World Academy of Science, Engineering and Technology
International Journal of Environmental and Ecological Engineering
Vol:10, No:4, 2016

Indoor Air Pollution of the Flexographic Printing Environment

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Open Science Index, Environmental and Ecological Engineering Vol:10, No:4, 2016 publications.waset.org/10004129/pdf

Abstract**—**The identification and evaluation of organic and inorganic pollutants were performed in a flexographic facility in Novi Sad, Serbia. Air samples were collected and analyzed *in situ*, during 4-hours working time at five sampling points by the mobile gas chromatograph and ozonometer at the printing of collagen casing. Experimental results showed that the concentrations of isopropyl alcohol, acetone, total volatile organic compounds and ozone varied during the sampling times. The highest average concentrations of 94.80 ppm and 102.57 ppm were achieved at 200 minutes from starting the production for isopropyl alcohol and total volatile organic compounds, respectively. The mutual dependences between target hazardous and microclimate parameters were confirmed using a multiple linear regression model with software package STATISTICA 10. Obtained multiple coefficients of determination in the case of ozone and acetone (0.507 and 0.589) with microclimate parameters indicated a moderate correlation between the observed variables. However, a strong positive correlation was obtained for isopropyl alcohol and total volatile organic compounds (0.760 and 0.852) with microclimate parameters. Higher values of parameter F than $F_{critical}$ for all examined dependences indicated the existence of statistically significant difference between the concentration levels of target pollutants and microclimates parameters. Given that, the microclimate parameters significantly affect the emission of investigated gases and the application of eco-friendly materials in production process present a necessity.

Keywords**—**Flexographic printing, indoor air, multiple regression analysis, pollution emission.

I. Introduction

ARTIFICIAL casings were developed at the beginning of the 20th century when, in some countries, the supply of natural casings could no longer cope with the demand from the growing meat industries. Following the development of highly automated sausage filling equipment, artificial casings proved to be better suited to those systems, mainly due to their uniformity. Also, from the hygienic point of view, there were certain advantages of artificial casings because the microbiological contamination is negligible, and there is no problem of deterioration during transport and storage. Nowadays, artificial casings are the material of choice, while for smaller calibre products, artificial and natural casings remain equally important [1]. Materials mostly used for the artificial casings manufacture are cellulose, collagen or synthetic materials. The collagen casings are extensively used in the meat industry. Their origin, composition and characteristics are the most similar to the natural ones. A great advantage of collagen casings is their uniform diameter, they are very elastic and stronger than the natural casings. They follow the stuffing and can easily be removed from it. Also, they can be colored, printed, and cut to a certain length [2].

Flexographic printing was developed to print the flexible materials such as paper, cellophane, polypropylene, polyester, polyamide, aluminum foil, natural and collagen casings. At the same time, with the technological advances in printing process, small flexographic printing facilities may present diffuse and permanent polluters, mainly due to the material used during the printing.

Solvent-based inks are widely used in flexographic printing process. Solvents are primarily volatile organic compounds (VOCs), which caused concerns for health and safety, as they are usually very flammable [3]–[7]. The most commonly used solvents are ethanol, isopropyl alcohol (IPA) and mixture of ethyl acetate and ethanol. VOC emissions in flexograhic indoor may occur from the printing process and cleaning operations, the solvent recovery system, and the drying processes. The flow of hazardous substances in manufacturing plant is presented in Fig. 1 [4].

In order to achieve better paint adhesion on artificial casings material, corona treatment is necessary. The usage of corona will contribute to the increased ozone emission in indoor environment, which is impossible to prevent [8]. Partly because of the all mentioned concerns and important reduction of the VOCs concentration in indoor environment, water-based and UV inks were introduced in the world's flexographic facilities, but not completely in the Serbian facilities. Because of these facts, it is interesting and useful to investigate and monitor the flexographic printing process, as well as the parameters of pollutants, in order to provide better management for the environmental protection. Therefore, the objective of this study was to determine the presence of various hazardous pollutants: isopropyl alcohol, acetone, total volatile organic compounds (TVOCs) and ozone during the printing on collagen casings for meat products. In addition, multiple regression analysis was applied in order to investigate the influence of microclimates parameters (temperature, relative humidity and light intensity) on target pollutants concentration.

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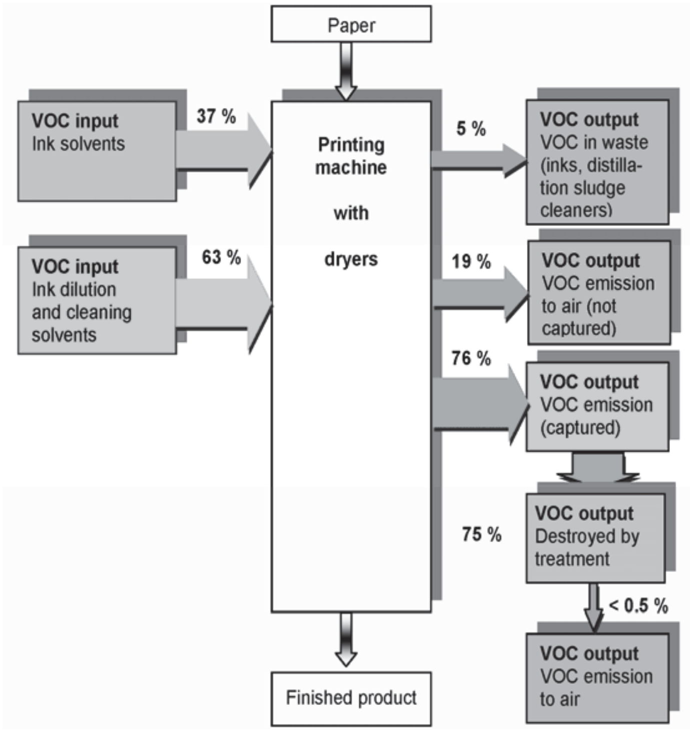

Fig. 1 The flow of hazardous substances in flexography printing (arrows indicate the relevance of the VOC amount) [4]

II. Materials and Methods

A. Site Description, Sampling and Analysis

The air sampling was carried out in a flexographic facility in Novi Sad (Fig. 2).

This facility comprises surface area of approximately 70 m^2 and employs four workers. A technological scheme of flexographic procedure with marked equipment (A, C and D) and auxiliary (B, E, F and G) is presented in Fig. 2.

Five sampling points (Table I) were selected according to technological printing process, as well as to the locations of machines as target gas sources.

TABLE I
DESCRIPTION OF SAMPLING POINTS

Sampling point	Description of sampling point
1	Sampler (X)
2	First set of aggregate for printing process (A)
3	Second set of aggregate for printing process (A)
4	Device for winding printed material (B)
5	Front door (C)

Air was discontinuously sampled in situ for six times, once per 40 minutes, during 4 hours by mobile gas chromatograph (Voyager, PerkinElmer Photovac Inc) and ozonometer (Aeroqual Series 200).

Microclimate parameters were monitored simultaneously with the air pollutants by an instrument Mannix DLAF-8000. Temperature, relative humidity and light intensity were in ranges from 27.4 to 30.8 °C, 22.0 to 25.8% and 45 to 172 lx, respectively (Table II).

TABLE II
VALUES OF MICROCLIMATE PARAMETERS IN PHOTOCOPYING ENVIRONMENT

Sampling point	Microclimate parameter	Sampling time (min)					
		40	80	120	160	200	240
1	t [°C]	27.5	28.0	29.0	30.2	29.9	30.0
	RH[a][%]	24.1	23.5	22.2	23.2	25.2	23.4
	LI[b] [lx]	145	116	88	154	94	124
2	t [°C]	27.4	28.0	28.9	30.5	30.0	29.5
	RH[a][%]	23.9	22.8	22.5	23.0	24.8	22.7
	LI[b] [lx]	92	92	172	113	99	153
3	t [°C]	27.9	27.9	29.2	30.6	29.9	30.1
	RH[a][%]	24.1	24.0	25.3	23.3	25.5	23.2
	LI[b] [lx]	45	65	48	66	53	63
4	t [°C]	27.7	28.8	29.9	30.5	30.1	29.9
	RH[a][%]	24.4	23.5	23.0	24.5	258	25.4
	LI[b] [lx]	72	81	92	100	88	93
5	t [°C]	27.8	28.6	30.1	30.8	30.2	30.0
	RH[a][%]	24.9	22.0	22.7	23.2	24.2	22.9
	LI[b] [lx]	125	112	148	166	91	130

[a]Relative humidity, [b]Light intensity

Voyager's powerful mobile gas chromatograph is composed of a built-in three-column configuration with an isothermal oven for fast gas chromatography analysis with a miniaturized dual detection system of Photoionization detector/Electron capture detector. The basic purpose of this instrument is to provide flexibility of analysis and reliable results [9]. In this study, Voyager was used for the concentrations measurement of IPA, TVOCs and acetone.

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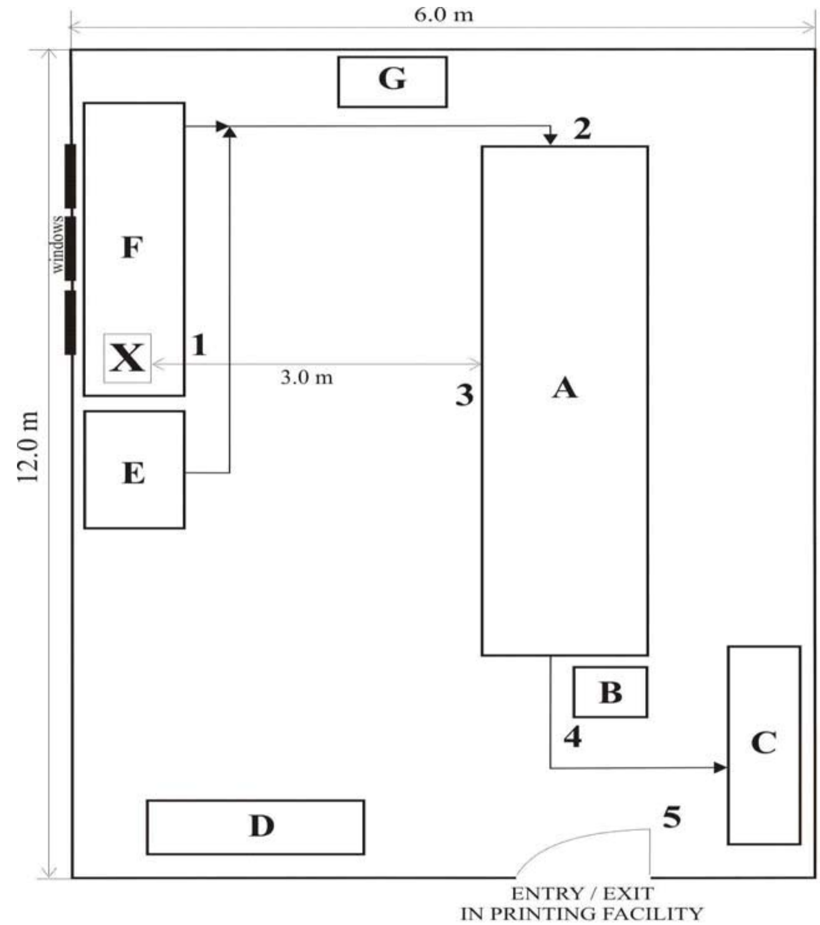

Fig. 2 Technological scheme of a flexographic procedure, A – flexo printing machine, DM FLEX C18-400; B – corona generator, CLNE 015-130-1KB4; C – shelf with printed products; D – shelf with raw printing materials; E – printing plate mounting machine; F – desk with raw printing materials; G – wash sink for flexo plates; 1 - 5 – location of sampling point for discontinuous measurement; X – location of air sampler (placed on the height of 1.2 m)

Portable Ozonometer Aeroqual Series 200 (Aeroqual Ltd.) is designed for an easy and affordable measurement of indoor and industrial air quality. The portable gas detectors can be configured with a wide range of single or multiple gas sensor heads, designed specifically for indoor air quality monitoring. In this investigation one sensor head, precisely for ozone, was used. The operation principle of ozonometer is based on the usage of gas semiconductor sensor technology. This technology presents a combination of smart measurement techniques and mixed metal oxide semiconductor sensors that exhibit an electrical resistance change in the presence of a target gas. A detection limit of instrument for ozone is 0-50 ppm whereas the precision of instrument is 1 ppb [10].

Mannix DLAF-8000 mini airflow meter can be used for monitoring of ambient temperature, light intensity, pressure, humidity and gas flow. The instrument is distinguished with the low-friction ball bearing mounted wheel design, which provides high accuracy at high and low gas flow. It is characterized by high precision thin-film capacitance humidity sensor with fast response to the humidity changes. Built-in microprocessor circuit assures accuracy while multi-channel display at the same time measures values of relative humidity, temperature, light intensity or gas flow [11].

B. *Multiple Linear Regression Analysis*

Statistical method that describes the relationship between different phenomena is well known as a regression analysis. As an extension of simple linear regression, multiple linear regression analysis allows researchers to answer questions that consider the roles of multiple independent variables in accordance with a single dependent variable. In the other words, it attempts to model the relationship between two or more explanatory variables and a response variable by fitting a linear equation to observed data [12].

Phenomena on which the prediction is obtained, X_1, X_2, ..., X_k, population, are independent (deterministic) variables or factors and their occurrence depend on dependent (stochastic) variable Y. Therefore, every value of the independent variable x is associated with a value of the dependent variable y [12], [13].

The relationship between the dependent variable and the independent variables is represented by (1) [14], [15]:

$$y_i = \beta_0 + \beta_1 x_{1i} + \beta_2 x_{2i} + ... + \beta_p x_{pi} + e_i \quad (1)$$

where: β_0 is the constant term; β_1 to β_p are the coefficients relating the p independent variables to the variables of interest; and e_i is the stochastic gradient.

In this study, multiple linear regression analysis was used to evaluate the influence of microclimates parameters as independent variables on target pollutant concentration, as dependent variable.

III. Results and Discussion

The concentrations of isopropyl alcohol, acetone, total VOCs and ozone were measured in the flexographic environment. The results are presented as average values by sampling points in Table III. The concentrations of all pollutants varied during the sampling time. Formation of isopropyl alcohol and total VOCs was dominant at 200 minutes from the production start with the highest average concentrations of 94.80 ppm and 102.57 ppm, respectively. Such high concentrations were influenced by increasing of light intensity during the 4-hour working time (I=45 lx to 172 lx, Table II). Also, higher production volume contributed to the VOCs accumulation in flexographic facility due to absence of ventilation system (turn-off ventilation). As a result of the corona generator work, ozone concentration reached the maximum level in 80 minutes of measurement (1.00 ppm), which conditionally increased the VOCs concentrations in the flexographic indoor environment.

TABLE III
Average Concentrations of Air Pollutants in a Flexographic Facility

Substance	Average concentration (ppm)					
	Sampling time (min)					
	40	80	120	160	200	240
Isopropyl alcohol	0.00	59.60	33.70	86.20	94.80	90.70
Acetone	0.00	0.58	0.57	0.11	0.0	0.43
TVOCs	0.76	21.22	32.08	79.61	102.57	96.87
Ozone	0.21	1.00	0.47	0.43	0.03	0.66

Further results analysis is based on the usage of multiple regressions analysis in order to investigate the mutual correlation between the pollutants concentration (IPA, acetone, TVOCs and ozone) and the microclimates parameters (temperature, relative humidity and light intensity). Dependent variables are IPA, acetone, TVOCs and ozone concentrations, whereas microclimates parameters present independent variables. Data analysis was performed by using the program package STATISTICA version 10 and the results are presented in Table IV.

Based on the obtained values of multiple coefficient of determination (0.507 and 0.589) it was established a moderate correlation between microclimates parameter and ozone, as well as between microclimates parameter and acetone, respectively. In contrast, a direct, strong correlation was determined between microclimates parameter with IPA and TVOCs, as indicated by the multiple coefficient of determination values of 0.760 and 0.852, respectively.

TABLE IV
The Results of Multiple Linear Regression Analysis

Mutual influence	R^c	F^d	t^e
t, RH, LI and IPA	0.760	11.861	-4.184
t, RH, LI and acetone	0.589	4.622	2.841
t, RH, LI and TVOCs	0.852	22.939	-6.877
t, RH, LI and ozone	0.507	3.002	2.506

[c]Multiple coefficient of determination; [d]Parameter obtained from *F*-statistic, [e]Parameter obtained from Student's *t*-test

Considering the obtained values of parameter *F* for all observation, it has been found that investigated dependence is statistically significant, since all *F* values are greater than $F_{critical}$ (2.98).

Values of parameter $|t|$ higher than $t_{critical}$ (1.706), appeared with all four investigated dependences, pointed out that microclimates parameters, in terms of independent variables, have significant influence on the emission of target pollutant, as dependent variable.

Within the results of multiple regression analysis corresponding regression models are obtained (2)–(5):

$$y = 1733{,}149x - 1907{,}065y + 40{,}305z \quad (2)$$

$$y = 10{,}539x - 11{,}891y + 0{,}170z \quad (3)$$

$$y = 1805{,}597x - 1922{,}081y + 13{,}524z \quad (4)$$

$$y = 13{,}970x - 15{,}098y + 0{,}218z \quad (5)$$

Figs. 3–6 present the spatial dependence of various pollutants concentrations on three independent variables - temperature, relative humidity and light intensity. This regression surfaces are obtained from regression models (2)–(5).

In the three-dimensional graphs, x-axis and y-axis represent the microclimates parameters while z-axis represents the pollutants concentration. The resulting surface shows an average stacking variation of investigated variables, presented by the general regression model: $y = b_0 + b_1xi_1 + b_2xi_2$ (Table IV). In this way, a quantitative agreement between the change of variables is possible and thus a more accurate knowledge of their mutual relations is obtained.

Based on the obtained results, it was confirmed that solvent-based printing materials (inks, thinners, retarders) are the main sources of VOCs emission. In order to provide properly controlled exposure of ozone and VOCs and to eliminate health risks, it is necessary to ensure the adequate substitution of solvent-based inks with water-based or UV-curing inks. VOC emission reduction can also be achieved with improved handling, VOC-free cleaning and efficient capture and treatment of waste gas.

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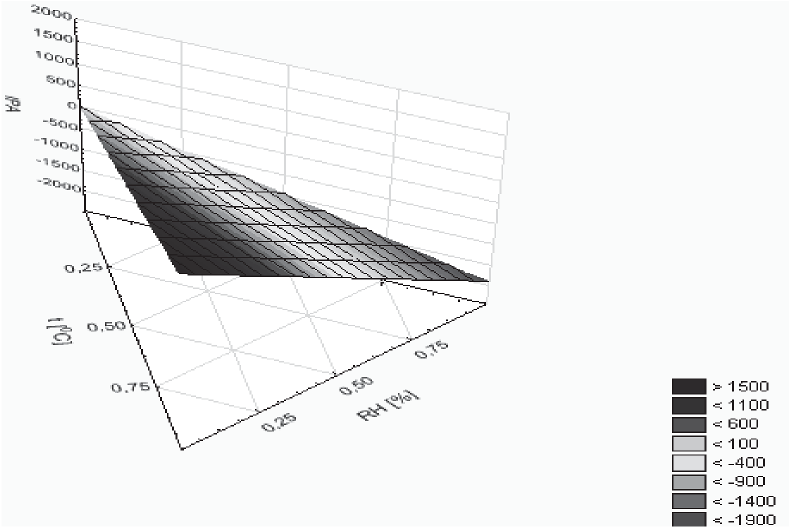

Fig. 3 The dependence of microclimates parameters and IPA

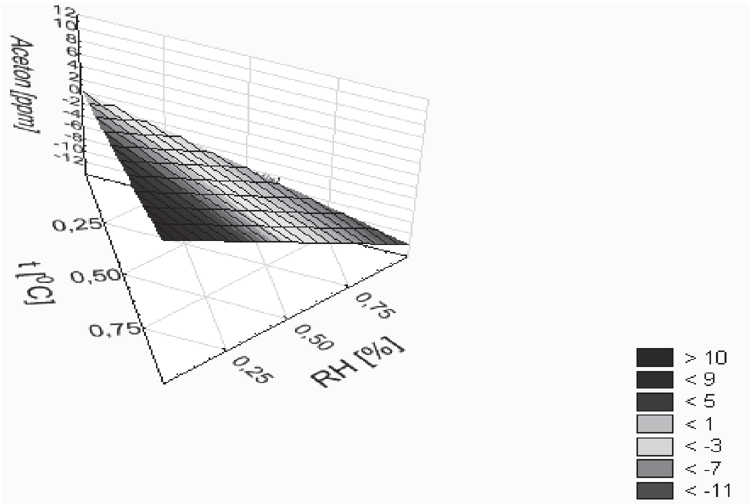

Fig. 4 The dependence of microclimates parameters and acetone

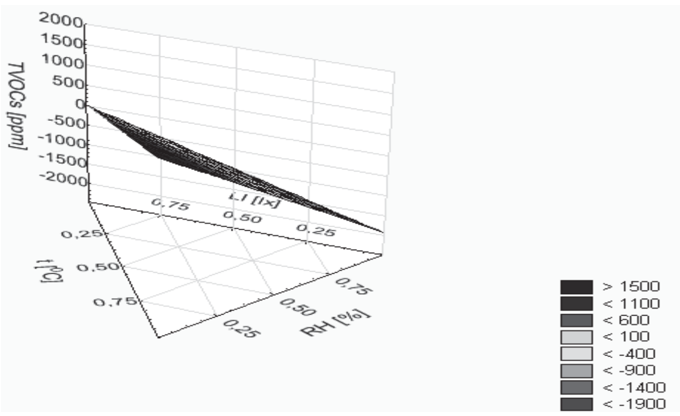

Fig. 5 The dependence of microclimates parameters and TVOCs

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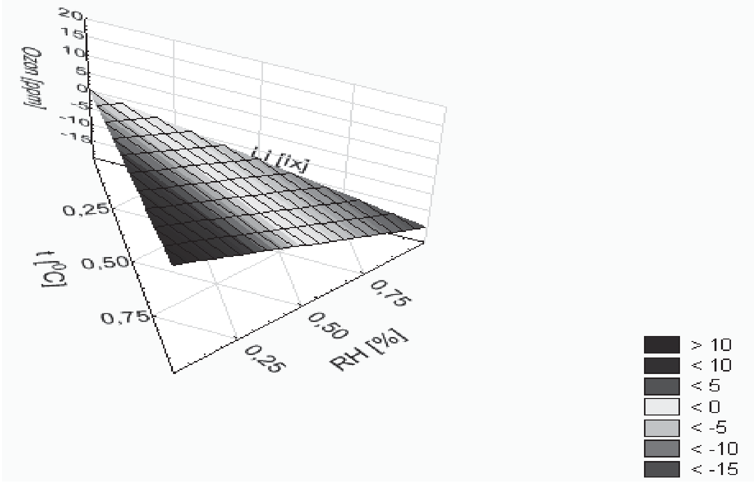

Fig. 6 The dependence of microclimates parameters and ozone

IV. Conclusion

This study provides experimental data concerning the indoor air pollution in flexographic facility in Novi Sad. The identification and evaluation of occupational hazards confirmed the presence of isopropyl alcohol, acetone, TVOCs and ozone in indoor air during the printing of collagen casing.

Analyzing data showed that the average concentrations of pollutants varied during sampling time. Increasing of light intensity affected the faster solvent evaporation from ink, which significantly contributed to the evaporation of organic pollutants. Additionally, the usage of corona generator influenced increasing of ground ozone concentrations in time interval from 40 to 120 minutes and after 200 minutes from starting the printing production. A multiple linear regression analysis was performed in order to predict and determine the mutual correlation between dependent variables (pollutants concentration) and independent variables (microclimates parameters). The obtained multiple coefficient of determination pointed out a moderate and strong positive correlation between the investigated variables. Also, a great influence of microclimate parameters on the emission of target gases was proved. Nonetheless, it demonstrates the existence of statistically significant differences between the tested parameters.

Based on the results and data classification collected under real conditions of the printing production, the introduction of adequate eco-friendly replacements for certain toxic substances must be suggested. As the key environmental issue for flexographic printing facilities is solvent emissions to indoor air, the application of eco-friendly materials must be a priority.

Acknowledgment

The authors acknowledge the financial support of the Ministry of Education, Science and Technological Development of the Republic of Serbia within the Projects No. TR 34014.

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