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Use of Micro/Nanobubbles for the Treatment of Polluted Effluents: A Systematic Review and Meta-analysis in Relation to BOD and COD

Carlos A. Castañeda-Olivera*, Renato L. Belahonia Talledo, Lucero M. García Espejo, Rita J. Cabello Torres, Elmer G. Benites Alfaro, Geraldine K. Guadalupe Sandoval

Professional School of Environmental Engineering, Universidad César Vallejo, Lima, Perú
ccastanedao@ucv.edu.pe

Deterioration of water quality is a serious problem that continues to face the environment due to the discharge of untreated wastewater into water bodies. The present systematic review and meta-analysis research evaluated the efficacy of micro/nano bubbles (MNB) in the treatment of polluted effluents in relation to BOD and COD. The research was of quantitative approach, applied type and non-experimental design. The scientific articles were collected from Scopus and Web of Science databases from the period of January 2011 to November 2021. The results showed that only five investigations were included and subjected to meta-analysis. Those studies indicated that treatment with MNBs achieved removals between 69 to 100% and 68 to 99% for BOD5 and COD, respectively. The best removal efficiency (97% COD and 100% BOD5) was achieved in the scientific investigation of Rameshkumar (Rameshkumar et al. 2019). Finally, it is concluded that MNBs effectively reduce the pollutants present in wastewater.

1. Introduction

Cities, industries, hotels, and mining, agricultural and livestock farms produce large amounts of polluted wastewater that cause great harm to the environment (Valdiviezo Gonzales et al. 2021; Benites Alfaro et al. 2022; Nicho Chavez et al. 2022). Eighty percent of these effluents are discharged into rivers, lakes, seas, and even into the subsoil, through septic tanks and landfills. This mainly affects watersheds, leading to the alteration of the trophic cycle of nature and harming both the environment and public health (WWAP 2017; Álvarez Huamán & Rios Bujaico 2020).Peru is no stranger to this, as the contamination of rivers and seas due to the mining and oil industry is constant in the country, bringing obvious consequences. For example, in 2020, the district of Pacora in Lambayeque, Peru was declared in a state of emergency due to arsenic contamination of drinking water, establishing permanent monitoring of water quality and epidemiological surveillance (Defensoría del Pueblo 2021). Another clear example is the Rimac River, with more than 900 points of contamination generated by the population due to land clearing, industrial activities, wastewater and other types of pollutants (Gestión 2019) The use of micro/nanobubbles (MNBs) in water treatment has become more relevant in the last decades due to their multiple applications in different fields such as biomedical engineering, agriculture, nanomaterials, industrial, environmental, among others (Takahashi, 2009). These MNBs have a high stability, which helps in decreasing the buoyant force and the presence of repulsive forces of the bubbles among themselves (Vásquez Benavides 2020), i.e. MNBs improve gas dissolution and mass transfer, which provides greater oxygenation in the effluents and accelerates the process of oxidation of organic matter. Furthermore, the application of MNBs in environmental remediation has recently attracted attention due to their special characteristics such as large specific surface area, a negatively charged surface, a long residence time in water, high mass transfer efficiency and the generation of highly reactive free radicals (Haris et al. 2020).

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The application of MNBs in the wastewater reclamation process is considered an effective and accepted method since it is a process that does not generate negative impacts on the environment. On the other hand, the use of this technology represents an improvement in public health, reducing the populations affected by the consumption of contaminated water. Therefore, the present research through a systematic review and meta-analysis evaluated the effectiveness of the application of MBMs in the treatment of polluted effluents in relation to BOD and COD, identifying the pollutant with the highest removal efficiency and the optimal characteristics that the MBMs should present to obtain favorable results in the treatment.

2. Materials and methods

2.1 Type of study

The present research had a quantitative approach and was of the applied type, since it is evidential and sequential given that the results are presented immediately (Muntané Relat 2010). The scientific articles were extracted from reliable sources of information such as Scopus and Web of Science.

2.2 Sources of information and search strategy

The systematic review of the present research was developed based on the PRISMA methodology and the statistical approach of meta-analysis (Aquije Morey et al. 2021; Acruta Paredes, Leyva Lira & Castañeda Olivera 2022). The search for research was limited from January 2011 to November 2021. This was performed in the English language and systematically according to keywords such as nanobubbles, microbubbles, effluent treatment, systematic review and meta-analysis. In addition, review articles were taken into account as a reference to analyze the most relevant studies.

2.3 Inclusion and exclusion criteria for scientific investigations

Duplicate documents were eliminated for the research. Then the abstracts of the selected articles were reviewed, excluding all the documents that did not meet the criteria for the development of the research. Subsequently, the articles were downloaded for review, and the different inclusion criteria defined were evaluated as explained below:

1. all research showing the reduction of pollutants from treated effluents with the application of MNBs were included. The selection of this criterion was based on the fact that the research should be focused on the environmental field.
2. All the articles selected were those that had a reduction in the parameters studied during treatment, including biochemical oxygen demand (BOD5), biochemical oxygen demand (BOD), pH, electrical conductivity, metals, microbiological pollutants and others.
3. All investigations with insufficient data on operational conditions were excluded.
4. All investigations that considered treatment technologies other than MNBs or bubbles with a diameter greater than 100 µm were excluded.
5. All investigations that did not describe the characteristics of the MNBs used in the effluent treatment were excluded.

2.4 Selection of articles and data extraction

Each potentially eligible article was reviewed to obtain information and data (using validated data collection forms). The following aspects were considered: (a) author(s) and year of publication, (b) types of effluent treated, (c) characteristics of the MBMs, (d) parameters evaluated and (e) reduction of the main pollutants.

2.5 Data meta-analysis

RevMan 5.4.1. software was used for data analysis. Dichotomous data were used and compared with the Odds Ratio (OR) meta-analysis criterion (Bonett & Price 2015; Aquije Morey et al. 2021). Effect estimates were developed with 95% confidence intervals. Heterogeneity of the investigations was assessed through visual analysis of the forest plot, where the Chi2 statistic indicates the differentiation of the results, as well as the compatibility of the results by performing a random selection of data. The p-value determines the heterogeneity of the investigations.

3. Results and discussion

3.1 Search and selection of studies

A total of 193 articles were identified through the search in the databases described above. Applying the different inclusion criteria defined; of the 193 selectable articles, 71 research studies were excluded as reviews, letter, conference paper, data paper and others, 46 articles were excluded for having duplicates in the Scopus and Web of Science databases, 75 research studies were included for the full text review, 66 research studies were excluded for having insufficient data, technologies not applied to the environmental field, presence of additional technology and technology with bubbles greater than 100 µm. A total of 10 articles were registered for the final evaluation.

3.2 Systematic review and meta-analysis of scientific investigations

The results presented are part of an undergraduate thesis (Belahonia Talledo & Garcia Espejo 2021). The research mentions the technique or methodology used in each selected study. In addition, the characteristics assessed in the MNBs are mentioned and the main findings are presented.

3.2.1 Description of the studies

Table 1 presents a summary of the efficiency of MNBs in the removal of the parameters studied in the selected studies. Only the studies that had at least two treatments with the use of MNBs were considered.

Table 1: Efficiency of the MNBs in the removal of the parameters studied in each scientific investigation

	Removal efficiency (%)										
No.	**BOD5**	**COD**	**Turbidity**	**TSS**	**Fe**	**Pb**	**Si**	**TCE**	**Total coliforms**	**Thermotolerant coliforms**	**Authors**
1	94.2	71.6	56.6	63.8	-	-	-	-	98.1	99.2	(Cruz & Valverde Flores 2017)
	95.6	85.8	65.2	79.3	-	-	-	-	98.9	99.6	
	97.1	95.0	77.3	81.3	-	-	-	-	99.9	99.9	
2	-	-	95.0	-	99.0	-	-	-	-	-	(Etchepare et al. 2017)
	-	-	-	-	94.3	-	-	-	-	-	
	-	-	-	-	66.0	-	-	-	-	-	
3	-	-	-	-	-	-	-	99.7	-	-	(Hu & Xia 2018)
	-	-	-	-	-	-	-	99.9	-	-	
	-	-	-	-	-	-	-	99.8	-	-	
	-	-	-	-	-	-	-	99.8	-	-	
	-	-	-	-	-	-	-	99.9	-	-	
4	90.2	92.5	91.1	79.1	-	-	-	-	-	-	(Menendez & Valverde Flores 2017)
	83.5	97.9	92.6	88.2	-	-	-	-	-	-	
5	70.5	68.5	84.1	-	-	-	-	-	99.9	99.9	(Ventura & Valverde Flores 2018)
	68.8	70.0	73.3	-	-	-	-	-	99.9	99.9	
6	-	-	77.4	-	-	95.3	57.9	-	-	-	(Valenzuela & Valverde Flores 2018)
	-	-	55.8	-	-	99.8	80.9	-	-	-	
	-	-	63.6	-	-	98.3	81.7	-	-	-	
7	99.7	97.1	70.4	-	-	-	-	-	-	-	(Nuñez Álvaro & Valverde Flores 2019)
	99.8	99.1	71.8	-	-	-	-	-	-	-	
	99.9	98.5	71.9	-	-	-	-	-	-	-	
8	36.7	64.5	-	30.7	-	-	-	-	-	-	(Rameshkumar et al. 2019)
	88.9	96.7	-	78.9	-	-	-	-	-	-	
	98.1	98.9	-	99.6	-	-	-	-	-	-	
	99.9	97.3	-	99.1	-	-	-	-	-	-	
9	-	-	89.3	-	-	-	-	-	-	-	(Kim et al. 2020)
	-	-	97.8	-	-	-	-	-	-	-	
	-	-	96.6	-	-	-	-	-	-	-	
10	-	-	-	-	33.3	-	-	-	-	-	(Sun et al. 2021)
	-	-	-	-	40.0	-	-	-	-	-	
	-	-	-	-	90.0	-	-	-	-	-	

With respect to the parameters evaluated, nine of the ten investigations measured physical-chemical parameters such as BOD5, COD, turbidity and TSS, while only two evaluated inorganic parameters (iron levels in one of them, and lead and silicon in the other), only one investigation contemplated an organic parameter (trichloroethylene) and two of them measured microbiological parameters (total coliforms and thermotolerant coliforms). The parameters evaluated that had the highest removal efficiency were trichloroethylene (TCE), total coliforms and thermotolerants coliforms, reaching up to 99.9% removal efficiency.

3.2.2 Meta-analysis

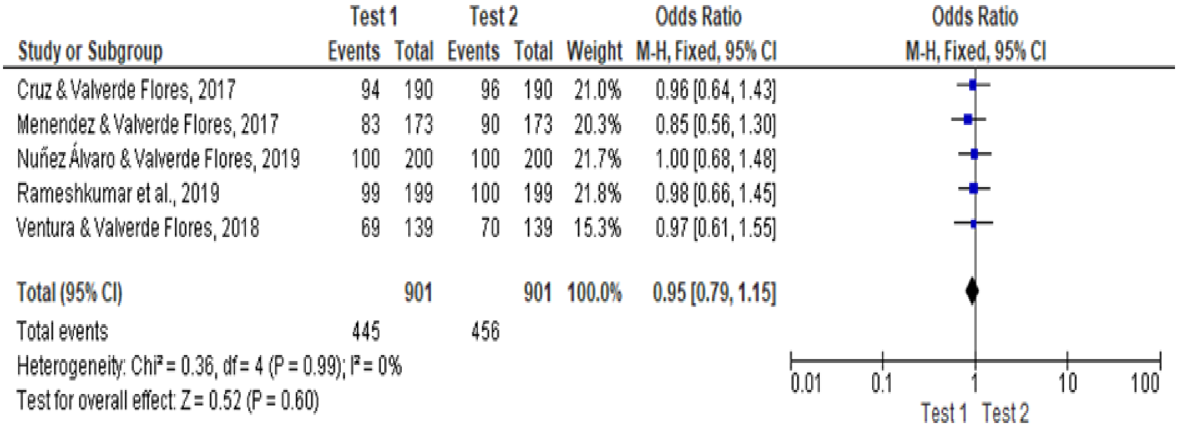
Table 2 and Table 3 show the studies that worked with biochemical oxygen demand (BOD5) and chemical oxygen demand (COD), respectively. All studies show the removal efficiency of BOD and COD considering at least two experimental tests, i.e. for each parameter (BOD and COD) evaluated there were two different values of removal efficiency which could be influenced by the initial concentration of the pollutant.

Table 2: Efficiency of MNBs in BOD5 removal

		BOD5				
No.	**Effluent treated**	**Concentration of sample 1 (mg/L)**	**Removal efficiency (%)**	**Concentration of sample 2 (mg/L)**	**Removal efficiency (%)**	**Authors**
1	Domestic effluent	342	94	342	96	(Cruz & Valverde Flores 2017)
2	Hospital effluents	127	83	132	90	(Menendez & Valverde Flores 2017)
3	Sanguaza (from the fish stalls found in a market)	410	69	474	70	(Ventura & Valverde Flores 2018)
4	Industrial washing effluents	2133	100	2136	100	(Nuñez Álvaro & Valverde Flores 2019)
5	Wastewater from different sources	2620	100	105	99	(Rameshkumar et al. 2019)

From Table 2, it was observed that the investigations of Rameshkumar et al. (2019) and Nuñez Álvaro & Valverde Flores (2019) stand out for having BOD removal efficiency values of 100%. Ventura & Valverde Flores (2018) presented the lowest removal efficiency, only reaching 70%. However, these values continue to be optimal percentages that demonstrate the efficiency of treating contaminated effluents with the nanobubbles. Other research such as Etchepare et al. (2017), combined microbubbles and nanobubbles to obtain a high removal efficiency of Fe+3 contaminant, reaching maximum values of 66% and 99%. Similarly, Sun et al. (2021) used the nanobubbles technique for iron removal, showing results of 79% and 100%. However, in this research it can be highlighted that when the aeration pressure increased, the size of the micro/nanobubbles decreased.

Figure 1 shows the meta-analysis of the five articles included, with respect to the BOD5 pollutant. In each of them, an analysis of the removal efficiency after treatment with MNBs was performed. In the first and second test, slightly approximate removal efficiency values are evidenced.



	Test 1		Test 2			Odds Ratio
Study or Subgroup	Events	Total	Events	Total	Weight	M-H, Fixed, 95% CI
Cruz & Valverde Flores, 2017	94	190	96	190	21.0%	0.96 [0.64, 1.43]
Menendez & Valverde Flores, 2017	83	173	90	173	20.3%	0.85 [0.56, 1.30]
Nuñez Álvaro & Valverde Flores, 2019	100	200	100	200	21.7%	1.00 [0.68, 1.48]
Rameshkumar et al., 2019	99	199	100	199	21.8%	0.98 [0.66, 1.45]
Ventura & Valverde Flores, 2018	69	139	70	139	15.3%	0.97 [0.61, 1.55]
Total (95% CI)		**901**		**901**	**100.0%**	**0.95 [0.79, 1.15]**
Total events	445		456			

Heterogeneity: Chi² = 0.36, df = 4 (P = 0.99); I² = 0%

Test for overall effect: Z = 0.52 (P = 0.60)

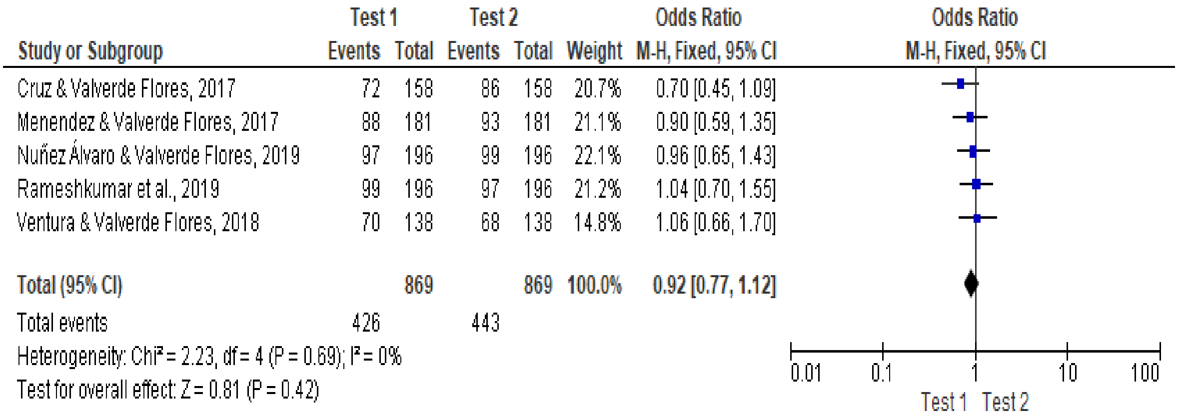
Figure 1: Meta-analysis of BOD5 pollutant removal efficiency after treatment with MNBs

The odds ratio was 0.95, indicating that the treatment with MNBs in the first sample presented a lower removal efficiency of 5%. On the other hand, the scientific articles included did not show statistical heterogeneity (P = 0.99 and I² = 0%), which indicates that the results of tests 1 and 2 show zero variability.

Table 3: Efficiency of MNBs in COD removal

		COD				
No.	**Effluent treated**	**Concentration of sample 1 (mg/L)**	**Removal efficiency (%)**	**Concentration of sample 2 (mg/L)**	**Removal efficiency (%)**	**Authors**
1	Domestic effluent	704	72	704	86	(Cruz & Valverde Flores 2017)
2	Hospital effluents	297	88	374	93	(Menendez & Valverde Flores 2017)
3	Sanguaza (from the fish stalls found in a market)	475	70	503	68	(Ventura & Valverde Flores 2018)
4	Industrial washing effluents	3514	97	3681	99	(Nuñez Álvaro & Valverde Flores 2019)
5	Wastewater from different sources	399	99	4520	97	(Rameshkumar et al. 2019)

Table 3 also showed Rameshkumar et al. (2019) and Nuñez Álvaro & Valverde Flores (2019) as the authors who achieved between 97% and 99% COD removal. In the research of Hu & Xia (2018) treated trichloroethylene (TCE) pollutant in groundwater using ozone micro/nanobubbles for its removal, which withstood highly saline conditions, and showed removal values of 97% and 100% in the samples worked Figure 2 shows the meta-analysis of the five articles included, with respect to the COD pollutant. The analysis of the removal efficiency after treatment with MNBs shows that both treatments present approximate removal values, indicating that there is an influence of the concentration of the initial sample.



Study or Subgroup	Test 1 Events	Test 1 Total	Test 2 Events	Test 2 Total	Weight	Odds Ratio M-H, Fixed, 95% CI
Cruz & Valverde Flores, 2017	72	158	86	158	20.7%	0.70 [0.45, 1.09]
Menendez & Valverde Flores, 2017	88	181	93	181	21.1%	0.90 [0.59, 1.35]
Nuñez Álvaro & Valverde Flores, 2019	97	196	99	196	22.1%	0.96 [0.65, 1.43]
Rameshkumar et al., 2019	99	196	97	196	21.2%	1.04 [0.70, 1.55]
Ventura & Valverde Flores, 2018	70	138	68	138	14.8%	1.06 [0.66, 1.70]
Total (95% CI)		**869**		**869**	**100.0%**	**0.92 [0.77, 1.12]**
Total events	426		443			

Heterogeneity: Chi² = 2.23, df = 4 (P = 0.69); I² = 0%

Test for overall effect: Z = 0.81 (P = 0.42)

Figure 2: Meta-analysis of COD pollutant removal efficiency after treatment with MNBs

The odds ratio was 0.92, indicating that the treatment with MNBs in the first sample presented a lower removal efficiency of 8%. On the other hand, the scientific articles included also showed no evidence of statistical heterogeneity (P = 0.69 and I² = 0%), indicating zero variability between the two tests

4. Conclusions

The ten articles selected through the systematic review and subsequent meta-analysis indicated that the treatment of polluted effluents with the use of MNBs is efficient for organic matter indicator pollutants (BOD5 and COD). The pollutant removal efficiency depends on the size of the MNBs used and the characteristics of the effluent to be treated, among the most relevant results:

1. It was demonstrated that MNBs in the treatment of polluted effluents is efficient in all the selected investigations, reaching BOD5 and COD removal values from 68% to 100%.
2. The treatment showed higher removal efficiency in inorganic and microbiological pollutants such as trichloroethylene (TCE), total coliforms and thermotolerant coliforms. The MNBs reduced the concentration of these pollutants in the effluents up to 99.9%.
3. The diameter of the MNBs used in the treatments ranged from 0.01 µm to 70 µm.

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