

Aldehydes and Acetone in Indoor Air of 19 Houses from Bucharest (Romania)

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Indoor air concentrations of aldehydes and acetone in nineteen houses located in Bucharest (Romania) were determined in this study. Passive sampling was done in bedrooms or living rooms, the air samplers being collected on DSD-DNPH cartridges. Aldehydes and acetone were quantified using a DNPH-derivatization followed by liquid chromatography coupled to UV detection analytical method. Results obtained showed the presence of aldehydes in all the houses, with formaldehyde having the highest concentration. Rooms with new furniture (under 1 year) showed the highest concentrations of aldehydes.

Keywords: indoor air; aldehydes, formaldehyde, DNPH

Aldehydes are pollutants of indoor and outdoor air that caught the attention of scientists around the world due to their potential impact on human health. Among them, formaldehyde is considered to be the most abundant, most studied and most harmful to humans.

Formaldehyde is listed as probable human carcinogen by US EPA [1, 2] and *Carcinogenic to humans (Group 1)* by IARC [3]. Recent studies on the effect of exposure to aldehydes and especially to formaldehyde on humans and animals mainly showed effects on the respiratory system, like: modification of nose cells [4, 5], buccal cell alteration [6], nasopharyngeal cell modifications [7], effects on lung function [8], sensory irritations [9, 10] and accentuate asthma symptoms [11]. Other effects in humans include menstrual disorders [12, 13], mutagenic effect and different forms of cancer [14].

Sources of aldehydes in homes are: building materials, laminate floorings, adhesives, paints and solvents, household products, furniture made by particleboard, fiberboard and plywood with formaldehyde resin glue [15-17] to this we can add activities like smoking and open fire stoves.

In recent years, a lot of attention was given worldwide to indoor air quality and indoor air pollutants; thus, studies were done also in Romania regarding indoor air quality and the concentrations of aldehydes found in indoor air spaces like schools [18], offices [19] and houses [20].

The aim of this study was to determine the indoor air concentrations of aldehydes and acetone in nineteen houses located in Bucharest (Romania). The targeted aldehydes and ketones in indoor air included: formaldehyde, acetaldehyde, acetone, acrolein, propionaldehyde, crotonaldehyde, butyraldehyde, benzaldehyde, isovaleraldehyde, valeraldehyde, *o,m,p*-tolualdehyde, hexaldehyde and 2,5-dimethylbenzaldehyde.

Experimental part

The sampling points consisted in nineteen apartments situated in Bucharest, a city of about 2 million inhabitants. Sampling was done in bedrooms and living rooms with new and old furniture on a period of time of around ten days. The inhabitants had to fill a form that contained data about the start time and the end time of the sampling. The questions in the form gave us information also about the

type of furniture in the house, the age of the furniture, the carpet and the floor material, the type of material the windows were made of and the surface covered by furniture. No air conditioning system or open fire stoves were used during the sampling period from 22th of October to 3rd of November 2015.

The indoor air samples were collected by passive sampling methods in order to not disturb the daily lives of the ones living in the selected houses. DSD-DNPH cartridges design for sampling aldehydes from Sigma-Aldrich were bought and used as sampling devices. DSD-DNPH cartridges consist in silica and DNPH (2,4-dinitrophenylhydrazine) packed in the sampler as the absorbent. The aldehydes present in air react with DNPH in the absorbent to form hydrazone derivatives. The passive sampler consists of a porous sintered polyethylene tube (PSP-diffusion filter), which acts as a diffusive membrane, and a small polypropylene syringe (PP-reservoir) for elution of analytes from the absorbent [21].

A calibration standard: TO11/IP 6A Aldehyde/Ketone-DNPH Mix certified reference material, with 15 µg/mL concentration of aldehydes and ketones, was purchased from Sigma-Aldrich. The calibration standard contains the hydrazone derivatives of the fifteen targeted aldehydes and ketones.

Aldehydes and acetone were analyzed and determined according to US-EPA methods

TO-11A and IP-6 and sampled according to US EPA 0100 [22-24].

A high performance liquid chromatograph model Agilent 1200, coupled with a UV detector was used for the determination of aldehydes and ketones. Analytical conditions included two Acclaim Carbonyl C18 columns (250 mm * 4.6 mm, 5 µm,) coupled in series, a diode array detector (DAD) set at 365 nm wavelength, 2 mL/min flow rate, 25 µL injection volume, 25°C column temperature and a gradient mobile phase of acetonitrile / water as seen in table 1.

All the samplers were refrigerated after sampling and analyzed within 10 days. After collection, the PP-reservoir of the sampling device was re-moved from the diffusion filter and connected to a clean 5mL syringe, who acted as a reservoir for the eluent. DNPH derivatives were eluted

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Time (minutes)	ACN (%)	Water (%)
0.0	60	40
30.0	75	25
50.0	100	0
55.0	100	0
55.1	60	40
70.0	60	40

Table 1
GRADIENT USED IN THE HPLC ELUTION OF THE ALDEHYDES AND KETONES

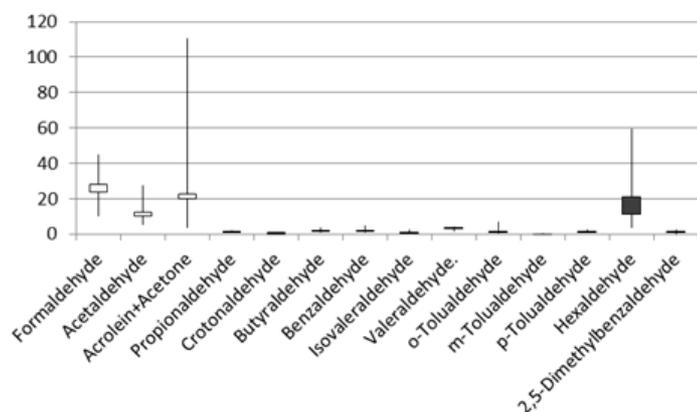


Fig. 1. Distribution of the targeted pollutants concentrations ($\mu\text{g}/\text{m}^3$) in indoor samples

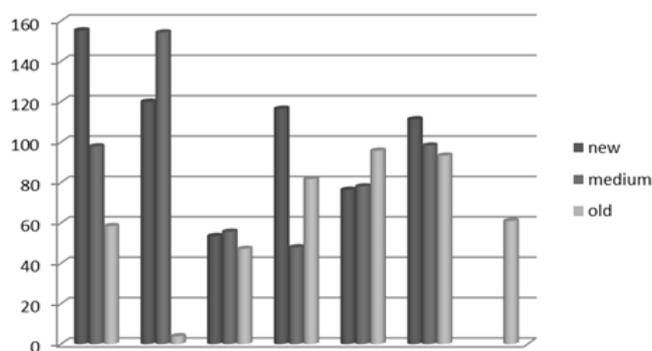


Fig. 2. Total aldehydes concentrations ($\mu\text{g}/\text{m}^3$) in homes with new, medium and old furniture

from the PP-reservoir absorbent in a volumetric flask with a known volume by passing acetonitrile; all of them were analyzed in the same day of extraction.

Results and discussions

Concentration of all the carbonyls were quantified by external standard calibration, the calibration curves covered the range of interest and showed good linearity - $r^2 > 0.999$.

The DNPH method is unsuitable for complete separation of acrolein from acetone [25]; in this study will be referred as acrolein+acetone.

Figure 1 shows the stock chart data analyse of the results obtained regarding the aldehydes concentrations in all the indoor air samples. The highest concentration was found in the case of acrolein+acetone. The most compact values were obtained for formaldehyde; and the highest variation in the concentration determined were found for acrolein+acetone.

In figure 2 and figure 3, are presented the results taking into consideration the age of the furniture. In this purpose, based on the questionnaire filled by the inhabitants regarding the type and the age of the furniture, the indoor air samples were classified in three categories: first category with furniture lower than one year old but not older than five years (described as *new* in the graphics); second category with furniture older than five years but not older than ten years (described as *medium* in the graphics); and a third category with furniture older than ten years (described as *old* in the graphics). Based on three

Formaldehyde

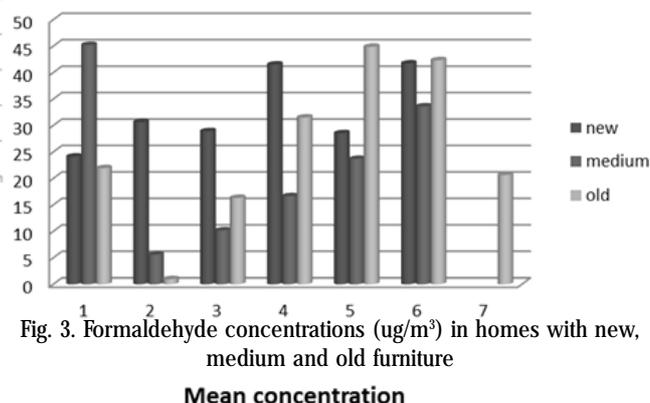


Fig. 3. Formaldehyde concentrations ($\mu\text{g}/\text{m}^3$) in homes with new, medium and old furniture

Mean concentration

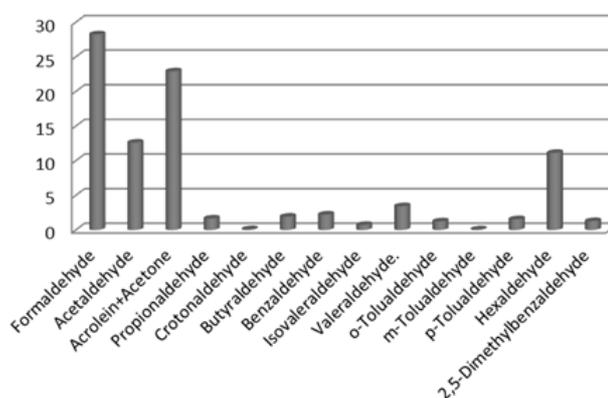


Fig. 4. Mean concentrations ($\mu\text{g}/\text{m}^3$) of the aldehydes from all the indoor air samples.

categories, the highest concentration of aldehydes were obtain in indoor air samples taken from rooms with new furniture, but not all the houses participating in this study followed this pattern.

In figure 4 are presented the mean concentrations ($\mu\text{g}/\text{m}^3$) of the aldehydes from all the indoor air samples. Concentration of the targeted aldehydes tend to follow the following pattern: formaldehyde > acrolein+acetone > acetaldehyde > hexaldehyde > valeraldehyde > benzaldehyde > butyraldehyde > propionaldehyde > *p*-tolualdehyde > 2,5-dimethylbenzaldehyde > *o*-tolualdehyde > isovaleraldehyde > crotonaldehyde and *m*-tolualdehyde. The most abundant aldehydes were: formaldehyde, acrolein+acetone, acetaldehyde and hexaldehyde; the other concentrations of aldehydes were much lower.

Conclusions

All the fifteen carbonyls (aldehydes and acetone) were found in quantifying quantities in all the samplers, with formaldehyde having the highest concentrations.

The highest concentrations of aldehydes and acetone were found in indoor air samples from rooms with new furniture (under one year old).

The most abundant aldehydes were: formaldehyde, acrolein+acetone, acetaldehyde and hexaldehyde; the other concentrations of aldehydes were much lower.

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