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Trends, new insights and perspectives in the treatment of hospital effluents
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Abstract
Recently, investigations of hospital effluent management and treatment have not only interested research groups with acquired experience in the field, but have also attracted the interest of new groups over the world. The most recent literature provides new insights into the occurrence of pharmaceuticals and other contaminants of emerging concern, pathogens, viruses, and antibiotic-resistant bacteria and genes in hospital effluent in various new developing and developed countries. It also provides information on the effective removal of key compounds (mainly antibiotics, analgesics, beta-blockers and chemotherapy drugs) by means of enhanced biological treatments and advanced oxidation processes. The current debate among the scientific community is mainly about the proper treatment to reduce the spread of antibiotic-resistant bacteria and genes and about the feasibility (from a technical and economic point of view) of treatment trains tested at lab and pilot scales.

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Introduction
The management and treatment of hospital effluent are issues of current concern in an increasing number of countries worldwide. At the same time, the increasingly widespread awareness that the effluent produced in healthcare structures may contain conventional pollutants and micropollutants — because of the different activities occurring within the structure (diagnosis, laboratories, clinics, surgeries, medications and presence of patients in different types of wards) — has led to discussion on how to improve the management and enhance the treatment of such effluent. A snapshot of the current situation is provided in a recently published collection by Verlicchi [1], which contains worldwide experiences of the management and treatment of hospital effluent. Nevertheless, despite the shared recommendations that hospital effluent must be treated (in a dedicated plant or together with urban wastewater) [2], in some countries, it is still directly discharged into rivers or the sea without any treatment. This is the case in Nepal, Bangladesh, Pakistan, Indonesia, India and Colombia [3–5]. Chemical substances, including residues of pharmaceuticals excreted within the hospitals, mainly by in-patients, have been the subject of recent monitoring investigations both in developed (among them the studies by Niemi et al. [6], Sörenård et al. [7], Kosma et al. [8], Azuma et al. [9] and Lasek et al. [10]) and developing countries (among them the studies by Khan et al. [11], Wielens Becker et al. [12] and Al-Qaim et al. [13]). However, increasing attention has recently been paid to the occurrence of (i) antibiotic-resistant bacteria (ARB) and antibiotic-resistant genes (ARGs) present in raw and treated hospital effluent, (ii) pathogens, and (iii) viruses [14], including coronavirus in hospital effluent after the recent public health emergency [15–17]. There is an ongoing debate about the efficiency of the well-known treatment technologies that have been adopted and evaluations are underway of the need for further and/or dedicated steps regarding disinfection to control or remove viral contamination from domestic and hospital wastewater [17].

The current survey aims to highlight these issues, analyse the main results achieved and identify any gaps in current knowledge.

Scope of the selected recently published investigations
A great number of the investigations included in this survey deals with the occurrence of a selection of conventional contaminants (the so-called macropollutants) and contaminants of emerging concern (CECs) (mainly pharmaceuticals) in hospital effluent, including ARGs and ARB (among them [18,19]). More recently, the
efficacy of pathogen and virus removal was at the centre of many debates and studies, and efforts were made to evaluate whether (further) measures should be adopted, and if so which, to reduce the risk of environmental contamination and to guarantee a higher level of hygienisation in the hospital effluent treatment [17,20]. Many studies aimed to improve the removal of key CECs by means of enhanced biological treatments or by combining biological, chemical and physical technologies.

Some investigations aimed to better understand the removal mechanisms of key compounds and their interactions with the treatment environment. This is the case of the investigations by Vo et al. [21] who tried to correlate the concentrations of acetaminophen and peroxidase enzymes in a pilot vertical subsurface flow system fed with hospital effluent to evaluate if the selected enzymes could be assumed as indicators of the occurrence of acetaminophen, thus reducing its monitoring costs.

**Perspectives in the treatment of hospital effluent**

Over the last two to three years, interesting investigations on the treatment of hospital effluent have been carried out worldwide: consolidated and modified/enhanced technologies as well as new solutions have been tested on the effluent of specific wards, the whole healthcare structure, nursing homes specialised in psychiatric diseases [22,23] or service units such as laundry [24]. They have not only taken place in countries where studies or investigations on hospital effluent have also been carried out, but also in new ones, such as Ethiopia [25], Columbia [5], India [26], Iran [27,28], Costa Rica [29] and Thailand [21]. This demonstrates the increasing interest in the management and treatment of healthcare structure effluent by researchers, practitioners, policy makers and administrative staff, both in developed and developing countries.

Many studies have tested the efficacy of removing key compounds of combined biological processes, consisting of membrane bioreactors (MBRs) coupled with advanced oxidation processes or membrane technologies. Paulus et al. [30] noted the effective removal of a selection of antibiotics and ARGs achieved by applying MBRs (equipped with microfiltration membranes) + O3 + granular activated carbon + UV to the hospital effluent of Harlev in Denmark.

The ozonation of the effluent of a nursing home specialised in psychiatric diseases in Germany was tested by Mousel and Pinnekamp [22]. The effluent was initially pretreated by an MBR equipped with ultrafiltration membranes. It was found that the best results were achieved by applying 5 mg O3/L (corresponding to 0.5 g O3/(g dissolved organic carbon DOC) with a contact time of 12.8 min. The poor removal of DOC during ozonation showed that any remaining recalcitrant compounds were not completely removed or mineralised and could produce transformation products requiring further investigations, as suggested by Bourgin et al. [31].

Interesting results have been achieved in the removal of a selection of antibiotics with the system tested by Vo et al. [32] consisting of an enhanced MBR (equipped with hallow fibre membranes) coupled with ozonation. The MBR system is that originally proposed by Ngo et al. [33] and then investigated by Nguyen et al. [34] for the effluent of Trung Vuong Hospital in Vietnam. Its novelty consists of the presence of polyester-urethane sponge media with high porosity in its aerobic compartment to promote micropollutant removal and at the same time reduce membrane fouling, which is still the most critical operational factor for this biological system. It operated with a biomass concentration of around 5000 mg/L, a ratio between volatile and total suspended solids of 0.79, and a sludge retention time of 20 days.

Some interesting investigations have dealt with enhanced biological processes based on the combination of MBRs coupled with granular activated carbon or powder activated carbon (PAC), or characterised by modified membranes, or operating under specific and controlled conditions. The Spanish group at the University of Santiago of Compostela [35] applied their patented system SEMPAC© to hospital effluent and evaluated its performance with regard to five CECs: ibuprofen, 17α-ethynylestradiol, diclofenac, carbamazepine, and trimethoprim. It consists of a sequential batch reactor followed by an external submerged microfiltration membrane tank, where PAC is added to enhance the sorption of residues of CECs. They found that the addition of PAC improves the removal of all the compounds, including the most recalcitrant carbamazepine and trimethoprim. The results also showed that it is important to identify when PAC must be added because of powder saturation to guarantee a constant removal of all the key compounds (every 20 days in their investigation).

Promising results have been obtained with moving-bed biofilm reactors (MBBRs) for the removal of specific pollutants such as anionic detergents as in the study by Shokoohi et al. [36] regarding effluent from an Iranian hospital (removal achieved around 92% of linear alkil-benzene sulfonates (LAS) with a filling rate of 70% of Kaldness carriers, a biomass concentration of 3000 mg/L and hydraulic retention time of 24 h). The MBBR coupled with ozonation was investigated by Tang et al. [37] in Denmark, and it emerged that an increment in the removal of recalcitrant compounds (propranolol,
Tramadol and trimethoprim) was due to ozonation. A following MBBR step showed that it was able to guarantee the removal of the residual toxicity of the ozonated effluent.

With regard to the beta-blocker metoprolol and its metabolite metoprolol acid, the study by Jaén-Gil et al. [38] investigated their degradation transformation and sorption in a fungal fluidised batch reactor fed with real hospital effluent. Different fungi were tested. The best results were achieved with Ganoderma lucidum treatment: after 15 days of treatment, the achieved removal was around 51% for metoprolol and 77% for its metabolite.

The combination of a conventional biological step with sonochemical treatment (375 kHz and 88 WL) was investigated by Serna-Galvis et al. [39] in Colombia to evaluate the removal of fifteen pharmaceuticals in real hospital effluent. They found that the chemical (radical attacks) and physical (suspended solids disaggregation) effects due to the sonochemical step improved the removal of the recalcitrant compounds showing a pondered average removal of around 59%. The reduction of the pharmaceutical load increased up to 83% when Fe$^{2+}$ (5 ppm) and UVC light (4 W) were added to the sonochemical system, thus generating a sono–photo–Fenton process. The study showed the promising results that can be achieved by these treatment trains but also the high energy costs: if future research is able to reduce them, this solution could become economically feasible.

Electrochemical technologies have also been tested for the degradation and mineralisation of specific compounds occurring in hospital effluent. In this context, Moreno et al. [40] investigated the degradation of the chemotherapy drug doxorubicin (which could reach concentrations as high as 1 μg/L) by nanostructured graphite electrodes with metallic oxides (graphite, TiO$_2$@graphite and AuO–TiO$_2$@graphite electrodes). The lab-scale experiments showed complete degradation with the AuO–TiO$_2$@graphite electrode, and no undesired compound was formed. As to ecotoxicological effects, the tested treatment did not cause any effect on embryo-larval development of zebrafish but DNA damage was observed after 96 h’ exposure.

**Threats and challenges in hospital effluent management and treatment**

Antimicrobials are commonly found in wastewater, particularly in hospital effluent, and are often still present in treated effluents, exerting continuous pressure on ARB [41]. This is something that worries the scientific community and has resulted in the World Health Organization including some ARB in the critically important priority list of pathogens for which new antibiotics are necessary [42]. Some of the selected ARB are *Acinetobacter baumannii*, carbapenem-resistant; *Pseudomonas aeruginosa*, carbapenem-resistant; *Enterobacteriaceae*, carbapenem-resistant; third generation cephalosporin-resistant (with priority one) and *Enterococcus faecium*, vancomycin-resistant (with priority two).

Many investigations have been carried out in recent years ([118,43–46]) demonstrating the increasing efforts made by research groups in this field and the increasing amount of data collected on their occurrence. The cited investigations concluded that the resistance to antibiotics is generally higher in hospital wastewater rather than in domestic wastewater, and if hospital effluent is not properly disinfected it may strongly contribute to the spread of ARB in the environment. An efficient disinfection step is thus necessary, and future research should provide new insights and recommendations in this direction.

Another hot topic of increasing interest is the removal of pathogens and viruses from healthcare structure effluent [17,47,48]. The recent public health emergency caused by the coronavirus highlights this aim, and many studies have been carried out in recent months with the first results already being published. According to the investigation by Zhang et al. [47] carried out on hospital effluent that was pretreated in a simple septic tank, the chemical disinfection of raw hospital effluent by means of high doses of sodium hypochlorite (up to 800 g/m$^3$) is not sufficient to remove SARS-CoV-2 viral RNA. The authors highlight that the current recommended doses by the World Health Organization (free chlorine ≥0.5 mg/L for at least 30 min) and China Centers for Disease Control and Prevention (free chlorine above 6.5 mg/L after 1.5-h contact) are not sufficient to reduce the content of the virus in hospital effluent. Overdoses of the disinfectant seem to be able to remove the virus, but they lead to significant concentrations of disinfectant byproducts.

Suggestions provided by Wang et al. [15], Barcelo [16] and Foladori et al. [17] include the importance of the proper management of waste and wastewater within a hospital, which becomes absolutely necessary during a public health emergency.

**Conclusive considerations**

The debate on the most suitable treatment of hospital effluent is ongoing and constantly fed by new insights. It allows identifying the fields that require urgent research and recommendations for a sustainable hospital effluent management and treatment. The coronavirus disease 2019 pandemic has highlighted the need for further investigations into how to reduce the risk of spreading coronaviruses and, more generally, that the reduction of ARGs, ARB and pathogens should be among priority research areas to preserve and protect the water environment, improve sanitation, and guarantee clean and
safe water for a wide range of activities, in accordance with the United Nations Sustainable Development Goals (https://www.unsd.org/content/undp/en/home/sustainable-development-goals.html).

Declaration of competing interest
The author declares that she has no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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