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# Correlation between odour and hydrogen sulphide concentrations from municipal wastewater treatment plant immissions

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#### Abstract

The study focused on odours generated by hydrogen sulphide ( $H_2S$ ) immissions from a municipal wastewater treatment plant in a city of 2 million people. Odour awareness was vital to monitor environmental pollution and human's safety.  $H_2S$  was released from decomposition of organic matter, sulphites ( $SO_3^{2-}$ ) and sulphates ( $SO_4^{2-}$ ) by anaerobic microorganisms used in the biological stage of wastewater treatment.  $H_2S$  was responsible for odours coming from the treatment plant. Long exposure (8 hours) to 2-5 ppm causes headaches and watery eyes, and 50 ppm irritates the airways. Maximum permissible concentrations for  $H_2S$ are 0.006 ppm (24 hours) and 0.011 ppm (30 minutes). In Romania there are no limits on the concentration of odours. The sludge pumping station from the wastewater treatment plant registered the highest odour concentration at 36.000  $OU_E/m^3$ , with significant levels of  $H_2S$  primarily found there. Nevertheless, the activated sludge aeration system effectively eliminated around 99% of the  $H_2S$  through degradation and absorption processes. The correlation between  $H_2S$  and odour has been proved by a direct proportional relationship with a ratio of 25.000.

Experimental determinations and mathematical modelling, using the Aermod View program, contributed to a deeper understanding of the impact of odours on the environment and on sensitive receptors, showed maximum odour concentrations of 38.8  $ou_E/m^3$  at the site of the municipal wastewater treatment plant and 5  $\div 7 ou_E/m^3$  at receptors, but without affecting residential areas.

Keywords: hydrogen sulphide, odour concentration, wastewater treatment plant

# INTRODUCTION

Domestic and industrial wastewater treatment processes have an effect on air quality. The pollutants released from the different operating processes of the wastewater treatment plant are:  $NH_3$ ,  $H_2S$ , volatile organic compounds, suspended particles ( $PM_{2.5}$ ,  $PM_{10}$  and total suspended particulates), greenhouse gas emissions ( $CO_2$ ,  $CH_4$ ), bioaerosols but also odour immissions [1]. Wastewater treatment plants (WWTP) are specialized in removing pollutants such as substances, microorganisms and other impurities from wastewater so that the resulting effluent can be discharged into the environment [2].

In 2021, a total volume of wastewater of 1,801 million  $m^3$  was generated at the national level of which: 844 million  $m^3$  came from economical activities and 957 million  $m^3$  from domestical activities. There are many types of industries that discharge wastewater into the sewerage network operated by this treatment plant have the following fields of activity: animal husbandry, food

industry, cigarette manufacturing, pig breeding, installations for the disposal or recovery of animal carcasses and animal waste, etc (Fig. 1). At the level of regions in Romania, the largest share of the population connected to the sewage systems was recorded in the municipality targeted in this study (90 %), followed by the Center (70.0%) and West (64.2%) regions of the country (Fig. 2) [3].



Fig. 1. Percentage of specific wastewaters generated in Romania in 2021 [3]

Fig. 2. Regions in Romania with the largest share of population connected to the sewerage system [4]

The treatment system of this studied wastewater station had two distinct treatment stages: the first stage, called *Primary treatment*, aimed to remove solids from the raw wastewater and included the use of screens, the sand retention basin (grit chamber), the grease separator and the primary clarifier. The second stage, known as *Secondary treatment*, aimed to remove non-sedimentable organic solids, whether dissolved or colloidal, as well as to stabilize the organic matter in the sludge. This process involved the use of anaerobic and aerobic reactors.

The main odorous compounds emitted into atmospheric air from wastewater treatment plants were sulphur-containing substances such as hydrogen sulphide, methyl mercaptan, dimethyl sulphide, dimethyl disulphide, ethyl mercaptan, carbon disulphide and carbonyl sulphide [5]. Although all of these odorous compounds might have been present in atmospheric air, the most significant odorous compound was hydrogen sulphide (H<sub>2</sub>S, being a dominant odorant gas). The biological degradation of organic matter generated compounds such as sulphites (SO<sub>3</sub><sup>2-</sup>) and sulphates (SO<sub>4</sub><sup>2-</sup>) [6]. High concentrations of hydrogen sulphide can lead to unpleasant and disturbing odours in the vicinity of treatment plant. The main chemical reactions leading to the formation of H<sub>2</sub>S in the treatment plant were (equation 1 and 2): decomposition of organic matter and sulphate reduction reactions in anaerobic environment, according to [7]:

$$SO_4^{2-} \xrightarrow{anaerobic} S^{2-} + H_2O + CO_2 \tag{1}$$
$$S^{2-} + 2H^+ \to H_2S \tag{2}$$

Depending on the immission of  $H_2S$  into the ambient air, the evolution of odour concentration took place in the municipal wastewater treatment plant.

To control these odours, wastewater treatment plants must implement strategies for efficient gas management, such as the use of gas capture and neutralization equipment and the optimisation of wastewater treatment processes [8].

Increasing the monitoring of odour immissions through dynamic olfactometry was important to control their impact on the environment and to ensure safe working conditions for WWTP personnel [9].

The odour is a chemical sense that is defined as the sensation that occurs when volatile substances interact with an organism's olfactory system, Sensory nerve endings located on the surface of the olfactory epithelium are sensitive to chemical stimuli, induced by volatile compounds. So an odorant that reaches the nose makes contact with the sensory endings of olfactory receptor neurons in the nasal epithelium [10].

The impact assessment of odour immissions on the site of the municipal wastewater treatment plant was made based on mathematical modelling by using dispersion programs. These programs simulated the effect of atmospheric dispersion occurring as odours move from the point of release to the receiver [11].

The regulations regarding air quality conditions in protected areas were provided in STAS 12574-87. The maximum permissible concentrations provided in this standard for  $H_2S$  in ambient air, for a period of 24 hours was 0.006 ppm, and for a period of 30 minutes it was 0.015 mg/m<sup>3</sup> 0.011 ppm, so the population was protected from the harmful effects [12].

Measures that obliged companies to install their own odour monitoring systems but also to develop and comply with the plan of measures to reduce this olfactory discomfort. Olfactory discomfort was defined by the law of odour as *the effect generated by an activity that may have an impact on the health of the population and the environment, which was subjectively perceived on different scales of odours or was objectively quantified according to national, European and international standards in force* [13].

The main purpose of the work was odours monitoring and correlation with hydrogen sulphide immissions from a municipal wastewater treatment plant with a population of about 2 million.

# MATERIALS AND METHODS

#### Sampling campaign

The sampling campaign covered the year of 2020, for a period of two weeks in early autumn at the municipal wastewater treatment plant, located in south region of Romania.

The sample of residual gaseous collected during the sampling campaigns was divided into two samples, one for hydrogen sulphide concentration quantification and the other sample to determine the odour concentration by dynamic olfactometry.

The sampling equipment used in the determination of hydrogen sulphide in immissions was a Gil Air type sampling pump, capable of ensuring a sampling flow between  $0.01 \div 0.03$  m<sup>3</sup>/h (Fig. 3), absorption, made of glass or material resistant to corrosion of absorbent solution/analyte, CINTRA 5 spectrophotometer.



Fig. 1. Type sampling pump Gil Air [14]

Therefore, both  $H_2S$  and odour were monitored at the following sources: screens, grit chamber, primary clarifier, sludge pumping station, aeration tank, secondary clarifier, storage platform, digester, co-generation, desulphurisation, period which average atmospheric air temperatures were between  $11\div21^{\circ}C$  (Fig. 4). The sampling points of the odour samples on the site of the municipal wastewater treatment plant are illustrated in Fig. 5.

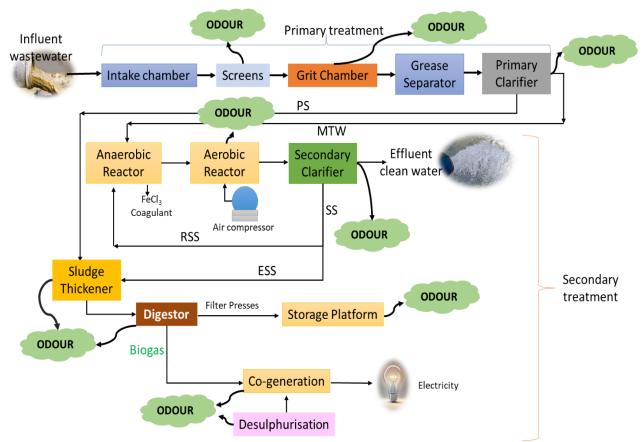


Fig. 2. Scheme of municipal wastewater treatment process and the sampling points: Primary Sludge-PS, Mechanically Treated Water- MTW, Secondary Sludge-SS, Recycled Secondary Sludge-RSS, Excess Secondary Sludge-ESS



Fig. 3. The sampling points of the odour samples on the site of the municipal wastewater treatment plant

#### **Odour Measurements**

Odour sampling involved collecting a volume of 10 L of odorant gas with a vacuum device, in containers that did not affect the quality of the sample (special certified nalophan bags). The odour analysis was performed by the dynamic olfactometry method, EN 13725 (2003) based on sample dilution until the detection threshold was reached by all four human evaluators.

The concentration at which an odour was detected was assigned as the lower limit of analytical determination, representing the concentration of the odorous substance which had a probability of 0.5% being detected. The concept of detection threshold was the basis of dynamic olfactometry, in which a quantitative sensory measurement was used to define the concentration of an odour.

This methodology used a dilution instrument, called a TO8 olfactometer (Fig. 6), which functioned as a high-accuracy diluter and used the human nose as a detector. The odour sample was diluted with nitrogen or air free from odorous substances. Examiners were selected according to the standardised procedure using reference gases (n-butanol, C<sub>4</sub>H<sub>9</sub>OH), only assessors meeting predetermined criteria of variability and sensitivity are selected [15].

The result was expressed in odour concentration (noted  $c_{od}$ ), as  $ou_E/m^3$ , an European unit of odour. According to European standardisation, the European odour unit has been defined as the quantity of odorous substance which, when evaporated in one  $m^3$  of neutral gas under standard conditions (0°C, 1 atmosphere), generates a psychological response from a group of assessors (detection threshold).



**Fig. 4.** Dynamic olfactometer T08 4-station [16]

#### H<sub>2</sub>S detection

Sampling for hydrogen sulphide in ambient air was carried out by retention in specific absorbent solution (zinc hydroxide,  $Zn(OH)_2$ ), bubbling a sampling rate of 1,5 L/min. Detection of the H<sub>2</sub>S concentration was carried out according to the *Handbook Environmental Technology method*, which laid the foundation for the internally validated method. In which a measured volume of residual gaseous effluent is passed through absorbent solution (alkaline aqueous zinc hydroxide solution) by suction of a measured volume of H<sub>2</sub>S gas and the S<sup>2-</sup> ions retained in the absorbent solution react acidically in the presence of Fe<sup>3+</sup> ions with paradimethylphenylenediamine, forming tetramethyldiaminodiphenylthiazine chloride as the reaction product, exhibiting maximum absorbance at 670 nm. The results are expressed in mg/m<sup>3</sup> and are calculated by the equation 3 [17]:

$$C_{H_2S} = \frac{c}{v} \qquad [mg/m^3] \tag{3}$$

where:  $C_{H_2S}$  - was the concentration of H<sub>2</sub>S sampled ( $mg/m^3$ ), c - was the H<sub>2</sub>S content read off the absorption calibration curve, V-volume of air sampled (Litri)

#### Mathematical modelling

To assess the impact of odour on sensitive receptors located in areas adjacent to the wastewater treatment plant, the Aermod View program was used. Program estimated on the basis of the

Gaussian model, the expected odour immissions around the municipal wastewater treatment plant. Figure 7 shows the spatial distribution of sensitive receptors in the nearest inhabited areas in the vicinity of the site.

The dispersion of odour concentration in the ambient air atmosphere varied significantly depending on the location of the treatment plant, prevailing weather conditions and areas of odour accumulation.

Thus, weather data provided by the National Meteorological Administration were used to carry out mathematical modelling of the dispersion of pollutants in atmospheric air. These data aimed to characterize weather parameters in the form of hourly averages: temperature, pressure, humidity, cloudiness, wind speed and direction.



Fig. 5. Location of points where sensitive receptors were inserted, where: point 1- distance of about 50 m, point 2- distance of about 100 m, point 3- distance of about 1500 m, point 4- distance of about 1000 m, 5-distance of about 1100 m

# **RESULTS AND DISCUSSION**

The production capacity of the municipal wastewater treatment plant had a flow rate of 10  $m^3/s$ , for total urban and meteoric wastewater.

In order to assess the impact of  $H_2S$  concentration and odour on sensitive receptors, many steps such: identification of odour sources on the site of the treatment plant, measurement of odour and  $H_2S$  concentration, characterization of all weather parameters, development of dispersion maps and interpretation of the results, were steps schematically shown in the Fig. 8.

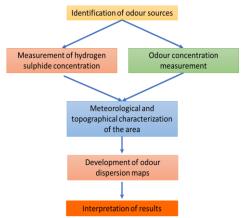


Fig. 6. Block diagram for impact assessment due to exposure to odour and hydrogen sulphide in municipal wastewater treatment plant

In terms of odour concentrations, a maximum value of approximately 36 000  $ou_E/m^3$  was recorded in *the Sludge pumping station* Fig. 9. The major source of high H<sub>2</sub>S concentrations was the sludge pumping station, followed by the digester. These elements played an important role in the efficient treatment of sewage sludge and in managing organic waste in a sustainable way.

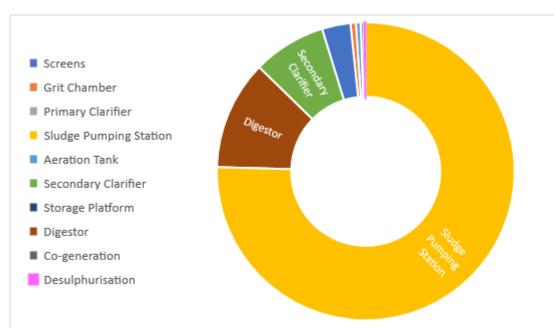


Fig. 7. Major odour sources identified on wastewater treatment plant areal

A direct relationship can be observed between the level of  $H_2S$  present in the atmospheric air and the odour concentration in the area of the municipal wastewater treatment plant.  $H_2S$  was a key contributing factor in determining odour. An increase in the concentration of  $H_2S$  will lead to an intensification of the smell. The results obtained in Fig. 10 showed the correlation between hydrogen sulphide and odour concentrations, indicated a significant increase in odour concentration with increased in hydrogen sulphide immission concentrations. The main major sources of odour within the treatment plant were associated with admission works, biological decomposition processes of organic matter or sludge processing. However, *the aeration activated sludge system* has successfully achieved desulfurization of approximately 99% of  $H_2S$  through degradation and absorption processes.

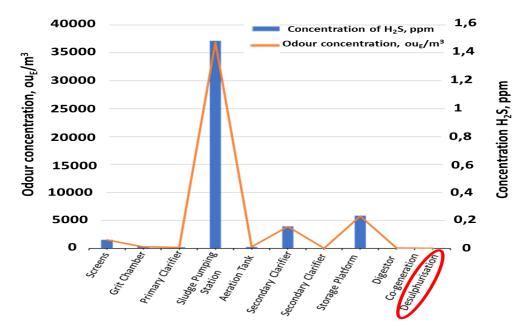


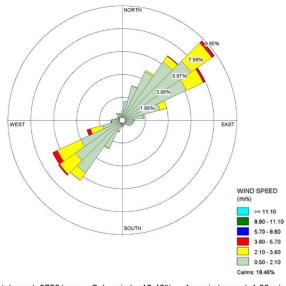
Fig. 8. Contribution of hydrogen sulphide in the determination of odour concentration

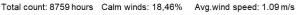
The processes of decomposition of organic sulphur and reduction of inorganic compounds, by anaerobic microorganisms, generated H<sub>2</sub>S imissions to the surrounding air [18]. The detection threshold for H<sub>2</sub>S concentration is 0.0014 ppm, but the presence of the odor has been identified at levels below 1 ppm [19]. Prolonged human exposure (over 8 hours) to concentrations ranging from 2 to 5 ppm can lead to symptoms such as headaches, nausea, and excessive tearing, whereas a concentration of 50 ppm can result in respiratory tract irritation [20]. Experimental found have indicated that the obtained results do not pose risks to human health.

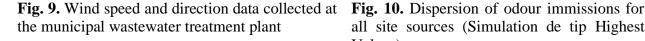
#### *Meteorological data*

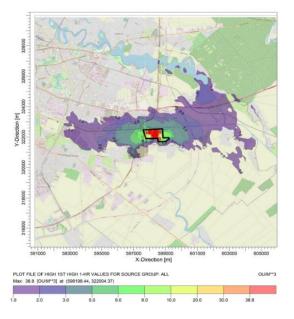
Meteorological data were necessary to carry out mathematical modelling. The average annual temperature was 13.1°C and the average relative humidity was 71.6%. Fig. 11 shows the wind rose. The prevailing wind direction was NE, and the average wind speed was 1.09 m/s, the percentage of hours of atmospheric calm was 18.46% and 1617 hours, respectively.

Thus, for all sources of odor existing on the site, the estimation of the highest concentrations that can be reached during a year in the analysed area was also made. In which a maximum odour concentration of 38.8  $ou_{E/M}^3$  was revealed on site and concentrations between 5÷7  $ou_{E/M}^3$  around the site, without affecting residential areas (Fig.12).









all site sources (Simulation de tip Highest Values)

The Highest Values function involved the processing of emission data, topography and hourly values for meteorological parameters (temperature, humidity, atmospheric pressure, cloudiness, wind direction and speed). This function did not characterize the usual odour level in the studied area, but the values reached in these conditions had a unique character (1 h of the year), corresponding to the most unfavourable meteorological and emission conditions. Highest Values were useful for assessing the potential for exposure to high levels of pollution and may have been important in assessing environmental impacts or public health risks.

In addition to the estimation of the odour level, the placement of sensitive receptors was carried out, by placing them in the closest residential areas, near the treatment plant. Table 1 highlighted the results obtained following the introduction of these sensitive receptors. The highest concentrations were found in point 4 (4.04  $OU_E/m^3$ ), followed by point 1 (3.87  $OU_E/m^3$ ). Although point 4 was located at a distance of 1000 m, it is located on the prevailing wind direction, NE. The predicted values for the concentration of  $H_2S$  were calculated based on the ratio between the concentrations determined by the smell and those of  $H_2S$ , 25 000. The estimated values of  $H_2S$  in residential, they were far below the maximum allowed concentrations. The safety level of the residents was respected.

Point	Coordinates (Stereo 70)		Distance to the unit (m)	Odour concentration, OU <sub>E</sub> /m <sup>3</sup>	Predicted concentration H <sub>2</sub> S, ppm
	Х	У		"Highest values"	
1	598198.68	321882.88	50	3.87026	1.55E-04
2	599430.70	321449.07	1000	0.24823	9.93E-06
4	597402.87	323144.08	1000	4.04557	1.62E-04
5	599552.47	323244.76	1100	1.89741	7.59E-05
3	596572.92	321347.08	1500	0.20362	8.14E-06

The prevailing wind direction significantly influenced the perceived odour concentrations at the receptors. If odours were carried directly to the receiver via wind and were not diluted or deflected by obstacles or atmospheric conditions, this could lead to high odour concentrations at the receiver. Wind speed and direction data collected at the municipal wastewater treatment plant were important in managing odour pollution and assessing impacts on communities and the environment.

An odour at a concentration of  $1 \text{ ou}_E/\text{m}^3$  was a weak concentration that could be only detected in a controlled environment of an odour laboratory (only 50% of the population could sense the presence of this odour).

Weather conditions played an important role, as was showed by the obtained results. Under the worst weather conditions, concentrations in the range of  $1\div 5 \text{ OU}_{\text{E}}/\text{m}^3$  could occur.

# CONCLUSIONS

In conclusion, the analysis of odour concentrations within the municipal wastewater treatment plant correlated with a major source of  $H_2S$  immissions such as a sludge pumping station, where maximum value of 36 000 ou<sub>E</sub>/m<sup>3</sup> was recorded. The sludge pumping station has been a crucial element in sewage sludge treatment and sustainable organic waste management. Ambient temperature played a significant role in the volatilization of  $H_2S$ , thereby influencing odour concentrations, and the correlation between hydrogen sulphide and odour was evident.

Although  $H_2S$  had a very low detection limit, experimental results indicated the absence of risks to human health. Based on mathematical modelling, the maximum odour concentrations that could be found at sensitive receptors were between  $0.2 \div 4$  ou<sub>E</sub>/m<sup>3</sup>, and the highest odour concentrations were located 1000 meters from the site, in the wind direction (Point 4) and 50 meters from the location (Point 1).

The location of the sensitive receptors demonstrated that the main wind direction had a significant impact on the odour levels perceived by them.

It was confirmed a direct link between the degree of air loading with  $H_2S$  and the determination of the odour concentration in the municipal wastewater treatment plant, which was essential in the process of odour assessment and quantification. In terms of olfactory perception, there was a directly proportional relationship between the concentration of odour and that of  $H_2S$ , with a ratio of 25 000. This makes the odour perceived by humans more intense as the concentration of  $H_2S$  increases, and this relationship is characterized by a constant and predictable increase in odour intensity with increasing  $H_2S$  concentration.

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