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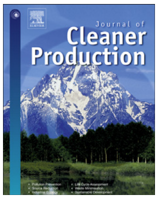

Evaluation of indoor air pollutants in 100 retrofit residential buildings from Romania during cold season

Mihail Simion Beldean-Galea [a], Tiberius Dicu [a, *], Alexandra Cucoş [a], Bety-Denissa Burghele [a], Tiberiu Catalina [a, b], Marius Botoş [a, c], Ancuţa Ţenter [a], Kinga Szacsvai [a], Alexandru Lupulescu [a], Istvan Pap [a], Gabriel Dobrei [a], Mircea Moldovan [a], Arthur Tunyagi [a,d], Ştefan Florică [a,e], Vlad Pănescu [a], Carlos Sainz [a,f]

[a] *Babeş-Bolyai University, Faculty of Environmental Science and Engineering, "Constantin Cosma" Radon Laboratory (LiRaCC), Cluj-Napoca, Romania*
[b] *Technical University of Civil Engineering of Bucharest, Faculty of Engineering Installations, Bld. P. Protopopescu 66, Bucharest, Romania*
[c] *Tehnical University of Cluj-Napoca, Faculty of Civil Engineering, Str. C. Daicoviciu 15, Cluj-Napoca, Romania*
[d] *Babeş-Bolyai University, Faculty of Physics, Str. M. Kogălniceanu 1, Cluj-Napoca, Romania*
[e] *Babeş-Bolyai University, Faculty of Biology and Geology, Department of Geology, Str. M. Kogalniceanu 1, Cluj-Napoca, Romania*
[f] *University of Cantabria, Department of Medical Physics, Faculty of Medicine, Santander, Spain*

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ABSTRACT

The indoor air quality (IAQ) was in the last decades a major topic of scientific research due to evidences of adverse effects of different chemicals on the human health. The retrofit of the existing buildings in order to increase the energy performance without proper implementation may have a negative effect on IAQ. This paper is focused on a comprehensive evaluation of indoor air pollutants in 100 retrofit houses from five Romanian cities, during cold season. Radon, volatile organic compounds (VOCs) and carbonyl compounds were determined using passive samplers. At the same time, continuous, real-time monitoring of radon and carbon dioxide concentrations, as well as indoor physical parameters, such as temperature, atmospheric pressure and relative humidity was carried out. In 95% of the houses the formaldehyde concentration was higher than the guidevalue. The most present volatile organic compounds were limonene, heptane, carbon tetrachloride, tetradecane and a-pinene. A statistically significant difference in radon concentration was observed between the two sampling methods, which highlights the importance of temporal variability of indoor pollutants. High radon values were correlated with the lack of adequate ventilation, an aspect underlined by high CO_2 concentrations. Additionally, the study finds that indoor air pollutants can be attributed to three main factors: the physical characteristics and usage of the indoor environment (volatile organic compounds and carbonyl compounds), the properties of the sub-slab soil (radon) and the occupational factor (CO_2). Each factor should be treated separately when indoor air quality management is addressed.

1. Introduction

In the last decades, the worldwide rush towards technological and economic developments has led to a deterioration of the environmental air quality. In this trend, the anthropogenic air pollution is the second global environmental threat after the climate change (Podstawczyńska and Chambers, 2019). The World Health Organization (WHO) has long pointed out that the household air pollution, ranked 9th in the Global Burden of Disease risk list, represents the leading cause of disease and premature death in low- and middle-income countries as well as in the developing countries (Ferguson et al., 2020; Stanaway et al., 2018; WHO, 2016). This outcome is partly due to the urge for energy efficiency of buildings (ex: Directive, 2012/27/EU, 2012) that has led to post implementations issues. Numerous researchers observed that the requirements for high airtightness of buildings lead to a „health pitfall" due to the degradation of the indoor air quality, particularly in buildings where additional mechanical ventilation was not included in the retrofit works (Cucoş Dinu et al., 2015; Dales and Raizenne, 2004; Fisk et al., 2020; Földváry et al., 2017;

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* Corresponding author.
E-mail address: tiberius.dicu@ubbcluj.ro (T. Dicu).

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Jiránek and Kačmaříková, 2014; Pampuri et al., 2018; Sharpe et al., 2015a; Wallner et al., 2017a; Ye et al., 2017). Wallner et al. (2015) following a study conducted in 123 buildings in Austria, found that the indoor air quality in highly energy-efficient, mechanically ventilated homes was higher than that of conventional homes with natural ventilation. Moreover, the perception of the indoor air quality was better for the inhabitants of energy efficient buildings with mechanical ventilation compared with those who lived in dwellings with natural ventilation only (Wallner et al., 2017b). The indoor air pollution has been associated with impaired health and performance in children and adults (Kim and Hwangbo, 2018; Meiboudi et al., 2016; Wyon, 2004). An increased risk of developing asthma has been associated with increased energy efficiency in homes without mechanical ventilation (Sharpe et al., 2015b). The interest for regulating residential pollutants is in high demand yet still underdeveloped due to the limited data available (Fisk et al., 2020; Stamatelopoulou et al., 2019; WHO, 2010; WHO, 2019). Although various studies on indoor air quality address volatile and semi-volatile organic compounds (Angulo Milhem et al., 2020; Fang et al., 2019; Fernández-Agüera et al., 2019; Park and Ikeda, 2006; Rösch et al., 2014; Yang et al., 2020), only a limited number of compounds have been addressed in the grey literature regarding exposure recommendations (ANSES, 2017, 2013; IARC, 1995; Umweltbundesamt, 2019; WHO, 2010). The indoor carbon dioxide (CO_2) has yet to receive the pollutant title, although the synergistic effect between CO_2 and VOCs has already drawn the scientific spotlight (Yang et al., 2020). Moreover, the indoor radon exposure is being addressed separately from that of other indoor pollutants (European Commission, 2014). Effects and risks for most common indoor pollutants are known to the extent that strategies to mitigate the problems can be created. However, new sources and accumulation issues have emerged for “old” pollutants in energy efficient buildings (Fisk et al., 2020), and some of them may react to produce secondary products whose effects are poorly defined (Billionnet et al., 2012). There is still a general drawback on addressing the quality of the indoor air as a whole, due to the lack of representative data.

Improving the IAQ requires knowledge on type of pollutants, sources, quantities and synergistic effects (Fernández-Agüera et al., 2019; Yang et al., 2020). Comprehensive studies are required in order to take the most appropriate decisions for mitigation. Conventional methods for IAQ monitoring often use short-term integrated measurements and complex apparatus (Yang et al., 2020). Although known for high accuracy these methods are unable to provide real-time temporal variability, essential for prevention and mitigation measures. Therefore, in order to implement a successful management of indoor air quality in the age of speed the spotlight is shifting towards real-time monitoring (Kelly and Fussell, 2019; Tunyagi et al., 2020).

The aim of this work was the systematic surveillance monitoring of indoor air in order to support the development of health-based guideline values and other guidance for key pollutants. In this context, the levels of VOCs, carbonyl compounds and radon have been investigated in 100 retrofit residential buildings. In order to investigate the relationship between the investigated parameters, indicators other than concentrations of the pollutants (i.e. ventilation rate, with CO_2 as a related marker, relative humidity, atmospheric pressure and temperature) have also been monitored.

2. Materials and methods

2.1. Eligibility and recruitment

The interest for home working has seen a constant increase during the recent years (Cerqueira et al., 2020). Consequently, the time spent indoors in the same environment has also increased. Moreover, the building retrofit of old residential buildings in Romania hardly ever includes mechanical ventilation, aspect of high relevance in qualifying the indoor air quality (Ferguson et al., 2020). Additionally, epidemiological studies (Bade and Dela Cruz, 2020) have observed an urban-related factor associated with an escalation of lung cancer deaths. Taking into consideration all these aspects, a large-scale radon research study has been carried out in 1000 energy efficient homes from five major cities of Romania (Cluj-Napoca, Timișoara, Bucharest, Iași and Sibiu) where, additionally to indoor radon risk (Todea et al., 2013), the outdoor pollution often exceeds the recommended threshold (IQAir, 2019). The buildings were selected among those representatives for each region’s architecture and based on randomized stratified sampling according to population size. The criteria regarding houses selection were as follows: (1) the age of the houses was greater than 2 years in order to avoid excessive emission of the target compounds; (2) the presence of thermal insulation of the walls or an upgrade of the windows, done within more than one year previous to the study; (3) volunteered participation in the study.

Following the application of detailed questionnaires on indoor air quality and passive radon measurements carried out in two campaigns of six months each, 100 houses were selected for detailed analysis (VOCs, carbonyl compounds, radon, CO_2 and indoor physical parameters). The selection criteria of the 100 houses (the object of the present study) were: (1) radon concentration higher than 250 Bq/m^3 in at least one investigated room; (2) poor indoor air quality, assessed qualitatively on the basis of the interviews with the owners of the houses about health symptoms and perceptions of IAQ.

2.2. Chemicals

For the qualitative and quantitative analyses of carbonyl compounds a mixture containing 2,4-dinitrophenylhydrazone derivatives of formaldehyde, acetaldehyde, acetone, acroleine, propionaldehyde, butyraldehyde and benzaldehyde, in concentration of 100 μg/mL of each compound, acquired from Supelco, was used. For VOCs analysis, a mixture containing 52 organic compounds (Japanese indoor air standard MIX 52) of different families (hydrocarbons, aromatic compounds, halogenated compounds, alcohols, esters, aldehydes, ketones, terpenes) in concentration of 100 μg/mL of each compound in methanol was acquired from Sigma Aldrich. From MIX 52, 10 compounds e.g. (n-butanol, 2-butanone, 1,2-dichloroethane, dichloromethane, 2,4-dimethylpentane, ethanol, ethyl acetate, hexane, 1 propanol and 2 propanol) were not quantified due to their overlap with CS_2 chromatographic pick. Acetonitrile of HPLC grade purity (Merk Darmstad, Germany) and Milli-Q water of 18.2 MΩ cm resistivity, prepared using a Milli-Q-Plus ultra-pure water system (Millipore, Milford MA, USA) were used for HPLC analysis. Helium in purity of 99.9999% (Linde Gas, Romania) was used for gas chromatographic analysis. The CR-39 passive detectors for radon measurements were etched in a solution of 6.25M NaOH (Merk).

2.3. Instrumental analysis

2.3.1. Carbonyl compounds

The determination of the selected carbonyl compounds were performed by high performance liquid chromatography (HPLC) coupled with UV–Vis detector (ISO 16000-4, 2011) using a HPLC model Shimadzu equipped with 10LC module pump, a 10LSD UV/Vis detector and a manual injection valve containing a sample loop of 5 μL. The separations were carried out on a reverse-phase column type NOVA-Pack C18 (300 × 3.9 mm, 4 μm particle size) purchased from Waters (USA). A mixture of acetonitrile and Milli-Q water (68:32, v/v) at a flow rate of 1.0 mL/min, in isocratic elution mode, was used as mobile phase. The detection was carried out at 360 nm. The LOD for formaldehyde was 0.1 μg/m^3 and for other carbonyl compounds was ranged between 0.1 and 0.9 μg/m^3 (Sigma Aldrich).

2.3.2. Volatile organic compounds

The determination of volatile organic compounds were achieved according to ISO 16200-2 (2000) and ISO 16017-2 (2003). For this purpose, a gas chromatograph model Focus GC equipped with a DSQII mass spectrometer and a TriPlus Autosampler (Thermo Electron Corporation) was used for the analysis. The separation was performed on DB-5MS column (25 m × 0.25 mm x 0.25 μm) with a gradient of temperature and using helium as carrier gas at a flow rate of 1.0 mL/min. The temperature of inlet, transfer line and ion source were set at 280, 250 and 220 °C respectively and for ionization a voltage of −70 eV was used. The detection of the compounds was performed in selected ion monitoring (SIM) mode and the quantification was done using the calibration curves method. Under the analytical conditions used in measurements, the limit of quantitation for 7 days exposure ranges from 0.001 to 0.1 μg/m^3 air, depending on the compound (Sigma Aldrich).

2.4. Samples collection and preparation

For the analysis of carbonyl compounds and VOCs, the air samplers were simultaneous collected using specific Radiello diffusive samplers purchased from Supelco (Brdaric et al., 2019; Langer et al., 2016; ISO 16000, 2011; Villanueva et al., 2018). Radiello cartridge adsorbents with 2,4-dinitrophenylhydrazine coated Florisil® were used for aldehydes sampling, while Radiello cartridge adsorbents with activated charcoal (CS_2 desorbtion) for VOCs sampling (ISO 16200-2, 2000; ISO 16017-2, 2003).

All the houses were monitored for a 1-week period, from October to December 2018. During the field test, natural ventilation was the only way for fresh air to enter the residences. The diffusive samplers were placed in the centre of living rooms at a height of 2.2 m above the floor and exposed for seven days. After exposure, the samples were collected and kept in refrigerator at 4 °C until their analysis. For carbonyl compounds the cartridges were desorbed in 2 mL of acetonitrile and analysed as 2,4-dinitrophenylhydrazone derivatives by HPLC, while for VOCs the desorption was done with 2 mL of CS_2 and the analyses were performed by GC-MS.

2.5. Passive radon measurements and analysis

The indoor radon measurements were performed by using CR-39 track detectors, provided by Radosys (Hungary). The detectors were placed in two successive campaigns of 6 months each, which allowed direct calculation of the annual indoor radon activity concentration. The detectors were installed by a member of the research team in the bedroom or living room on the ground floor of the house at a distance of about 0.5 m from the wall and 1.5–2 m from the floor, away from any direct airflow currents. At the end of the exposure period, the detectors were transported to the laboratory for processing and analysis, using the Radosys System. The used processing procedures have been previously described in detail by Cucoş et al. (2015).

The quality assurance and quality control of passive radon measurements in indoor air were performed by participating in several international intercomparison exercises and by the annual calibration of the Radosys System by the manufacturer (Rabago et al., 2020). The total uncertainty of the measurement was calculated to be between 8 and 15%, depending on radon concentration.

2.6. Active measurements

During the time when the diffusive samplers for VOCs and carbonyl compounds were exposed, an active device, called ICA system, was installed in each of the 100 houses. The ICA system presents sensors for continuous, real-time monitoring of radon and CO_2 concentrations, as well as indoor ambient conditions, such as temperature, pressure and relative humidity (RH). The ICA system was developed by the research team, the calibration of the sensors being performed in a special chamber within the laboratory; afterwards the ICA device was internationally validated (Tunyagi et al., 2020). The main characteristics of the sensors from the ICA system are listed in Table S1.

2.7. Energy evaluation of the houses

The energy performance of the building (EPC) is calculated considering the estimated energy under normal conditions of use of the building, including in this case, energy for heating, air conditioning (if it is the case), domestic hot water and indoor lighting. When calculating the energy performance of the building, multiple parameters are taken into account like: the thermal insulation of the envelope, type of glazing, heating and domestic hot water system, lighting fixtures, the location of the building - climatic factors, outdoor shading, and indoor physical required conditions. The energy evaluation is based on an energy audit using the national calculation method (MC 001, 2006).

2.8. Statistical analysis

The statistical analysis of the data was performed using SPSS 24, GraphPrad Prism 5 and OriginPro 2020b software. The statistical distribution of the data was evaluated using the Shapiro-Wilk test. For the comparison of the samples, the *t*-test or Mann-Whitney test (M-W test) was performed for two samples or Kruskal-Wallis test (K–W test) with Dunn's post-hoc analysis for more than two samples. In order to evaluate the intensity of the relationship between the examined variables, the Pearson or Spearman correlation coefficients were calculated. In order to evaluate the correlation between two variables by maintaining constant the effect of another variable, the partial correlation was calculated. As such, zero order correlation means the correlation without controlling for any other variable, first order for controlling the effect of one variable and second order correlation with control for two variables. The significance level α was chosen at 0.05.

The multivariate analysis was conducted using log-transformed data of pollutant as dependent variable and "ID of investigated city", "year of construction", "cellar under investigated room", "concrete screed", "type of floor", "type of ceiling", "construction material", "insulation type", "use of candles", "use of fireplace", indoor temperature, RH and CO_2 concentration as independent variables. Because the CO_2 concentration can be assimilated as an indicator for the ventilation rate, it has been included in the list of predictors (Fisk et al., 2020). The Stepwise regression procedure was used and only the factors that had a statistically significant influence on dependent variable ($p < 0.05$) were taken into account.

Principal Component Analysis (PCA) was performed in order to identify structures in the relationship between monitored variables. In order to have normally distributed variables, the raw or log-transformed data were used. Of the total monitored variables, those with a correlation coefficient greater than |0.3| were kept for PCA. For factor extraction, taken into account the sample size, the scree plot was used, the point of inflexion being used as the cut-off for retaining factors (Field, 2013). The oblique rotation using Direct oblimin method, with delta equal to 0, was selected in SPSS in order to calculate the variable loadings on each factor. Finally, the factor loadings with a value greater than |0.4| were considered.

3. Results and discussion

3.1. Building characteristics and energy performance of the building

The houses involved in the analysis were built between 1800 and 2015, the median of the construction year being 1972. Although most of the houses were built before 2000 (62%), this percentage is not representative for the general situation due to the preferential selection of those houses that had retrofit works. About 90% of dwellings present a thermal insulation applied on the outside of the walls, the main type of material used being polystyrene (94%). A similar percentage of homes have upgraded their windows, but only two houses have installed a mechanical ventilation system. In 34% of the cases, the cellar is present under the investigated room. About 80% of dwellings present a concrete slab under the floor, the main floor type being laminate flooring (62%). In 18% of cases, the participants mentioned that they use air fresheners, electrical room perfumes or scented candles in the investigated room. The main building characteristics of the houses involved in this study are summarized in Table S2.

Most of the studied building are well insulated, as such 43% of these are classified in the energy class B (125–201 kWh/m^2/year), 45% in class C (201–291 kWh/m^2/year), while only 3% are A class (less than 125 kWh/m^2/year). Among them there are also some buildings with deficient heating system (e.g. stove with low energy efficiency), thus their class is either D (5%) or E (2%), with a maximum energy consumption of 474 kWh/m^2/year. The energy classes correspond to the Romanian National reglementation according to the energy methodology (MC 001, 2006). For 99 houses an energy certificate was delivered presenting the energy consumption, greenhouse emissions and the proposed measures to improve the energy efficiency. To calculate the emission expressed in kg CO_2, the final energy calculate in the EPC was transformed firstly into a primary energy using the national coefficients (1.17 for gas and 2.62 for electricity) and afterwards this primary energy was multiplied by a factor of kg CO_2/kWh (0.205 for gas, 0.299 for electricity). It can be concluded that the average emission of the 100 houses is 56 kg CO_2/m^2. The total heated area of the 100 houses is 14,772 m^2, thus the total emissions of these buildings is 827 tonnes of CO_2/year.

3.2. Indoor air quality

3.2.1. Continuous IAQ monitoring

As was mentioned before, during the time when the diffusive samplers for VOCs and carbonyl compounds were exposed, in each house, using the ICA system, were continuously monitored parameters such as CO_2 and radon concentrations, temperature, pressure and relative humidity (RH). Generally, a diurnal pattern was identified with high radon and CO_2 concentrations during the nights and mornings, respectively low values in the afternoon (Fig. 1).

The CO_2 concentration depends mainly on the number of occupants, being a good indicator of the human bioeffluents emissions. Taken into account the fact that the measurements took place predominantly in the master bedroom, in 96% of cases two people were present during the measurements period with an average occupancy factor of 0.35 in the analysed room. As can be seen in Table 1, the mean CO_2 concentration is higher than 1000 ppm in over 50% of the houses. These results can be used as a surrogate of the occupational patterns (time spend indoor and the ventilation patterns). As such, for most of the monitored houses (46%), a common pattern was noticed, which involved the ventilation of the room by opening the windows once a day, either in the morning or in the evening. In less than 10% of the houses, the pattern showed a double ventilation period at an interval of about 12 h. In about 25% of the houses, insignificant ventilation periods were identified, behaviour that led to high indoor radon and CO_2 concentrations (up to 3083 Bq/m^3 and 4072 ppm, respectively).

A geometric mean of 22 °C was calculated for the indoor temperature, with a minimum temperature of 15 °C (three houses) and a maximum value of 30 °C (5 houses). The relative humidity showed a wide variation between 16% and 70%, with an arithmetic mean of 42%, the measured values being normally distributed.

3.2.2. Radon concentration

The indoor radon concentration was measured with passive detectors in two successive 6-month campaigns, which allows calculating the annual concentration. The annual radon concentrations show a log-normal distribution with a geometric mean of 309 Bq/m^3 (Table 1). In 52% of the investigated dwellings, the annual radon concentration is higher than the reference value of 300 Bq/m^3, established by national and EU legislation. This high percentage is due to the preference to select for detailed analyses those houses with radon concentration higher than 250 Bq/m^3. As can be seen in Fig. 2, a moderate correlation was obtained between the annual radon concentrations, through passive method and temporal radon measurements, by ICA system ($r_S = 0.65$, $n = 98$, $p < 0.01$). On average, the radon concentrations obtained with active method are higher (AM = 516 Bq/m^3) than those for passive method (AM = 356 Bq/m^3). This difference was statistically significant $t(97) = 6.39$, $p < 0.001$, and presented a medium-sized effect ($d = 0.55$). Taking into account the duration (one week) and the season (end of autumn and early winter) of the active measurements, this difference can be largely attributed to the temporal correction factor, which must be applied to any short-term measurement.

3.2.3. Carbonyl compounds concentration

The total carbonyl concentrations were calculated as a sum of the carbonyl compounds identified for each house. With a range between 94 and 1735 μg/m^3, the total carbonyl compounds presented a log-normal distribution with a geometric mean of 363 μg/m^3. For formaldehyde, the concentrations ranged from 25 to 332 μg/m^3, with a median value of 103 μg/m^3 and an identical geometric mean (Fig. 3a).

In the case of acetaldehyde, the range was between 14 and 581 μg/m^3, with a median of 70 μg/m^3. A median concentration of 125 μg/m^3 was obtained for acetone with a wide range of values between 5 and 1036 μg/m^3 and a geometric mean of 138 μg/m^3. Propionaldehyde was found in 59 dwellings, with a median concentration of 16 μg/m^3 and a maximum concentration of 150 μg/m^3. While acrolein was not detected in any investigated house, benzaldehyde and butyraldehyde were found in 25 dwellings with a median of 3.5 μg/m^3 and 13 μg/m^3, respectively (Table 1).

The distribution of formaldehyde concentrations was log-normal, while for acetaldehyde and acetone the distributions become log-normal only after eliminating the outliers (between 2 and 5% of raw data). From the weight of the carbonyl compounds to total carbonyl concentrations, acetone has a median contribution of 38%, followed by formaldehyde with 31%, respectively acetaldehyde with 22%. As such, their contribution to the total concentration of carbonyl compounds exceeds 90% in almost all studied houses, which could be a consequence of high emission rate of these

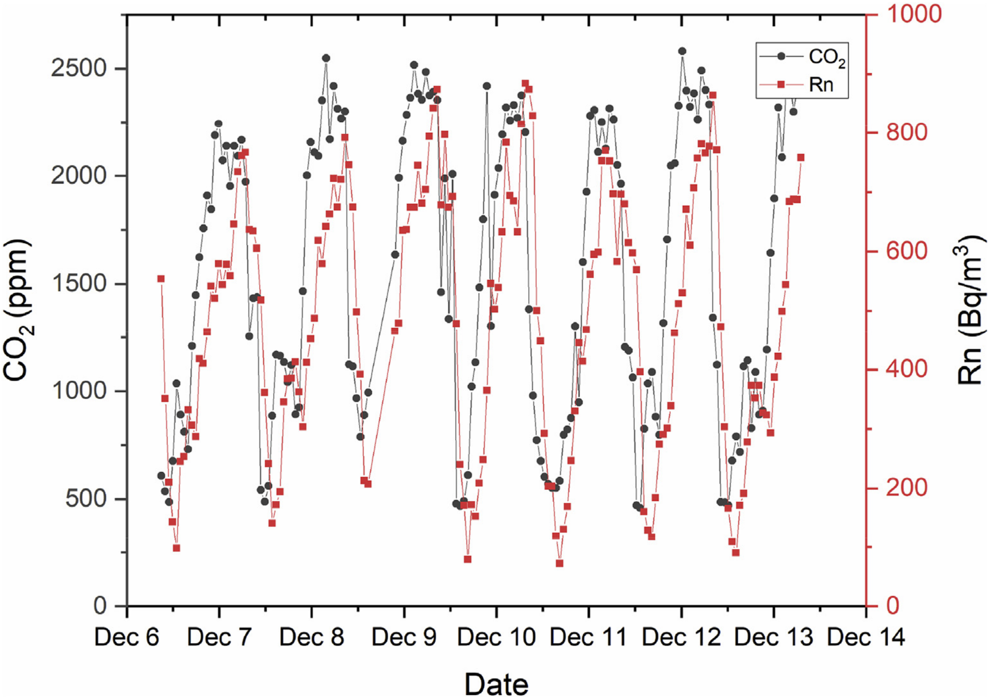

Fig. 1. Typical diurnal variation of radon and CO_2 concentrations during the measurement period.

Table 1
Descriptive statistics of some carbonyl compounds, CO_2 and radon concentrations, as well as indoor physical factors measured in the investigated retrofit houses.

Parameter	N[a]	Min.	Q1	Mdn.	Q3	Max.	GM	GSD	AM	SD
Annual radon conc. - passive method (Bq/m^3)	98	73	225	301	465	1709	309	1.7	356	213
Temporal radon conc. - active method (Bq/m^3)	98	99	311	428	689	1482	445	1.8	516	287
Formaldehyde (μg/m^3)	100	25	77	103	135	332	103	1.5	111	47
Acetaldehyde (μg/m^3)	100	14	49	70	97	581	73	1.8	89	76
Acetone (μg/m^3)	100	5	81	125	260	1036	138	2.2	188	167
Propionaldehyde (μg/m^3)	59	3	10	16	27	150	17	2.0	24	26
Butyraldehyde (μg/m^3)	25	1.7	4	13	70	92	17	4.0	35	34
Benzaldehyde (μg/m^3)	25	0.1	2.9	3.5	4.7	23.8	3.3	2.6	4.7	4.8
Total Carbonyl (μg/m^3)	100	94	258	360	486	1735	363	1.6	412	238
CO_2 (ppm)	95	432	696	1076	1355	3375	1064	1.6	1196	633
RH (%)	95	16	35	42	47	70	41	1.3	42	10
Temp (°C)	95	15	20	22	23	30	22	1.2	22	3.2
Energy consumption (kWh/m^2/year)	99	102	168	209	236	474	206	1.3	213	61

[a] N indicates the number of houses were the investigated parameter was eligible for analysis (value higher than detection limit or the detectors have been recovered); Q1 – First quartile (25% percentile); Mdn. – Median (second quartile), Q3 – Third quartile (75% percentile); GM – Geometric Mean; GSD – Geometric Standard Deviation; AM – Arithmetic Mean; SD – Standard Deviation.

compounds from building materials correlated with an inefficient ventilation of the room. As was mentioned before, only in two houses a mechanical ventilation system exists, as such the mainly way to ventilate the houses being achieved by opening the windows.

At a first glance, it can be noticed that the concentrations of the carbonyl compounds are generally high. Because in Romania there are no established threshold limits for the carbonyl compounds in residential buildings, the guidevalues recommended by different agencies such as Umweltbundesamt from Germany (Umweltbundesamt, 2018) or French Agency for Food, Environmental and Occupational Health and Safety (ANSES, 2013, 2017) were used. As such, in 95 houses, formaldehyde concentration, which is classified as a human carcinogen, is higher than the guideline value of 50 μg/m^3 (Umweltbundesamt, 2018), respectively 23% than the recommended value of 100 μg/m^3 (ANSES, 2017). The acetaldehyde, another carcinogenic compound, in about 25% of the investigated houses is higher than 100 μg/m^3, the guide value according to German recommendation (Umweltbundesamt, 2018). For 10% of the houses, the acetaldehyde is higher than 160 μg/m^3, the guidevalue recommended by ANSES for long indoor exposure (ANSES, 2013). A comparative analysis with other studies indicates that the values obtained in the present study are in line with those reported in China (Chen et al., 2017; Huang et al., 2017; Liang and Yang, 2013), in Taiwan (Wu et al., 2003), in Malaysia (Sakai et al., 2017) or in Portugal (Canha et al., 2017). However, the obtained concentrations for carbonyl compounds are twice as high as in Lithuania (Kauneliene et al., 2016) or other European countries (Salthammer, 2019), in Canada (Gilbert et al., 2005), in China (Dai et al., 2018), in Japan (Kishi et al., 2018) or even more than reported in Iran (Delikhoon et al., 2018), in Sri Lanka (Chan et al., 2018), in China (Cheng et al., 2018; Fang et al., 2019) in France (Langer et al., 2016) or in Sweden (Langer et al., 2015).

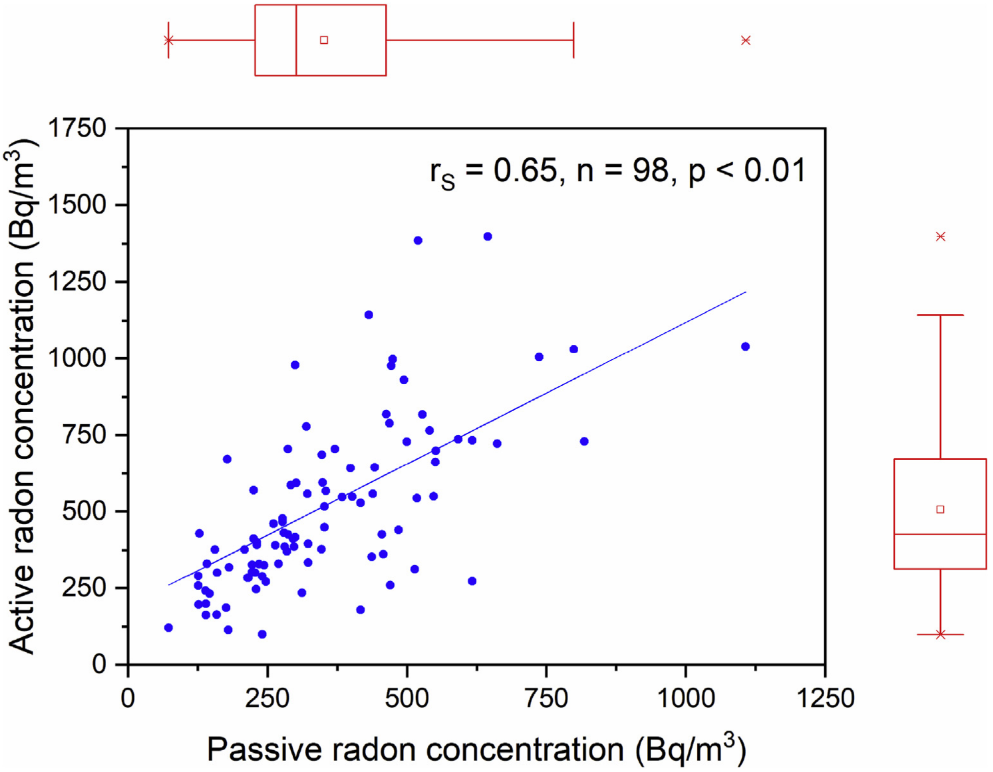

Fig. 2. The relationship between the annual radon concentration, measured by the passive method, and the radon concentration actively monitored for 1-week time period, using ICA system.

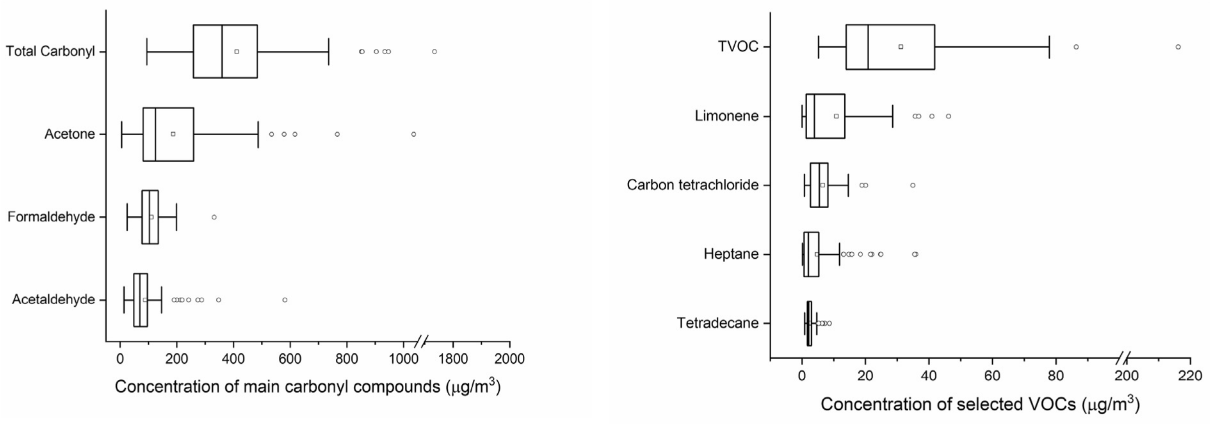

Fig. 3. The Box-plot representation for: (a) the total carbonyl and main carbonyl compounds and (b) total VOCs and main volatile organic compounds found in the selected houses (the dots represent the outlier values, greater than 1.5 × IQR, while the arithmetic mean is represented by square).

3.2.4. VOCs concentration

The results of the analysed samplers showed that from 41 selected compounds only 35 have been identified, from which 24 had an occurrence frequency greater than 90%. Trichlorethane had the lowest presence in investigated houses (49%). A detailed descriptive statistics of levels of VOCs is presented in Table S3. Only 12 (34%) volatile organic compounds showed a log-normal distribution. Among the VOCs with log-normal distribution are carbon tetrachloride, which had the highest GM (4.72 $\mu g/m^3$), followed by limonene (GM = 3.74 $\mu g/m^3$), tetradecane (GM = 2.27 $\mu g/m^3$) and heptane (GM = 1.97 $\mu g/m^3$) (Fig. 3b). In fact, for 83% of the monitored houses these four organic compounds, which can be mainly associated with odorants and cleaning products, represent more than 50% of the total VOCs (TVOC). Li et al. (2019) after a monitoring period of more than 24 months in 3524 Canadian houses identified limonene as the most abundant VOCs, but the calculated GM is about six times higher than in our study.

The comparison of the measured VOCs concentrations with the threshold limits for indoor atmosphere applied worldwide indicates that there is no exceeding of the recommended value neither for individuals compounds or TVOC. The presence of a high TVOC could indicate the existence of an important source, as well as a low ventilation of the investigated environment (Becerra et al., 2020). A wide range of values were calculated for the TVOC (5.2–216.23 $\mu g/m^3$), with a geometric mean of 23.23 $\mu g/m^3$. By comparing the obtained results with other reports, it can be observed that our results are two or three time lower with those reported in dwellings in China (Chen et al., 2017; Cheng et al., 2018; Fang et al., 2019; Huang et al., 2018) or in Malaysia (Sakai et al., 2017). Our results are comparable to those reported for the dwellings in Sweden (Langer et al., 2015), in China (Huang et al., 2019), in Japan (Kishi et al., 2018), in energy-efficient houses from France (Langer et al., 2016), in Lithuania (Kauneliene et al., 2016), in schools in Croatia (Brdaric et al., 2019) or are higher than reported in schools in Spain (Villanueva et al., 2018), in dwelling under ventilation conditions in Portugal (Canha et al., 2017) or France (Langer et al., 2016).

If the investigated compounds are classified according to their classes, it can be observed that the most prevalence compounds are esters and terpenes (36–43%), followed by aliphatic hydrocarbons (29–43%), halogenated compounds (13–20%), aromatic hydrocarbons (3–5%) and aldehydes and ketones (0.2–11.6%). Terpenic VOCs at mass concentration of up to 5% w/w were identified in cleaning products, limonene being a major compound in air fresheners, electrical room perfumes or scented candles (Milhem et al., 2020).

3.3. Multivariate analysis

3.3.1. City level

According to the non-parametric K–W test, the indoor temperature was significantly affected by the investigated city, $H(4) = 37.93$, $p < 0.01$. Pairwise comparisons with adjusted p-values revealed that the indoor temperature in Sibiu is significantly higher compared to those in the other four investigated cities (all $ps < 0.01$). This difference can be mainly attributed to the period in which the measurement took place and, secondly, to the microclimate particular to each city. The passive measurements of VOCs and carbonyl compounds were made successively in the five cities, as such Sibiu was the first selected city (mid-October), followed by Timişoara, Iaşi, Cluj-Napoca (November) and Bucharest (mid-December). A similar trend was found by K–W test applied to relative humidity, $H(4) = 33.0$, $p < 0.01$, in Sibiu the measurements for this parameter being significantly lower than in Cluj-Napoca, Timişoara or Bucharest (all $ps < 0.01$).

The formaldehyde concentrations were significantly affected by the investigated city as well, $H(4) = 13.9$, $p < 0.01$. Pairwise comparisons with adjusted p-values revealed that the formaldehyde concentrations in Sibiu are significantly higher compared to Cluj-Napoca ($p = 0.03$) or Timişoara ($p = 0.01$). By applying K–W test, a significant difference was found for TVOC concentrations, $H(4) = 35.9$, $p < 0.01$. The follow-up analysis showed a TVOC concentrations significantly higher for Bucharest than for Cluj-Napoca, Timişoara or Sibiu (all $ps < 0.01$). A detailed analysis of individual volatile organic compounds, indicated a ratio of approx. 3 times higher in Bucharest than in the rest of the cities for mesitylene, 1,2,3-trimethylbenzene, chloroform, limonene and carbon tetrachloride, 5 times higher for 2-ethyltoluene, methylethylcetone, durene and decane, 7 times for 1,2,4-trimethylbenzene, 3-methylpentane and styrene, and 10 times higher for m-,p-xylenes. In order to explain these results, looking at the period in which the measurements took place, can be found that the measurements in Bucharest were made in the middle of December, the period in which the households used cleaning products before the winter holidays. Taking into account the sample size of the results after their division according to the city in which they were performed, the results of this analysis should be viewed with caution.

3.3.2. House level

In order to highlight the impact of the house characteristics on the investigated parameters, the non-parametric M-W test was applied. Thus, the values of TVOC were significantly higher in rooms with concrete ceiling ($Mdn = 28\ \mu g/m^3$) than for those with wooden ceiling ($Mdn = 17\ \mu g/m^3$), $p < 0.05$. By contrary, the median of CO_2 concentrations was 1.2 times smaller in the rooms with concrete ceiling than those with wooden ceiling, the difference being statistically significant ($p = 0.03$). From the analysis of the data it was observed that the wooden ceiling corresponds mainly to the old houses, built around 1960, while those with concrete ceilings to the houses built after 1990. In fact, statistical significant differences were also recorded for heptane, TVOC and acetone concentrations, these being approx. 2 times higher for the houses built after 2000 compared to the oldest ones. Similar results were obtained in other studies (Park and Ikeda, 2006; Shin and Jo, 2013), in general high concentrations of TVOC being recorded in newly built houses as a result of construction materials and furniture used. On average, the TVOC concentration was 1.5 times higher in the houses that reported the use of candles, volatile oils or odorises in the investigated room compared to those that do not use such products.

A statistically significant difference was found for radon concentrations (passive and active measurements) in those rooms with concrete slab under the floor ($Mdn = 287\ Bq/m^3$), the median of radon concentrations being 1.5 times less than for the rooms with the floor placed directly on the ground. An inverse pattern was observed for TVOC concentrations, a median of approx. 1.5 times higher being specific to those with concrete slab. This inverse relationship could be explained by the fact that the main source of radon (soil) is outside to the investigated room, while for TVOC it is inside the room. A statistically significant difference was observed for those houses with cellar below the monitored room from the perspective of the TVOC concentration ($p = 0.03$), radon by passive method ($p = 0.05$) and relative humidity ($p < 0.001$), the calculated medians being significantly smaller in these type of houses compared to those that do not have a cellar under the investigated room. Naturally, this feature can be attributed to the period of construction of the house, usually the old houses having built the cellar under the house.

Moreover, statistically significant positive correlations were obtained between relative humidity and radon concentrations by the active method ($r = 0.45$, $p < 0.001$), limonene ($r = 0.39$, $p < 0.001$), respectively with energy consumption ($r = 0.26$, $p < 0.05$). The values of Pearson correlation coefficients performed for the main investigated parameters are shown in Fig. 4. The highlighted values are statistically significant for a p-value < 0.001. A moderate positive correlation was found between CO_2 concentration and RH ($r = 0.56$), limonene ($r = 0.50$), aldehyde ($r = 0.41$), acetone ($r = 0.40$) and radon concentrations, by active method ($r = 0.39$). A negative good correlation was recorded between relative humidity and temperature ($r = -0.56$). Moderate positive correlations were also obtained between indoor temperature and tetradecane ($r = 0.30$) and formaldehyde ($r = 0.33$).

As temperature, RH and CO_2 concentration, influence each other, was evaluated the intensity of the dependence between the concentration of CO_2 and the other investigated pollutants by maintaining constant the effect of relative humidity (first order correlation), respectively of relative humidity and temperature (second order). The obtained results are represented in Fig. 5. By maintaining constant the effect of RH, it can be observed a reduction of the correlation coefficients for limonene (0.50–0.28), heptane (0.20–0.12) and radon (0.39–0.24) an aspect that suggests that the control variable (RH) mediates the interaction between the CO_2 and investigated pollutants. By maintaining constant the impact of RH and indoor temperature, the correlation coefficient values undergo a reduction for formaldehyde (0.32–0.23), acetaldehydes (0.41–0.29), limonene (0.50–0.18) and tetradecane (0.20–0.14). Despite the general downward trend, only for the

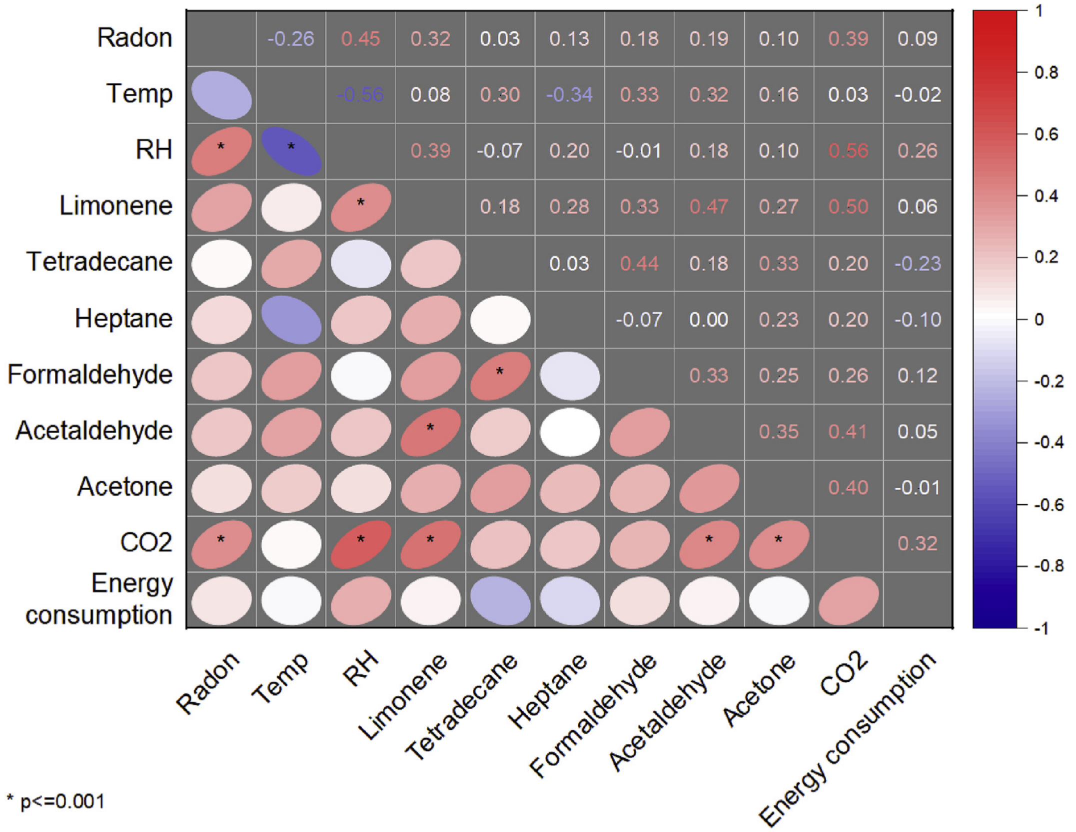

Fig. 4. Person correlation coefficients for the main investigated parameters.

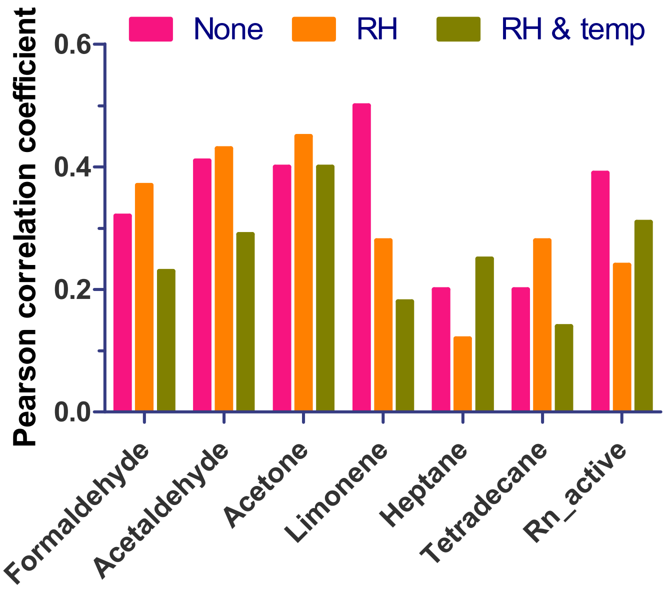

Fig. 5. The Pearson correlation coefficients between CO_2 and investigated parameters by maintaining constant the effect of RH, respectively RH and indoor temperature.

correlation coefficients between CO_2 and limonene, the addition of the control variable (RH, respectively RH & temperature) led to a statistically significant reduction ($p = 0.04$, respectively <0.01). These results indicated that it must be paid special attention to possible sources of multicollinearity in multivariate analysis. As such, for stepwise multiple linear regression analysis, the multicollinearity was investigated using Variation Inflation Factor (VIF) and the average of VIF values, while for the evidence of bias by checking the residuals, a Cook's distance >1 and a Mahalanobis distance > 25 were set as thresholds. The assumption of homoscedasticity of the variance of the regression errors was tested using White's test. If the assumption is violated, in order to reduce the effects of heteroscedasticity on inference process, the heteroscedasticity consistent covariance matrix estimators (HCCME) was applied.

The Stepwise multiple linear regression was used in order to model the relationships between indoor pollutants and indoor physical parameters, as well as house characteristics. As such, for formaldehyde concentrations, the indoor temperature and CO_2 concentrations, as a surrogate for ventilation, are the main explanatory variables, followed by the use of candles ($R^2_{adj.} = 0.23$). According to Ahn et al. (2015), formaldehyde concentrations were emitted in large quantities from some scented candles. As Fisk et al. (2020) concluded, an increase in the concentration of formaldehyde in retrofit buildings is expected, taking into account that these works result in a reduction of the ventilation rate, as well as an increase of the indoor temperature, which facilities an increase in the emission rates and the accumulation of the pollutant. A similar impact was found for acetaldehyde concentrations ($R^2_{adj.} = 0.26$) due to indoor temperature and CO_2 concentrations. The explanatory variables for the acetone concentrations are the presence of the fireplace, as well as CO_2 concentrations ($R^2_{adj.} = 0.24$). Guillaume et al. (2013) pointed out that the modern trend in using decorative ethanol fireplaces has as a downside the fuel, which may incorporate many compounds such as denaturants, including acetone. In addition, acetone is used as solvent, being present in many household products (Huang et al., 2019). Noticeable emissions of formaldehyde, acetaldehyde, TVOC and CO_2 were found during the combustion processes for wood burning fireplaces (Salthammer et al., 2014). As such, for the total carbonyl compounds, the explanatory variables are the presence of the fireplace, CO_2 and indoor temperature ($R^2_{adj.} = 0.38$). For the radon concentrations, measured by ICA system, the main explanatory variables are the CO_2 concentration, respectively the indoor temperature and the presence of the concrete screed ($R^2_{adj.} = 0.20$). For aliphatic hydrocarbons (heptane and tetradecane) concentrations, the ID of the city is the main explanatory variable, followed by temperature, year of construction and RH ($R^2_{adj.} = 0.45$, respectively 0.20). For terpenes (limonene and a-pinene) the explanatory variable are CO_2 concentration and RH ($R^2_{adj.} = 0.27$, respectively 0.22). In order to explain the maximum amount of total variance using the smallest number of explanatory constructs the Principal Component Analysis (PCA) was applied.

3.3.3. Principal component analysis

The PCA was applied onto log-transformed data of the main contributors of VOCs (such as limonene, heptane, tridecane, tetradecane and a-pinene), carbonyl compounds (formaldehyde, acetaldehyde and acetone), as well as for CO_2, temporal radon concentrations (active measurements), temperature, relative humidity and year of construction of the house. As such, a principal axis factor analysis was conducted on 13 variables with oblique rotation. The Kaiser-Meyer-Olkin (KMO) measure verified the sampling adequacy for the analysis and has a value of 0.68, while the KMO values for individual variables, extracted from anti-image matrices, are all above the acceptable limit of 0.5. Three factors were selected from the scree plot, each having eigenvalue >1 Table S3, and in combination explained 58% of the variance. The factor loadings after oblique rotation are shown in Table S4. The first factor, with the highest load (9 of 13 parameters), showed a combination between the carbonyl compounds (formaldehyde, acetaldehyde and acetone), some VOCs (aliphatic hydrocarbons, such as tri- and tetradecane and terpenes, such as limonene and a-pinene), temperature and CO_2. The terpenes were identified in household products (Milhem et al., 2020), while aliphatic hydrocarbons could be assigned to renovation works (Rösch et al., 2014) or floor coverings (Shin and Jo, 2013). Formaldehyde is presented in houses mainly as an adhesive resin for wood products, but can be found in insulating materials, cleaning agents, pesticide, parquet or carpets (Sarigiannis et al., 2011). Marchand et al. (2008) suggested that the aldehydes concentrations depend on both the characteristics of the building and the behaviour of home users (ventilation, cleaning products used). The carbonyl compounds are directed correlated with an increase of indoor temperature, respectively a poor ventilation, indicated by means of the CO_2 concentration. The second factor was controlled by five parameters (temperature, RH, CO_2, Rn and limonene). This factor could represent the impact of the indoor environment on the outdoor source (radon). Finally, the third factor explained 12% of the total variance and showed an association between heptane, tridecane and the year of construction, which can be attributed to renovation works/traffic emissions.

4. Limitation of the study

The main limitation of the study is the fact that the measurements, except for radon by passive method, were of short duration (7 days) during winter. Thus, any comparison with other season cannot be performed. Taking into account that the cold season is the period with lower ventilation rates, the concentration of indoor pollutants is expected to reach the annual maxima. In this sense, the obtained values can serve as a preliminary estimation of the highest annual values expected for each of the measured agents. The impact of other ventilation pathways on indoor pollutants cannot be assessed, the natural ventilation being the single way for the air exchange. In addition, as the multivariate analysis shows, more information is needed on the characteristics of the house in order to have a complete picture of the emission source and the factors that lead to the accumulation of pollutants inside homes.

5. Conclusion

This study represents one of the few international initiatives to monitor at the same time integrated levels of organic compounds and real-time radon and CO_2 concentrations, as well as physical parameters (RH and indoor temperature). The obtained results provide a good indicator of indoor air quality in retrofit buildings without indoor mechanical ventilation. As such, out of the total of 100 monitored houses, 52% presented an annual radon concentration above the reference value of 300 Bq/m^3. Moreover, 95% of the houses have a formaldehyde concentration higher than 50 $\mu g/m^3$, recommended by the German authorities (Umweltbundesamt, 2018). For acetaldehyde, 10% of the houses had values higher than those recommended by ANSES (ANSES, 2013).

In fact, from the analysis of the obtained data it can be drawn two house profiles. One is of the houses built mainly during the communist period (1950–1989) which, in general, have a cellar under the house, floor is placed directly on the ground and have a wooden ceiling. For this type of house, it is more likely that the TVOC, respectively the concentrations of heptane and acetone will be lower compared to the newly built houses (especially after 2000). From the radon perspective, the main source being the soil, the presence of the cellar under the monitored room generally conducts to a low indoor radon concentration, while the absence of a barrier in the form of a concrete screed under the floor leads to a high radon concentration. Given that the measurements of VOC and carbonyl compounds were performed successively in the five cities, with some caution, it can be concluded that a temporal variation can be observed due to human activity. As such, increased concentrations of both TVOC and individual volatile organic compounds were recorded in Bucharest, where the passive measurements were carried out before the winter holidays. Multivariate statistical analysis indicated that certain behaviours, such as the use of fireplaces, candles and volatile oils lead to an increase in the concentration of carbonyl compounds. In addition, a special attention must be paid to mutually influencing parameters such as temperature, relative humidity and CO_2 concentration. The computation of the zero order, first order (constant the effect of RH) and second order (constant the effect of RH and indoor temperature) correlation coefficients between the CO_2 concentration and the investigated parameters indicated that in some cases this correlation is mediated, which means that the impact of the CO_2 concentration, as a surrogate of the degree of ventilation, is lower than expected. As such, in order to efficiently implement retrofit works, it is highly recommended to design residential energy improvement programs adapted to the household, provide adequate ventilation and address summer and winter conditions. Intervention measures should pay attention to sources and accumulation patterns of indoor pollutants. Continuous, real-time monitoring provides valuable information on the time intervals at which high concentrations of pollutants are more likely to accumulate inside the home. Consequently, real-time indoor monitoring will improve IAQ management, which will implicitly improve indoor health outcomes. The temporal coverage offered by sensor technologies, as opposed to integrated measurements can provide the missing link to optimize the management of ventilation strategies, preventing wrong decisions and subsequent adverse health effects. The present study has been performed in the context of a research project where different mitigation strategies have been studied and implemented. However, the description of the results of these strategies are out of the scope of the present work.

CRediT authorship contribution statement

Mihail Simion Beldean-Galea: Writing - review & editing, Writing - original draft, Supervision, Conceptualization, Formal analysis. **Tiberius Dicu:** Project administration, Funding acquisition, Writing - review & editing, Writing - original draft, Supervision, Visualization, Conceptualization, Formal analysis. **Alexandra Cucoş:** Project administration, Funding acquisition, Conceptualization. **Bety-Denissa Burghele:** Writing - review & editing, Funding acquisition. **Tiberiu Catalina:** Writing - review & editing, Funding acquisition. **Marius Botoş:** Formal analysis. **Ancuţa Ţenter:** Funding acquisition, Writing - review & editing. **Kinga Szacsvai:** Funding acquisition. **Alexandru Lupulescu:** Writing - review & editing, Funding acquisition. **Istvan Pap:** Funding acquisition. **Gabriel Dobrei:** Funding acquisition. **Mircea Moldovan:** Funding acquisition. **Arthur Tunyagi:** Funding acquisition. **Ştefan Florică:** Funding acquisition. **Vlad Pănescu:** Funding acquisition. **Carlos Sainz:** Conceptualization, Supervision, Writing - review & editing, Funding acquisition, Project administration.

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.jclepro.2020.124098.

Unlisted references

OriginPro 2019b .

References

Ahn, J.H., Kim, K.H., Kim, Y.H., Kim, B.W., 2015. Characterization of hazardous and odorous volatiles emitted from scented candles before lighting and when lit. J. Hazard Mater. 286, 242–251.

Angulo Milhem, S., Verriele, M., Nicolas, M., Thevenet, F., 2020. Does the ubiquitous use of essential oil-based products promote indoor air quality? A critical literature review. Environ. Sci. Pollut. Res. 27 (3), 14365–14411. https://doi.org/10.1007/s11356-020-08150-3.

ANSES, 2013. Propositions de Valeurs Guides de qualité de l'Air intérieur - Acétaldéhyde.

ANSES, 2017. Updated Indoor Air Quality Guide Values - Formaldehyde.

Bade, B.C., Dela Cruz, C.S., 2020. Lung cancer 2020: epidemiology, etiology, and prevention. Clin. Chest Med. 41 (1), 1–24. https://doi.org/10.1016/j.ccm.2019.10.001.

Becerra, J.A., Lizana, J., Gil, M., Barrios-Padura, A., Blondeau, P., Chacartegui, R., 2020. Identification of potential indoor air pollutants in schools. J. Clean. Prod. 242 https://doi.org/10.1016/j.jclepro.2019.118420.

Billionnet, C., Sherrill, D., Annesi-Maesano, I., 2012. Estimating the health effects of exposure to multi-pollutant mixture. Ann. Epidemiol. 22, 126–141.

Brdaric, D., Kovac-Andric, E., Sapina, M., Kramaric, K., Lutz, N., Perkovic, T., Egorov, A., 2019. Indoor air pollution with benzene, formaldehyde, and nitrogen dioxide in schools in Osijek, Croatia. Air Qual. Atmos. Heal. 12, 963–968.

Canha, N., Lage, J., Candeias, S., Alves, C., Almeida, S.M., 2017. Indoor air quality during sleep under different ventilation patterns. Atmos. Pollut. Res. 8, 1132–1142.

Cerqueira, E.D.V., Motte-Baumvol, B., Chevallier, L.B., Bonin, O., 2020. Does working from home reduce CO_2 emissions? An analysis of travel patterns as dictated by workplaces. Transport. Res. Transport Environ. 83 https://doi.org/10.1016/j.trd.2020.102338.

Chan, C.S., Ranasinghe, R.S.A., Ho, S.S.H., Ho, K.F., Yim, S.H.L., Sugathapala, A.G.T., Lee, S.C., Hung, W.T., Huang, Y., Zhang, H., 2018. Evaluation of hazardous airborne carbonyls in five urban roadside dwellings: a comprehensive indoor air assessment in Sri Lanka. Atmos. Pollut. Res. 9, 270–277.

Chen, X.Y., Li, F., Liu, C.Y., Yang, J., Zhang, J.D., Peng, C.L., 2017. Monitoring, human health risk assessment and optimized management for typical pollutants in indoor air from random families of university staff, wuhan city, China. Sustainability 9. https://doi.org/10.3390/su9071115.

Cheng, Z., Li, B.Z., Yu, W., Wang, H., Zhang, T.J.W., Xiong, J., Bu, Z.M., 2018. Risk assessment of inhalation exposure to VOCs in dwellings in Chongqing, China. Toxicol. Res. (Camb). 7, 59–72.

Cucoş Dinu, A., Dicu, T., Cosma, C., 2015. Indoor radon exposure in energy-efficient houses from Romania. Rom. J. Phys. 60, 1574–1580.

Dai, X.L., Liu, J.J., Yin, Y.H., Song, X., Jia, S.S., 2018. Modeling and controlling indoor formaldehyde concentrations in apartments: on-site investigation in all climate zones of China. Build. Environ. 127, 98–106.

Dales, R., Raizenne, M., 2004. Residential exposure to volatile organic compounds and asthma. J. Asthma 41, 259–270.

Delikhoon, M., Fazlzadeh, M., Sorooshian, A., Baghani, A.N., Golaki, M., Ashournejad, Q., Barkhordari, A., 2018. Characteristics and health effects of formaldehyde and acetaldehyde in an urban area in Iran. Environ. Pollut. 242, 938–951.

European Commission, 2012. DIRECTIVE 2012/27/EU OF THE EUROPEAN PARLIAMENT AND OF THE COUNCIL of 25 October 2012 on energy efficiency, amending Directives 2009/125/EC and 2010/30/EU and repealing Directives 2004/8/EC and 2006/32/EC. Off. J. Eur. Union.

European Commission, 2014. Council directive 2013/59/euratom of 5 december 2013. Off. J. Eur. Union. https://doi.org/10.3000/19770677.L_2013.124.eng.

Fang, L., Norris, C., Johnson, K., Cui, X.X., Sun, J.Q., Teng, Y.B., Tian, E.Z., Xu, W., Lig, Z., Mo, J.H., Schauer, J.J., Black, M., Bergin, M., Zhang, J., Zhang, Y.P., 2019. Toxic volatile organic compounds in 20 homes in Shanghai: concentrations, inhalation health risks, and the impacts of household air cleaning. Build. Environ. 157, 309–318.

Ferguson, L., Taylor, J., Davies, M., Shrubsole, C., Symonds, P., Dimitroulopoulou, S., 2020. Exposure to indoor air pollution across socio-economic groups in high-income countries: a scoping review of the literature and a modelling methodology. Environ. Int. 143 https://doi.org/10.1016/j.envint.2020.105748.

Fernández-Agüera, J., Domínguez-Amarillo, S., Alonso, C., Martín-Consuegra, F., 2019. Thermal comfort and indoor air quality in low-income housing in Spain: the influence of airtightness and occupant behaviour. Energy Build. 199, 102–114.

Field, A., 2013. Discovering Statistics Using IBM SPSS Statistics, fourth ed. SAGE, London.

Fisk, W.J., Singer, B.C., Chan, W.R., 2020. Association of residential energy efficiency retrofits with indoor environmental quality, comfort, and health: a review of empirical data. Build. Environ. 180 https://doi.org/10.1016/j.buildenv.2020.107067.

Földváry, V., Bekö, G., Langer, S., Arrhenius, K., Petráš, D., 2017. Effect of energy renovation on indoor air quality in multifamily residential buildings in Slovakia. Build. Environ. 122, 363–372.

Gichner, T., 1995. IARC monographs on the evaluation of carcinogenic risks to humans. Volume 62. Wood dust and formaldehyde. Biol. Plant. (Prague) 37 (500). https://doi.org/10.1007/bf02908826.

Gilbert, N.L., Guay, M., Miller, J.D., Judek, S., Chan, C.C., Dales, R.E., 2005. Levels and determinants of formaldehyde, acetaldehyde, and acrolein in residential indoor air in Prince Edward Island, Canada. Environ. Res. 99, 11–17.

Guillaume, E., Loferme-Pedespan, N., Duclerget-Baudequin, A., Raguideau, A., Fulton, R., Lieval, L., 2013. Ethanol fireplaces: Safety matters. Saf. Sci. 57, 243–253.

Huang, S.D., Wei, W.J., Weschler, L.B., Salthammer, T., Kan, H.D., Bu, Z.M., Zhang, Y.P., 2017. Indoor formaldehyde concentrations in urban China: preliminary study of some important influencing factors. Sci. Total Environ. 590, 394–405.

Huang, K.L., Song, J.S., Feng, G.H., Chang, Q.P., Jiang, B., Wang, J., Sun, W., Li, H.X., Wang, J.M., Fang, X.S., 2018. Indoor air quality analysis of residential buildings in northeast China based on field measurements and longtime monitoring. Build. Environ. 144, 171–183.

Huang, Y., Su, T., Wang, L.Q., Wang, N., Xue, Y.G., Dai, W.T., Lee, S.C., Cao, J.J., Ho, S.S.H., 2019. Evaluation and characterization of volatile air toxics indoors in a heavy polluted city of northwestern China in wintertime. Sci. Total Environ. 662, 470–480.

IQAir, 2019. World air quality report. 2019. World Air Qual. Rep. https://www.iqair.com/world-most-polluted-cities/world-air-quality-report-2019-en.pdf.

ISO 16000-4, 2011. Indoor Air — Part 4: Determination of Formaldehyde — Diffusive Sampling Method. ISO/TC 146/SC 6 Indoor Air.

ISO 16017-2, 2003. Indoor, Ambient and Workplace Air — Sampling and Analysis of Volatile Organic Compounds by Sorbent Tube/thermal Desorption/capillary Gas

Chromatography — Part 2: Diffusive Sampling. ISO/TC 146/SC 6 Indoor Air.

ISO 16200-2, 2000. Workplace air quality — sampling and analysis of volatile organic compounds by solvent desorption/gas chromatography — Part 2: diffusive sampling method. In: ISO/TC 146/SC 2 Workplace Atmospheres.

Jiránek, M., Kačmaříková, V., 2014. Dealing with the increased radon concentration in thermally retrofitted buildings. Radiat. Protect. Dosim. 160, 43–47.

Kauneliene, V., Prasauskas, T., Krugly, E., Stasiulaitiene, I., Ciuzas, D., Seduikyte, L., Martuzevicius, D., 2016. Indoor air quality in low energy residential buildings in Lithuania. Build. Environ. 108, 63–72.

Kelly, F.J., Fussell, J.C., 2019. Improving indoor air quality, health and performance within environments where people live, travel, learn and work. Atmos. Environ. 200, 90–109.

Kim, J., Hwangbo, H., 2018. Sensor-based optimization model for air quality improvement in home IoT. Sensors 18. https://doi.org/10.3390/s18040959.

Kishi, R., Ketema, R.M., Bamai, Y.A., Araki, A., Kawai, T., Tsuboi, T., Saito, I., Yoshioka, E., Saito, T., 2018. Indoor environmental pollutants and their association with sick house syndrome among adults and children in elementary school. Build. Environ. 136, 293–301.

Langer, S., Beko, G., Bloom, E., Widheden, A., Ekberg, L., 2015. Indoor air quality in passive and conventional new houses in Sweden. Build. Environ. 93, 92–100.

Langer, S., Ramalho, O., Derbez, M., Riberon, J., Kirchner, S., Mandin, C., 2016. Indoor environmental quality in French dwellings and building characteristics. Atmos. Environ. 128, 82–91.

Li, Y., Cakmak, S., Zhu, J., 2019. Profiles and monthly variations of selected volatile organic compounds in indoor air in Canadian homes: results of Canadian national indoor air survey 2012–2013. Environ. Int. 126, 134–144. https://doi.org/10.1016/j.envint.2019.02.035.

Liang, W.H., Yang, X.D., 2013. Indoor formaldehyde in real buildings: emission source identification, overall emission rate estimation, concentration increase and decay patterns. Build. Environ. 69, 114–120.

Marchand, C., Le Calvé, S., Mirabel, P., Glasser, N., Casset, A., Schneider, N., de Blay, F., 2008. Concentrations and determinants of gaseous aldehydes in 162 homes in Strasbourg (France). Atmos. Environ. 42, 505–516.

MC 001, 2006. National Methodology for the Calculation of Energy Performance of Buildings MC001/2006. Official Gazette of Romania.

Meiboudi, H., Lahijanian, A., Shobeiri, S.M., Jozi, S.A., Azizinezhad, R., 2016. Creating an integrative assessment system for green schools in Iran. J. Clean. Prod. 119, 236–246.

Pampuri, L., Caputo, P., Valsangiacomo, C., 2018. Effects of buildings' refurbishment on indoor air quality. Results of a wide survey on radon concentrations before and after energy retrofit interventions. Sustain. Cities Soc. 42, 100–106. https://doi.org/10.1016/j.scs.2018.07.007.

Park, J.S., Ikeda, K., 2006. Variations of formaldehyde and VOC levels during 3 years in new and older homes. Indoor Air 16 (2), 129–135. https://doi.org/10.1111/j.1600-0668.2005.00408.x.

Podstawczyńska, A., Chambers, S.D., 2019. Improved method for characterising temporal variability in urban air quality part I: traffic emissions in central Poland. Atmos. Environ. 219 https://doi.org/10.1016/j.atmosenv.2019.117038.

Rabago, D., Fuente, I., Celaya, S., Fernandez, A., Fernandez, E., Quindos, J., Pol, R., Cinelli, G., Quindos, L., Sainz, C., 2020. Intercomparison of indoor radon measurements under field conditions in the framework of MetroRADON European project. Int. J. Environ. Res. Public Heal. Sect. Environ. Sci. Eng. 17 https://doi.org/10.3390/ijerph17051780.

Rösch, C., Kohajda, T., Röder, S., von Bergen, M., Schlink, U., 2014. Relationship between sources and patterns of VOCs in indoor air. Atmos. Pollut. Res. 5, 129–137.

Sakai, N., Yamamoto, S., Matsui, Y., Khan, M.F., Latif, M.T., Mohd, M.A., Yoneda, M., 2017. Characterization and source profiling of volatile organic compounds in indoor air of private residences in Selangor State, Malaysia. Sci. Total Environ. 586, 1279–1286.

Salthammer, T., 2019. Data on formaldehyde sources, formaldehyde concentrations and air exchange rates in European housings. Data Br 22, 400–435.

Salthammer, T., Schripp, T., Wientzek, S., Wensing, M., 2014. Impact of operating wood-burning fireplace ovens on indoor air quality. Chemosphere 103, 205–211.

Sarigiannis, D.A., Karakitsios, S.P., Gotti, A., Liakos, I.L., Katsoyiannis, A., 2011. Exposure to major volatile organic compounds and carbonyls in European indoor environments and associated health risk. Environ. Int. 37, 743–765.

Sharpe, R.A., Thornton, C.R., Nikolaou, V., Osborne, N.J., 2015. Higher energy efficient homes are associated with increased risk of doctor diagnosed asthma in a UK subpopulation. Environ. Int. 75, 234–244.

Shin, S.H., Jo, W.K., 2013. Longitudinal variations in indoor VOC concentrations after moving into new apartments and indoor source characterization. Environ. Sci. Pollut. Res. 20, 3696–3707.

Stamatelopoulou, A., Asimakopoulos, D.N., Maggos, T., 2019. Effects of PM, TVOCs and comfort parameters on indoor air quality of residences with young children. Build. Environ. 150, 233–244.

Stanaway, J.D., Afshin, A., Gakidou, E., Lim, S.S., Abate, D., 2018. Global, regional, and national comparative risk assessment of 84 behavioural, environmental and occupational, and metabolic risks or clusters of risks for 195 countries and territories, 1990-2017: a systematic analysis for the Global Burden of Disease Study 2017. Lancet 392, 1923–1994.

Todea, D., Cosma, C., Dicu, T., Roşca, L., Dinu, A.C., Rişteiu, M., Iancu, D., Papuc, I., Rădulescu, D., 2013. Lung cancer risk induced by residential radon in Cluj and Alba Counties, Romania. Environ. Eng. Manag. J. 12, 1281–1285.

Tunyagi, A., Dicu, T., Cucos, A., Burghele, B.-D., Dobrei, G., 2020. An innovative system for monitoring radon and indoor air quality. Rom. J. Phys. 65.

Umweltbundesamt, 2018. Guide Values for the Concentration of Specific Substances in Indoor Air, 2019.

Villanueva, F., Tapia, A., Lara, S., Amo-Salas, M., 2018. Indoor and outdoor air concentrations of volatile organic compounds and NO_2 in schools of urban, industrial and rural areas in Central-Southern Spain. Sci. Total Environ. 622, 222–235.

Wallner, P., Munoz, U., Tappler, P., Wanka, A., Kundi, M., Shelton, J.F., Hutter, H.P., 2015. Indoor environmental quality in mechanically ventilated, energy-efficient buildings vs. Conventional buildings. Int. J. Environ. Res. Publ. Health 12, 14132–14147.

Wallner, P., Tappler, P., Munoz, U., Damberger, B., Wanka, A., Kundi, M., Hutter, H.P., 2017. Health and wellbeing of occupants in highly energy efficient buildings: a field study. Int. J. Environ. Res. Publ. Health 14. https://doi.org/10.3390/ijerph14030314.

World Health Organization, 2010. WHO Guidelines for Indoor Air Quality: Selected Pollutants.

World Health Organization, 2016. Household Air Pollution and Health.

World Health Organization, 2019. Air Pollution.

Wu, P.C., Li, Y.Y., Lee, C.C., Chiang, C.M., Su, H.J.J., 2003. Risk assessment of formaldehyde in typical office buildings in Taiwan. Indoor Air 13, 359–363.

Wyon, D.P., 2004. The effects of indoor air quality on performance and productivity. Indoor Air 14, 92–101. Suppl.

Yang, S., Pernot, J.G., Jörin, C.H., Niculita-Hirzel, H., Perret, V., Licina, D., 2020. Energy, indoor air quality, occupant behavior, self-reported symptoms and satisfaction in energy-efficient dwellings in Switzerland. Build. Environ. 171.

Ye, W., Zhang, X., Gao, J., Cao, G.Y., Zhou, X., Su, X., 2017. Indoor air pollutants, ventilation rate determinants and potential control strategies in Chinese dwellings: a literature review. Sci. Total Environ. 586, 696–729.