UNIVERSITY OF BUCHAREST
FACULTATY OF CHEMISTRY
DOCTORAL SCHOOL OF CHEMISTRY

ABSTRACT OF THE PhD THESIS

INDOOR AIR POLLUTION STUDY

PhD student:
Elena COMŞA (BUCUR)

PhD Supervisor:
Prof. Univ. Dr. Andrei Florin DĂNEŢ

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CONTENT *(The numbering of pages corresponds to the PhD thesis.)* Pag.

THEORETICAL PART

ORIGINAL PART

Ambient air quality has always been an area of great interest due to its implications on the health and welfare of the population, but also because of its potential effect on other environmental factors and historical monuments. Especially in the last decades, studies have brought to the forefront of scientific concerns a new field - indoor air quality (IAQ) - giving it a status that is sometimes priority in terms of influence on the health and comfort of people (Franklin, 2007; Harrison, 1997; Nazaroff, 2013).

Assessing air quality in a building and impacts on receptors of interest is a process of high complexity due to the large amount of data to be analyzed and interpreted, the large number of variables and possible reciprocal, synergistic or antagonistic influences with effect on materials and / or the perception of human discomfort.

In this sense, throughout the thesis, based on literature and case studies, we have tried to answer three major questions: i) how do we approach and identify the interest indicators for assessing the air quality in a building? ii) by what methods do we measure, process and interpret effectively the series of monitoring data to identify specific sources of pollution and air quality assessment? iii) how do we quantify the impact of air quality on receptors?

We can find the answer to the first question by going through the first two chapters of the thesis, in which are presented selectively, in the two major fields of interest (air quality in the offices - Chapter 1 and museums - Chapter 2) useful literature for carrying out case studies assessment of air quality in buildings. These chapters familiarize us with the main chemical pollutants that can be identified in the two types of spaces and the specific sources of emissions, with the level of air concentrations and the effects they may have on the health or the materials from which various exhibits are made.

Chapter 3 provides information on the best known methods of assessing air quality in buildings and measuring the concentration of chemical pollutants present in the air, but also how a number of specific indicators such as *the I/O ratio, the infiltration factor,* F_{in}*, or penetration factor,* P, can be used to interpret the measurement results. Also in this chapter a series of statistical correlation and regression information used in the experimental part of the thesis are systematized for the characterization and interpretation of data series for the monitoring of the concentration of pollutants in the air. We also respond to the second question: *"By what methods do we measure, process and interpret effectively the series of monitoring data to identify specific pollution sources and assess air quality?"*

Chapter 4 is dedicated to the two case studies on indoor air quality assessment: the first one, carried out in a new office building, and the second in the exhibition spaces belonging to a museum. For the assessment of air quality in the office building, both the method based on health insurance and the comfort-based method were used. The main aspects of the air quality assessment process were the verification of the concentration of the pollutants within the limits imposed by the regulations in force and the identification of the sources of pollution in order to reduce the effects on the persons / exhibits.

The answer to the third question, *"How do we quantify the impact of air quality on receptors*?" is given in Chapter 5 through an example of a binary logistic regression application for assessing the possible cummulated impact of air quality on the exhibits from one of the museum rooms. There are step by step explanations regarding the methodology for using the binary logistic regression for the calibration and validation of the mathematical model for the prediction of the efect probability, and how the results obtained from the analysis can be interpreted and used.

The probability values can also be interpreted as *the possible environmental impact on the exhibits, taking into account a cumulative effect of all parameters*; thus, for an easier interpretation, an impact assessment system based on the calculated value of the probability (p) is proposed: i) very low impact for $0 < p < 0.2$; ii) low impact on $0.2 < p < 0.4$; iii) moderate impact on $0.4 < p < 0.6$; iv) strong impact for $0.6 < p < 0.8$ and v) very strong impact for $p > 0.8$.

The results of the application demonstrate the utility of statistical analysis methods in interpreting the data series obtained by monitoring environmental parameters. Thus, from the regression coefficients $ß_i$ and the test results for the statistical significance, the pollution indicators with significant effect on the exhibits can be identified and ranked, allowing a prioritization of the pollution reduction measures and, implicitly, the costs of implementing these measures. At the same time, with the mathematical model developed, predictions can be made about the effect that a particular context of pollution can induce on exhibits. By calculating the probability p and assimilating it as a possible cumulative impact, it is a useful indicator in assessing the air quality for the spaces for storing or displaying patrimony objects.

The method may also be useful in deciding on the reduction measures to be implemented in an exposure space by simulating some theoretical situations, analyzing the results obtained and by including the mathematical model in the continuous monitoring system of museums can be calculated in real time the effect of the environment on the exhibits.

Objectives of the thesis

1. Air quality assessment inside two types of buildings: an office building and exhibition spaces belonging to a museum by:

    - long-term monitoring of concentrations for specific chemical compounds (SO_2, NO_2, O_3, CO, CO_2, CH_2O, H_2S), particulate matter in the air and microclimate parameters (temperature and humidity);
    - comparing the results obtained from monitoring with the limit values set or recommended by the specific regulations in force to identify chemical compounds with potential impact on the health of the staff involved in office activities or the integrity of exhibits in a museum;
    - determination of metal content, water-soluble anions and PAHs from particulate matter in the air in order to identify / demonstrate the existence of indoor sources of air pollution with dusts;
    - establishing the possible sources of indoor air pollution;

2. Using statistical analysis methods to interpret the results of the indoor air quality monitoring process, taking into account in particular:

    - correlation analysis between the parameters inside and outside the buildings for the purpose of identifying the sources of pollution;
    - linear regression to identify possible internal sources of airborne air pollution;
    - binary logistic regression applied to the monitoring data obtained in the exhibition spaces of a museum and interpretation of the results with the purpose: i) establish a hierarchy of the chemical compounds present in the air depending on the potential effect on the exhibits; ii) an assessment of the cumulative effect of air pollutants on the exhibits, iii) development and validation of a predictive mathematical model for objective assessment of the impact of indoor air quality on objects exhibited in museums;

Aspects of originality and novelty of the thesis

The scope of the thesis - the air quality in office and museum spaces - is novel, being little studied in our country and also abroad.

In terms of originality, the thesis presents for the first time the use of binary logistic regression for assessing the indoor air quality in a museum. To date, according to our information, no work has been published to address this topic in specialized scientific literature from abroad or across the country (search: Web of Science and Google, June 20, 2017). The use of logistic regression allows, among other things, a hierarchy of pollutants present in the air depending on the effect on the exhibits in a museum, a prediction of their cumulative effect, and the choice of the most appropriate measures to reduce the effects of pollutants present in the air on the exhibits.

4.1. Assessment of air quality in office spaces

The air quality tests in the office spaces were first focused on assessing the level of air pollution in urban areas: particulate matter ($PM_{2.5}$, PM_{10} and total suspended particles), NO_2, SO_2, CO, O_3 and CO_2, as an indicator of air quality in the spaces where people are present. In the process of identifying the sources of pollution, additional tests were required to determine the concentration of formaldehyde and H_2S in the indoor air and the presence of some compounds in the dusts taken inside and outside the building: metals, polycyclic aromatic hydrocarbons and water-soluble anions. The air quality assessment in the building was carried out by checking the concentration of the compounds present in the air within the limits set by the regulations in force *(method based on health and the comfort regime according to ISO 16814: 2008).*

Air quality studies in office spaces were conducted during 2013-2017 in a new building located on the outskirts of Bucharest, in an area with low road traffic and no major sources of industrial pollution.

In order to monitor the concentration of pollutants in the air inside and outside the building, calibrated sampling / measuring equipment was used in compliance with the provisions of the method standards and referential SR EN ISO/CEI 17025:2005 regarding validation/ verification of analytical methods and quality assurance of results. Statistical tests were performed using the AnalyiseIt and SPSS 20.0 programs.

In order to determine *the level of particulate matter pollution of the indoor air* and the identification of the specific sources of pollution, the following tests were carried out: i) assessment of $PM_{2.5}$ pollution level; ii) the distribution of particulate matter by dimensional fractions; iii) analysis of some compounds present in particulate matter (metals, PAHs and water soluble anions).

Four rooms were selected for the $PM_{2.5}$ particulate matter pollution assessment: two offices (E3-13 and E1-23), a weighting room and a transit space; based on the preliminary results, the E3-13 office space was selected for the air quality assessment within an office.

The results of the tests for outdoor particulate matter during 2 - 24.04.2013 revealed the exceedance of the limit value for approx. 70% (15 values out of 23) of daily averages (Fig. 4.8), although road traffic is low in the area and there are no significant sources of industrial pollution. According to the literature data (Gradon, 2009; Harrison şi colab., 2012; Nicholson, 1988) the high concentration of particulate matter are due to resuspension phenomenum of sedimented particulate matter, favored by the presence of Northwest wind during the test and the lack of precipitation.

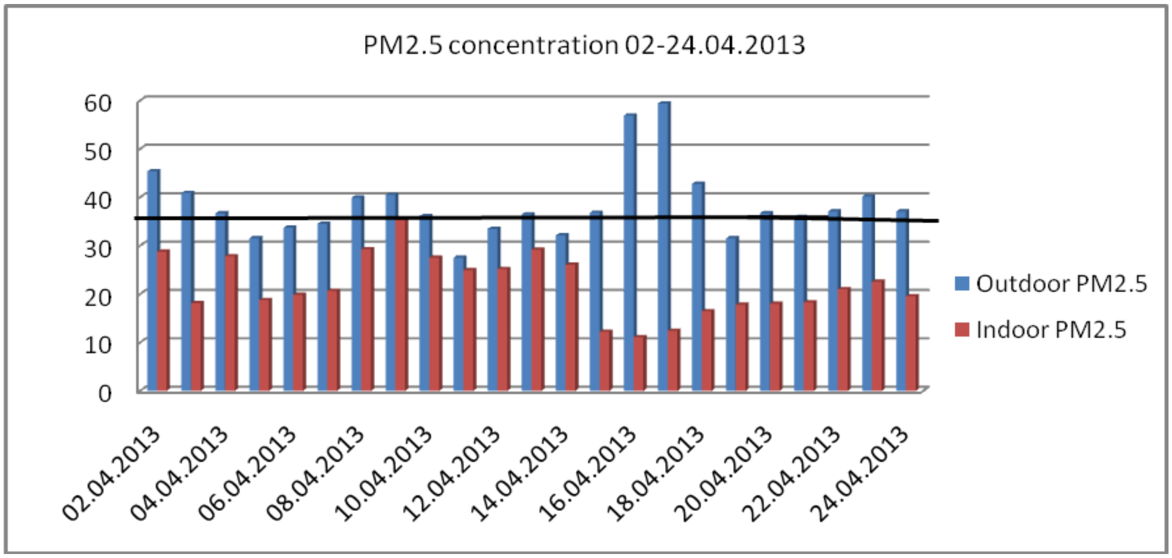


Fig. 4.8 Time variation of $PM_{2.5}$ concentration indoor and outdoor, 2 - 24.04.2013 (Bucur and Danet, accepted publication *Environ Eng Manag J*).

At the same time, $PM_{2.5}$ concentrations were consistently lower indoor than outdoor; constantly smaller values in the interior demonstrate a good insulation of the building and the lack of important cracks in the building that allow the penetration of the outside.

Regarding the dimensional fractionation of dusts, the tests carried out in the E3-13 office during 14-23.06.2013 revealed the presence in the air of especially small dust particles, $PM_{2.5}$; thus, on average, total suspended particulate matter (TSP) contain 91% of $PM_{2.5}$ particles, which determines us to consider the hypothesis of the existence indoor of small dust pollution sources.

This hypothesis is supported by the results of the linear regression applied to the concentration of particulate matter indoor and outdoor, respectively the high values of the slope and the regression line intercept, as well as of the I/O ratio in the office E3-13 compared to the other spaces (Table 4.11). Also, Pearson correlation coefficients (r), the determining coefficient (r^2), but also the values of I/O ratio indicate outdoor air as a major source of air pollution in the building.

Table 4.11. The results of the Pearson correlation analysis and linear regression applied to $PM_{2.5}$ concentration monitoring results inside and outside the building (Bucur and Danet, accepted publication, *Environ Eng Manag J*).

Room/Period	**Pearson correlation coefficient, r**	**Liniar regresion**			**Ocupancy degree, pers/100m^3**	**I/O**	**Diference betweeen I/O şi F_{in}**
		r^2	**slope, F_{in}**	**intercept**			
Reception/ 02-08.04.2013	0.58	0.34	0.608	0.512	1.7	0.62	0.012
Office E3-13/ 09-14.04.2013	0.86	0.74	0.759	1.955	6.5	0.82	0.061
Office E1-23/15-18.04.2013	-0.43	0.19	-0.093	17.58	2.5	0.28	-
Weighting room, E3-22/19-24.04.2013	0.79	0.62	0.535	0.047	0.9	0.54	0.005

The Pearson correlation analysis applied to the occupancy degree and intercept value demonstrates a very good direct correlation (r = 0.99) of the two parameters, indicating the presence of staff in the room (expressed as occupancy) as a possible source of pollution with $PM_{2.5}$ transferred to air on their clothing and footwear. The presence of staff can also cause the resuspension of already sedimented particles on furniture or flooring (Gradon, 2009; Thatcher and Layton, 1995), which can be reduced by proper maintenance and sanitation of the premises. This hypothesis is also supported by the time variation (24 hours) of particulate matter concentration in E3-13 office, which shows a continuous increase in particulate matter concentration during nighttime.

The specificity of the activities carried out in this office, typing, printing and copying documents suggests the printing / copying process as a possible source of internal pollution with particulate matter.

Additional information useful in the process of identifying indoor sources of particulate matter has been provided by the results of chemical composition of particulate matter tests taken from the office indoor air: the concentrations of metals, anions and polycyclic aromatic hydrocarbons (PAH).

Regarding the metals (Table 4.13) it is worth noting the large amount of Fe from particulate matter, knowing that the iron oxides are one of the main components of black toner used in copiers, along with Mn, Zn, Cr, Ti and As (Bai et al., 2010). The barium also comes into the composition of the special paper for the copier, which confirms the hypothesis that in the E3-13 office the copying process can be an important internal source of small particle pollution.

The ion chromatography analysis of the extracts in distilled water at 20°C led to the identification of the following anionic quantities in the three dimensional particle fractions in the same order: $NO_3^- > SO_4^{2-} > Cl^- > F^- > PO_4^{3-} > NO_2^-$.

In the largest quantity the NO_3^- anion was identified, the concentration of which is approx. 40 times greater than the next anion, SO_4^{2-}. The presence of NO_3^- in the largest quantity can be explained by the intake of particulate matter from outdoor air, the adjacent area being a former agricultural area, where chemical fertilizers have been used in excess.

Tabel 4.13. Composition in metals and anions corresponding to particulate matters: $PM_{2.5}$, PM_{10} and TSP, E3-13 (Bucur and colab., 2014)

Metal	**Concentration, µg/g**			**Ratios**			**Anion**	**Concentration, mg/g**		
	$PM_{2.5}$	**PM_{10}**	**TSP**	**PM_{10} / TSP**	$PM_{2.5}$ / **PM_{10}**	$PM_{2.5}$ / **TSP**		$PM_{2.5}$	**PM_{10}**	**TSP**
As	32	27	20	1.38	1.17	1.62	**F^-**	0.23	0.38	0.30
Cu	69	58	60	0.97	10.87	10.56	**Cl^-**	7.11	5.56	6.82
Cr	13	31	19	1.67	0.42	0.71	**NO_2^-**	0.005	0.011	0.004
Fe	1068	1521	1935	0.79	0.70	0.55	**NO_3^-**	**397.6**	**389.6**	**382.7**
Mn	21	29	41	0.72	0.72	0.52	**PO_4^{3-}**	0.18	0.22	0.25
Ni	10	10	90	0.11	1.09	0.12	**SO_4^{2-}**	8.21	9.13	10.87
Pb	146	193	274	0.70	0.76	0.53				
Ti	23	39	47	0.83	0.60	0.50	**Anion**	**Ratios**		
Zn	185	418	164	2.55	0.44	1.13		**PM_{10} / TSP**	$PM_{2.5}$ / **PM_{10}**	$PM_{2.5}$ / **TSP**
Al	207	801	837	0.96	0.26	0.25				
Ba	143	161	75	2.14	0.89	1.90	**F^-**	1.27	0.61	0.77
Na	493	765	751	1.02	0.65	0.66	**Cl^-**	0.82	1.28	1.04
K	3171	3377	3363	1.00	0.94	0.94	**NO_2^-**	2.75	0.45	1.25
Ca	643	801	2143	0.37	0.80	0.30	**NO_3^-**	1.02	1.02	1.04
Mg	133	254	427	0.59	0.52	0.31	**PO_4^{3-}**	0.88	0.82	0.72
r, Pearson				**0.93**	**0.96**	**0.88**	**SO_4^{2-}**	0.84	0.90	0.76

Another possible source of particulate matter pollution can be the the smoking spot located on the same floor of the building; the tests performed for HPLC-FLD determination of the presence of PAHs in particulate matter indicate the highest concentrations of PAHs in the particulate matter collected from outdoor air, 36.30 ng/m^3 (expressed as sum of PAHs), and the lowest in E3-13 office: 21.82 ng/m^3. Among the analyzed compounds, benzo(a)pyrene has the highest concentrations in the E3-13 office and the crisene in the outdoor air and smoking site.

The fact that we find PAHs in the particulate matter from the outdoor air is not a novelty; urban air pollution with PAHs is unanimously recognized (Querol et al., 2007, Khan et al., 2010, Khaiwal et al., 2006, Wingfors et al., 2001, Baek et al., 1991) and Bucharest is no exception (Bucur and Danet, 2016*b*).

Their presence in indoor particulate matter may be due to the influence of smoking-related emissions but also of smokers (Castro et al., 2011, Nicula et al., 2014, Gundel and Mahanama, 1995); Ren et al. (2006) also identified the presence of PAHs in particulate matter that accumulate in computers and other office equipment containing components from plastic materials.

The results make us consider smoking and, implicitly, smoking-specific emissions a less important source of dust than originally assumed, with little influence on the air quality in office E3-13.

Tests to measure the *level of air pollution in office buildings with gaseous compounds* and identification of sources of pollution were carried out in two monitoring campaigns (13-27.07.2016 and 19-27.01.2017) and the results corresponding to the warm season are presented in Table 4.17. Urban pollution-specific compounds were determined: NO_2, SO_2, CO, O_3, and same indicators of indoor air quality: CO_2, H_2S and formaldehyde.

We find a low level of pollution with these compounds, the values being below the recommended limit values both indoor and outdoor.

The sub-unit values of the I/O ratio for all indicators also demonstrate the lack of indoor sources for these compounds. The outdoor air thus remains the most important source for indoor air with NO_2, SO_2, CO and O_3, sustained by the values of the Spearman correlation coefficients (q) presented in Table 4.18. Thus, the correlation between indoor and outdoor is good for NO_2 and SO_2 ($q_{NO2} = 0.627$ and $q_{SO2} = 0.605$) and moderate for CO ($q_{CO} = 0.449$).

With regard to the air quality assessment of the office by the comfort method, taking into account the CO_2 concentration, the values of the differences between the measured concentrations inside and outside the building, the air quality in office E3-13 is in the quality class IDA 1 – high quality indoor air - for 97% of the measurements carried out between 19-26 January 2017, the remaining 3% of the cases falling under the appropriate conditions of IDA 2, namely moderate air quality.

Table 4.17 Air quality monitoring results in E3-13 office and outdoor during 13-27.07.2016

	I- NO$_2$ μg/m^3	O-NO$_2$ μg/m^3	I-SO$_2$ μg/m^3	O-SO$_2$ μg/m^3	I-CO mg/m^3	O-CO mg/m^3	O-temp ^{0}C	O-RH %
Number of measurements	157	184	156	184	156	184	340	340
Mean	15.47	22.2	4.2	4.72	0.184	0.235	25.44	56.01
Median	15.45	20.15	4.23	4.72	0.185	0.23	25.3	50.2
Standard Deviation	5.716	10.05	0.745	0.905	0.031	0.055	4.81	20.55
Skewness	0.311	0.819	0.405	0.385	-0.047	0.822	-0.04	0.502
Standard Deviation of Skewness	0.194	0.179	0.194	0.179	0.194	0.179	0.132	0.132
Kurtosis	0.490	0.741	-0.314	-0.185	-0.129	0.696	-1.05	-0.955
Standard Deviation of Kurtosis	0.385	0.356	0.386	0.356	0.386	0.356	0.264	0.264
Minimum	3.36	4.42	2.75	3.06	0.11	0.14	15.9	26.7
I/O Ratio	0.70		0.89		0.78		-	-
Maximum	33.71	54.31	6.40	7.29	0.27	0.43	34.7	99.6
Limit values, ISO 16814:2008	200(1h)	-	125(24h)	-	10(8)	-	-	-
Limit values, Low 104/2011	-	200(1h)	-	125(24h)	-	10(8)	-	-

Table 4.18 Spearman's correlation coefficients for indoor (I) and outdoor (O) air quality parameters, 13-27.07.2016

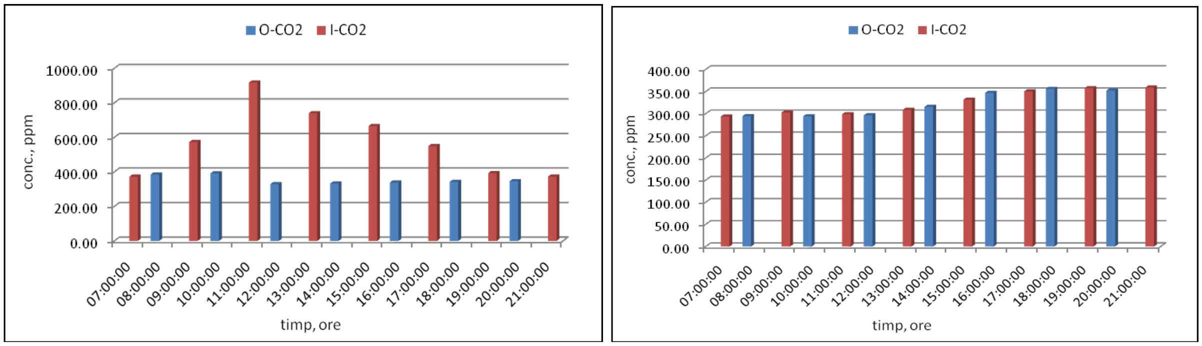
	I- NO$_2$	I-SO$_2$	I-CO	O-NO$_2$	O-SO$_2$	O-CO	O-temp	O-RH
I- NO$_2$	1.000							
I-SO$_2$	-0.012	1.000						
I-CO	0.302**	0.231**	1.000					
O-NO$_2$	0.627**	-0.022	0.226**	1.000				
O-SO$_2$	0.005	0.605**	0.271**	0.049	1.000			
O-CO	0.352**	0.115*	0.449**	0.537**	0.310**	1.000		
O-temp	0.111*	-0.233**	-0.041	0.256**	-0.333**	0.175**	1.000	
O-RH	0.193**	0.318**	-0.021	0.017	0.266**	-0.204**	-0.050	1.000

*Semnificative corellation, level of probability: 0,01

**Semnificative corellation, level of probability: 0,05

Colour	🟦	🟦	🟦	🟦	
Semnification of corellation	very weak	weak	moderated	good	very good

The contribution of the presence of the personnel in the analyzed space is also evidenced by the results obtained by monitoring the concentration of CO_2 (Figure 4.14), H_2S and formaldehyde which show substantial increases of the concentration in the indoor air during the work program (Figures 4.15 and 4.16), reaching the recommended limit value of 3.5 times in case of formaldehyde.



(a) (b)

Fig. 4.14 Time varying the concentration of CO_2 in the office and outside the building on a working day (a) and weekend (b).

Thus, we find a large variation in the CO_2 concentration in the office (the difference between the maximum and the lowest value of 656.5 ppm) compared to the outside air of the building (136.8 ppm) and a higher I /O ratio (I/O ratio = 1. 2) which highlights the presence of CO_2 sources inside, represented by the staff present in the office.

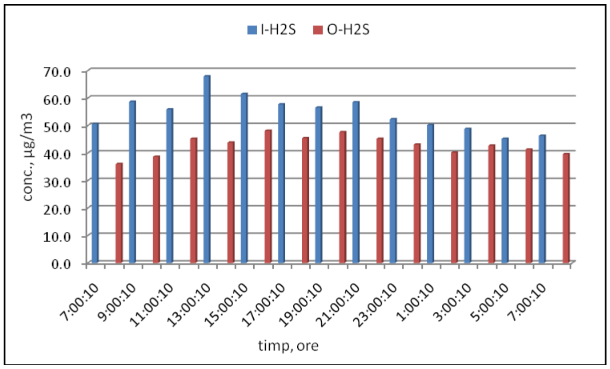


Fig. 4.15 Time varying H_2S concentration in the office and outside the building, 03-04.02.2017, µg/m^3.

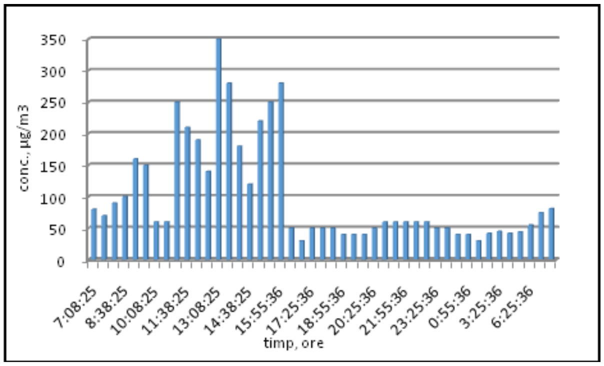


Fig. 4.16 Diagram of time variation of CH_2O concentration in the office, 03-04.02.2017, µg/m^3.

In conclusion, the air quality of the E3-13 office does not pose any special health problems if appropriate measures are taken to reduce the formaldehyde concentration. An efficient method can be frequent ventilation of the space or replacement of the air conditioning system with a more efficient one to allow adequate freshening of the air introduced into the room. Reducing office occupancy can also be a solution for improving air quality and comfort.

4.2. Assessment of air quality in areas where heritage objects are displayed.

Tests to assess indoor air quality and the effect pollutants present in the air on exhibits were conducted in 2014-2015 in two exhibition halls belonging to the National Museum of Romanian Aviation (Hangar 1 and Hangar 2) and in a wooden church located on the same site. Taking into account the location of the museum, urban area, and the peculiarities of the exhibits, in order to assess the air quality inside and outside the museum, the concentrations of NO_2, SO_2, O_3 and $PM_{2.5}$ were monitored, in parallel, in two campaigns during the hot and cold season. At the same time, the parameters temperature and humidity (RH) indoor and outdoor were monitored; consideration was given to assessing daily variations in temperature and humidity that can generate repeated contraction / expansion processes and, implicitly, a fragility in the structure of the exhibits. The limit values recommended by the ASHRAE Handbook - Chapter 21, 2007 for general collections (Table 4.26) were used to assess the air quality in the museum.

Table 4.26. Limit values recommended for NO_2, SO_2, O_3, $PM_{2.5}$ in museums (ASHRAE Handbook - Chapter 21, 2007).

Pollutants	**Suggested limits for collections**				**Invervention limits**			
	sensible materials		**general collection**		**urgent**		**extremely urgent**	
	ppb	µg/m^3	ppb	µg/m^3	ppb	µg/m^3	ppb	µg/m^3
NO_2	<0.05 – 2.6	<0.1-5.3	2-10	4-20	26-104	110-210	>260	>530
SO_2	<0.04 – 0.4	<0.1-1	0.4-2	1-5.7	8-15	23-43	15-57	43-160
O_3	0.05	0.1	0.5-5	1-10	25-60	50-130	75-250	160-530
$PM_{2.5}$	-	<0.1	-	1-10	-	10-50	-	50-150

The results of the tests carried out in the two hangars revealed a good indoor air quality with average concentrations of NO_2, SO_2 and $PM_{2.5}$ lower than the recommended ASHRAE limits for general collections (Table 4.34 presents the results of the air quality monitoring in

Hangar 2, between 23.02.2015-19.03.2015); but in the case of ozone, frequent exceedances of the recommended limit value (10 μg/m^3) have been identified, however, values below the lower limit of intervention (50 μg/m^3). Also, for Hangar 2, exceedances of the $PM_{2.5}$ concentration in the warm season were identified with an average of 10.7 μg/m^3 vs. 10 μg/m^3, the recommended limit value.

Analyzing the information provided by the Spearman's correlation analysis (Table 4.35 refers to Hangar 2, period 23.02.2015-19.03.2015) we observe a very good correlation between indoor and outdoor in the case of CO, O_3 and NO_2 ($q_{CO} = 0.904$; $q_{NO2} = 0.823$; $q_{O3} = 0.805$ according to Table 4.36), which determines us to consider the outdoor air as the main source of indoor air pollution with these compounds. However, the possibility of additional sources of pollution due to restoration works or the presence of visitors and/or museum staff may not be neglected.

Concerning the wooden church indoor air pollution, except for ozone whose average concentration (10.14μg / m^3) exceeds the recommended limit (10μg/m^3) with approx. 1.5%, period averages for other compounds fall within the limits recommended by ASHRAE for general collections.

However, during the days when religious services are performed, an increase of the NO_2, SO_2, CO, CO_2 and $PM_{2.5}$ concentration is recorded over the average values of the other days of the week (Figure 4.20a); the growth may be due to candle burning and parish attendance in the church.

Table 4.34. Monitoring results and indicators of the central trend, dispersion and form of the data series distribution for Hangar 2 and ambient air (Bucur et al., 2017).

	NO_2, µg/m^3			SO_2, µg/m^3			O_3, µg/m^3			$PM_{2.5}$, µg/m^3			Temp, ^{0}C		RH, %	
	I	O	I/O	I	O	I/O	I	O	I/O	I	O	I/O	I	O	I	O
Number of measurements	794	794		794	794		794	794		794	794		794	794	794	794
Mean	12.36	18.39	0.92	4.03	5.17	0.84	10.78	31.05	0.35	9.88	20.51	0.49	12.9	10.96	57.83	77.74
Median	10.19	12.43		3.48	4.35		9.96	31.13		9.54	20.12		9.6	8.0	56.4	80.48
Standard Dev.	7.23	15.53		1.24	2.94		1.86	5.79		1.32	2.103		6.94	7.92	7.69	19.58
Skewness	1.54	1.71		2.65	5.96		1.29	-0.147		0.564	-0.079		1.12	1.16	0.282	-0.505
Standard Dev. of Skewness	0.087	0.087		0.087	0.087		0.087	0.087		0.087	0.087		0.087	0.087	0.087	0.087
Kurtosis	2.57	3.2		11.2	54.14		0.956	-0.925		-0.927	-0.379		-0.554	0.147	-0.507	-0.827
Standard Dev. of Kurtosis	0.173	0.173		0.173	0.173		0.173	0.173		0.173	0.173		0.173	0.173	0.173	0.173
Minimum	2.98	0.78	0.14	2.93	3.13	0.15	8.42	19.9	0.24	7.98	15.42	0.34	4.2	0.3	37.8	30.0
Maximum	43.7	86.1	1.6	12.9	36.6	1.66	19.49	43.16	0.54	12.65	24.54	0.78	27.6	33.4	73.7	100.0
ASHRAE*	4-20	-		1-5.7	-		1-10	-		1-10	-		< 25	-	25-75	-
Directiva 2008/50/EC**	-	200		-	350		-	120		-	-		-	-	-	-
US EPA***	-	-		-	-		-	-		-	35		-	-	-	-

Table 4.36. Spearman's correlation coefficient (q) for the indicators monitored in Hangar 2 and in ambient air between 23.02-19.03.2015

	I-NO_2	I-SO_2	I-CO	I-O_3	I- $PM_{2.5}$	I-temp	I-RH	O-NO_2	O-SO_2	O-CO	O-O_3	O- $PM_{2.5}$	O-temp	O-RH
I-NO_2	1.000													
I-SO_2	0.408**	1.000												
I-CO	**0.729****	0.366**	1.000											
I-O_3	-0.377**	0.096*	-0.568**	1.000										
I- $PM_{2.5}$	-0.060	0.114**	0.049	-0.063	1.000									
I-temp	-0.068	0.185**	-0.095*	0.005	-0.172**	1.000								
I-RH	-0.157**	0.053	-0.073	-0.155**	0.157**	.0325**	1.000							
O-NO_2	**0.823****	0.232**	**0.697****	-0.557**	-0.063	-0.048	-0.042	1.000						
O-SO_2	0.498**	**0.657****	0.384**	0.114**	0.101*	-0.054	-0.176**	0.300**	1.000					
O-CO	**0.661****	0.318**	**0.904****	-0.576**	0.056	-0.144**	-0.080	**0.790****	0.348**	1.000				
O-O_3	-0.430**	-0.066	**-0.639****	**0.805****	-0.052	-0.057	-0.178**	**-0.730****	0.057	**-0.748****	1.000			
O- $PM_{2.5}$	-0.183**	-0.287**	-0.179**	-0.065	-0.190**	-0.005	0.207**	-0.061	-0.290**	-0.150**	-0.074	1.000		
O-temp	0.184**	0.273**	-0.098*	0.433**	-0.137**	-0.090*	-0.425**	-0.070	0.398**	-0.129**	0.407**	-0.102*	1.000	
O-RH	-0.102*	-0.252**	0.243**	-0.429**	-0.103*	0.100*	0.262**	0.116**	-0.486**	0.265**	-0.469**	0.312**	-0.512**	1.000

* Semnificative corellation, level of probability: 0,05

** Semnificative corellation, level of probability: 0,01

This hypothesis is also supported by the graphical representation of the time evolution of the mean hourly concentrations of NO_2, SO_2, CO, CO_2 on Sunday 06.07.2014 inside and outside the church (Figure 4.21a).

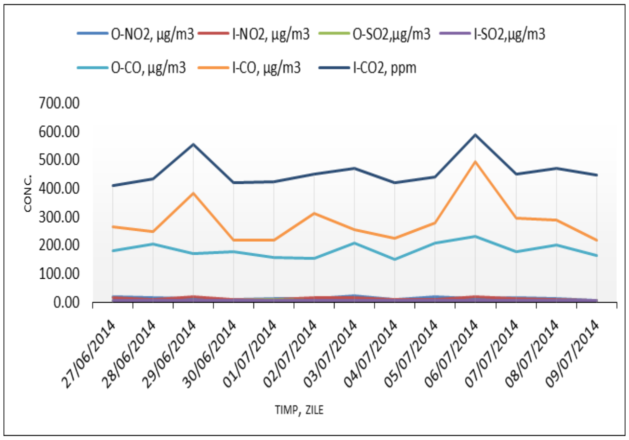


Fig. 4.20a. Evolution in time for NO_2, SO_2, CO and CO_2 concentrations in wooden church and ambient air between 27.06.2014 - 9.07.2014

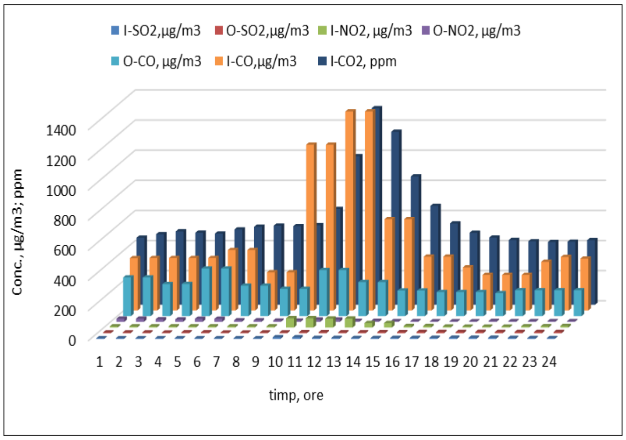


Fig. 4.21a. Evolution in time for NO_2, SO_2, CO and CO_2 concentrations in the wooden church and ambient air, Sunday 06.07.2014.

In conclusion, we can assume that the results of the tests indicate good air quality in the three tested areas, the concentrations of the monitored compounds are, on average, below the recommended values of ASHRAE for the prevention of degradation and conservation of heritage objects. Exceptions to ozone and $PM_{2.5}$ (Hangar 2 in the hot season), which may exceed the recommended limit value and, during religious services, concentrations of NO_2, SO_2, CO, CO_2 in the wooden church. The airing of the worship place after the completion of the religious service reduces the risk of degradation of the patrimony objects present here or of the wood from which the church is made. Higher concentrations of NO_2 and CO have also been observed in the cold season in Hangar 2, possibly due to restoration activities carried out inside or in the immediate vicinity of the access road.

The values of temperature and humidity microclimate parameters and their diurnal variations fall within a low risk range for most exhibits (ASHRAE control class C).

Regarding pollution sources, information from the values of I/O Ratio, and Spearman correlation analysis of monitoring data series, we consider outdoor air as the source with the most important contribution to indoor air quality. The restoration activities carried out in this area and the presence of visitors may also represent minor sources of pollution whose contribution may in the short term influence the indoor concentration of pollutants but without long-term effect.

An air quality assessment based only on reporting to the set / recommended limit values for each indicator does not provide the most complete picture of the effect on the exhibits. Through such an approach we can obtain information on the possible effect that each of the monitored parameters can induce on exhibits without taking into account the potential cumulative effect.

Further information we try to obtain by binary logistic regression of the data series resulting from the long-term monitoring of the chemical and microclimate parameters.

5. Assessing the environmental impact of a museum on exhibits through binary logistic regression

The logistic regression was developed by the statistician David Cox in 1958 and was initially used in medicine and epidemiology to analyze risk factors associated with some diseases and /or for early diagnosis (Callas et al., 1998, Wang et al., 2002, Reisner et al. ., 2015; Hailpern and Visintainer, 2003; Whittemore and Halpern, 2003). Over time, he has found himself useful in other fields of activity, such as sociology, tourism, economics and financial economics. No applications of logistic regression in the field of air quality have been identified to date, although we believe that it would be of interest at least to determine the cumulative effect of the environment on the exhibits in a museum or on the health of the population. This could be the first step towards using the binary logistic regression to assess the effect of the environment on exhibits in a museum.

For the application we used the data obtained in the air quality monitoring process in Hangar 2 both in the hot and cold season (Tabel 4.34). Out of the 794 data combinations obtained, 715 were used to calibrate the mathematical model, the rest (79 combinations) representing the values used for model validation (10% of the data combinations).

All six monitored parameters were considered as predictors of variables: the concentration of NO_2, SO_2, O_3, $PM_{2.5}$, temperature and humidity, and as a dependent (categorial) variable *the effect on the exhibits.*

The department criterion of the values of the dependent variable was the reporting to the limit values recommended by the ASHRAE Handbook - Chapter 21, 2007 for general collections ($20\mu g/m^3$ for NO_2, $5.7\mu g/m^3$ for SO_2, $10\mu g/m^3$ for O_3, $10\mu g/m^3$ for $PM_{2.5}$; 25^0C for temperature and 75% for humidity). Under these conditions, the dependent variable was assigned to the values:

- 1, *possible effects on exhibits* if at least one of the values of the six predictor variables exceeds the recommended limit;
- 0, *no effect on the exhibits*, if the values of all six predictor variables are below the recommended limits;

In these conditions the mathematical model will be of the form:

$$\ln(odds\_ratio) = \beta_0 + \beta_1 x_{NO2} + \beta_2 x_{SO2} + \beta_3 x_{O3} + \beta_4 x_{PM2.5} + \beta_5 x_{temp} + \beta_6 x_{RH} \qquad (5.5)$$

where: - $ß_{1}...ß_{6}$ are the logit coefficients corresponding to the six variables (NO_2, SO_2, O_3, $PM_{2.5}$, temperature and humidity);

- $ß_0$ represents the constant (similar to linear regression);

- $x_{NO2}.......x_{RH}$, are the concentrations of NO_2, SO_2, O_3, $PM_{2.5}$, respectively the values for the microclimate, temperature and humidity parameters (RH).

- *odds_ratio,* expresses the ratio between the probability of realization and the probability of not realizing the event;

The results obtained by applying the binary logistic regression, the Enter method, to the monitoring data series for determine the logite coefficients corresponding to the six parameters and their statistical significance too, are found in Table 5.7.

Thus we observe that of the six logit coefficients, only four have statistical significance: the corresponding concentrations of NO_2, O_3, $PM_{2.5}$ and humidity (RH), and the other two, the temperature and SO_2 concentration are not statistically significant (Sig. 0.05); Under these conditions, their contribution is not significant, they can be removed from the model without significantly affecting the accuracy of the prediction.

Table 5.7. Results of binary logistic regression - the Enter method (Block 1) applied to the six predictor variables (Bucur şi colab., 2017).

Model Summary

Step	-2 Log likelihood	Cox & Snell*	Nagelkerke*
1	231.852	0.564	0.824

*the values of *Pseudo-* R^2coefficients(see Annex 3, point. A3.6)

Classification Table

Observed			Predicted		
			Effects on exhibits		Percentage Correct
			0,00	1,00	
Step 1	Effects on exhibits	0.00	176	13	93.1
		1.00	19	507	96.4
	Overall Percentage				95.5

Variables in the Equation

		B	S.E.	Wald	df	Sig.	Exp(B)
Step 1	NO_2	0.360	0.052	48.090	1	0.000	1.434
	SO_2	0.472	0.767	0.379	1	0.538	1.604
	temp	-0.038	0.153	0.061	1	0.805	0.963
	RH	-0.081	0.025	10.302	1	0.001	0.922
	O_3	6.102	0.689	78.484	1	0.000	446.565
	PM	3.422	0.365	88.133	1	0.000	30.633
	Constant	-90.931	9.389	93.795	1	0.000	0.000

By eliminating the two variables, the model equation is simplified, becoming:

$$\ln(odds\_ratio) = \beta_0 + \beta_1 x_{NO2} + \beta_3 x_{O3} + \beta_4 x_{PM2,5} + \beta_6 x_{RH} \tag{5.6}$$

The values of the coefficients $ß_{0}…ß_{4}$ are obtained by applying the binary logistic regression, the Enter method, only to the data series corresponding to the four predictive variables with statistical significance. The results of the final prediction stage, with only four variables (*Block 1)*, can be found in Table 5.8.

Table 5.8. Results of binary logistic regression - the Enter (Block 1) method, without SO_2 and temperature (Bucur et al., 2017).

Model Summary

Step	-2 Log likelihood	Cox & Snell	Nagelkerke
1	232.269	0.564	0.823

Classification Table

Observed			Predicted		
			Effects on exhibits		Percentage Correct
			0,00	1.00	
Step 1	Effects on exhibits	0.00	174	15	92.1
		1.00	20	506	96.2
	Overall Percentage				95.1

Variables în the Equation

		B	S.E.	Wald	df	Sig.	Exp(B)
Step 1	NO_2	0.372	0.046	64.674	1	0.000	1.450
	RH	-0.083	0.025	10.929	1	0.001	0.921
	O_3	6.169	0.674	83.872	1	0.000	477.720
	PM	3.468	0.350	98.137	1	0.000	32.061
	Constant	-90.813	9.367	93.989	1	0.000	0.000

By eliminating from the model the SO_2 and temperature threre are no changes identify regarding the accuracy of the prediction (*Overall Percentage* has a lower decrease from 95.5% of the 95.1%) or variability (the value of the Nagelkerke indicator is decreasing insignificant from 0.824 to 0.823).

By introducing the values of *ß* regression coefficients (0,372 for *ß*$_1$; 6.169 for *ß*$_2$; 3.468 for *ß*$_3$; -0.083 for *ß*$_4$; -90.813 for *ß*$_0$) in equation (5.6) we obtain the final form of the mathematical model of prediction:

$$ln\ (odds\_ratio) = 0.372x_{NO2}+6.169x_{O3}+3.468x\mathrm{PM}_{2.5} - 0.083x_{RH} - 90.813 \tag{5.7}$$

that allows the calculation *odds_ratio value* with reltion (5.8) and the probability value of effect on the exhibits, *P*, using the relation (5.9):

$$odds\_ratio = p/(1-p) = Exp\ (0{,}372x_{NO2}+6.169x_{O3}+3.468x\mathrm{PM}_{2.5} - 0.083x_{RH} - 90.813) \tag{5.8}$$

$$P = \frac{e^{0,372x_{NO2}+6,169x_{O3}+3,468x_{PM2,5}-0,083x_{RH}-90,813}}{1+e^{0,372x_{NO2}+6,169x_{O3}+3,468x_{PM2,5}-0,083x_{RH}-90,813}} \quad (5.9)$$

The probability value can also be calculated using the SPSS 20.0 program; this feature was applied to validate the mathematical model by running the 79 unused data combinations in the model calibration process. Following the validation process, 78 results were correct (98.7%), one being erroneous; the result obtained corresponds to the level of accuracy (*Overall Percentage*) calculated by the program (95.1%).

We can appreciate in these conditions that the predictive mathematical model developed by the binary logistic analysis of the monitoring data can be used with confidence for the purpose it was developed, estimating with a very good accuracy the probability that the exhibits in Hangar 2 are affected due to the air quality from the building.

The usefulness of the results obtained through the binary logistic analysis applied to the monitoring data series is not limited to prediction of the probability of effect on the exhibits.

Starting from regression coefficient values or from the values *exp(B)* we can establish a hierarchy of indoor air pollution parameters depending on the potential effect on the exhibits; we can easily identify the measures to be taken to mitigate possible damage to the exhibits for preventive preservation. In our case the concentration of O_3 ($ß_{03}$ = 6.169) has the most powerfull effect, followed in order by: the concentration of $PM_{2.5}$ ($ß_{PM2.5}$= 3.468), the concentration of NO_2 ($ß_{NO2}$ = *0.372*) and the level of humidity ($ß_{RH}$ = *-0.083*).

The probability values calculated using relation (5.9) can also be interpreted as *a possible environmental impact on the exhibits taking into account a cumulative effect of all monitored parameters*; for this purpose, for an easier interpretation we can introduce an impact assessment system based on the calculated probability value *(p)*: i) very low impact for 0 <p <0.2; ii) low impact on 0.2 <p <0,4; iii) moderate impact on 0.4 <p <0,6; iv) strong impact for 0.6 <p <0.8 and v) very strong impact for p > 0.8.

To illustrate the usefulness of knowing the cumulative impact of chemical pollutants and statistically significant microclimate parameters on the exhibits we randomly chose six combinations, representing as many pollution situation, and for them was calculated the probability that, in this pollution context, the exhibits were affected and the possible impact was estimated. I mention that in this case we are talking about a cumulative impact of the four indicators on the exhibits. The results are presented in Table 5.9.

Table 5.9. Predictions regarding the cumulated effect on the exhibits in Hangar 2.

Analyzed situations	**The indoor air pollution context, Handar 2**				**Calculated Probability, %**	**Impact**
	O_3, µg/m^3	**$PM_{2.5}$, µg/m^3**	**NO_2, µg/m^3**	**RH, %**		
1	9.38	8.42	13.36	71.40	0.912	very low
2	**10.49**	9.21	9.66	60.5	98.82	very strong
3	**10.33**	9.21	4.28	53.20	41.33	moderate
4	**10.16**	8.79	4.05	52.2	38.56	low
5	9.98	8.52	8.50	53.50	27.56	low
6	9.45	9.56	11.11	58.9	47.35	moderate
Recommended ASHRAE limit values	10.0	10.0	20.0	25.0-75.0	-	-

If we look at situation 2, when only the O_3 concentration exceeds the recommended value by only 5% and the other three values are below the recommended limits, we find a very strong cumulative impact of the pollution context, the probability being 98.82%;

In cases 3 and 4, however, where the O_3 concentration remains above the recommended value (by 3.3% and 1.6%), the potential impact on the exhibits becomes moderate (case 3) or low (case 4). Hence the particular influence of other pollutants present in the air and the importance of determining the cumulative impact in assessing the effect of air quality on the exhibits. In cases 2 and 3, the passing of the cumulative effect from one impact class to another was largely due to the decrease in NO_2 concentration and humidity.

Similarly, in cases 1, 5 and 6 where all the pollution indicators are below the recommended values, the cumulative effect may cause different levels of impact on the exhibits: very low for 1, low for 5 and moderate for 6.

Even if the values of all the parameters involved are below the recommended values, the exhibits may be affected to a greater or lesser extent, depending on each input and positioning in their hierarchy as a hazard. Thus, small variations in the concentration of pollutants can cause large variations in the impact of the environment on the exhibits.

The results of the binary logistic regression can not be used only to assess the impact of the environment inside the museum on the exhibits and the hierarchy of parameters depending on their hazard probability.The mathematical model developed can be very useful for decision-making, and various simulations can be made to establish the most effective mitigation measures.

For example, we started from the pollution context corresponding to case 2 of Table 5.9, with a very strong impact on the exhibits (with a probability of 98.82%), where the O_3 concentration exceeds the recommended value by 5%, the other three parameters, NO_2, $PM_{2.5}$ and RH is below the limit value. At first sight the concentrations of O_3 and $PM_{2.5}$ seem to determine this impact, and the simplest method of reducing pollution could be to equip the air-conditioning system with a filtration system to reduce the concentration of particulate matter; assuming that under these circumstances the particulate matter concentration in the air would reduce by 10%, the impact on the exhibits and implicitly the probability of affecting the exhibits will decrease to 77.46% (case 2, Table 5.10) and a reduction of 20 % of the particulate matter concentration would lead to a very low impact (case 3, Table 5.10) with a probability of only 12.39%.

Table 5.10. Predictions on the effect of measures to reduce air pollutant concentrations on Hangar 2 exhibits.

Analyzed situations	**Context of the indoor air pollution in Handar 2**				**Calculated Probability, %**	**Impact**
	O_3, μg/m^3	$PM_{2.5}$, **μg/m^3**	**NO_2, μg/m^3**	**RH, %**		
1	10.49	9.21	9.66	60.5	98.82	very strong
2	10.49	8.29	9.66	60.5	77.46	strong
3	10.49	7.37	9.66	60.5	12.39	very low
4	9.97	8.29	9.66	60.5	2.69	very low
Recommended ASHRAE values	10.0	10.0	20.0	25.0-75.0	-	-

Case 4, where the impact is only 2.69%, can be ensured by lowering the particulate matter concentration by 10% (case 2) while reducing the O_3 concentration by 5% (by using reducing substances in the filtration system or by improving sealing of the building, O_3 being almost twice as high).

It should also be noted that we are not talking about a mathematical model generaly valid; even in the case of spaces located on the same site (the characteristics of the outdoor air quality being the same) considering the multitude of parameters that can influence the indoor air quality, there may be differences regarding the pollutants with statistical significance, the values of the β_i coefficients or the pollutant hierarchy from the danger point of view.

For exemplification, in relations (5.9-5.11) are presented the equations of the mathematical models obtained by binary logistic regression for the wooden church (eq. 5.9), Hangar 1 (eq. 5.10) and Hangar 2 (eq. 5.11).

We find differences in both the type of pollutants with statistical significance and in their hierarchy. Referring to Hangar 2 (eq. 5.11), in the case of the wooden church (eq. 5.9) the most important change occurs in the case of microclimate parameters. Humidity does not have statistical significance, it is necessary to introduce the temperature in the equation, which seems even more important in these conditions than the NO_2 concentration. Changes in Hangar 1 (eq. 5.10) target the pollutant hierarchy of pollutants but also the introduction of the SO_2 concentration into the equation.

$$ln\ (odds\_ratio) = 7{,}634x_{O3} + 0{,}901x\mathrm{PM}_{2.5} - 0{,}852x_{Temp} + 0{,}144x_{NO2} - 103{,}647 \qquad (5.9)$$

$$ln\ (odds\_ratio) = 1{,}961x\mathrm{PM}_{2.5} + 1{,}698x_{SO2} + 1{,}304x_{O3} + 0{,}368x_{NO2} + 0{,}160x_{RH} - 49{,}618 \qquad (5.10)$$

$$ln\ (odds\_ratio) = 6{,}169x_{O3} + 3{,}468x\mathrm{PM}_{2.5} + 0{,}372x_{NO2} - 0{,}083x_{RH} - 90{,}813 \qquad (5.11)$$

We appreciate that the use of the mathematical model obtained through the binary logistic regression analysis allows for a more accurate and complete picture of how a particular context of pollution can affect the exhibits in a museum than simply referring to the limit values for each indicator, provided it is built on a large number of monitoring data covering as much as possible the vast majority of potential situations.

CONCLUSIONS

Through the doctoral dissertation we wanted to draw the attention of the scientific environment once again to a domain that is less approached in Romania - indoor air quality - due to the possible impact on human health or the integrity of museum exhibits, with the purpose of providing optimal comfort conditions for staff or for preventive preservation of heritage objects.

The data presented in the original part of the thesis were obtained in two large case studies organized and carried out in:

- office spaces in the ECOIND building (E3-13 representative office)

- three exhibition spaces belonging to the National Museum of Romanian Aviation, in which the following aspects were followed:

1. Long-term monitoring in the hot and cold season of the following parameters:

- NO_2, SO_2, CO, $O_3$and particulate matter in both locations and, additionally,

- H_2S, CO_2 and formaldehyde in the office space

- parameters of microclimate temperature and humidity in the museum;

2. Statistical characterization of the monitoring data series: verification of distribution normality, internal/external correlation, linear regression of particulate matter concentration and frequency of exceedance of limit values;

3. Identification of compounds with the *strongest impact on staff health / integrity of exhibits*: formaldehyde for office E3-13; ozone in the event of exposure to the museum.

4. Identification of the main sources of indoor air pollution:

- in office spaces (E3-13): outdoor air, printing / copying and staff;

- in the museum: outside air and occasional restoration work;

- in the wooden church: outdoor air and, occasionally, emissions of NO_2, SO_2, CO, CO_2 and particulate matter during religious services;

5. Binary logistic regression analysis of the monitoring data in Hangar 2 in order to:

- development of a mathematical model validated by prediction of the indoor air quality effect on the exhibits;

- establishing the relationship of calculating the probability of effect on the exhibits;

- quantifying the cumulative impact of airborne compounds and microclimate parameters on the exhibits by assimilating it with the probability of effect on the exhibits;

- the hierarchy of pollutants present in the air in Hangar 2 depending on the potential effect on the exhibits;

Being the *first application of binary logistic regression in the field of air quality assessment* in museums and quantification of possible impacts on exhibits, we consider this research as a first step for a range of future projects applicable in related areas concerning air quality within residential, office and ambient air and assessing the effect of the environment on the health of the population.

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LIST OF SCIENTIFIC PAPERS

A. Original articles published in the thesis field :

1. **Bucur E**., A.F. Danet, C.B. Lehr, E. Lehr, M. Nita-Lazar. 2017. Binary logistic regression – instrument for assessing museum indoor air impact on exhibits. *Journal of the Air&Waste Management Association*. 67(4): 391-401. FI(2016):1,57.
2. **Bucur E**., A. Vasile, M. Petrescu, A. Danet. 2016. Indoor air quality assessment in spaces designed for office activities: PAH's and phenols. *Journal of Environmental Protection and Ecology*.17(1): 9-17; FI(2016): 0,774.
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B. Participation at scientific events:

1. Petrescu M., **E. Bucur**, A. Vasile, R. Diodiu, G. Vasile, V. Danciulescu, M. Bratu, G. Tanase. 2016. Characterizatioin of the chemical composition of atmospheric deposition. *International Symposium SIMI 2016" The Environment and Industry"* proceedings book: 376-382 - prezentare orala.
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4. **Bucur E.**, A. Vasile, M. Petrescu. How protected from pollution are we indoor? - method for vinyl chloride from indoor air determining. *International Symposion „The Environment & The Industry", SIMI 2011*, 16-18.11. 2011, Bucharest, vol II, pg. 221-226; prezentare orala.

C. Other scientific articles:

1. Diodiu R.**,** T. Galaon, **E. Bucur**, D. S. Stefan, L. F. Pascu. 2016. Aldehydes, acetone, formic and acetic acids in indoor air of an office building. *Revista de Chimie*. 67 (12): 2396-2399.
2. Diodiu R., T. Galaon, **E. Bucur**, L.F. Pascu. 2016. Aldehydes and Acetone in Indoor Air of 19 Houses from Bucarest (Romania). *Revista de Chimie*. 67(8):1466-1468;
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