Formaldehyde as Screen Printing Indoor Pollutant

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Abstract. The presence of formaldehyde in indoor air samples has been detected in five screen printing facilities of Novi Sad, Serbia. Air was sampled five times during one working week for each screen printing facility. The sampling was conducted continuously during 4 hours. The concentrations of formaldehyde were determined by UV-VIS spectrometry at 580 nm. The average concentrations of formaldehyde were in the range from 0.413 to 0.836 ppm. Comparison of the detected concentration levels with the permissible exposure limit of 0.75 ppm (the OSHA standard) and 0.016 (the NIOSH standard) indicated that the average formaldehyde concentration in facility 5 was 1.11 and 52.25 times higher than prescribed values, respectively.

Keywords: formaldehyde, indoor pollution, screen printing, statistical regression analysis

1 Introduction

Formaldehyde is arguably the most common and the best-known indoor air pollutant. Over the years, the release of formaldehyde from building products has been decreasing. On the other hand, formaldehyde concentrations in ambient air are increasing continuously, especially in the urban and industrial environments [1, 2]. Industrial releases of formaldehvde can occur at any stage during the production, use, storage, transportation, or disposal of products with residual formaldehyde. Formaldehyde has been emitted from chemical manufacturing plants, pulp and paper mills, forestry product plants, tire and rubber plants, petroleum refining and coal processing plants, textile mills, automotive manufacturing plants, metal products industry and printing facilities [3]. Screen printing is possibly the most versatile of all printing processes. It can be used to print on a wide variety of substrates, including paper, paperboard, plastic, textile, glass, metal, fabric, and many other materials [4]. Liquid materials (printing ink, cleaning solution, varnish, adhesive, etc.) used in screen printing process generate numerous toxic, hazardous substances of organic origin, especially contaminants from the group of volatile organic compounds (VOCs), such as formaldehyde, benzene, toluene, xylene, and methanol, which are well known to cause the so-called building-related sickness [5, 6]. The amounts of formaldehyde vary depending on the type of used compound, the printing and drying processes, substrates and end-use application requirements [7, 8].

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Because of its toxicity, formaldehyde is classified as a hazardous substance able to affect the workers' health [3, 9]. The high solubility of formaldehyde in water causes rapid absorption in the respiratory and gastrointestinal tract. The biological half-life is extremely short at about 1 min [10]. The lowest observable adverse effect levels (LOAEL) for human sensory irritation range from 0.4 ppm (rhinitis) to 3 ppm (eye, nasal, and throat irritation) [11]. A recent study of formaldehyde and sensory irritation in humans showed that eye irritation is the most sensitive parameter. A no observed effect level (NOEL) of 0.5 ppm was derived in the case of constant exposure [12].

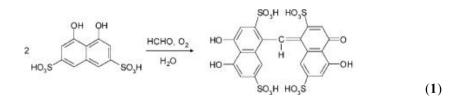
The research has been conducted for the first time in Serbia with the aim to monitor formaldehyde concentration in screen printing indoor. The objective was to determine the correlations between the formaldehyde concentrations and microclimate parameters.

2 Materials and Methods

The concentrations of formaldehyde were measured in five screen printing facilities (SPF 1 - 5) in Novi Sad, Serbia. The investigated facilities are small and similar in area (50 - 70 m²), number of employees (3 - 4), production volume (50 - 70 products per hour). They have not installed a ventilation system. Their production covers a range of products including labels, folders, planners, posters, lighters and t-shirts. The main equipment is a desk with a screen holder. The most often used materials were a porous mesh stretched tightly over a metal frame, paper, cardboard, textile, screen emulsion, screen printing ink, solvent and adhesive.

Measurements were carried out in the morning, five times during one working week. The sampling period was 4 hours within continuous printing process. Air was sampled from one sampling point using air sampler PRO-EKOS AT. 401X. The position of sampling point was determined according to the technical characteristics of the screen printing desk.

The indoor air was aspirated through the Drechsel bottles with formaldehyde reagent (95 cm³ cc sulfuric acid and 0.5 cm³ 1% hromotropic acid). The airflow was 0.5 dm³/min. In the presence of concentrated sulfuric acid, chromotropic acid (1,8 dihydroxynaphthalene- 3,6-disulfonic acid) reacts with formaldehyde to give a red-violet hydroxydiphenylmethane derivative (Equation 1).



The resulting chromophore solution is analyzed by UV/VIS spectrometry at 580 nm (UV/VIS spectrophotometer DR 5000 HACH LANGE). The concentrations of

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formaldehyde were determined from calibration curve with standard formaldehyde solution, 1 mg/cm³ [13].

Microclimate parameters were measured with a direct reading instrument Mannix DLAF 8000. The instrument accuracies were ± 0.5 °C, $\pm 0.5\%$ and ± 1 1x for temperature, relative humidity and light intensity, respectively. The average values of temperature, relative humidity and light intensity were measured *in situ* 3 times, during 4 hours.

3 Results and Discussion

Experimental results confirmed the presence of formaldehyde in screen printing environment. The daily concentrations of formaldehyde in screen printing facilities were in the range from 0.275 to 0.879 ppm, while the average formaldehyde concentrations varied from 0.413 to 0.836 ppm (Table 1). Only facility 5 has the daily and average formaldehyde concentrations above 0.75 ppm as the PEL value prescribed by OSHA standard [14]. The average formaldehyde concentration levels were almost 1.11 to 1.82 times lower than PEL (facilities 1-4), whereas in facility 5 the average formaldehyde concentration was 1.11 times higher than PEL value. According to the STEL value, the formaldehyde concentrations in all investigated facilities were below 2 ppm [14]. In printing facility 5, the daily and average formaldehyde concentrations were almost 50 to 55 times higher than the REL value (0.016 ppm) prescribed by NIOSH standard [15]. The concentration values of formaldehyde were obtained by registering current situation of the working environment of screen printing facilities in Novi Sad, Serbia. The daily, average and maximum allowed concentration (MAC) values of formaldehyde in studied screen printing facilities are presented in Table 1.

Screen		Concentration of formaldehyde (ppm)				MAC (ppm)				
printing		Da	y of the w	eek		Average	¹ O	SHA	² NI	OSH
facility	1	2	3	4	5	value	³ PEL	⁴ STEL	SREL	⁶ IDLH
SPF 1	0.275	0.299	0.455	0.637	0.399	0.413				
SPF 2	0.567	0.762	0.638	0.755	0.668	0.678				
SPF 3	0.476	0.599	0.433	0.639	0.673	0.564	0.75	2	0.016	20
SPF 4	0.388	0.464	0.364	0.503	0.536	0.451				
SPF 5	0.807	0.855	0.799	0.840	0.879	0.836				

Table 1. Daily, average and MACs of formaldehyde in screen printing facilities

¹Occupational Safety and Health Administration (OSHA)

²National Institute for Occupational Safety and Health (NIOSH)

³Permissible Exposure Limit (PEL)

⁴Short-Term Exposure Limit (STEL)

⁵Recommended Exposure Limit (REL)

⁶Immediately Dangerous to Life and Health (IDLH)

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Based on the detected formaldehyde concentrations it was confirmed that printing raw materials as volatile compounds (inks, cleaning solution, adhesive) are the main sources of formaldehyde emission in screen printing indoor. The concentrations of free formaldehyde in these products are generally less than 2%. Additionally, unsaturated VOCs from conventional cleaning solutions in the reaction with ambient ozone contribute to the formaldehyde release in screen printing indoor.

Beside the chemical composition of raw materials the variations in the formaldehyde concentrations in screen printing facilities were related also to the ambient conditions, volume of production, and distance from the printing desk, as well as the presence of ventilation installed in workplace. Numerous studies have reported that the indoor emission of formaldehyde increases with the increase of temperature and relative humidity [9, 16]. In accordance with the literature, our research confirmed the dependence of formaldehyde concentration on temperature and relative humidity. Therefore, the increased concentration of formaldehyde in facility 5 was caused by a significant increase of temperature and relative humidity (Table 2).

The average temperature, relative humidity and light intensity in screen printing facilities were presented in Table 2. Average temperature varied from 19.3 to $31.1 \,^{\circ}$ C, relative humidity varied from 42.0 to 65.4% and light intensity varied from 85 to $1163 \, \text{lx}$.

Screen printing	Microclimate Days of the week					
facility	parameters	1	2	3	4	5
	t [°C]	19.5	20.5	19.6	19.3	19.3
SPF 1	RH [*] [%]	42.0	43.1	45.6	44.3	43.2
	LI** [lx]	99	238	357	408	870
	t [°C]	21.8	20.5	19.1	20.2	19.6
SPF 2	RH [*] [%]	38.9	40.3	46.4	42.1	52.1
	LI** [lx]	85	96	298	302	340
	t [°C]	24.2	24.1	23.8	23.6	23.7
SPF 3	RH [*] [%]	38.3	39.2	50.3	40.6	40.3
	LI** [lx]	409	420	447	279	265
	t [°C]	26.9	25.8	26.3	27.7	27.6
SPF 4	RH [*] [%]	49.8	51.1	53.9	49.3	50.5
	LI** [lx]	493	549	1163	1032	1025
	t [°C]	28.8	29.9	27.8	29.2	31.1
SPF 5	RH [*] [%]	54.7	62.5	53.7	58.6	65.4
	LI** [lx]	1076	1012	992	951	1062

Table 2. Average values of microclimate parameters in screen printing facilities

I ** Light intensity

LI** - Light intensity

The obtained results indicate that formaldehyde concentrations in press department of screen printing facility, especially in printing facility 5, have a potential risk factor to workers' health. Many researches also confirm that formaldehyde can be toxic for human health and considered to be carcinogenic, mutagenic, or teratogenic [17, 18].

Thus, nowadays environmental regulations and EU Directives are increasingly pushing printing industry to reduce emission of formaldehyde, or find alternative nonformaldehyde materials. The use of alternative materials is an environmentally responsible choice that will reduce the generation of hazardous formaldehyde in screen printing environment.

3.1 Correlation analysis

Mutual dependence of parameters (temperature / relative humidity / light intensity) in the printing facilities. In the study of formaldehyde concentration (F) dependence on temperature (t), relative humidity (RH) and light intensity (LI), the question of mutual dependence of the observed parameters is posed.

The mutual dependence of these parameters was studied in each of the five individual printing facilities, as well as on pooled data from all the printing facilities, although it should be noted that in each printing facility specific conditions prevail. Contrary to our expectations, the examination of the obtained linear correlation coefficients shows that they are all very small (Table 3). Only the coefficient 0.963139 of the linear correlation of temperature on relative humidity is statistically significant, which can be explained by production volume.

 Table 3. The coefficients of mutual linear correlation of temperature, relative humidity and light intensity in SPF 5

Facility	Correlation t - RH	Correlation t - LI	Correlation RH - LI
SPF 5	0.963139	0.364247	0.171673

From the Table 3 we can not detect any other correlation between the parameters. This justifies our choice to study the dependence of the formaldehyde concentration on temperature, relative humidity and light intensity individually.

Dependence of the formaldehyde concentration on the temperature, relative humidity and light intensity in printing facilities. Dependence of the formaldehyde concentration (F) on temperature (t), relative humidity (RH) and light intensity (LI) was studied in each of the five individual printing facilities as well as on pooled data from all facilities. In the consideration of the results obtained from interpreted data, one should bear in mind that specific conditions present in each of the printing facilities. Reviewing the linear correlation coefficients obtained for formaldehyde concentrations respectively with the temperature, relative humidity and light intensity, we see that they are all small. Statistically significant are only the linear dependence coefficients 0.963562 and 0.992551 of formaldehyde concentration on temperature and relative humidity, respectively, in SPF 5 (Table 4). This may be a coincidence, or it may be caused by an indirect factor present only in SPF 5.

 Table 4. The coefficients of linear correlation of formaldehyde concentration with temperature, relative humidity and light intensity

Facility	Correlation F - t	Correlation F - RH	Correlation F - LI
SPF 5	0.963562	0.992551	0.115759

Figure 1 shows the regression line of the formal dehyde concentration (F) compared to the temperature (t) in SPF 5, with a correlation coefficient 0.963562, and the equation of regression line F = 0.0260013t + 0.0726014.

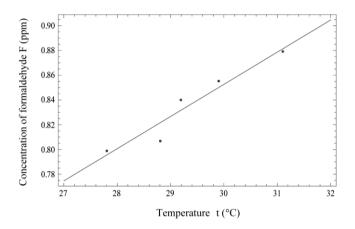


Fig. 1. Graph of linear regression formaldehyde concentration - temperature in SPF 5

Figure 2 shows the regression line of the formaldehyde concentration (F) compared to the relative humidity (RH) in SPF 5, with a correlation coefficient 0.992551, and the equation of regression line F = 0.00661244RH + 0.445998.

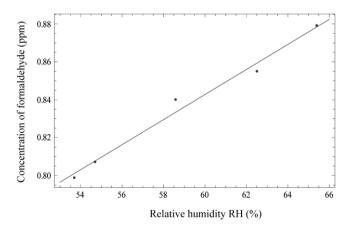


Fig. 2. Graph of linear regression formaldehyde concentration - relative humidity in SPF 5

From the Tables 3 and 4, we could conclude that there is a statistically significant linear correlation with the formaldehyde concentration on temperature and humidity only in the case of printing facility 5.

4 Conclusions

This study provides experimental data concerning the indoor air pollution in screen printing facilities in Novi Sad for the first time. The main indoor sources of formaldehyde were conventional printing inks and cleaning solutions.

The results indicate that the manual screen printing processes generate formaldehyde due to the higher production volume (25-50 printed products during 1 hour) and application of conventional raw printing materials.

The measured formaldehyde concentration in SPF 5 exceeded 50 times the REL prescribed by NIOSH standard. Such high concentrations suggest that formaldehyde is the indoor air pollutant that could seriously affect the workers health.

Based on statistical regression analysis it was concluded that in the case of SPF 5 a statistically significant linear correlation exists between formaldehyde concentration, temperature and relative humidity.

The findings obtained in this study would significantly enhance our understanding of the levels, emission sources and factors which affect indoor concentrations of formaldehyde in screen printing environment.

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