughcast analysis could be a new, reliable source of knowledge about the anthropogenic impact that has been occurring for many years in the city and influenced the well-being of both, inhabitants and the environment.

In this paper, the analysis of samples of roughcasts collected in 6 European cities was conducted to compare the results and evaluate the findings. This study aims to show the purpose of carrying out this kind of study and to open the discussion of interpretation possibilities. However, due to the novelty of the method, not much of the source or reference material is available.

STUDY AREA

Each sample was collected in a city that was historically part of the Soviet Union. Their economy back in those days was based on a high-emission industry that mainly had been processing raw materials – hard coal or metal ores – which often had been reaching 50% of GDP [4]. Samples were collected in the centers of the cities, where the roughcast layer is the oldest, therefore the exposure to the pollutants and different weather conditions that last the longest. Also, the buildings in the near vicinity of the main or heavily used roads were chosen as they are prone to accumulate more harmful substances.

Cracow is claimed to be the most polluted city in the European Union [5]; through the years the air quality had been lowered by industrial plants (i.e. ironworks of Tadeusz Sendzimir) and furnaces used on daily basis by people living in the city. This situation has changed recently - in 2019 usage of the fossil fuels and heavy-burning oil was banned [6]. Burning fossil fuels had been a big problem; in the city center (and the Old Town) the furnaces had been powered by hard coal, oil, or wood. Also, installations, that had let exhausts out were old and most of them had no filters applied. Non-trivial sources of occurrence and type of pollutants are two international communication trails that lead through the city and in its near vicinity (Fig. 1). Excessive traffic, well-developed public transport, and traffic jams are sources of significant amounts of a particular matter, heavy metals and other

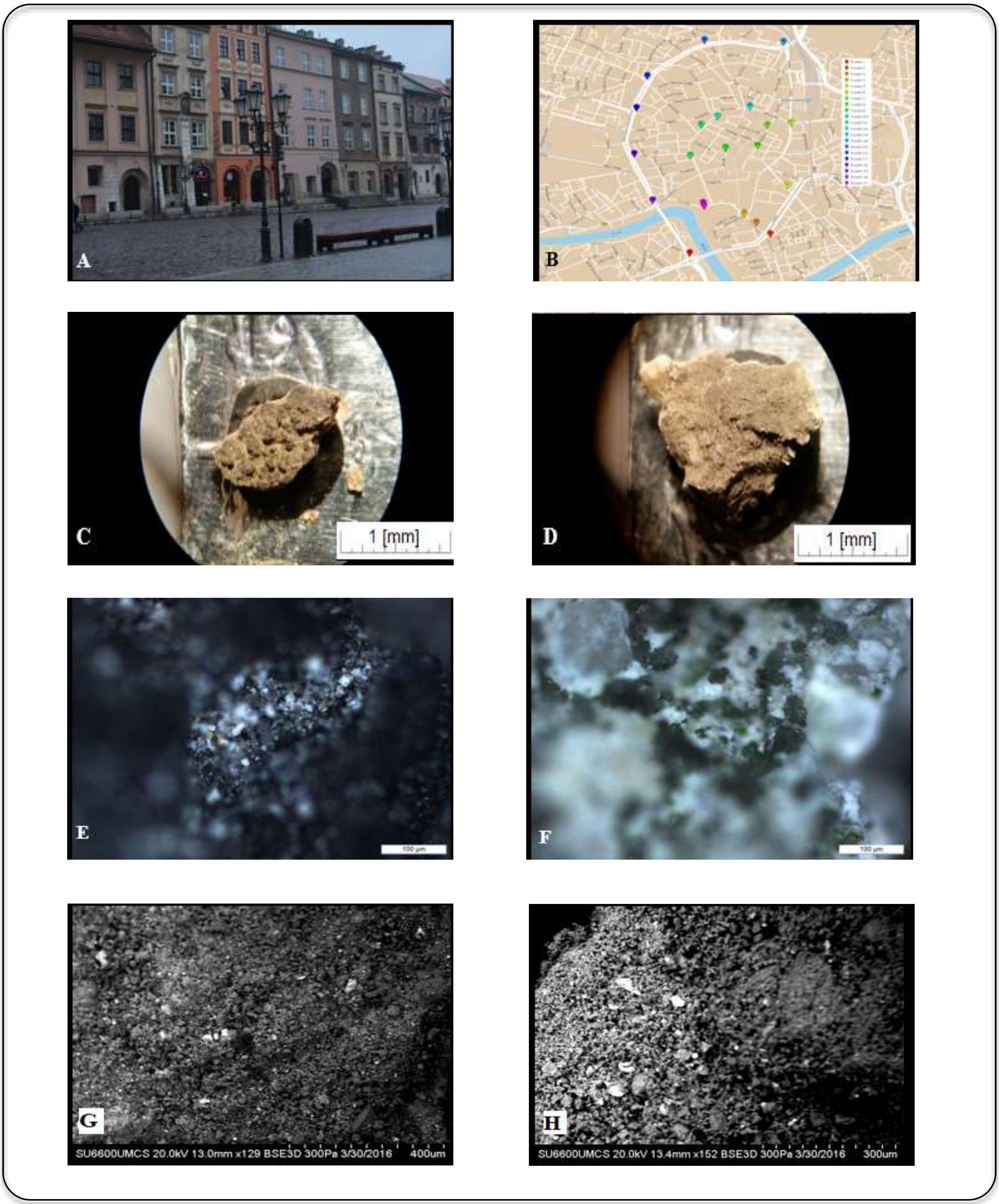

Fig. 1: *The photograph of Cracow landscape (A) and situated sketch with samples localization (B); Microphotographs with binocular-magnifier of plaster samples with dust (C, D), optical–polarized microphotographs (reflected light, crossed polars) with plaster surface, dust, and pigments; Backscattering electron BSE) microphotographs of plaster samples with scoot, metallic dust and microflora (G, H).*

pollutants [5]. In the case of Cracow, the air quality is also lowered by geographical location □ the city is located in a hollow which causes decreased ventilation conditions.

No big ironworks nor metal processing plants were found in Lublin (Fig. 2). Its industry had been based on manufacturing work (i.e. iron foundry, sugar mill, gasworks, mechanistic factories) [7]. Nowadays, air pollution in Lublin has its source in traffic (exhaust, aerosols) and household furnaces; the air quality is also lowered by local electrical and heating power. The noteworthy fact is, the location of Lublin on hills increases efficient ventilation of the city and dilution of particulate matter and chemical compounds in the air.

In the capital city of Poland, Warsaw, the main source of air pollution comes from transportation and traffic. There is also ironworks in the northwest part of the city which can decrease the quality of air. Household furnaces likewise have a great impact on air pollution in the city, not to mention that more and more people from Warsaw are locating their houses outside the city, in the neighboring towns. According to this situation, the real issue to address is the advection of pollutants from those residing places over the capital city [8] (Fig. 3).

In Lviv, there are a couple of sources that decrease air quality in the city □ heavy industry plants, transportation, and traffic, and fossil fuels burning in household furnaces [9]. Because of the soaring prices of gas, many people have given up on having gas furnaces, buying and using solid fuel (hard coal or wood) powered ones instead, which leads to an incredible increase of particular matter pollution in the city. Because of legal and illegal logging for heating purposes, local forests are disappearing, and, in conjunction with loess substrate, this situation creates a threat of accelerated soil erosion as well as progressive deterioration of air quality through an increased amount of soil particles suspended in the air [20]. The center of Lviv is located in the relief, which causes non-effective ventilation of the city; during the time of influence of the high-pressure zone, the cumulation of pollutants above the city often occurs [10] (Fig. 4).

Murmansk is a port city and also the main mining and metal ores processing center of Russia (Fig. 5). The pollution in the region (Murmansk Oblast) is caused especially by mining complexes and this pertains to air quality issues as well as water and soil contamination [11]. The quality of air is also lowered by the local electrical and heating powerplant. Poor insulation of buildings deepens the problem of air pollution □ significant losses of energy generate greater consumption of fuel to obtain enough energy to heat the buildings.

A similar situation, as in Murmansk occurs in Monchegorsk, located in the same region (Fig. 6). The main sources of air pollution are ironworks of ferrous and non-ferrous metal ores and mining [12]. The area of Monchegorsk is claimed to be a hot spot of contamination, due to intensive nickel, cobalt, and copper mining and also because of processing often sulfated metal ores [13]. However, the official documents report that the emission of pollutants decrease**s** yearly [14].

EXPERIMENTAL SECTION

The roughcast samples were collected from buildings of different ages and covering preservation states. Corrosion of elevation was also detected in the field studies through architectural detail observation that allowed to set apart different types of corrosion, leading to naming the types that are caused by human activity [1,15-24]. Collected samples were divided into groups □ the first group had microscopic observations done, they were also prepared as thin plates cross-sections of the surface layer of roughcast, and fragments of mortar) to further analysis. The second group of samples was used to conduct observation in microscale and chemical analysis.

The second group of materials was first analyzed by a binocular magnifier (in this process microphotographs were taken to identify the samples) and then in an optical microscope Leica DM2500P in reflected light. Microscale observations were conducted in a scanning electron microscope in a low vacuum using Hitachi SU6600 with EDS (Thermo) add-on [25,26]. Samples were placed on a carbon, string partitioned with paper [27]. Every swatch was put in place with extreme caution with a spatula under a binocular magnifier. A representative sample of no more than 1g of substance was used in this process. After the weighted swatches were placed in an electron microscope chamber, analysis without sputtering under conditions of low vacuum (approx. 10hPa) was conducted. To the probe, 15kV beam was applied with a time of 180 s of analysis for every sample. Two types of microanalysis were processed - firstly, the chemical composition of the swatch was examined by exposing all grains at once, and secondly, grains were analyzed separately (approx. 150 analyses per sample).

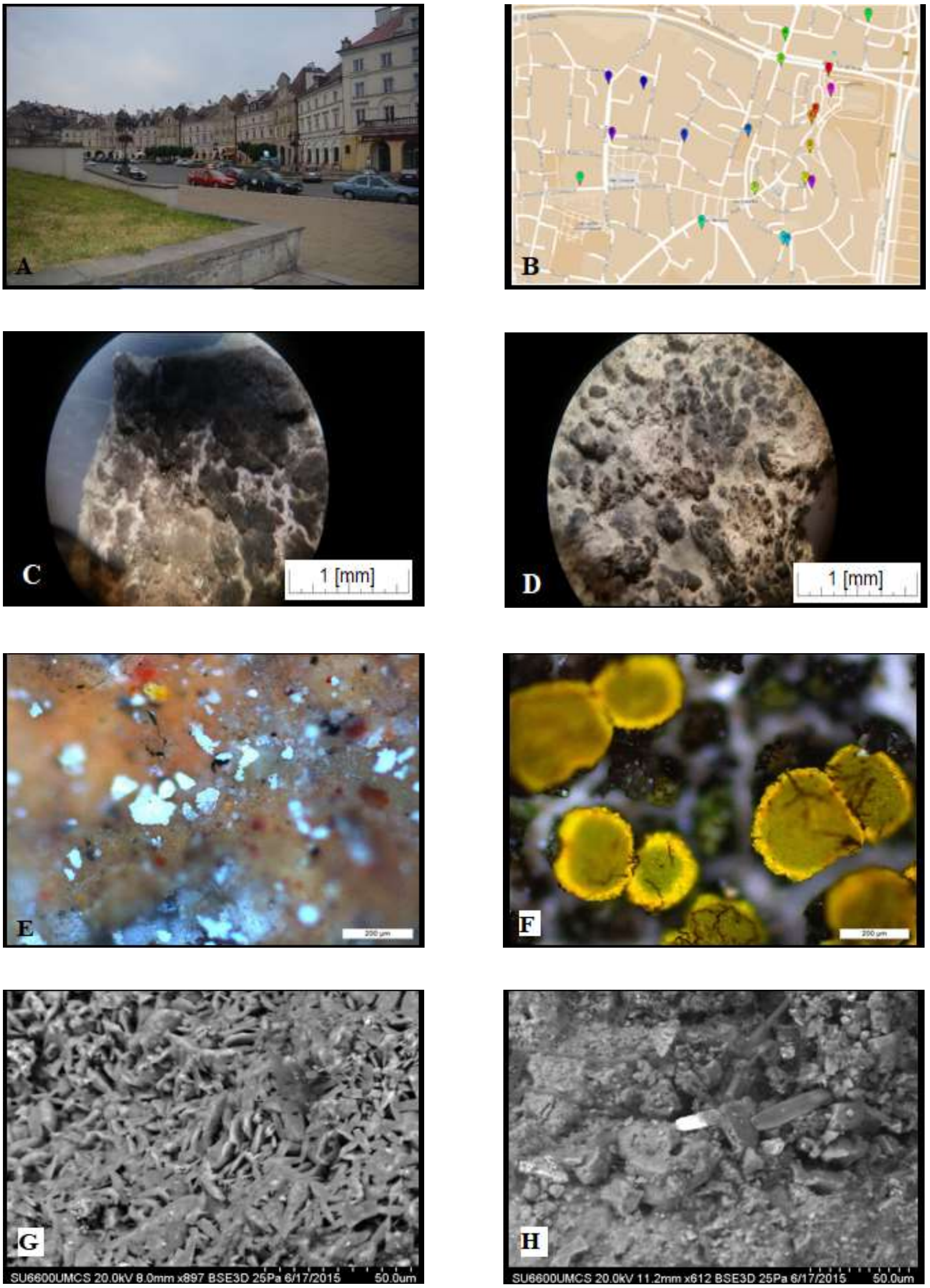

Fig. 2 The photograph of Lublin landscape (A) and situated sketch with samples localization (B); Microphotographs with binocular-magnifier of plaster samples with dust (C, D), optical–polarized microphotographs (reflected light, crossed polars) with plaster surface, dust, and pigments; BSE microphotographs of plaster samples with scoot, metallic dust, and microflora (G, H).

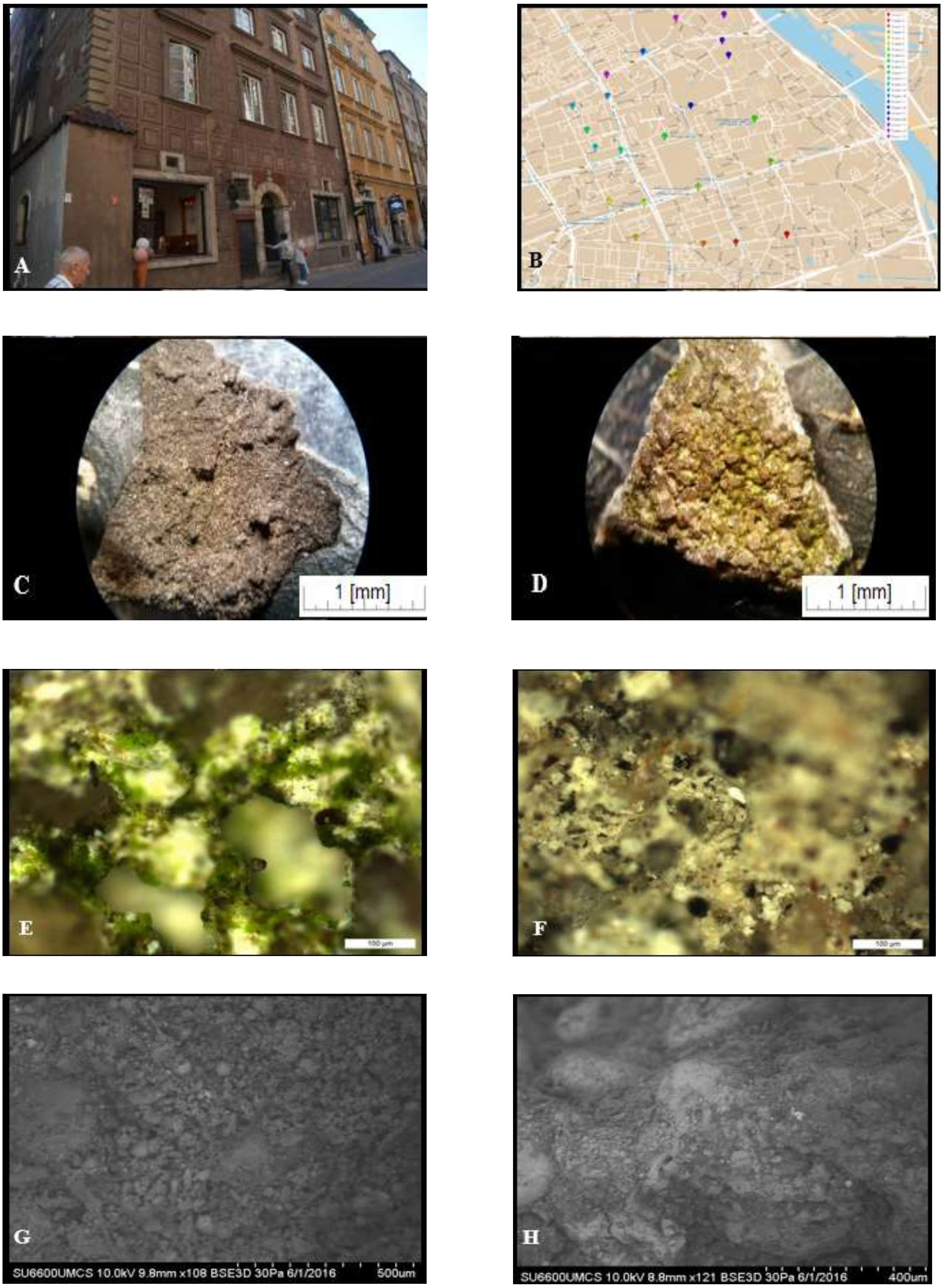

***Fig. 3:** The photograph of Warsaw landscape (A) and situated sketch with samples localization (B); Microphotographs with binocular-magnifier of plaster samples with dust (C, D), optical–polarized microphotographs (reflected light, crossed polars) with plaster surface, dust and pigments; BSE microphotographs of plaster samples with scoot, metallic dust, and microflora (G, H).*

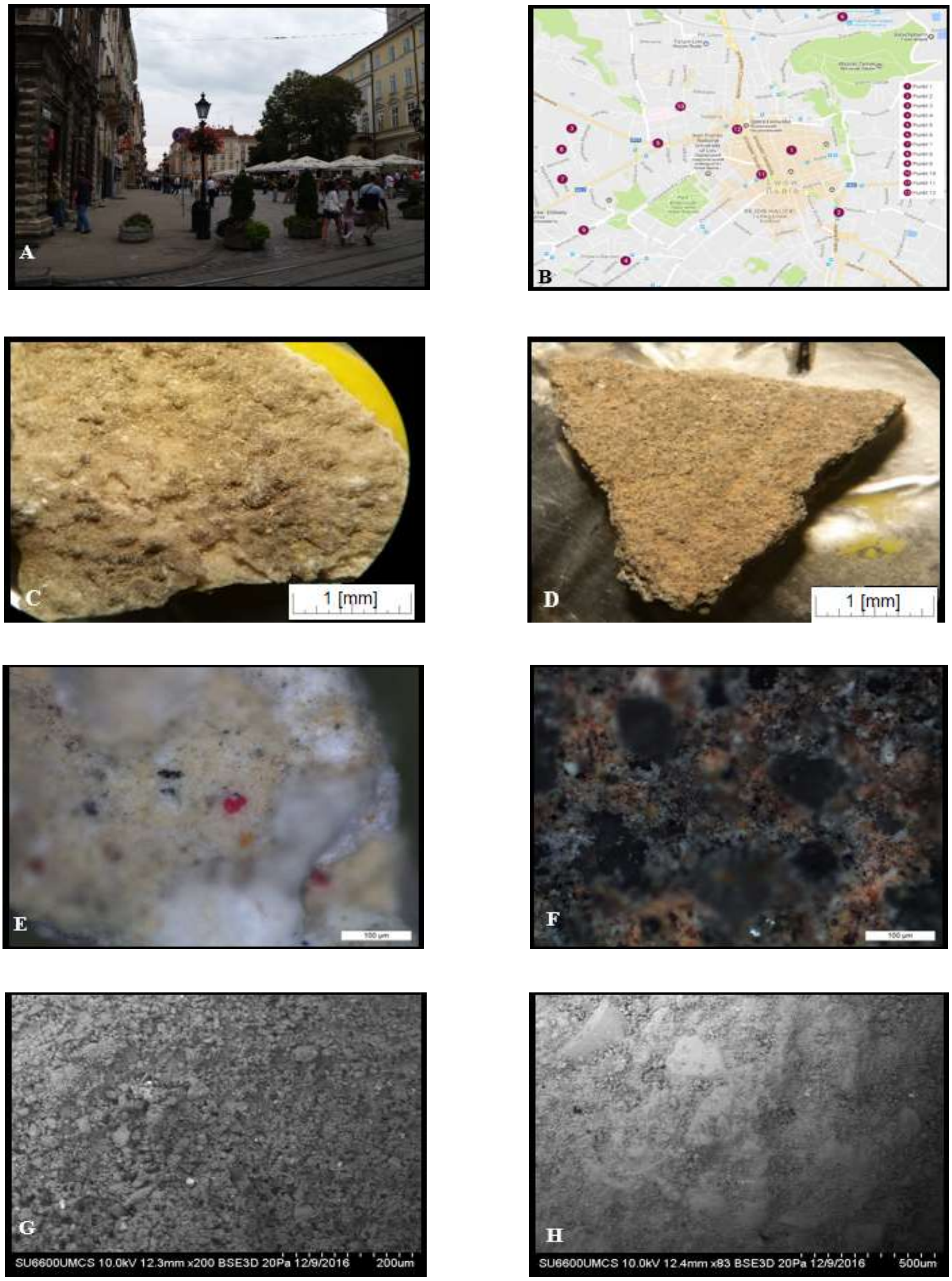

***Fig. 4** The photograph of Lviv landscape (A) and situated sketch with samples localization (B); Microphotographs with binocular-magnifier of plaster samples with dust (C, D), optical–polarized microphotographs (reflected light, crossed polars) with plaster surface, dust and pigments; BSE microphotographs of plaster samples with scoot, metallic dust, and microflora (G, H).*

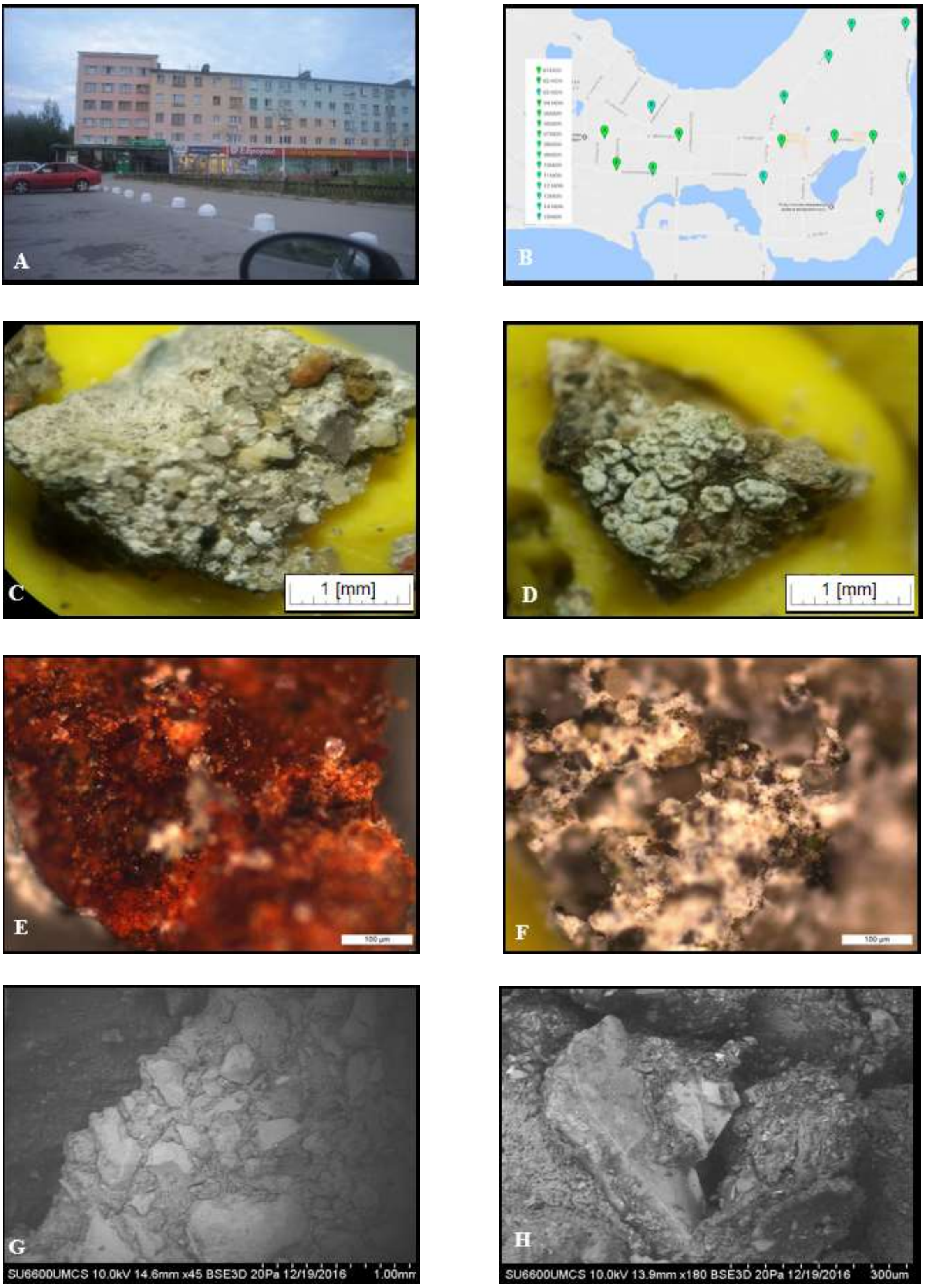

Fig. 5 The photograph of Monchegorsk landscape (A) and situated sketch with samples localization (B); Microphotographs with binocular magnifier of plaster samples with dust (C, D), optical–polarized microphotographs (reflected light, crossed polars) with plaster surface, dust, and pigments; BSE microphotographs of plaster samples with scoot, metallic dust, and microflora (G, H).

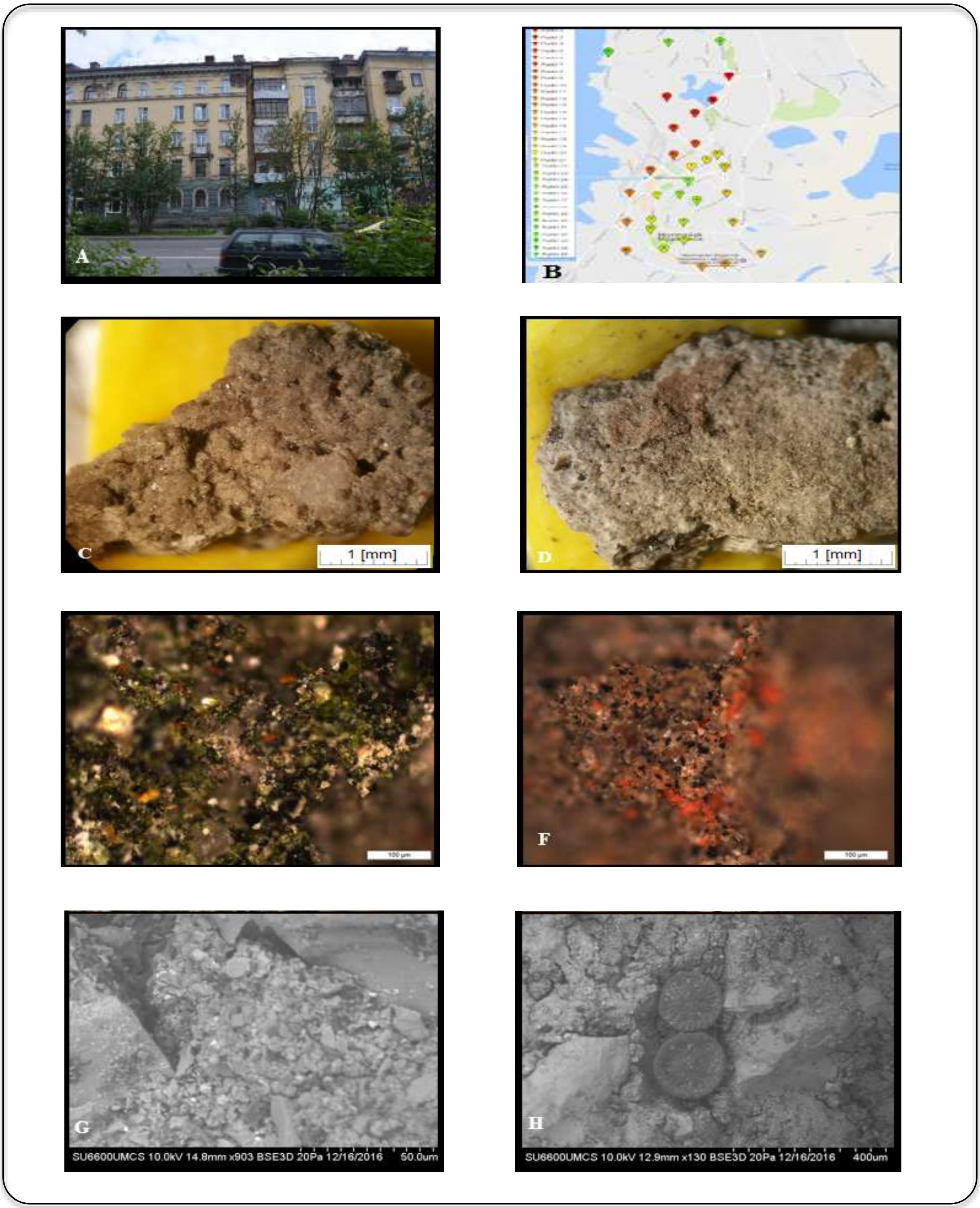

***Fig. 6** The photograph of Murmansk landscape (A) and situated sketch with samples localization (B); Microphotographs with binocular-magnifier of plaster samples with dust (C, D), optical–polarized microphotographs (reflected light, crossed polars) with plaster surface, dust, and pigments; BSE microphotographs of plaster samples with scoot, metallic dust, and microflora (G, H).*

The findings of observations were placed in MS Excel and then processed mathematically. The surfaces of roughcast samples were also studied using XRF technics.

For chemical composition examination, samples were diluted by microwaves and checked for the presence of heavy metal particles, connected with pollutants emissions. ICP-OAS and ASA methods were implemented. Using the X Series 2 (Thermo Scientific) spectrometer with ionization in an induction-coupled plasma (ICP-OAS), a chemical analysis of samples was conducted. Swatches were filtered with a 0,45 µm porosity membrane filter; 0,5 mL was the minimum volume that was used to determine one element. The time of one determination equaled about 1 minute and the sensitivity of the device for iron, calcium, and magnesium were respectively: 10;1;1 µg/dm^3. ICP-OAS analyses were calibrated using the “C Loam 7004” soil standard. Analysis was conducted by *Lesia Lata*, Ph.D. on the Varian emission spectrometer (ICP-OAS). Similar to the findings of the microanalysis, the outcome of chemical composition research was processed mathematically in MS Excel and Surfer programs. For uncluttered outcomes, data on charts were presented without outliers. The values of all examined heavy metals concentrations with the outliers can be found in Table 1 as Supporting information.

All the observations above, including the microscopic analysis, ICP-OAS, and ASA were conducted at the Laboratory of the Department of Geology, Soil Science and Geoinformation of Maria Curie-Skłodowska University in Lublin, Poland.

RESULTS AND DISCUSSION

The outcome of 21 samples from the center of Cracow shows a high content of non-ferrous metals and nickel; cadmium and arsenic contributions were measured up to a few parts per million (Fig. 1). Lead content was not detected in every sample (no. 2, 4, 9, 14, and 16) and their share was no higher than over a dozen parts per million. In many swatches over a dozen, chromium content was detected. Zinc and manganese parts fluctuated up to 500 ppm, whereas the average amount of copper equate**s** to 15 ppm. In examined samples, the content of titanium was detected up to 100 ppm, when iron encompassed even over a dozen or so percentages by weight of measured samples (Fig. 7). Distribution of contents of the elements in swatches was quite approximate near the center, which means that examined additions of heavy metals are particles absorbed from the air, especially when different roughcast structure and texture is remarked.

15 samples have been taken from the city center of Monchegorsk (Fig. 8). Cadmium content was comparable to that measured in Cracow, as the situation applied to arsenic and chromium. The concentration of copper was significantly higher, compared to Cracow, and equated from 50 to 200 ppm (Fig. 2). The lead load did not differ between Cracow and Monchegorsk, except for one sample where its content reached a few percentages by weight; this level of lead has been caused by the artificial addition of this element as a component of the roughcast. In other cases presence of lead was from accumulated material. Zinc and manganese levels were comparable in both cities as well as titanium and iron content**s**.

In Lublin and Lviv, the content of heavy metals is significantly lower, which is a corollary of the agricultural character of the regions and cities**’** geographical locations. In Lublin 18 samples were examined and in Lviv – 12. Arsenic, cadmium, and lead contents in swatches from Lublin were below device sensitivity, also they did not appear in the ICP analysis outcome (Fig. 9). Nickel and chromium loads fluctuated in the range of 5-8 (max. 18) ppm. The content of copper in some samples was very high, reaching 400 ppm. The reason for this level of copper occurrence in the roughcast is the location of the sampling place, near the copper roof of **a** historical building – the 12th-century defensive wall. Precipitation had caused the washing off of the copper from the roof and then the element had been accumulating in the roughcast. Zinc and manganese content reached up to 100 ppm (max. 540 ppm), but iron load attained about 4 percent by weight (Fig. 9).

Lviv had a higher level of detected heavy metals in roughcast than Lublin, which is related to the high pollution of the city center. In 5 samples cadmium levels reached up to 8 ppm; arsenic content equated up to 2,5 ppm (Fig. 10). Average chromium content in samples was a few parts per million, but in one of the swatches (no. 9) the noticed content reached over 50 ppm.

Lead load in roughcasts from Lviv ranged from approximately 25 ppm, while its maximum reached up to 212 ppm (sample no. 9). Its copper content was up to 10 ppm, but zinc was much higher - from 200 to 1000 ppm. Manganese and titanium concentrations were comparable - they reached up to 100 ppm. Iron, like in other cities, comprises up to a few percentages by weight in each sample.

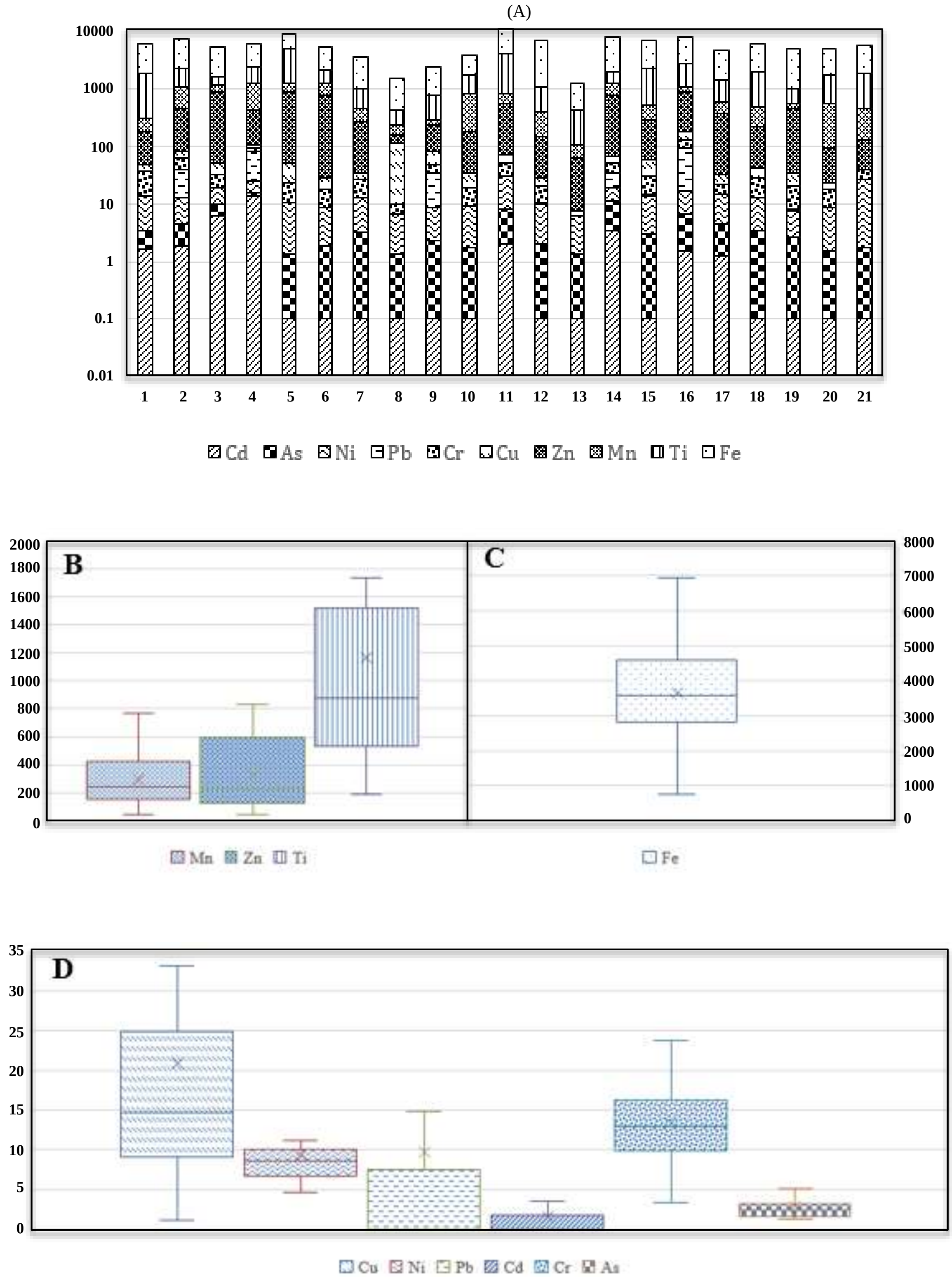

Fig. 7: Contents of heavy metals in the samples from Cracow [ppm] (A) and their range [ppm] (B, C, D) without outliers.

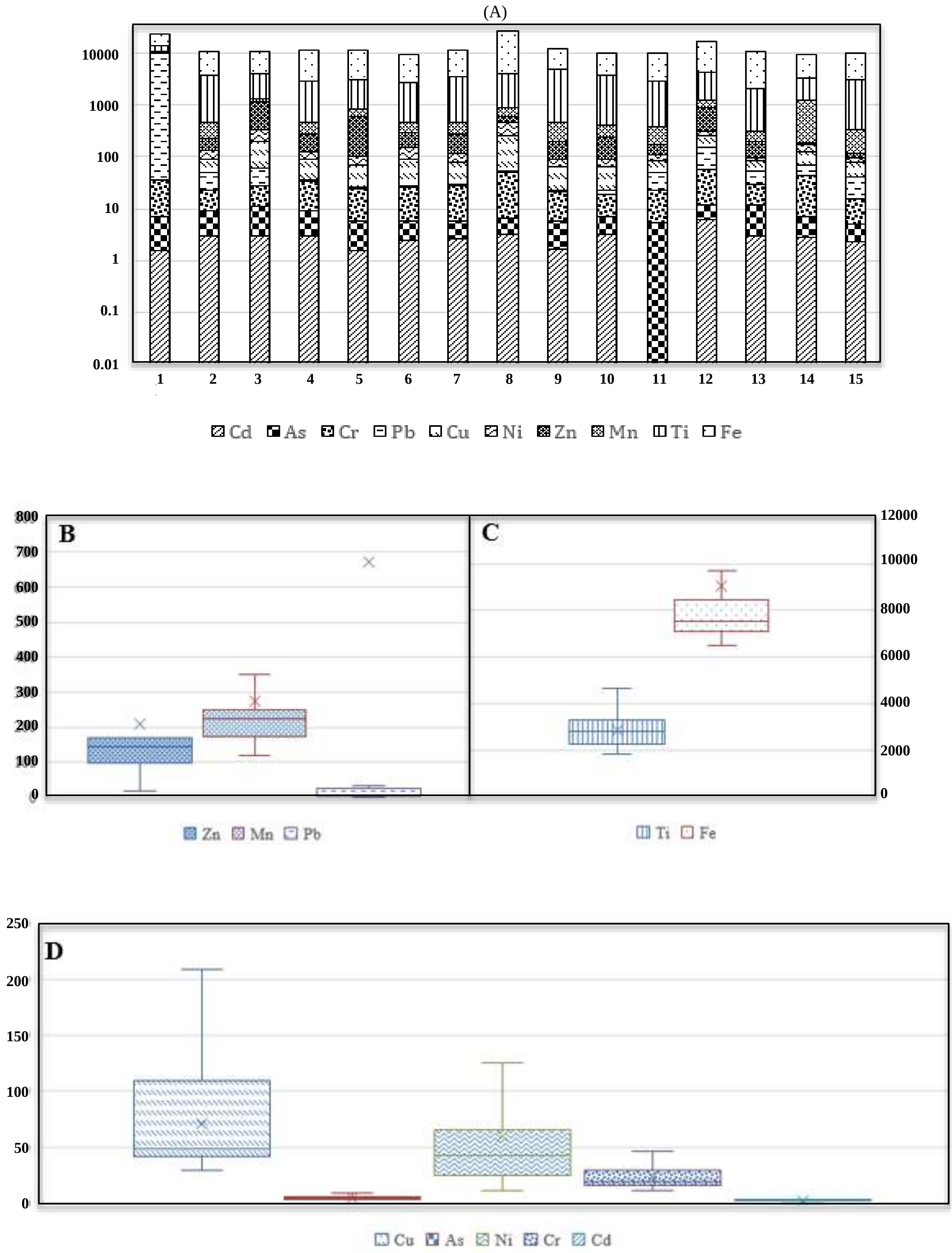

Fig. 8: Contents of heavy metals in the samples from Monchegorsk [ppm] (A) and their range [ppm] (B, C, D) without outliers.

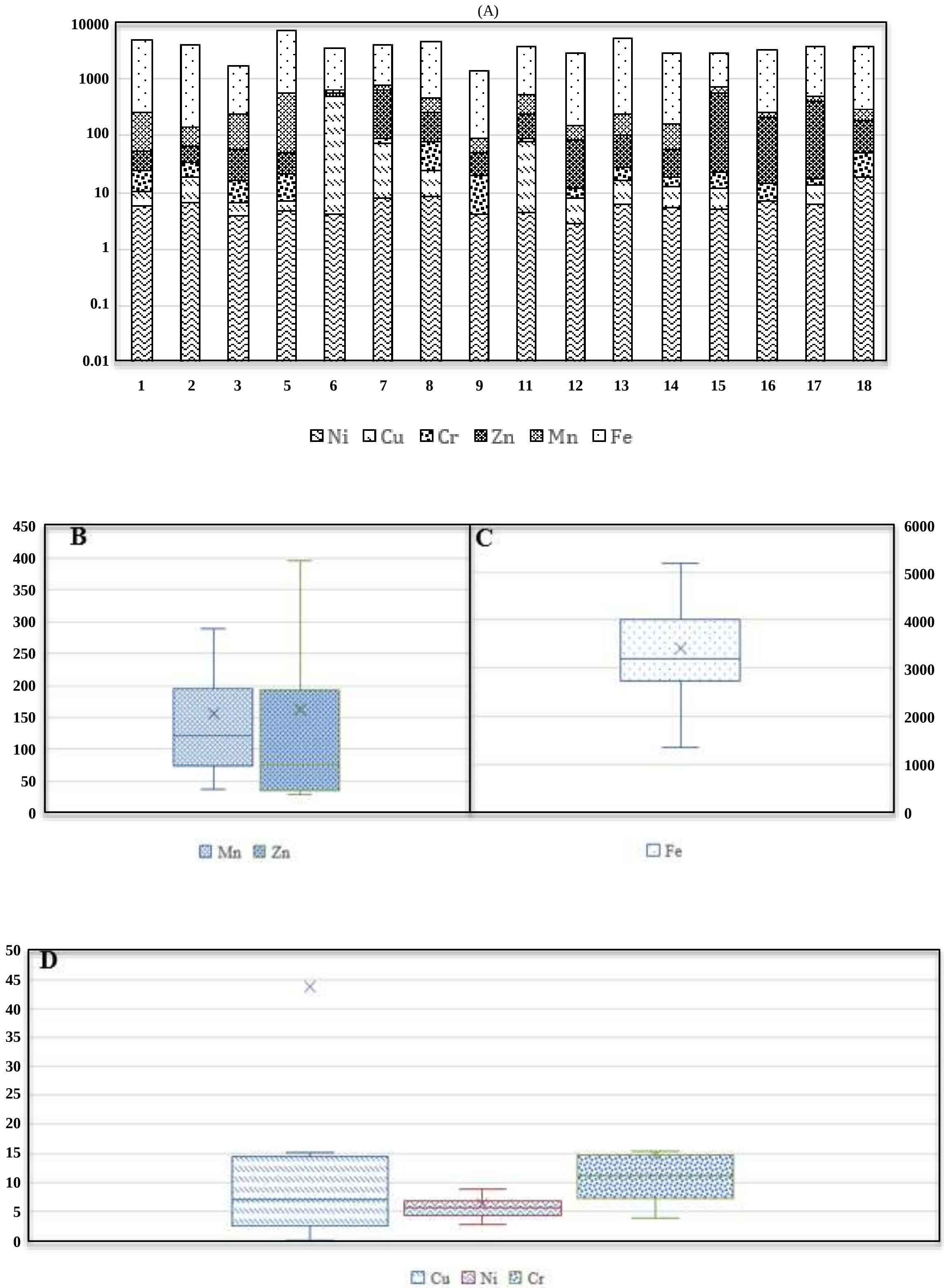

Fig. 9: Contents of heavy metals in the samples from Lublin [ppm] (A) and their range [ppm] (B, C, D) without outliers.

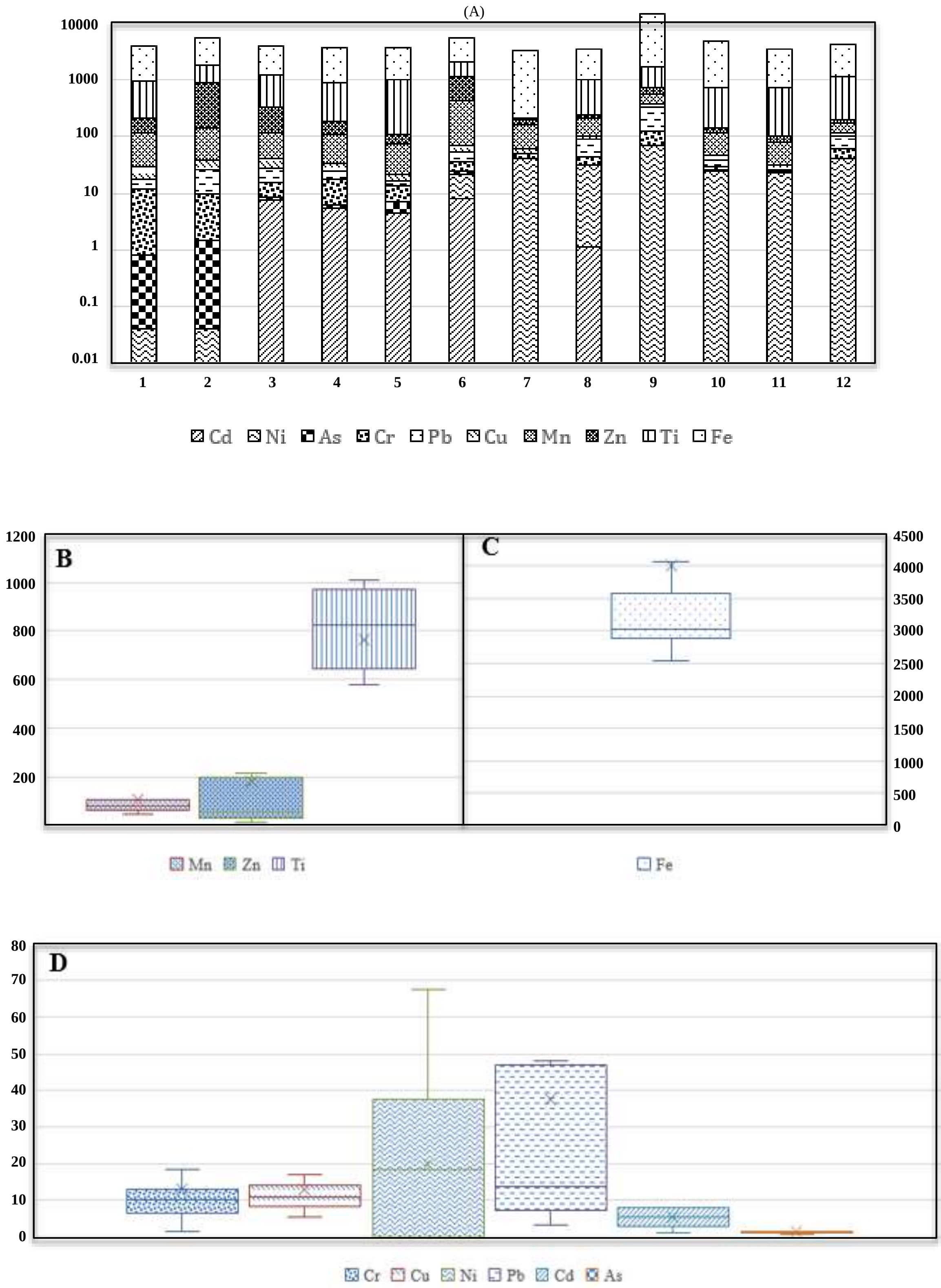

Fig. 10: Contents of heavy metals in the samples from Lviv [ppm] (A) and their range [ppm] (B, C, D) without outliers.

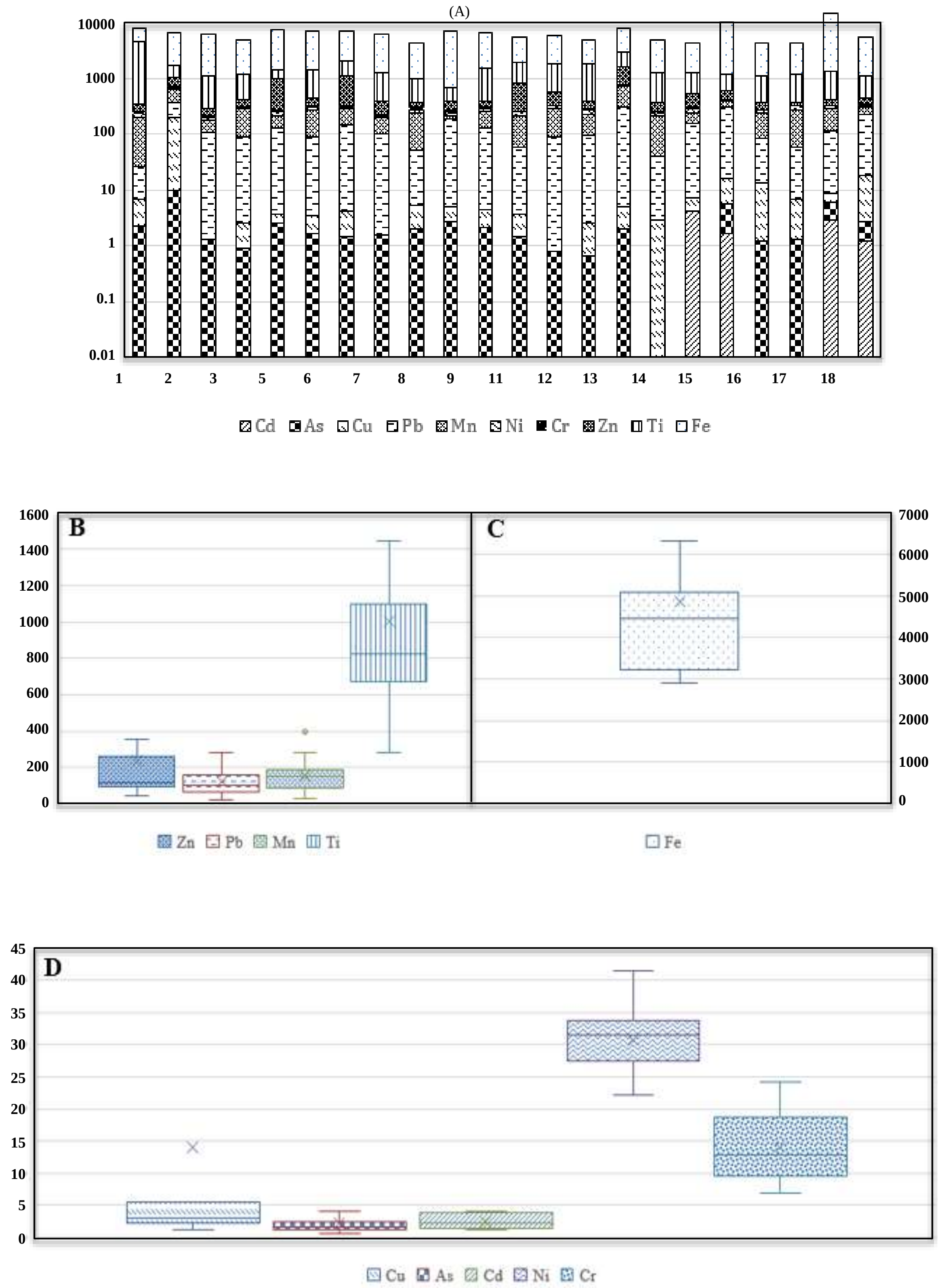

Fig. 11: Contents of heavy metals in the samples from Warsaw [ppm] (A) and their range [ppm] (B, C, D) without outliers.

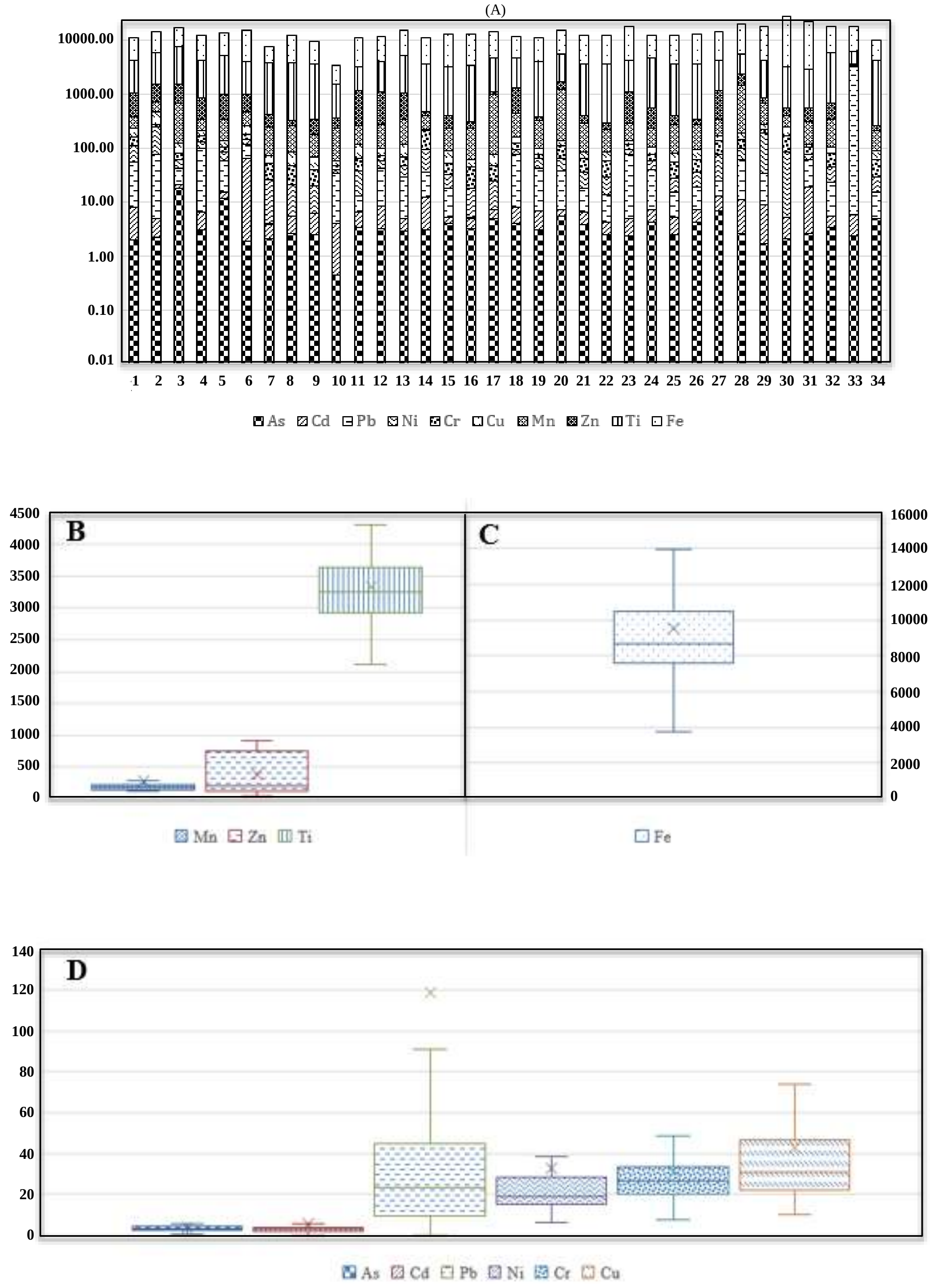

***Fig. 12:** Contents of heavy metals in the samples from Murmansk [ppm] (a) and their range [ppm] (b,c,d) without outliers.*

In the capital city of Poland, Warsaw, the share of heavy metals in the roughcast was similar to that examined in Cracow and Lviv. In just 4 of 22 samples cadmium content was defined at 1 ppm level, but arsenic came out in much more of them (17 samples), but its content never exceeded 5 ppm (Fig. 11). Copper concentration was approximately up to 10 ppm, but, on the contrary, a lead level reached up to 100 ppm and it was high in every sample. Manganese load was rather minor and not exceeding tens of ppm, likewise nickel and chromium. The content of zinc reached up to 500 ppm, titanium - 100 ppm, and iron - a few percentages by weight.

Very high shares of arsenic and cadmium were found in samples from Murmansk-both metals had content up to 10 ppm and above that level (Fig. 6). The average lead load was about 50 ppm, but in one sample it reached above 3158 ppm (no. 32). Contents of nickel, copper, and chromium ranged from 20 to 30 ppm and for manganese - 50 ppm. Zinc share reached up to 100 ppm. On the other hand, titanium content was a few thousand parts per million at the maximum level and iron share took a few percentages by weight. Those results from Murmansk, in comparison with results from *Lviv* and *Warsaw*, lead to the conclusion that high pollution is correlated with traffic in the city.

DISCUSSION AND CONCLUSIONS

Even though deposition takes place mostly on the soil's top layer, not walls [28], the roughcast is the recipient of pollutants in the first place, even before they reach the surface. The highly porous structure and rough texture of the coating of the building favor settlement of the particular matter and toxic chemical compounds embedded in them. It is presumed that roughcast acidity (pH) can be related to the concentration of the substances accumulated (i.e. Cu) [1,17,29]. Possibly, the chemical composition and texture of the roughcast itself can condition the deposition of specified types of impurities, which states we are going to investigate in future papers.

Increased deposition, especially of pollutants with some specific characteristics like heavy metals, takes place in areas with whetted production of aerosols, particular matter, and chemical compounds. These are in general: the main streets in the cities, industrial districts, or residential areas [19,30-34]. Trackways and urbanization have a non-negligible influence on concentrations of heavy metals- lead, zinc, copper, nickel, and chromium in particular.

The highest concentrations of lead and zinc are signs of intensive automotive traffic in past years and nowadays - exhausts of leaded gasoline, worn-out tires, and corrosion of electroplated car parts - as well as a high share of metal processing industry in the city's economy [20-24, 32, 35]. Also, exhausts from Diesel engines have a significantly negative impact on the air quality of the cities as they consume oil. Traffic fumes cause getting impurities like zinc, nickel, copper, and cadmium into the atmosphere, not only mention the products of combustion but also car-maintaining substances like grease, antioxidants, or anti-corrosion agents [35,36].

Heavy metal (i.e. nickel or copper) compounds are typical for cities with historically or currently working smelters of ferrous and non-ferrous metals (see examples of Murmansk and Cracow) [35-39]. Intriguing sources of copper in the cities are tractions of city trams and municipal railways. Moving trains cause lifting rust dust from tracks which later settles on nearby objects and buildings; the closer the location of the object to the tram or railway tracks, the higher the final concentration of copper or iron compounds [35]. A significant source of copper and nickel in cities is also waste residues whose particles could be dislocated by wind shears and deposited on the covering of the buildings [40].

The disproportion of heavy metal contents is connected to the location of buildings and their proximity to metallurgic plants. Contamination of roughcast in Monchegorsk and Cracow is sourced mainly in heavy industry plants; in the case of Cracow, the traffic and transportation overlap mentioned issue. Also, geographical location has a crucial role in pollutant distribution - Cracow lies in a hollow, and heavy metal compounds are hardly blown outside the city. The alike situation occurs in Monchegorsk: the disproportion of the distribution of the pollutants is connected to the directions of wind shears as well as building orientation against them. Noteworthy is the fact that Monchegorsk is a young city (70 years old) and pollutants have accumulated in a relatively short period.

A significantly high share of pollutants in roughcast has been found in Lviv, which is mostly a result of automotive traffic in the city; most of the roads run directly across the city center. Moreover, the intensity of the traffic is much higher in Lviv than in i.e. Lublin, where a lot of main roads are located in suburbs or at a greater distance from the city center.

A supremely important factor of heavy metal pollution in urban areas in moderate and polar climate zones is incinerating fossil fuels for heating purposes. This process leads to releasing compounds like arsenic, cadmium, and nickel, among others into the atmosphere, which then stick to the aerosols, soots, and ashes [37] and later settle on buildings.

Roughcast analysis allows examining the state of the environment in the places, where industrial plants or other enterprises, that had a significant impact on the environment, were located and their activity has had a long-term influence on the well-being of the population of the city. Through this analysis new possibility is open-pollutants, emitted from old industrial plants through other activities, can be determined even though the factory is already closed.

Chemical compounds and particular matter from air accumulate on the coverings of the buildings and can be an important factor in the examination of the state of the environment in the city, but some processes can lower the concentration of heavy metals in roughcast. Facades are continuously exposed to changing weather conditions; rain and snow, especially heavy rainfall or blizzards cause the washing and diluting of deposited pollutants. Nevertheless, also temperature and insolation, that change not only through the day but also through the year, have an impact on the covering of the building affecting changing chemical bounding and accelerating the roughcast erosion process.

Particular matter of eroded roughcast dust with bounding-changed impurities trapped inside comes across the soil and is an additional source of i.e. heavy metals. Erosion increase**s** the bioavailability of elements and chemical compounds [29, 41, 42] - through this process, plants and vegetables absorb detrimental substances easier and that leads to an increased supply of i.e. heavy metals to the human body. The changes in chemical bonding are also a reason why crumbed roughcast, walls or bricks should not be used as low-cost ballast or as a way to fill the unevenness of a field. For chemical analysis of the bioavailability of heavy metals in roughcast, the *Tessier et al.* (1979) method can be used [43]. Sequential extraction allows not the only examination of the bioavailability of element and chemical compound but also for determination of their origin, type of occurrence, and why it is transported [43].

Analysis of specific layers of roughcast could be used as proxy data to specify the climate in the past-inclusions of oxygen could tell us what the temperature was when the roughcast was applied. Consequently, paleoclimate analysis through roughcast examination could be conducted. However, most of the samples we possess are from the XX century or the end of the XIX century-there have been more precise methods already used for paleoclimate investigation. Also, there is an issue of the impossibility to determine the margin of error in the examination of oxygen inclusion in roughcast samples. Nevertheless, historically the oldest roughcast comes from ages when weather conditions had been noticed and written down.

All the examples of the cities of Poland, Ukraine, and Russia show that regardless of the climate zone, heavy metal pollution is caused mostly by anthropogenic activities (powerplants, ironworks, smelters, household furnaces) and is deepened by the character of the terrain (evenly polluted Cracow and differently polluted Monchegorsk). In the cities in agricultural regions, the concentration of heavy metals is significantly lower than in cities in regions where the economy is based on heavy industry. But commonly, the highest content of heavy metals comes from traffic.

Analysis of roughcast from 6 cities shows that the initial value of heavy metals in the roughcasts are not having a substantial impact on the final concentrations. The advantage of this method is the possibility of multiannual analysis of air quality in cumulated long-term episodes. In contrast to low vegetation examination, which can be analyzed only during the period of vegetation and only in this one particular year, while roughcast analysis can be performed annually and the outcome will show the result providing data for all years [44]. Nevertheless, during the renovation, a new layer of roughcast is applied; this way the older layers are protected from weather erosion, washing, and rinsing of heavy metals and other impurities.

Conducted isotopic examination on precipitates collected on brick structures in Berno, Switzerland, shows that the sulfur isotope $BaSO_4$ derived from a precipitate sample gives $\delta^{34}S$ =+18.3‰ and it points to Jurassic evaporites as a sulfur source [45]. The result of SO_4 oxygen, $\delta^{18}O$ = + 8.0‰ vs. VSMOW, indicates the influence of the water from atmospheric precipitation. This confirms that impurities accumulate and infiltrate the surface of the building and exhibit validity of conduction of roughcast analysis.

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