Odour pollution assessment through indirect methods based on the monitoring of technological parameters - case study

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The odours in the ambient air, through the discomfort that they induce, represent an actual problem for the communities located nearby companies with technological processes that emit in the air different strong and unpleasant odorous substances.

The standard method for odour assessment involves measuring the concentration through dynamic olfactometry according with SR EN 13725:2003, a very complex method that requires specialized working staff and expensive equipment.

The paper presents an indirect method for odour assessment in the ambient air, based on monitoring the process and meteorological data - Predictive Emission Monitoring Systems (PEMS) and it is applied to a livestock farm. Using the multiple regression analysis of the monitoring data for the most important specific technological and meteorological parameters it can be developed a mathematical model that could be used for the calculation of odour concentration in air, without the necessity of direct measurement, after the initial step. For the case study presented in the paper, the distance between the slurry lagoon was identified as a significant statistical parameter that can determine in a proportion of 72% the concentration of odour in the ambient air nearby the farm; the margin of error for odour concentration assessment, according to the model validation tests, is ± 8%, acceptable value for an estimation method by mathematical modelling.

Keywords: *odour, dynamic olfactometry, multiple regression method, intensive livestock farming*

Odour can be defined as sensations resulting from the interaction of volatile chemical species inhaled through nose, making contact with the olfactory area and registering in the brain [1]; in this context, the standardized method for odour determination, the method based on dynamic olfactometry [2-4] uses the human nose, respectively a group of odour assessor connected to an olfactometer – a performant system that dilutes, present the samples to the human assessors, and does all the calculations using a data processing software. An automatic alternative based on electrochemical cells is represented by the *electronical nose*, used mainly in comparative studies [5]. Even though the direct methods are preferred, in the case when these analyses are very complex, involves very high costs or impracticable conditions, indirect methods can be used, methods based on statistical analysis of the parameters values that are correlated with the level of odour emission. In this regard, the use of statistical correlation and regression methods have been applied in various activity, including environmental protection. [6-10].

The paper presents an indirect method for odour assessment based on the monitoring of process, meteorological and geographical parameters - *Predictive Emission Monitoring Systems* [11] – applied to a livestock farm. Using the multiple regression analysis applied to specific monitoring parameters data it is intended to obtain a mathematical model that can be used for the odour concentration in air estimation, without direct measurement, except the initial phase.

EXPERIMENTAL PART

Location. The case study was conducted at a livestock farm operating at maximum capacity, which, according to Order no. 994/2018 [12] requires a 1500 m sanitary protection area. The farm is in a hilly area, in the immediate vicinity of a locality, characterized by winds in the direction of NV, as shown by the graphical expression of the wind rose (Figure 1).

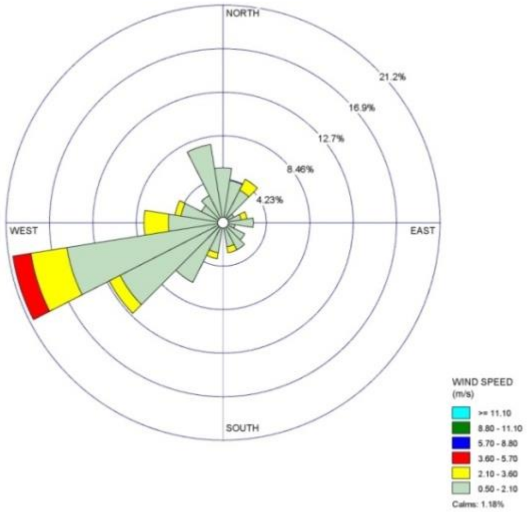

Fig. 1. Wind rose of the area

The very small distance, sometimes under 100 m from houses, determined over the years some problems due to the air pollution with NH3, H2S, and odours largely due to the activities carried out on the farm but also to the non-compliance with the sanitary protection area. Technological processes carried out on the farm meet the requirements of the BAT Conclusions [13], both in terms of the application of the best technologies and also regarding the methods for reducing pollutant emissions in the air. Thus, animal feeding and shelter ventilation systems are automated, the ventilation rate being set according to the temperature and humidity parameters of the shelter to ensure optimal animal development conditions. The slurry is automatically transferred to pre-sealed lagoons that prevent soil and ground infiltration and are covered with sealed geomembranes where they are enzymatically treated to reduce pollutant/odour emissions in the air.

Materials and Methods. The evaluation method is based on the statistical correlation and multiple regression analysis applied to the data series obtained by monitoring the odour concentration, the main specific odorous pollutants (NH_3 and H_2S) and the technological / geographic parameters and involves the following steps: i) identification the main sources of odour emission and their specific parameters, ii) monitoring of the identified parameters and of the odour concentration, iii) correlation and multiple regression analysis applied to the data series obtained by monitoring, iv) obtaining the equation of the mathematical model and its validation by direct measurements and by the mathematical modelling of the odour dispersion in the air.

Based on the analysis of technological processes from a livestock farm and the main BAT techniques [13] to reduce the odour level but also on the specific conditions of the farm in which the case study was conducted, three potential sources of odour pollution and strong smelling compounds have been identified: ventilation openings for animal shelters (NH_3, odour), the slurry transfer system (NH_3, H_2S, odour) and the lagoons for storage / treatment of slurry (NH_3, H_2S, smell) to which are added the sources of diffuse emissions. In these conditions *the parameters that can characterize the emissions of odour and strong smelling specific compounds, NH_3 and H_2S,* which were taken into account for the mathematical model development are: i) ventilation rate, expressed in % from the maximum capacity (vent-cap), ii) distance to shelters, m (dist), iii) distance to lagoons, m (dist), iv) wind direction, expressed in degrees N (dir-vant), v) wind speed, m/s (vit-vant), vi) atmospheric temperature, ^{0}C (temp), (vii) atmospheric humidity, % (RH) and viii) the concentration of NH_3 and H_2S in air (in the case of odour calculation) in µg/m^3 or mg/m^3. For concentration of NH_3 and H_2S determination, Horiba automatic analysers were used, with detection limits of 5 ppbv for NH_3 and 2 ppbv for H_2S. The odour analyzis was done following the dynamic olfactometry method according to

SR EN 13725:2003 using a Odournet T08 dynamic olfactometer, developed by Odournet GMBH, Germany. Meteorological parameters were monitored using a MetPack Wind Sonic Portable Weather Station. For the statistical interpretation of the data - Pearson correlation and multiple regression analysis - the SPSS 20.0 (Statistical Package for Social Sciences, version 20.0) software was used. The *correlation* analysis provides information regarding the intensity and direction of the statistical link between two or more variables without necessarily involving a causal link between them. In the case study Pearson correlation was used, whose coefficient (r) can take values in the range [-1, + 1]; the closer is the value of the r coefficient to ± 1, the better is the correlation between the two analyzed parameters. To assess the *statistical significance* of the correlation coefficient and the regression coefficients, the *p* value - the probability that the value of the correlation coefficient is equal to zero (the null hypothesis) - is used; if the probability is less than the *significance level* (ex. $p<0.05$ for a confidence level of 95%), then the correlation coefficient is statistically significant. Statistical correlation is always the first step in predictive studies preceding regression analysis.

The *regression* represents the process by which it can estimate the value of the dependent variable based on the values of the predictor variables and the mathematical relationship expressing their interdependence. Such an estimate is, in fact, a *prediction*. In this case study, the *multiple regression* was used, which basically relies on the same fundamental concepts as simple linear regression, but must deal with the specific problems of a multivariate model. The multiple regression equation (Ec. 1) is similar to the simple regression equation, applied to several predictors:

$$Y = a + b_1 x_1 \dots\dots + b_k\, x_k \qquad (1)$$

in which:

$x_1 \dots x_k$ = values of the *k* predictor variables

a = ordinate to origin

$b_1 \dots b_k$ = regression coefficients corresponding to the *k* predictor variables

Y = estimate values for the criterion variable (dependent)

RESULTS AND DISCUSSION

Starting from the actual conditions on the site, 9 points were identified outside the farm enclosure at different distances from animal shelters and lagoons for slurry manure handling and storage, around the site; at each point measurements of the concentration were made for NH_3, H_2S, odour and weather parameters, and the results of the measurements are found in Table 1. Analyzing the results of the measurements and compared with the limit values for the concentration of H_2S and NH_3 in the air [14] it can see that in all 9 points the concentration exceed the limit values for short-term measurements (0.015 mg/m^3 for H_2S and 0.300 mg/m^3 for NH_3); it should be noted that these points are located inside the sanitary protection area (at distances of 80 - 440 m from the farm), demonstrating once again the necessity of regard the sanitary protection zones imposed by Order no. 994/2018 [12].

In this case the 1500 m sanitary protection area is not respected, with housing reaching in some cases 100 m from animal shelters. With regard to odour concentrations, for which limit values are not set in the ambient air, it finds values that fall within the normal range for these types of activities, taking into account the small distance from the emission sources.

Table 1. Values of parameters monitored in the livestock farm

Point	Location	H_2S, µg/m^3	NH_3, mg/m^3	Odour, ou$_e$/m^3	Temp, ^{0}C	RH, %	vent-cap, %	Dist, m	dist-lag, m	dir-vant, grade	vit-vant, m/s
0	1	2	3	4	5	6	7	8	9	10	11
1	Farm gate	35.40	1.30	16.00	17.00	56.00	50.00	310.00	600.00	104.00	0.60
2	Close to lagoon and slurry distribution area	69.50	3.90	31.00	21.00	44.00	50.00	290.00	.00	150.00	0.70
3	25m from the pt 2. to farm	38.60	5.60	47.00	22.00	41.00	64.00	240.00	90.00	163.00	1.70
4	25m from pt 1. to N	39.50	1.51	19.00	23.00	41.00	64.00	370.00	660.00	220.00	1.90
5	On the road (house)	33.20	10.20	4.40	23.00	41.00	70.00	160.00	600.00	211.00	2.19

0	1	2	3	4	5	6	7	8	9	10	11
6	On the road (pension)	35.60	3.98	8.00	23.00	41.00	95.00	440.00	810.00	213.00	1.35
7	In the farm, close to the monitoring system	36.30	13.06	18.00	23.00	42.00	100.00	80.00	520.00	171.00	0.70
CMA (30 min)		15	0.300								
Average		41.15	5.65	20.48	21.71	43.71	70.42	270.00	468.57	176.00	1.305
Standard deviation		4.789	1.672	5.470	.837	2.089	7.55	46.291	114.775	15.87	.245
Median		36.30	3.98	18.00	23.00	41.00	64.00	290.00	600.00	171.00	1.35
Minimum value		33.20	1.30	4.40	17.00	41.00	50.00	80.00	.00	104.00	0.60
Maximum value		69.50	13.06	47.00	23.00	56.00	100.00	440.00	810.00	220.00	2.19
8	30m from pt. 2	46.5	3.21	29	18	54	35	220	200	110	0.9
9	On the road, close to the office	42.4	10.1	24	20	46	80	120	410	165	1.2

Statistical interpretation of data

First step in the statistical interpretation of data was the Pearson correlation analysis, whose results are found in Table 2 (the Pearson correlation coefficients, *r*) reveals for the interest indicators, odour, NH_3 and H_2S a series of very good and good correlations with some of the monitored parameters, as follows:

- in the case of the *odour concentration* a very good inverse correlation with the *distance from the* lagoon (r_{odour} = - 0.847) and moderate, direct correlation with the H_2S concentration (r_{odour} = 0.425) is observed;
- in the case of *H_2S concentration* a very good inverse correlation with the *distance to the lagoon* (r_{H2S} = - 0.718), and also moderate correlations with the *ventilation capacity of the shelters* (r_{H2S} = - 0.468) and *odour concentration* (r_{odour} = 0.425);
- for *the concentration of NH_3* we can observe a very good inverse correlation with *the distance from the animal shelters* (r = - 0.865), a good correlation with the *ventilation capacity of the shelters* (r_{H2S} = 0.618) and a moderate one with the *atmospheric temperature and humidity*.

Table 2. The values of the Pearson correlation coefficients, *r*

	odour	dist	dist-lag	dir-vant	vit-vant	temp	RH	vent-cap	H2S	NH3
odour	1.000									
dist	-.075	1.000								
dist-lag	-.847	.270	1.000							
dir-vant	-.376	.155	.432	1.000						
vit-vant	-.123	.068	.216	.754	1.000					
temp	-.107	-.154	.131	.891	.586	1.000				
RH	-.080	.116	.061	-.835	-.605	-.974	1.000			
vent-cap	-.379	-.209	.493	.506	.042	.640	-.510	1.000		
H2S	.425	.118	-.718	-.257	-.394	-.116	-.015	-.468	1.000	
NH3	-.169	-.865	-.014	.213	.065	.504	-.430	.618	-.239	1.000

Colour legend:

Colour			░	▒	▓
Correlation significance	very week	week	moderate	good	very good

Under these conditions, the results of the Pearson correlation analysis provide the first indices for predictor variables with significance for the value of the dependent variable (odour concentration). Once a correlation between the variables is established, *the statistical regression method* is used to establish the mathematical relationship. In our case, multiple regression, the Forward method, was applied to the monitoring data series obtained in the first 7 measurement points (Table 1), including the concentrations of NH_3 and H_2S; points 8 and 9 were used to validate the mathematical model.

The results of multiple regression (Table 3) for *odour concentration* as a dependent variable eliminate as statistically insignificant all variables from the equation, except *the distance from the lagoon*, the only statistically significant variable (Sig. = 0.016).

Table 3. The results of multiple regression analysis for odour concentration.

Model Summary[b]

Model	R	R Square	Adjusted R Square	Std. Error of the Estimate	Change Statistics				
					R Square Change	F Change	df1	df2	Sig. F Change
1	.847[a]	.718	.662	8.41942	.718	12.728	1	5	.016

a. Predictors: (Constant), dist-lag
b. Dependent Variable: odour

Coefficients[a]

Model		Unstandardized Coefficients		Standardized Coefficients	T	Sig.	Collinearity Statistics	
		B	Std. Error	Beta			Tolerance	VIF
1	(Constant)	39.408	6.185		6.371	.001		
	dist-lag	-.040	.011	-.847	-3.568	.016	1.000	1.000

a. Dependent Variable: odour

Excluded Variables[a]

Model		Beta In	t	Sig.	Partial Correlation	Collinearity Statistics		
						Tolerance	VIF	Minimum Tolerance
1	dist	.166[b]	.631	.562	.301	.927	1.078	.927
	dir-vant	-.013[b]	-.044	.967	-.022	.814	1.229	.814
	vit-vant	.064[b]	.236	.825	.117	.953	1.049	.953
	temp	.003[b]	.013	.990	.007	.983	1.017	.983
	RH	-.028[b]	-.105	.922	-.052	.996	1.004	.996
	vent-cap	.051[b]	.168	.875	.084	.757	1.321	.757
	H2S	-.378[b]	-1.141	.317	-.496	.484	2.064	.484
	NH3	-.181[b]	-.725	.509	-.341	1.000	1.000	1.000

a. Dependent Variable: odour
b. Predictors in the Model: (Constant), dist-lag

The value of R^2 = 0.718 indicates that this variable determines 71.8% of the odour concentration in the analyzed area. We mention that these results apply only to the conditions specific to the period in which the measurements and data sets were used.

Applying the same procedure in *the case of* H_2S, similar results have been obtained, demonstrating that odour and H_2S emissions are from the same source, i.e. the lagoon in which the slurry effluent is discharged, the only parameter with statistical significance. In this case, however, this parameter characterizes the H_2S emission only in a proportion of 51.6%. In the *case of* NH_3 concentration, the results of the multiple regression analysis reveal two statistically significant parameters, the distance from the animal shelters and the ventilation rate, which together account for 94.8% of the emission.

The equations of the mathematical models for the three indicators and statistically significant parameters can be found in Table 4.

Table 4. Equations of mathematical models for estimating odour concentration, NH_3 and H_2S

Indicator	Statistically significant indicators	Equation of the mathematical model
Odour	- distance from the lagoon, m (dist-lag)	$Y = 39.408 - 0.04x$ x = distance from the lagoon, m Y = estimated value for the odour concentration, ou_e/m^3
H_2S	- distance from the lagoon, m (dist-lag)	$Y = 55.198 - 0.03x$ x = distance from the lagoon, m Y = estimated value for the concentration of H_2S, $\mu g/m^3$
NH_3	- ventilation rate, % from maximum capacity (vent-cap) - distance from the shelters, m (dist)	$Y = 6.037 - 0.028 * dist + 0.101 * vent.cap$ x_1 = distance from the shelter, m x_2 = ventilation rate, % Y = estimated value for the concentration NH_3, mg/m^3

In order to validate the equations of the mathematical models, the data obtained in points 8 and 9 were used; the error margin for odour concentration assessment according to model validation tests is -8%, for NH_3 -6%, and for H_2S -15%, acceptable for an estimation method by mathematical modelling. Based on the mathematical equations developed for H_2S, NH_3 and odour, the concentrations of these pollutants can be calculated at any point on the site starting from the statistically significant parameters, respectively the distance from the lagoon for H_2S and odour and the distance from the shelter and ventilation rate for NH_3. Thus, the minimum distances from the animal shelters for NH_3 and from lagoon in the case of H_2S and odour were calculated that should be respected in order not to exceed the limit values in the legislation; for H_2S the estimated minimum distance from the lagoon is 1340 m and for NH_3, for a maximum ventilation rate of 100%, the distance from shelters must be at least 580 m; in the case of odour, at an estimated distance of 960 m from the lagoon, the odour concentration would be below the theoretical threshold of odour perception (the odour concentration perceived by 50% of the members of a panel constituted according to SR EN 13725:2003) of 1 ou/m^3. Considering the value of the Maximum Allowable Concentration for H_2S (15 $\mu g/m^3$) it can be concluded that at a distance of at least 1340 m from the lagoon the concentration of H_2S is below the limit value, i.e. if the sanitary protection zone is respected the concentration of H_2S in sensitive areas would fall within the limit values imposed by environmental legislation in force [14]. The same applies to the odour and NH_3 concentration.

CONCLUSION

Starting from the results obtained in the case study, we find once again a high level of chemical pollution (NH_3 and H_2S) and odour generated by the livestock farm activities, but also the need to comply with the requirements of the regulations in force regarding the sanitary protection areas related to these farms, to ensure comfort and reduce the effect on the health of the population.

The indirect method of assessing the concentration of odour, NH_3 and H_2S based on the analysis of the data series from the monitoring of the technological/geographic parameters through the multiple regression method is applicable to this type of activity; the method allows the estimation of odour, NH_3 and H_2S concentration at any point on the site using mathematical relationships resulting from multiple regression analysis with an assessment error margin of -8% for odour, -6% for NH_3 and -15% for H_2S, acceptable values for an estimation method by mathematical modelling. Using these relationships, the minimum values of the influence parameters on H_2S, NH_3 and odour emission (lagoon distance, shelter distance at an 100% ventilation rate) can be calculated for which the concentration of these indicators in air reaches the maximum concentration value admitted.

For the case study, the following results were obtained:

- for H_2S the minimum distance to the lagoon: 1340 m;
- in the case of NH_3, for the maximum ventilation rate (100%) distance to: minimum 580m;
- in case of odour at a distance of 960 m from the lagoon, the odour concentration

is below 1 ou/m^3 - the theoretical threshold of odour perception;

We can appreciate that, for the case studied, outside the 1500 m of the sanitary protection area, the estimated odour concentrations, NH_3 and H_2S are below the limit values; inside the sanitary protection areas these concentrations exceed the limit values for sensitive areas [14] in the case of the settlements that do not respect the minimum distance from the farm imposed by the legislation.

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