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ASSESSMENT OF CHEMICAL POLLUTANT LEVELS IN THE INDOOR AIR OF STRAW BALE HOMES. A CASE STUDY

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Abstract

More and more people are interested in environment protection and sustainable development, which appears in the demand to live in "zero-energy" buildings or natural buildings. Most of natural materials used in building constructions and some constructions techniques have been used for hundreds or even thousands of years, but the straw bale technology was developed in the 20th century and became popular not only in less developed world regions but also in the USA and Europe. The research related to natural buildings focused on thermal insulations or durability and very little is known about the indoor environment quality. The aim of this study was the assessment of indoor chemical pollutant levels in the buildings constructed in the straw bale technology located in the rural area of Eastern Poland (Lublin region). Investigation in the inhabited premised were conducted using passive sampling (Radiello dosimeters) followed by HPLC and UV spectroscopy. The data showed that the NO_2 and ozone concentrations were similar to the levels measured in typical buildings located in the same area, while the levels of several identified VOC and carbonyl compounds were lower. Despite the structure material, the human factor seems to be the most important parameter impacting indoor air quality.

***Keywords**: Natural buildings – natural materials, Environment protection, Sustainability, Indoor air quality, Chemical pollutants, Volatile organic compound, Carbonyl compounds*

Introduction

European citizens are more and more interested in the environment protection and sustainable development, including their way of living. It is reflected, among other activities, in the demand to live in "*green buildings*", "*low energy*" buildings or even "*zero energy*" buildings.

Zero energy buildings and green buildings are typically defined and categorized by different certification programs. The environmental responsibility of green buildings should cover their full life cycle from design and construction, through maintenance to deconstruction [1].

Another popular trend in the building industry corresponds to natural buildings. There is no certification program devoted to natural buildings. The main idea of natural buildings is the need to minimize the impact of buildings on the environment, without decreasing comfort and indoor air quality. Therefore, the natural buildings are a range of materials and building systems that focus on sustainability. One way to achieve sustainability is to use long-lasting materials; another way is to use the minimally processed, plentiful or renewable resources, or recycled

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products. The technologies used in natural buildings rely more on the usage of locally available materials as well as human labor, rather than on sophisticated technologies.

The list of usable natural materials is long: stone, earth, wood, straw, rice-hulls, bamboo, clay and sand. All of the above-mentioned materials have been used in different construction types for hundreds or even thousands of years. Among the techniques popular up till now, we have to mention: stone, adobe, cob, cordwood, earthbag, hempcrete and rammed earth. The examples of historical buildings constructed in these technologies can been found all over the world; there are also new investments, such as Copernicus Centre for Science in Warsaw, Poland, the longest rammed earth building in Europe, opened in 2010.

However, the highest interest lies in the straw bale technology. Straw bale buildings are usually erected by placing bales in rows on a foundation or footing including a moisture barrier between them. Surface wire meshes, bamboo, or wood are used to tie the bale walls together. Afterwards, they are plastered by means of a earth/clay render or lime-based formulation (Fig. 1).

Grasses and straw have been used for building since the prehistoric times; however, the modern straw bale technology emerged in the early 20th century in Nebraska, USA, with the introduction of first machine-manufactured modular bales onto the market [2].

Fig. 1. Photo of a straw bale house during construction (Lublin area, Poland, 2017)

Following the energy crisis of 1970s, there has been growing awareness pertaining to the environmental effect of the anthropogenic activity. The straw bale technology became popular in less developed world regions, where easily available local materials and low costs are the most important factors, but also in the USA and Europe, where the technology was the image of ecological awareness. The first modern buildings constructed in the straw bale technology in northern China were implemented in 1998 [3].

This technology is highly prospective for its contribution towards the building sector decarbonization [4]. Straw bales absorb carbon, whereas the walls made of this material are characterized by favorable thermal insulation properties, greatly reducing the heating/cooling energy loads in houses built using this technology. Research shows that straw bale buildings are durable [2, 3, 5], present good hydrothermal performance [3, 6-8] and thermal capacity [2].

Very little is known about the quality of indoor environment in the existing, inhabited houses constructed in the straw bale technology. In the developed countries, people spend more time indoors then outdoors; therefore, the indoor air quality has higher impact on their health, well-being and comfort, as well as efficiency. The time spent in houses easily exceeds 8 hours per day, and has increased significantly during the COVID-19 pandemic and due to the on-line work from private houses, what was commonplace for thousands of people. Therefore the quality of indoor air in residential premises is of growing importance.

There is limited information in the literature about basic inorganic pollutants (carbon dioxide) and bioaerosols in straw bale houses. *Ramez et al.* [8] examined the carbon dioxide and bioaerosol levels in two straw bale buildings in Estonia and measured the CO_2 levels from 569±164 to 680±130ppm. The fungi concentrations were higher indoors (compared to outdoors) in winter and autumn and lower indoor in spring.

The data about the chemical pollutant levels in natural buildings are very scarce. One can find general statement on the website (StrawBale.com, Oct.1, 2019):*"A straw bale house, if properly designed and built with the right materials, can be free from VOCs and other toxins*", but the general belief that if something is "natural", it should be good for humans, needs an empirical proof.

There is lack of information about the levels of Volatile Organic Compounds (VOCs), as it was stated at natural building website. Therefore, measurements of selected VOCs and carbonyl compound as well as NO_2 and ozone were undertaken in the Eastern part of Poland.

Materials and Methods

The straw bale technology became popular in the rural area in the Lublin region – Eastern Poland about 15 years ago. Poland has problem with the outdoor air pollution in cities and some southern heavily populated areas, but Eastern part of Poland is less developed. Despite the main city in the area (Lublin), the land in the north and south are agriculture areas with some national parks (natural conservation area). It is selected for living by ecologically concerned people. Some of those people, despite living in a less polluted region, decided to live in natural buildings. Therefore, it was possible to select a few homes in the straw bale technology for the examination of the indoor air quality. The location of houses under investigation is presented on the map in Figure 2.

Fig. 2. Location of examined houses in Eastern part of Poland

All the examined houses were located south or north from Lublin, built in the straw bale technology and equipped with mechanical ventilation with heat recovery, but differ in the number of inhabitants, heating systems (winter season), wall finishing (clay paints vs. traditional paints), and floor finishing (ceramic tiles, wood panels, laminate panels). All houses have a living room with open kitchen, but different type of ovens (electric or gas). All occupants claimed to be non-smokers.

Three series of sampling (with 7 days intervals) were performed in bedrooms and living rooms (with open kitchen) during the spring season.

VOCs, aldehydes, nitrogen dioxide and ozone were collected by a passive technique with RAD 130, RAD 165, RAD 166 and RAD 172 Radiello dosimeters, respectively, for 3-7 days, regardless of the pollutant and expected level. This method provides the average concentrations for the duration of samplings as results and does not generate noise; therefore, it was suitable for measurement in living premises during regular occupancy.

The determinations of the final compound concentrations covered by this measurement were made using the following analytical techniques. Volatile organic compounds were detected by gas chromatography with a flame ionization detector (Trace Ultra, Thermo Scientific). Carbonyl analysis was performed with high performance liquid chromatography (Water Action Analyzer). Nitrogen oxide and ozone were determined by ultraviolet spectrometry (U-1500, Hitachi). The analytical procedures used in this study are described in detail elsewhere [9, 10].

Thirteen VOCs, namely benzene, 1,2-dichloropropane, trichloroethylene, toluene, chlorobenzene, ethylbenzene, (m+p)-xylene and o-xylene, styrene, α-pinene, 1,2,4-trimethylbenzene, 1,4-dichlorobenzene and limonene were detected and quantified. The target carbonyls were metanal (formaldehyde), ethanal (acetaldehyde), 2-propenal (acrolein), propanal (propionaldehyde), butanal (butyraldehyde), benzaldehyde, pentanal (pentalaldehyde) and hexanal (hexaldehyde). The detection limits for VOCs ranged from 0.01 to 0.05μg/m^3 depending on the compound, as well as from 0.1 to 0.9μg/m^3 for carbonyl compounds, 0.9μg/m^3 for NO_2 and 1.0μg/m^3 for ozone.

The compounds covered by this research are considered to be the main gaseous pollutants of the indoor air. Long-term exposure to these substances may cause deterioration of health among residents. Most indoor VOCs negatively affect the functioning of the nervous and respiratory systems. Furthermore, BTEX chemicals (benzene, toluene, ethylbenzene and sum of xylenes) can be the cause of carcinogenesis [11, 12]. The World Health Organization (WHO) has recommended to monitore benzene but has not proposed any threshold level [13]. Some aldehydes and ketones (carbonyl compounds) are toxic, but formaldehyde at concentrations exceeding 1.0μg/m^3 is considered a problem [12]. Most of carbonyl compounds are known as eye irritants: therefore, they are believed to cause the sick building syndrome [14].

Opinions about carcinogecity of formaldehyde vary, although all authors agree that this carbonyl compound is eye and nose irritant [15]. In 2006, IARC published a report with strong statement that formaldehyde is carcinogenic to humans and listed HCHO in Group 1 [16].

Formaldehyde is the only carbonyl compound regulated in residential areas in some countries, but Polish regulations for indoor air premises established limit values for six carbonyl compounds: formaldehyde, acetaldehyde, acrolein, propanal, butanal and benzaldehyde [17].

U.S. EPA listed acrolein as one of 188 most hazardous air pollutants [18]; it was found to contribute to children's asthma and pulmonary toxicity. It was also confirmed that the women exposed to cooking fumes developed lung cancer [19]. In addition, studies proved that acetaldehyde (ethanal) causes eye and lung irritation [20].

NO_2 was reported to exacerbate asthma and cause wheezing [21, 22]. Although the WHO guidelines pertaining to NO_2 exposure were at 200μg/m^3 for 1 hour and 40μg/m^3 for per year, adverse health effects have been reported [13] at much lower levels than those for 1 hour average [23].

The selected pollutants were found as abundant during investigation in the same area in the houses built in traditional technologies, undertaken two years prior to this research.

Results and Discussions

Table 1 gathers the results of measurements in the examined houses as well as the results for the houses built in conventional technologies located in the same area, where a similar campaign was performed two years before by the same authors (unpublished data).

As it may be seen from the table, the highest concentrations were measured for carbonyl compounds, namely hexanal, metanal (formaldehyde) and etanal (acetaldehyde). This result might be expected in the inhabited houses, as hexanal comes from human effluents and cosmetic products, and formaldehyde is the most abundant pollutant, emitted from a number of finishing materials commonly used in indoor premises.

Table 1. Concentration of selected VOC, carbonyl compounds, NO_2 and O_3 ($\mu g/m^3$) in the examined premises and in the typical, conventional houses in the neighborhood

Compound	Straw bale houses	Limit Polish regulation [17]	Regular houses
benzene	0.1-0.46	10 /5*	1.20-5.60
1,2-dichloropropane	nd	-	1.10- 10.10
trichloroethene	nd	100	nd
toluene	0.68-1.13	200	9.25- 22.10
chlorobenzene	0.31-0.99	15	0.50-5.78
ethylbenzene	0.10-0.13	10	0.20- 1.25
xylenes (total)	0.46-1.10	100 /50*	2.10 – 10.20
styrene	0.18-0.50	20	0.80-4.60
1,4-dichlorobenzene	0.30-2.56	30	1.15-46.55
1,2,4-tri methylobenzene	0.25-1.45	30	nd – 4.0
a-pinene	0.15 – 0.30	-	0.10 – 0.35
limonene	0.10 – 0.30	-	0.20 – 0.35
formaldehyde	9.80-15.90	50 /20*	18.50-40.10
acetaldehyde	5.74-16.50	20	9.30 – 31.20
acrolein	nd – 5.51	10	nd – 7.30
propanal	nd – 3.36	100	nd – 19.40
butanal	2.50-5.30	50	nd – 19.85
benzaldehyde	nd – 4.50	100	nd – 5.30
pentanal	5.50-11.95	-	nd – 60.50
hexanal	12.50-25.55	-	0.60 – 75.80
nitrogen dioxide	1.0 – 5.7	-	no data
ozone	46.3 – 55.95	100	43.15 – 73.98

nd – not detected
/* - proposed changes

The main sources of formaldehyde in the indoor air are: building materials and household products, as well as open fire sources (fuel-burning appliances and tobacco smoking). Formaldehyde, alone or in combination with other substances, is added to many manufactured goods. It is used, for example, to add permanent-press qualities to textiles (clothing and draperies), as a component of glues and adhesives, as well as a preservative in some paints and coating products [24].

Among the most significant sources of formaldehyde, the following can listed: particleboard (used as sub-flooring and shelving in furniture); hardwood plywood paneling (used for floor covering), and medium density fiberboard (used for drawer fronts, cabinets, and furniture tops) [25, 26], which means that pressed wood products manufactured with the use of the urea-formaldehyde resins can be considered the most important source of HCHO indoors.

Surprisingly, low-VOC paints seem to be a significant source of formaldehyde. Due to the environmental-friendliness, latex paints have become popular, especially among the allergic people [27, 28]. Formaldehyde has been identified as an important secondary pollutant originating from cleaning products in the presence of ozone [29, 30].

Buildings under investigation are "natural" because of the construction material used, but the decorations and furniture in the examined houses are typical for this part of Europe in 20th and 21st century. Most of houses were finished with lime and clay paints, just after construction, but after a few years, during renovation and re-painting, owners did not pay much attention to use the same materials, and conventional latex paints were applied.

Acrolein constitutes another aldehyde which is widespread both indoors and outdoors. It is generated through incomplete combustion of organic matter, and oxidation of various atmospheric pollutants, including 1,3-butadiene, which is a constituent of vehicle exhaust gases. The most probable indoor sources of acrolein include tobacco smoke, cooking oil, incenses, as well as candle and wood burning [31]. Formation of acrolein might occur in the course of various oxidation processes involving indoor VOCs [32].

The indoor sources of acetaldehyde include environmental tobacco smoke, in addition to carpeting and particle board furniture [33].

Lower levels of formaldehyde, acrolein and acetaldehyde compared to traditional houses and literature data from different countries, e.g. France [33, 34] and Sweden [35, 36], support the information about their possible sources. Open fire sources were found in one house only, all occupants were non-smokers and they did not report any smoking visitors during the measurements period. It may also explain low levels of acrolein.

The highest concentrations were found for hexanal, the most abundant carbonyl compound originated from personal hygiene products and oxidation reactions involving ozone [37]. It might be supported by the data in figures 3 and 4, where the comparison of pollutant profiles for the house of the highest concentrations and the lowest concentrations are presented. Carbonyl compounds are presented in figure 3, while VOC in figure 4.

In both houses, the concentrations of hexanal were found to be at similar levels in bedrooms and in living rooms. Living rooms are larger open spaces used for limited time (occupants are at work or school during the day hours) and usually better ventilated. Bedrooms are of smaller volume, occupied for minimum 8 hours during the night. The measurements were undertaken in late spring, when temperatures were so moderate that mechanical ventilation was switched off, but windows were closed during the nights.

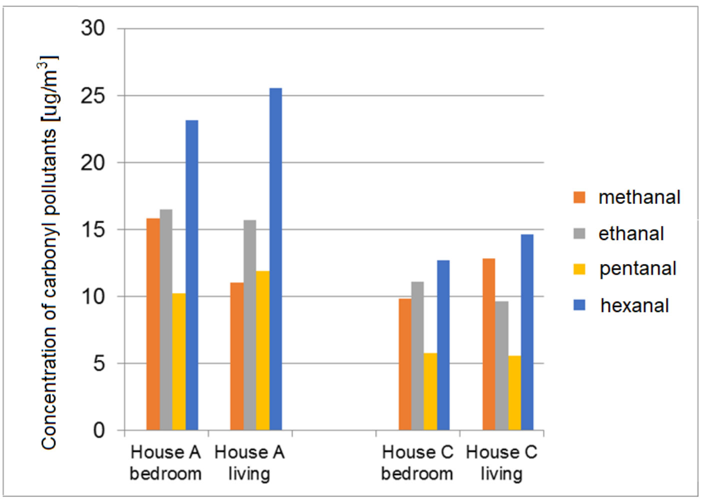

Fig. 3. Comparison of the concentrations (μg/m^3) of the examined carbonyl pollutants in the house with the highest concentrations (A) and the lowest concentrations (C) measured

The highest concentrations in the bedroom in house A were measured in the bedroom of a teenage boy, who spent more time in his room than in common area and who was very active in sports (his clothes were an additional source of human effluents). The lowest concentrations were in relatively new house (3 years old) with original clay finishing.

All the examined carbonyl pollutants were found in much lower concentrations than the permitted levels (Table 1).

The levels of benzene, toluene, styrene and ethylbenzene were very low. In one of the examined houses higher levels of 1,4-dichlorobenzene, compared with other houses were measured, but still much below the permitted ones, and much lower than in regular houses in the neighborhood. Most of the finishing products which were sources of these pollutants in the past are no longer in use. Nowadays, outdoor air and combustion in vehicle engines is considered the most important source of benzene, toluene and ethylbenzene indoors [38]. The lowest levels of VOC were found in the same house were carbonyl concentrations were the lowest (house C). But the highest concentrations of measured VOCs were determined in the house marked ‘B’, located close to regional road.

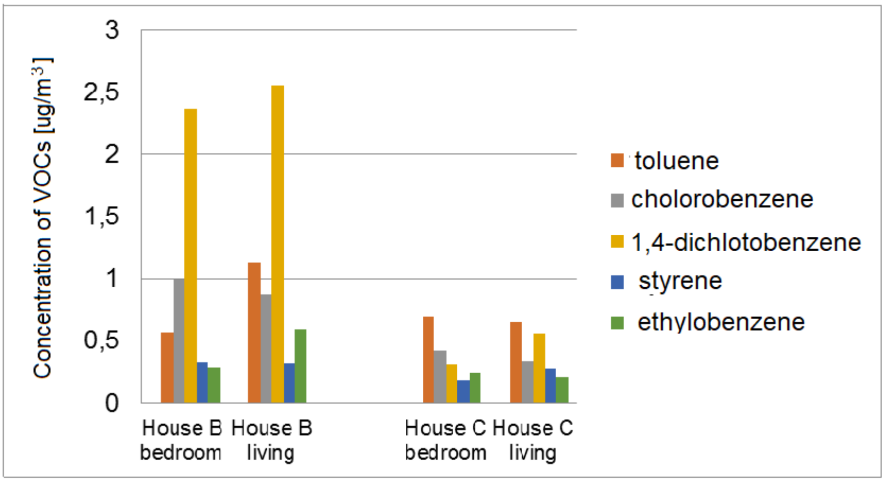

Fig. 4. *Comparison of the concentrations (μg/m^3) of the examined VOCs in the house with the highest concentrations (B) and the lowest concentrations (C) measured*

Limonene and a-pinene, two terpenes commonly used in the house cleaning products and personal hygiene products, were found above the detection levels in the all premises under investigation, although their concentration were low and at the similar levels like in regular houses.

The nitrogen dioxide concentrations were at low levels, within the range 1.0 – 5.7μg/m^3. The highest concentration was measured in the living room with open kitchen equipped in the gas stove. All other houses under examination were equipped with the electrical stoves. The ozone levels were very similar in the all examined houses (46.3 – 55.95μg/m^3) and reached about half of the permitted level (100μg/m^3). The main sources of ozone indoor are outdoor air and electronic equipment, mainly laser printers. Laser printers were not found in any of the examined indoor environments; therefore, outdoor air remains the suspected source. The nitrogen dioxide concentrations are impacted by outdoor air too.

It might be confirmed by the fact that ozone and nitrogen dioxide concentrations measured indoors were at very similar levels, like typical concentrations of these pollutants in the late spring season in the investigated area. Outdoor ozone was within 16-73μg/m^3, while outdoor nitrogen dioxide within 2.7-17.2μg/m^3.

The data from the previous campaign in the traditional houses located in the same area performed two years prior to the discussed measurements are gathered in table 1, too. As it may be seen, all examined VOC and carbonyl compounds were found at slightly higher or considerably higher levels in the traditional houses. However, a detailed examination is required to perform a comparison, because majority of the examined conventional houses were naturally ventilated and half of them were equipped in gas stoves.

This does not applies for terpenes (limonene and a-pinene), as they are originated from cleaning agents, personal cosmetic products and air fresheners, so their concentrations depend on human activities rather than materials used indoors. Moreover, the levels of ozone are very similar in the all straw bale and traditional houses.

Conclusions

All the targeted pollutants were detected in the examined houses although at low levels. The levels were lower than in a similar investigation undertaken two years earlier in the same area in the houses built in the traditional technologies. The number of examined houses is limited, and conclusion cannot be generalized, but one cannot state that natural buildings, namely straw bale buildings, are VOC free.

The outdoor air might be the main factor impacting the NOx and ozone levels. However, the VOC levels are highly dependent on finishing materials and occupants' activities.

All houses were painted using clay paint at the beginning, but when renovations were needed, latex paints were used. The furniture used was typical, wood or wood-like boards, which could explain formaldehyde levels. Cooking and oven type are among other important factors influencing the indoor air quality.

Despite the structure material, the human factor seems to be the most important parameter impacting the indoor air quality, in respect to habits and activities, as well as decisions about finishing materials and renovation.

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