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Hydrogen Sulphide and Odour Emissions Control in Wastewater Treatment Plants (WWTPs) by an Integrated Sustainable Biotechnological System

Federica Pasquarelli*, Aniello Mariniello, Fabiana Romano, Giuseppina Oliva, Vincenzo Belgiorno, Vincenzo Naddeo, Tiziano Zarra

Sanitary Environmental Engineering Division (SEED), Department of Civil Engineering, University of Salerno, via Giovanni Paolo II, Fisciano, SA, Italy
fpasquarelli@unisa.it

Wastewater treatment plants (WWTPs) play a crucial role in the water cycle. These facilities ensure the balance of the ecosystem and avoid significant negative effects on the environment and human health. However, they also exert negative pressures. Among these, odours and odorous compound emissions, principally hydrogen sulphide (H_2S), require more attention. Prolonged exposure to odour emissions produces negative effects on exposed populations, such as nausea, headaches, and other related respiratory problems. The perception of odours is also considered a sign of a polluted environment and therefore the first cause of complaints. In this context, there is a need to characterize and control odour emissions. Currently, among the odour treatment processes, biological ones show the greatest potential for future development, as they are cheaper and more sustainable for the environment. However, decomposition by microorganisms with oxidative properties produces carbon dioxide, which is one of the main greenhouse gases and contributes to global warming.
The research presents and discusses the application and validation of an advanced sustainable biological-based system aimed at treating odours and avoiding the release of GHG into the ambient air, thus ensuring clean air and mitigating climate change according to the Sustainable Development Goals (SDGs) of the Agenda 2030. In-depth and extensive experimental activities are carried out and reported to validate the proposed system, consisting of an integration of a Moving Bed Biofilm Reactor (MBBR) and an algal Photobioreactor (aPBR). System performance has been evaluated under different operating conditions, with reference to H_2S removal efficiencies and CO_2 abatement rates and subsequent conversion to usable biomass. The analyses also included the determination of the concentration of odours according to EN 13725:2022. The results obtained confirm the efficiency of the proposed system in reducing odours and biofixation of CO_2, promoting a novel advanced and environmentally friendly solution in odour treatment.

1. Introduction

Wastewater treatment plants (WWTPs) aims to remove pollutants from wastewater so that it can be discharged without harming the environment (Li et al., 2021). Given the nature and type of effluent treated, the treatments have the disadvantage of generating and release odours into the atmosphere (González et al., 2022). The different units, that make up the treatment plant, are responsible of the odour emissions with different odour concentration ranges. As reported by different authors, the main units of unpleasant odours are: arrival tank (100 – 100000 OU_E m^{-3}), grids (380 – 37000 OU_E m^{-3}), sand trap/oil separator (860 – 7000 OU_E m^{-3}), primary settler (410 – 800 OU_E m^{-3}), oxidation/denitrification (160 – 510 OU_E m^{-3}) and sludge dehydration (450 – 10000 OU_E m^{-3}) (Prudenza et al., 2023; Zarra et al., 2022).
Prolonged exposure to odours can lead to direct impacts on the human health of people, such as nausea, headaches, breathing and psycho-physical problems (Oliva et al., 2021; Zarra et al., 2022). As a result, the exposed population tends to reject the installation of WWTPs.This phenomenon is known as NIMBY syndrome (Not In My Back Yard) (Uji et al., 2021).

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Among the main odorous substances emitted by WWTPs there is hydrogen sulphide (H_2S), which can be particularly dangerous to human health and environment (Lee et al., 2023). H_2S is a flammable, colourless and toxic gas associated with the common smell of rotten eggs. The effects of human exposure to this gaseous compound range from eye and airway irritation (at concentrations between 10-20 ppm) to immediate loss of consciousness and death (1000-2000 ppm). At particularly high concentrations (>700 ppm), H_2S causes pulmonary oedema and risk of death (Senatore et al., 2021).
The problem of characterization, measurement and control of odour emissions caused by WWTPs is therefore receiving increasing attention, also linked to an increase in people's demand for life quality and an ever-increasing proximity of residences to the location of the plants. As regards the characterization and measurements, the most common technique for the determination of odour concentration is the dynamic olfactometry (DO), applied according to EN 13725:2022. DO is a sensory quantification method that uses an olfactometer and a group of trained evaluators. The instrument responses are recorded and processed statistically, so as to obtain the final measurement result expressed in OU_E m^{-3} (Burgués et al., 2022). Furthermore, recently, for the continuous monitoring of odours, Instrumental Odour Monitoring Systems (IOMS) are receiving the highest attention from all actors involved in odour issues, because they offer considerable application potential (Zarra et al., 2020). On the other hand, with reference to the odour emission treatment systems still many fields are unexplored or not completely analyzed and above all not congruent with the environmental sustainability and climate change policies promoted by the sustainable development objectives of the Agenda 2030 and by the circular economy and ecological transition approaches. The existing treatment processes are classified into chemical-physical and biological (Pasquarelli et al., 2024; Senatore et al., 2020). The latter, compared to physical and chemical, have the advantage of being more environmentally sustainable and for these more promoted. However, they are associated with emissions of greenhouse gases (GHGs), due to biological degradation (Wu et al., 2018). In order to minimize climate change, it is therefore necessary to develop biological systems that can also control GHGs emissions.
The paper presents the application and validation of an advanced biological-based integrated system for the sustainable treatment of odorous emissions, with the aim to avoid odour impacts and reduce anthropogenic climate change, avoiding the release of GHG into the atmosphere.

2. Material and Methods

2.1 Experimental set-up and operating conditions

The experimental set-up (Figure 1) consists of an MBBR (Mobile Bed Biofilm Reactor), referred to as R1, made of glass and with a working volume of 6.5 L (internal diameter of 165 mm and height of 520 mm), and an aPBR (algal photobioreactor), indicated as R2, made of plexiglass and with a working volume of 26 L (internal diameter of 200 mm and height of 1000 mm). Inside the R1 reactor there are, for one third of its volume, plastic support materials (Carrier, Kaldnes Ring), in order to promote bacterial biofilm. In addition, for the monitoring, there are three gas sampling ports in the system (GSP1, GSP2 and GSP3) and two liquid phase sampling ports (LSP1 and LSP2).
Three different concentrations of H_2S (Table 1) are used for the experimental activities and for the validation of the system. The H_2S is mixed with atmospheric air, supplied by a compressor and regulated by a flow meter (8.5 L min^{-1}), before introduced in the system. H_2S is produced in a mixing chamber, inside which are inserted 150 mg of iron sulfide (FeS), 5 ml of water and a solution of HCl (1.85 % for the I and II stage and 3.8 % for the III and IV stage). The gaseous flux with hydrogen sulfide is fed into the MBBR, through metal diffusers placed on the bottom of the reactor, and then into the aPBR. The light source of reactor R2 consists of 4 LED bulbs with luminous intensity of 8561 LUX and a 12-hour light/dark cycle is set. The mixture of H_2S and atmospheric air is fed into the plant for 6 hours a day.

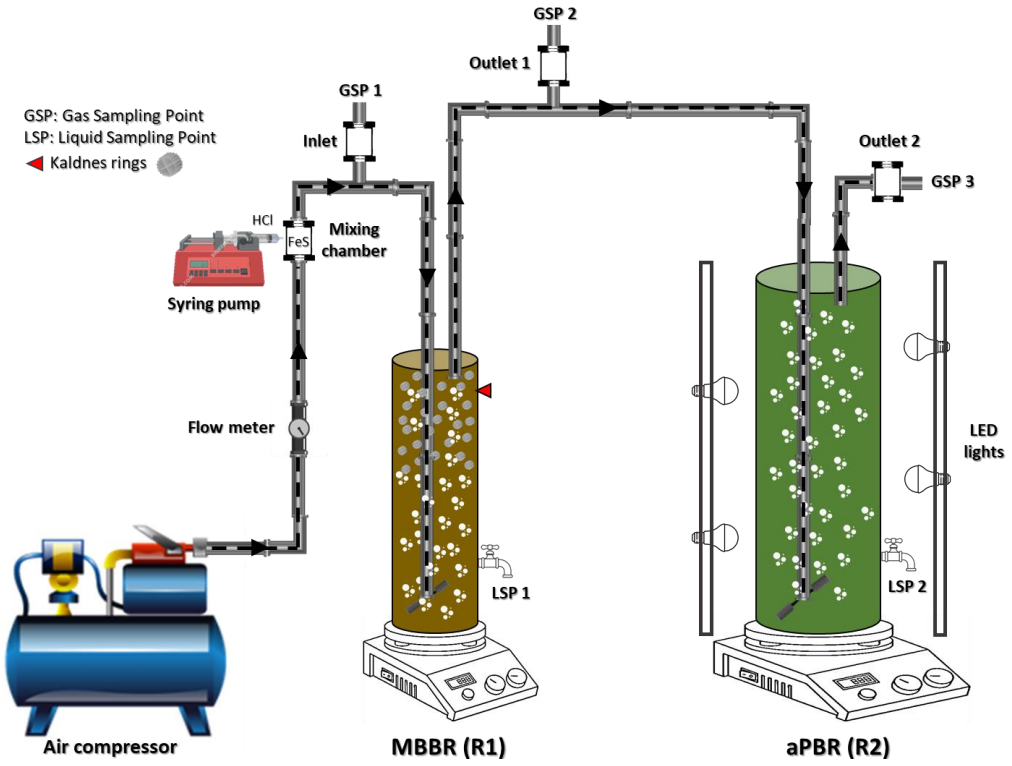

Figure 1: Experimental set-up

The experimental activities were divided into four stages (Table 1). For the first three stages the air flow rate (Q_g) was 0.001 m^3 min^{-1}, while in the last stage it was doubled to 0.002 m^3 min^{-1}, in order to test different operation conditions. As reported, the performance of the system was evaluated with three different concentrations of H_2S (C_{in}): 100 ppm for stages I and II; 300 ppm for stage III; 200 ppm for stage IV. During the entire test activities, liquid samples from R1 and R2 were taken three times a day to measure total suspended solids (TSS), dissolved oxygen (DO), temperature and pH. In addition, the efficiency of the system was evaluated in terms of the removal efficiency of the target odorous compound, CO_2 biofixation and the odour concentration. For mineral renewal, dairy wastewater was used in quantities of 100 (stage I) and 250 (stages II, III and IV) mL d^{-1} for R1 and 500 (stages I, II and III) and 1000 (stage IV) mL d^{-1} for R2.

Table 1: Experimental plan and program – main operating conditions

Stage	C_{in} (H_2S) [ppm]	Feeding time [h d^{-1}]	Q_g [m^3 min^{-1}]	Mineral Renewal R1 [mL d^{-1}]	Mineral Renewal R2 [mL d^{-1}]	Light Intensity [LUX]
I	100	6	0.001	100	500	8561
II	100	6	0.001	250	500	8561
III	300	6	0.001	250	500	8561
IV	200	6	0.002	250	1000	8561

2.2 Inoculum and culture medium

The bacterial sludge present in MBBR was collected at the real wastewater treatment plant located in the municipality of Salerno (Campania region, Italy). The sludge was centrifuged at 7000 rpm for 10 minutes and suspended in synthetic wastewater, similar to dairy wastewater. Chlorella Vulgaris CCAP 211/11B (Culture Collection of Algae and Protozoa, CCAP; Dunbeg, Scotland) was the strain of microalgae grown within aPBR; they were centrifuged at 7000 rpm for 10 minutes and pre-inoculated into a modified Bold Basal Medium solution (a stocks solution per 400 mL of (1) 10.0 g of $NaNO_3$; (2) 3.0 g of $MgSO_4 \cdot 7H_2O$; (3) 1.0 g of NaCl; (4) 3.0 g of K_2HPO_4; (5) 7.0 g of KH_2PO_4; (6) 1.0 g of $CaCl_2 \cdot 2H_2O$; (7) trace elements solution (g/L); $ZnSO_4 \cdot 7H_2O$ (8.82 g), $MnCl_2 \cdot 4H_2O$ (1.44 g), $(NH_4)_6MO_7)_4 \cdot 4H_2O$ (0.87 g), $CuSO_4 \cdot 5H_2O$ (1.57 g), $CoCl_2 \cdot 6H_2O$ (0.38); (8) H_3BO_3 11.42

g/L; (9) 50.0 g/L of EDTA and 31.0 g/L of KOH; and (10) 4.98 g/L of $FeSO_4 \cdot 7H_2O$). Throughout the experimental activities, as a culture medium for both reactors, a synthetic dairy wastewater with the following composition was prepared: per 1 L of wastewater (1) 2.5 g of milk powder; (2) 1.4 g of NH_4CL; (3) 0.05 g of $MgSO4 \cdot 7H_2O$; (4) 1.0 g of KH_2PO_4; (5) 2.0 g of $NaHCO_3$; (6) 0.038 g of $CaCl_2$.

2.3 Analytical method

The concentration of H_2S was determined at GSP1, GSP2 and GSP3 using the multigas instrument (MultiRAE, RECOM Industrial s.r.l) during stages I, II and IV, while during stage III, since the MultiRAE instrument cannot detect an H_2S concentration of more than 200 ppm, the GC - TCD (TRACETM 1300 Gas Chromatograph, Thermo Fisher) instrument was used. With the latter instrument, CO_2 concentrations were also evaluated at GSP1, GSP2 and GSP3.
The concentration of odour was determined by dynamic olfactometry, using a TO8 olfactometer (Ecoma, Gmbh, D) according to the EN13725:2022. The pH, dissolved oxygen and temperature trend were monitored using the multiparameter probe (HI9829, Hanna Instruments), while the monitoring of total suspended solids (TSS) was carried out according to the standard 2540 D method.
All analyses were carried out at the Sanitary Environmental Engineering Division (SEED) Laboratory of the Department of Civil Engineering of the University of Salerno.

3. Results and Discussion

3.1 H_2S removal efficiency

H_2S concentrations were monitored twice a day, at 3 h and 6 h after the start of plant feeding. Figure 2 shows the data of the Removal Efficiency (RE) of H_2S, in terms of mean value for stage, recorded in R1 and in the entire system R1 + R2, correlated to the respective Inlet Load (IL) and Elimination Capacity (EC) of R1 + R2, during the whole experimental analyses.

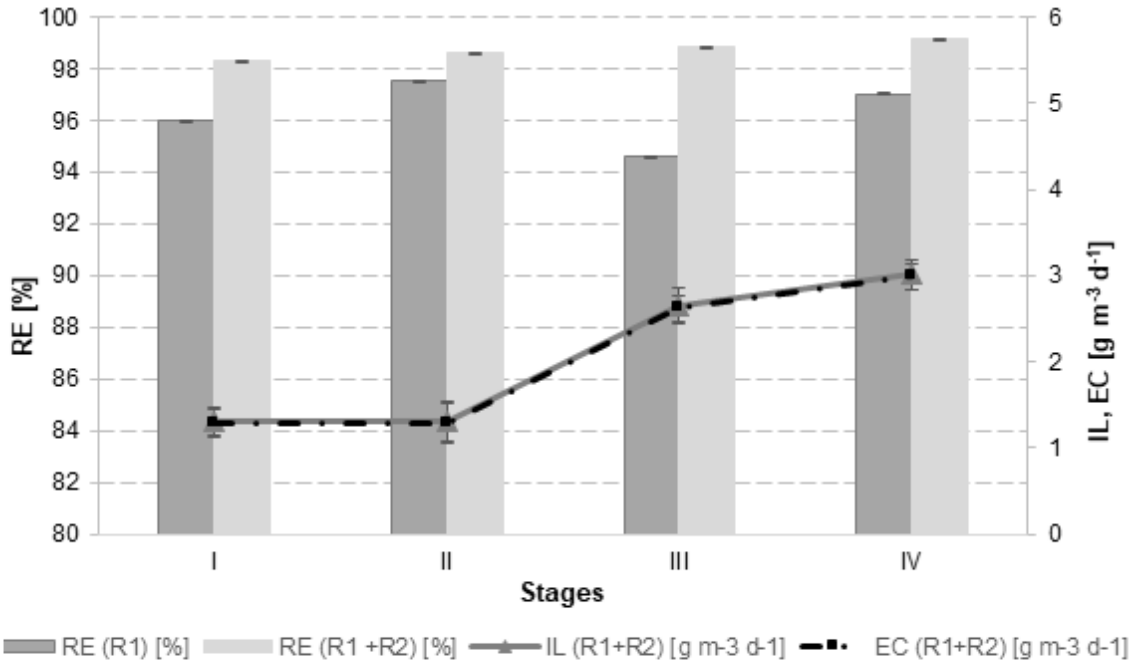

Figure 2: Average RE of H_2S in R1 (dark grey columns) and R1 + R2 (light grey columns), EC (dashed black line) and IL (grey line) in R1 + R2 during the different stages

The results highlight how R1 was able to cope with inlet load fluctuations and the recorded RE was very good, with values up to 97.56 ± 0.01 %. Furthermore, the synergistic work of R1 + R2 has been proved, allowing to obtain particularly high removal efficiencies: 98.34 ± 0.01 % in the stage I, 98.63 ± 0.00 % in the stage II, 98.83 ± 0.01 % in the stage III and 99.18 ± 0.01 % in stage IV. In fact, as can be seen from Figure 2, the inlet load was almost completely degraded, with EC equal to 2.99 ± 0.15 g m^{-3} d^{-1} for IL of 3.01 ± 0.17 g m^{-3} d^{-1} for stage IV where the maximum RE is obtained.

3.2 CO_2 biofixation efficiency

As regards the CO_2 analysis, concentrations were monitored twice a day, at 3 h and 6 h after the start of plant feeding. Table 2 reports the results in terms of removal efficiency (RE) as a function of CO_2 concentrations, input and output at the different sampling points, during the entire operating period. Results show how the removal efficiency achieved with R1 + R2 was 93.90 ± 0.07 % in stage I, 93.67 ± 0.07 % in stage II, 39.76 ± 0.27 % in stage III and 81.57 ± 0.15 % in stage IV.

Table 2: RE of CO_2 in R1 + R2, mean CO_2 concentrations in inlet at R1 (Ci_n), output from R1 ($C_{out,1}$) and output from R2 ($C_{out,2}$) during the different stages

Stage	C_{in} [g m^{-3}]	$C_{out,1}$ [g m^{-3}]	$C_{out,2}$ [g m^{-3}]	RE (R1+R2) [%]
I	0.78	1.55	0.04	93.90
II	0.77	1.82	0.05	93.67
III	0.89	1.91	0.53	39.76
IV	0.94	2.24	0.18	79.72

The highest concentration values were found at the exit of R1, as a result of efficient biodegradation of the target compound by the bacterial community, whereas the CO_2 concentration at the exit from R2 has always been lower than that measured at the inlet, indicating a high biofixation of algal biomass present in the algal photobioreactor. This has allowed for high algal biomass growth within the photobioreactor, which is considered a promising clean energy alternative carrier with a variety of uses in the production of sustainable biofuels, polymers, nutraceuticals and food supplements (Yen et al., 2019).

3.3 Odour abatement efficiency

Odour concentrations were monitored twice a week and Table 3 shows the results obtained for the olfactory tests carried out in accordance with EN 13725:2022 to correlate the H_2S concentrations introduced into the system during the whole activity with the corresponding inlet and outlet odour concentrations.

Table 3: Input concentrations of H_2S investigated during activity, corresponding odour inlet concentrations, mean odour outlet concentrations and mean removal efficiency

C_{in} (H_2S) [ppm]	$C_{od,in}$ [OU_E m^{-3}]	Mean $C_{od,out}$ [OU_E m^{-3}]	Mean RE [%]
100	2299	43	95
200	1448	45	96
300	1149	94	94

The results showed a decreasing linear tendency of the inlet odour concentration between samples 1 and 2, with values of 2299 OU_E m^{-3} and 1448 OU_E m^{-3} for H_2S concentrations of 100 ppm and 200 ppm respectively. Among samples 2 and 3, this decreasing trend is less marked than in the previous case, reaching a concentration value of 1149 OU_E m^{-3} with an H_2S concentration of 300 ppm. With regard to the outlet odour concentrations, average values of 43 OU_E m^{-3}, 35 OU_E m^{-3} and 94 OU_E m^{-3} have been detected, depending on the different removal efficiencies of H_2S in the different stages investigated. The average outlet odour concentrations throughout the activity were always lower than 300 OU_E m^{-3}, that represent the limit conventionally assumed in Europe for emissions into the atmosphere from odorous sources (Settimo and Avino, 2024). A maximum average odour abatement efficiency of 96 % was therefore determined.

4. Conclusions

Technologies for the integrated treatment of odorous compound and the simultaneous capture of carbon dioxide are solutions that scientific research must deepen to reduce environmental pressures and climate change. In this perspective, the advanced integrated biological-based system presented proves to be a valid solution. Removal efficiencies, in terms of reduction of H_2S, in the MBBR reactor were between 92 and 98 % for IL between 1,31 and 3,01 mg m^{-3} d^{-1}, with a further abatement from the aPBR reactor up to 99 %. While CO_2 produced in MBBR, with concentrations between 1,55 and 2.24 g m^{-3}, was effectively biofixed and converted into algal biomass within aPBR, with an overall biofixation efficiency of more than 93 %. Furthermore, also the

olfactory tests confirmed the validity of the integrated system, which achieved average removal efficiencies between 93.51 % and 96.08 % for input concentrations between 861 and 1448 OU_E m^{-3}. The results obtained confirm the efficiency of the proposed system in reducing odours and biofixation of CO_2, promoting a novel advanced and environmentally friendly solution in odour treatment, in accordance with the principles of ecological transition.

References

Burgués, J., Doñate, S., Esclapez, M.D., Saúco, L., Marco, S., 2022. Characterization of odour emissions in a wastewater treatment plant using a drone-based chemical sensor system. Science of The Total Environment 846, 157290. https://doi.org/10.1016/j.scitotenv.2022.157290

González, D., Colón, J., Sánchez, A., Gabriel, D., 2022. Multipoint characterization of the emission of odour, volatile organic compounds and greenhouse gases from a full-scale membrane-based municipal WWTP. Journal of Environmental Management 313, 115002. https://doi.org/10.1016/j.jenvman.2022.115002

Lee, J., Lee, S., Lin, K.-Y.A., Jung, S., Kwon, E.E., 2023. Abatement of odor emissions from wastewater treatment plants using biochar. Environmental Pollution 336, 122426. https://doi.org/10.1016/j.envpol.2023.122426

Li, R., Han, Z., Shen, H., Qi, F., Ding, M., Song, C., Sun, D., 2021. Emission characteristics of odorous volatile sulfur compound from a full-scale sequencing batch reactor wastewater treatment plant. Science of The Total Environment 776, 145991. https://doi.org/10.1016/j.scitotenv.2021.145991

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

Pasquarelli, F., Oliva, G., Mariniello, A., Buonerba, A., Li, C.-W., Belgiorno, V., Naddeo, V., Zarra, T., 2024. Carbon neutrality in wastewater treatment plants: An integrated biotechnological-based solution for nutrients recovery, odour abatement and CO2 conversion in alternative energy drivers. Chemosphere 354, 141700. https://doi.org/10.1016/j.chemosphere.2024.141700

Prudenza, S., Bax, C., Capelli, L., 2023. Implementation of an electronic nose for real -time identification of odour emission peaks at a wastewater treatment plant. Heliyon 9, e20437. https://doi.org/10.1016/j.heliyon.2023.e20437

Senatore, V., Zarra, T., Galang, M.G., Oliva, Giuseppina, Buonerba, A., Li, C.-W., Belgiorno, V., Naddeo, V., Oliva, Giusy, 2021. Full-Scale Odor Abatement Technologies in Wastewater Treatment Plants (WWTPs): A Review. Water. https://doi.org/10.3390/w13243503

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

Settimo, G., Avino, P., 2024. State-of-art of the legislation on odour emissions with a focus on the Italian studies. Environmental Pollution 123525. https://doi.org/10.1016/j.envpol.2024.123525

Uji, A., Prakash, A., Song, J., 2021. Does the “NIMBY syndrome” undermine public support for nuclear power in Japan? Energy Policy 148, 111944. https://doi.org/10.1016/j.enpol.2020.111944

Wu, H., Yan, H., Quan, Y., Zhao, H., Jiang, N., Yin, C., 2018. Recent progress and perspectives in biotrickling filters for VOCs and odorous gases treatment. Journal of Environmental Management 222, 409–419. https://doi.org/10.1016/j.jenvman.2018.06.001

Yen, H.-W., Hu, I.-C., Chen, C.-Y., Nagarajan, D., Chang, J.-S., 2019. Chapter 10 - Design of photobioreactors for algal cultivation, in: Pandey, A., Chang, J.-S., Soccol, C.R., Lee, D.-J., Chisti, Y. (Eds.), Biofuels from Algae (Second Edition). Elsevier, pp. 225–256. https://doi.org/10.1016/B978-0-444-64192-2.00010-X

Zarra, T., Galang, M.G.K., Oliva, G., Belgiorno, V., 2022. Smart instrumental Odour Monitoring Station for the efficient odour emission management and control in wastewater treatment plants. Chemosphere 309, 136665. https://doi.org/10.1016/j.chemosphere.2022.136665

Zarra, T., Mark, G., Galang, M.G., Ballesteros Jr, F., Belgiorno, V., Naddeo, V., 2020. Instrumental Odour Monitoring System Classification Performance Optimization by Analysis of Different Pattern-Recognition and Feature Extraction Techniques. Sensors 21, 114. https://doi.org/10.3390/s21010114