

VOL. 85, 2021



DOI: 10.3303/CET2185009

Continuous Characterization of the Odour Emissions by Advanced Instrumental Odour Monitoring System in Oil Refinery and Petrochemical Plant

Giuseppina Oliva^{a,b*}, Tiziano Zarra^{a,b}, Gianluca Pittoni^c, Marco Castellani^c, Vincenzo Belgiorno^{a,b}, Vincenzo Naddeo^{a,b}

^aSPONGE Srl, Accademic Spin Off of the University of Salerno, Laboratory SEED, Via Giovanni Paolo II, 132, 84084 Fisciano (SA), Italy

^bSEED - Sanitary Environmental Engineering Division, Department of Civil Engineering, University of Salerno, via Giovanni Paolo II, Fisciano, SA, Italy

^cSARTEC, Saras Ricerche e Tecnologie Srl, I Traversa 2^a strada Est, Macchiareddu, Assemini, CA, Italy goliva@spongeitalia.com

Oil refinery and petrochemical plants are of crucial importance to enhance world economy. Besides the related considerable economic impact of this industries, the population and governments pose towards a great attention which boosted the necessity of developing effective tools for the control of the associated environmental pressures, without hindering economy growth. Among the environmental aspects of key relevance in the refinery sector, air quality and atmospheric pollution in terms of odour emissions, especially in the last years, have become a priority concern in the management of the plants. The monitoring and control of the odour emissions are thus needed in order to prevent potential impact and avoid complaints from the exposed population. Nowadays, senso-instrumental methods by using Instrumental Odour Emissions Systems (IOMSs) represents the most attractive tool for the monitoring of environmental odours, allowing the possibility to obtain real-time and continuous information's, to support the decision-making process and for the implementation of proactive approach. No universally accepted regulation or standardized procedure exist at present and limited data are available in the scientific and technical literature with reference to these systems for the odour monitoring control in the oil refinery plants. In the CEN/TC264 'Air Quality' standardization committee, a specific working group (WG41) is currently working to produce a validation procedure for the IOMSs. The research presents and discusses an advanced instrumental odour monitoring system device useful for the continuous characterization of the odour emissions in complex oil refinery and petrochemical plants. The influence of the use of different feature extraction methods in the odour monitoring model elaboration, in terms of classification, is analysed. Results demonstrate the existence of considerable differences in terms of correct classification percentage, in the use of different feature extraction methods. The importance and usefulness of having a fully-developed flexible system that allows to select and compare automatically different settings options, as the different feature extraction methods, in order to guarantee the best training model, is highlighted. Results demonstrated the high reliability of the system in recognizing artificial gas samples, with characteristics similar to the emissions from refinery plants.

1. Introduction

The great attention which the population and governments pose towards the industrial sites of the Oil and Gas Industries (OGI) sector boosted the necessity of developing effective tools for the control of the associated environmental aspects. Among the environmental aspects of key relevance in the refinery sector, air quality and atmospheric pollution assume an important role related to the human health (Oliva et al., 2018; Xu et al., 2019). The correlated odour emissions may cause several problems, annoyance and discomfort to the exposed people even when they do not imply toxic effects (Belgiorno et al., 2012; Naddeo et al., 2016, Zarra et al., 2008). Odour has been identified as one of the atmospheric pollutant and, among them, it represents the major cause of

Paper Received: 18 December 2020; Revised: 19 February 2021; Accepted: 19 April 2021

Please cite this article as: Oliva G., Zarra T., Pittoni G., Castellani M., Belgiorno V., Naddeo V., 2021, Continuous Characterization of the Odour Emissions by Advanced Instrumental Odour Monitoring System in Oil Refinery and Petrochemical Plant, Chemical Engineering Transactions, 85, 49-54 DOI:10.3303/CET2185009

50

complaints (Senatore et al., 2020; Naddeo et al., 2016). Odour pollution is nowadays a serious hindrance, but the complexity of odour compounds characterization limited a shared approach for odour emissions regulation (Zarra et al., 2012; Fasolino et al., 2016). With a view at addressing suitable strategies for odour and air pollution abatement and control, the characterization of the volatile compounds responsible for possible environmental pressures is of key importance (Oliva et al., 2021). In that view, to overcome the limitation of sensory and instrumental methods for the detection of odourous compounds, in the last years the proposition of sensorialinstrumental technique is rapidly expanding (Capelli et al., 2014, Giuliani et al., 2012; Zarra et al., 2019). Instrumental Odour Emissions Systems (IOMSs) represents indeed an attractive tool for the monitoring of environmental odours, with a view at obtaining real-time information, to support the decision-making process and for the implementation of proactive approaches (Zarra et al., 2018). The use of these electronic devices, also known as electronic nose, for odour monitoring purposes is of key interest since they offer the strengths of both sensorial and analytical tools. Nowadays, among the most challenging problems related to the application of IOMSs, the adequate selection of the sensors array for the specific application, with high robustness, selectivity and reproducibility, assumes a key role (Blanco-Rodríguez et al., 2018). Moreover, the needs to face with the complexity of gaseous mixtures from industrial sources boosted the attention given to sampling technique, training phase and data analysis. These activities have to be adapted to the specific requirements. In particular, the attention paid to data processing resulted in different possibilities to implement, among which statistical pattern recognition. The pre-processing operation of the raw data is a key operation and the selection of the best procedure for the data reduction has an high impact on the accuracy of the results (Shi et al., 2008; Yan et al., 2015). In that view, the research presents and discusses the influence of the use of different preprocessing and extraction methods in the odour classification model elaboration, by using instrumental odour monitoring system, with the aim of optimizing and increasing the reliability of the measurements. The outcomes of this works has been discussed in order to implement the preparatory works needed to put the system in fields.

2. Material and method

2.1 Instrumental odour monitoring system (IOMSartec)

The experimental activities are carried out by using an advanced instrumental odour monitoring system, named IOMSartec, that allows to select and compare automatically different settings options, as the different feature extraction methods. The IOMSartec hardware is composed by two main parts, the measuring unit and the management unit (Figure 1).



Figure 1: IOMSartec hardware layout

The measuring chamber has the main function of ensuring the reproducibility of the chemical conditions of the air to analyse and allowing the array of sensors the contact time needed to carry out the measurement. The chamber has an internal volume designed to optimize the response time of the installed sensors. Sixteen sensors in total are distributed on two horizontal levels, eight for each one: first level, sensors ID1.1 – 1.8; second level, sensors 2.1 – 2.8. Three different types of sensors are located in the measurement chamber: metal-oxide-semiconductor (MOS, n.14), photoionization detector (PID, n.1, ID1.8) and control sensor (temperature-humidity, n.1, ID2.8). The gaseous mixture, to be analysed, is introduced into the measuring chamber by a single-stage membrane pump, specific for gas, with a constant air flow equal to 300 ml min⁻¹. The management system is used to generate zero air and span air by using respectively an ozone system and a permeate tube. The zero air is used to clean the system, while the span air to verify the sensor signals.

The IOMSartec software is designed to perform different operation modes (Figure 2) and allows to select and compare automatically different settings options, like the different feature extraction methods in the raw data pre-processing operation carried out to elaborate the odour monitoring model in terms of classification and quantification, for the training phase.



Figure 2: Operation modes

2.1 Experimental plan and program

Five different training classes, collected in odourless 7 litres volume bags, were used for the experimental activities (Table 1). Artificial odour samples were created with the contamination of pure air with toluene (CAS: 108-88-3) and ethylene (CAS: 74-85-1), which are compounds typically emitted by OGI sector plant (Qiu et al., 2019).

Table 1: Training classes

	Concentration of	
ID	Description	the odour substance
AF	air filtered with active carbon filter	-
Т	air filtered with active carbon filter artificially contaminated with toluene solution	1 ppm
PT	air filtered with active carbon filter without contamination analysed immediately after	_
	T samples	-
Е	air filtered with active carbon filter contaminated with ethylene solution	1 ppm
PE	filtered air without contamination analysed immediately after E samples	-

Table 2: Investigated pre-processing methods

ID	Name	Equation
DVR	Differential Value of resistance	$f_{i,j} = r_{i,j} - r_{i,0}$
RCR	Relative change in resistance	$f_{i,j} = \frac{r_{i,j}}{r_{i,0}}$
FCR	Fractional charge in resistance	$f_{ij} = \frac{r_{i,j} - r_{i,0}}{r_{i,0}}$
FCC	Fractional change in conductance	$f_{ij} = \frac{x_{ij} - x_{i,0}}{x_{i,0}}$
LCR	Logarithmic change in conductance	$f_{ij} = \ln(\frac{x_{ij} - x_{i,0}}{x_{i,0}})$
FCCN	Fractional change in conductance normalized	$\mathbf{f}^{*}_{ij} = \frac{\mathbf{x}_{ij} - \mathbf{x}_{i,0}}{\mathbf{x}_{i,0}}; \ f_{ij} = \frac{f^{*}_{ij}}{\sqrt{\sum_{i=1}^{n} f^{*}_{ij}^{2}}}$

PT and PE classes were introduced to simulate typical conditions which occurs immediately after an odour event, while AF class was used to define the baseline vector. A sequence of AF – T – PT – E – PE cycle was

acquired by the IOMSartec, using a sampling time of 3 minutes (T_F) for each class sample and a detection period of 2 seconds (T_A) for each data. A total of 25 cycle were carried out on different days, acquiring 90 data for each class and 450 data for each cycle.

For each cycle a baseline vector and the training dataset vectors were elaborated. The baseline vector was achieved by extracting the maximum value detected by each sensor in the T_F , in terms of resistance ($r_{i,0}$) or conductance ($x_{i,0}$). The training dataset vector of the feature extraction (f_{FE}) was obtained by calculating the mean value of the f_{ij} values acquired during the sampling time and pre-processed with six different pre-processing methods (Table 2). For each gaseous sample, the F_{FE} matrix was determined with a number of columns equals to the number of the sensors of the array and a number of lines equals to the number of the samples. Linear Discriminant Analysis (LDA) statistical elaboration was then applied to each matrix to create the qualitative classification prediction odour monitoring model. The data set was used to preliminarily validate the accuracy of the prediction model, by applying the predictive algorithms on the training dataset itself.

2.2 Comparison analysis

The comparison analysis was carried out by analyzing the results of the confusion matrix associated to each preprocessed training data. The confusion matrix is an output of the LDA statistical analysis and is useful to describe in terms of percentage of correct classification, the performance of the qualitative classification prediction model for odour monitoring.

Moreover, in order to identify the best methods for the discrimination among the classes, the f_{FE} , values of the different odour samples were evaluated, with a view at identify the solution able to amplify the distances among the odour classes.

3. Results and discussions

The analysis of the results of all investigated samples, in terms of differences of the values detected for odour and odourless samples and among the samples of the different odour classes, highlighted how, even if all the pre-elaboration applied methods demonstrated a good classification among the different investigated odour classes, the feature extraction method with the highest performance is FCCN, for the specific case. The FCCN pre-processing method resulted indeed able to better distinguish odour classes from odour-less class.

The analysis showed the influence of the choice of the feature extraction method in the accuracy of the classification model elaboration and as consequence the possibility to optimize the model with a preliminary selection study. This kind of approach is of prior importance, in particular when higher accuracy and sensitivity are required because of the higher probability of confusion among similar odour classes.

reports the results of the elaborated confusion matrix, calculated applying the predictive algorithms on the training dataset itself. The confusion matrix resulted the same for all the six different pre-processing methods investigated.

			Predicted odour classes (percentage of correct classification)				
		AF	Т	PT	E	PE	
~	AF	100%	0%	0%	0%	0%	
sed	Т	0%	100%	0%	0%	0%	
aly: SS	PT	0%	0%	100%	0%	0%	
C O C	Е	0%	0%	0%	100%	0%	
	PE	0%	0%	0%	0%	100%	

Table 3: Results of the elaborated Confusion matrix

Results highlighted the perfect classification of the investigated samples applying the created prediction models, with a 100% correctness for the corresponding class for all the investigated extraction methods (consisting of the mean values of the data pre-processed according to the formulations in Table 2) and, consequently, their non-influence, in the specific investigated case.

Figure 3 shows the extracted f_{FE} vectors by considering the different methods, for one investigated sample. The analysis of the results of all investigated samples, in terms of differences of the values detected for odour and odourless samples and among the samples of the different odour classes, highlighted how, even if all the preelaboration applied methods demonstrated a good classification among the different investigated odour classes, the feature extraction method with the highest performance is FCCN, for the specific case. The FCCN preprocessing method resulted indeed able to better distinguish odour classes from odour-less class.

52

The analysis showed the influence of the choice of the feature extraction method in the accuracy of the classification model elaboration and as consequence the possibility to optimize the model with a preliminary selection study. This kind of approach is of prior importance, in particular when higher accuracy and sensitivity are required because of the higher probability of confusion among similar odour classes.



Figure 3: fFE vectors calculated by applying the different feature extraction methods for one sample

4. Conclusions

The use of IOMS for the continuous characterization of the odour emissions is rapidly expanding as a preferred method since the high adaptability and the smart and proactive approach achievable. However, senso-instrumental device, as IOMS, have to be properly trained and set to optimize their performance according to the real application in which they would be used. To define the best strategy to implement in real application like as oil refinery sector, the influence of different extraction methods has been studied by using an advanced IOMS

that allows the automatically analysis by setting the different operational modes. Results highlighted how all the implemented methods provide consistent results, with correct classification percentages, of the five different investigated classes, of a 100%. The results showed how, for the specific investigated case, the FCCN method allowed to better differentiate the neutral class (negative values) to the odour artificial contaminated classes. The feature extraction vectors were the input to the LDA and PLS statistical methods and thus the optimization of the data reduction method can allow to increase the recognition scores also for the real applications. In that view, the identification of the most suitable feature extraction method is of key importance to optimize the prediction models. Flexible and smart approaches for IOMSs, supported by a specific software, are thus strongly recommended. The results referred only to the first phases of the creation of the plant.

References

- Belgiorno V., Naddeo V., Zarra T. (2012). Odour Impact Assessment Handbook. p. 1-312, John Wiley & Sons, Inc., ISBN: 9781119969280, DOI:10.1002/9781118481264
- Blanco-Rodríguez, A., Camara, V.F., Campo, F., Becherán, L., Durán, A., Vieira, V.D., de Melo, H., Garcia-Ramirez, A.R., 2018. Development of an electronic nose to characterize odours emitted from different stages in a wastewater treatment plant. Water Res. 134, 92–100. https://doi.org/10.1016/j.watres.2018.01.067
- Chen, H., Zhang, M., Guo, Z., 2019. Discrimination of fresh-cut broccoli freshness by volatiles using electronic nose and gas chromatography-mass spectrometry. Postharvest Biology and Technology 148, 168–175. https://doi.org/10.1016/j.postharvbio.2018.10.019
- Laura Capelli, Selena Sironi, Renato Del Rosso, 2014. Odour emission factors: fundamental tools for air quality management. Chemical Engineering Transactions 40, 193–198. https://doi.org/10.3303/CET1440033
- Naddeo V., Zarra T., Oliva G., Kubo A., Uchida N., Higuchi T., 2016. Odour measurement in wastewater treatment plant by a new prototype of e.nose: correlation and comparison study with reference to both european and japanese approaches. Chemical Engineering Transactions 54, 85–90. https://doi.org/10.3303/CET1654015
- Oliva G., Naddeo V., Zarra T., Belgiorno V., 2018. Abatement of odour emissions by UV/Ozone oxidation process. Global NEST Journal 20, 669-673. https://doi.org/10.30955/gnj.002798.
- Oliva, G., Zarra, T., Pittoni, G., Senatore, V., Galang, M.G., Castellani, M., Belgiorno, V., Naddeo, V., 2021. Next-generation of instrumental odour monitoring system (IOMS) for the gaseous emissions control in complex industrial plants. Chemosphere 271, 129768. https://doi.org/10.1016/j.chemosphere.2021.129768
- Qiu, W., Li, S., Liu, Y., Lu, K., 2019. Petrochemical and Industrial Sources of Volatile Organic Compounds Analyzed via Regional Wind-Driven Network in Shanghai. Atmosphere 10, 760. https://doi.org/10.3390/atmos10120760
- Senatore, V., Zarra, T., Oliva, G., Belgiorno, V., Naddeo, V., 2020. Volatile organic compounds (Vocs) removal by combining bioscrubber and ozone pretreatment. Global Nest Journal 22, 143–146. https://doi.org/10.30955/gnj.003298
- Shi, Z., Yu, T., Zhao, Q., Li, Y., Lan, Y., 2008. Comparison of Algorithms for an Electronic Nose in Identifying Liquors. Journal of Bionic Engineering 5, 253–257. https://doi.org/10.1016/S1672-6529(08)60032-3
- Wilson, A.D., Baietto, M., 2011. Advances in Electronic-Nose Technologies Developed for Biomedical Applications. Sensors 11, 1105–1176. https://doi.org/10.3390/s110101105
- Xu, M., Wang, J., Gu, S., 2019. Rapid identification of tea quality by E-nose and computer vision combining with a synergetic data fusion strategy. Journal of Food Engineering 241, 10–17. https://doi.org/10.1016/j.jfoodeng.2018.07.020
- Yan, J., Guo, X., Duan, S., Jia, P., Wang, L., Peng, C., Zhang, S., 2015. Electronic Nose Feature Extraction Methods: A Review. Sensors 15, 27804–27831. https://doi.org/10.3390/s151127804
- Zarra T., Naddeo V., Belgiorno V., Reiser M., Kranert M. (2008). Odour monitoring of small wastewater treatment plant located in sensitive environment. Water Science and Technology, vol. 58, p. 89-94, ISSN: 0273-1223, doi: 10.2166/wst.2008.330.
- Zarra T., Giuliani S., Naddeo V. and Belgiorno V. (2012). Control of odour emission in wastewater treatment plants by direct and undirected measurement of odour emission capacity, Water Science and Technology, 68(8), 1627–1633, ISSN: 0273-1223, doi: 10.2166/wst.2012.362.
- Zarra T., Cimatoribus C., Naddeo V., Reiser M., Belgiorno V., Kranert M. (2018). Environmental odour monitoring by electronic nose. Global NEST Journal, 20, 3, 664 668.
- Zarra, T., Galang, M.G.K, Ballesteros, F.J., Belgiorno, V., Naddeo, V. (2019). Environmental odour management by artificial neural network – A review. Environment International, 133, 105189. https://doi.org/10.1016/j.envint.2019.105189.

54