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Comparative Life-Cycle Cost Analysis of Alternative Technologies for the Removal of Emerging Contaminants from Urban Wastewater

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Abstract: Emerging contaminants (ECs) continue to threaten our fragile ecosystem, yet their mitigation remains limited by economic factors. Meanwhile, a relatively expensive material, Graphene Oxide (GO), has shown promise as a solution for EC removal following further development into three graphene-based materials (GBMs): Porous graphene adsorbent (PGa), Graphene-oxide foam adsorbent (GOFa), and the hybrid filter. Due to the nuances of each synthesis process, financial costs will differ throughout the GBMs' life cycle which have been quantified and compared in the present work at a range of possible breakthrough times. Finally, economic and environmental costs have been combined for each technology to compare eco-efficiency. Results demonstrated a substantial economic advantage of the GBMs when compared to alternative technologies, most notably the GOFa filter that incurred the lowest life-cycle costs at $1.73 \pm 0.09/m^3$. This was mainly attributed to the lower demand of GOFa on the most expensive material required for material synthesis, hydrazine. In addition, the material demands of GOFa were more evenly distributed which suggest a higher resilience of the overall costs to price hikes of individual materials required for synthesis. In terms of eco-efficiency the GOFa filter also demonstrated the greatest improvement when compared to the reference technology These results have provided robust total investment costs for several technologies that can now offer contrast to other EC-removal solutions.

Keywords: life cycle; sustainable development; emerging contaminants; economic aspects

1. Introduction

The increasing prevalence of emerging contaminants (ECs) in both rural and urban water systems continues to highlight the shortcomings in our current water management strategies [1]. While a range of technologies have demonstrated the capacity to remove ECs from the water stream [2–5], trade-offs present between each of the elements within the environmental impact–economic cost–pollution discharge nexus of wastewater management are yet to be overcome [6]. As such, continued efforts remain necessitated to pursue an optimal solution that is high-performing, while incurring minimal environmental and economic costs.

One method that offers promise is adsorption due to its high efficiency, simple operating design and low-cost implementation [7–9]. While having demonstrated considerably less environmental burden than alternative technologies [10], adsorption is also often shown to demonstrate improved economy in comparison [11–13]. For the purpose of desalination, Thu et al. [11] found adsorption incurred only half the expense of reverse osmosis (RO) across its life cycle, while Ng et al. [12] observed similar financial gains when adsorption was compared with RO in two different configurations. Adsorption has

also shown economic advantage over membrane technology during carbon dioxide (CO_2) removal in natural gas processing, demonstrating annual savings of up to 25% [13].

Due to its simplicity, adsorption can be achieved by many different materials which has driven focus towards finding more cost-effective sorbents such as activated and enhanced clay [14–17], waste fish scale [18], recycled powder coke [19], desiccated coconut waste [20], bio-chars and hydro-chars [21,22], and microalgae [23]. Despite many of these solutions suggesting better economy than more expensive, purpose-made alternatives [9,24], this will only hold true if performance is not compromised. This trade-off between cost and performance is a common theme in manufacturing [25], and in this case the theoretical costs that environmental degradation could incur from lesser treatment performance may nullify any economic gains made, although these are hard to quantify [26]. As such, efforts to minimize costs in known high-performing sorbents should not be neglected in search of low-cost alternatives that are rarely uncompromising [27].

One sorbent that is gaining traction in the field of EC adsorption is graphene oxide (GO), due to its favourable characteristics [7]. While GO offers high water permeance and stability [28–30], it is arguably its high theoretical surface area that offers the greatest advantage due to the enhanced adsorption capacity it affords [31,32]. The superior adsorption capacity of this carbon structure compared to more conventional adsorbent materials has already been demonstrated across a range of pollutant types including ECs [33–39]. However despite its strong removal performance, GO remains one of the more expensive sorbent options and efforts to improve its cost efficiency are warranted [40,41].

Due to the versatility of GO, recent work has investigated multiple graphene-based materials (GBMs) and the different formats that they can be presented in for the application of EC removal [31,32,42]. Each format entails different synthesis protocols and will therefore incur different costs. When considered across the technology's full-service life, these differences may be substantial, with a more cost-efficient format identifiable. Due to their novelty, no work has yet investigated the total financial outlay that would be required throughout the service life to provide EC removal by these technologies. It is therefore the aim of this work to quantify and compare the life cycle costs of the three GBM filter formats by way of a life cycle cost analysis (LCCA) for the first time. In order to provide a more comprehensive assessment of the relative economic viability of these filters, several further considerations were made.

First, despite its usefulness in economic optioneering of wastewater treatment processes [43–45], there is little evidence of the LCCA's application in investigating adsorption materials in a pollutant removal role. This may be due to the lack of comparability of results for adsorbents when considering different contaminants, as the adsorption capacity is known to differ substantially between them [46]. This will in turn affect the breakthrough time that has been shown to heavily govern the life cycle costs, as this will largely dictate the amount of material synthesis and maintenance (backwash/desorption) materials required [10]. Other factors may also influence breakthrough time such as the source of water or efficacy of the proceeding treatment [47,48]. It is therefore prudent with sorbents to perform life cycle analyses across a range of breakthrough times to provide a more robust set of results that allow comparability beyond the absorbates or water sources under investigation.

The second consideration is given to the relativity of the GBM filter costs to those incurred by other treatment types. For instance, while the GBM adsorbents rely on physiochemical filtration for EC removal, this generates hazardous waste (desorption material/removed ECs/spent adsorbents) that will need to be disposed of and therefore incur further expenditure. In contrast, an alternative treatment type is the advanced oxidation process by pulsed power treatment (AOP-PPT) that generates free radicals to degrade the ECs as well as other contaminants while producing no waste [49]. While each technology will be expected to demonstrate considerable cost disparity at different stages of the life cycle, this analysis will determine the total investment required for each for a more consoli-

dated comparison. This will better inform future decision-makers tasked with removing ECs from the wastewater stream within set budgets.

Finally, consideration is given to the relationship between the economics of the investigated technologies and their environmental costs, which can often conflict [2,50,51]. LCCAs have shown an increasing integration with life cycle impact assessments (LCIAs) over the last 5 years to better incorporate the environmental costs that are also incurred and provide a more holistic perspective of a product's sustainability [44,52,53]. In fact, Ilyas et al. [44] identified almost half of 83 LCCAs pertaining to wastewater treatment to be in support of LCIAs. A common approach is the use of the eco-efficiency index (EEI) that combines both economic and environmental costs to provide a more inclusive proxy of overall sustainability [2,50,51,54]. For example, Canaj et al. [54] used an EEI to compare the sustainability of integrating tertiary reuse water into irrigation with 100% groundwater use. While the reuse of water reduced environmental impact, it would require greater expenditure and reduce crop yield. Using the combined eco-efficiency approach, they showed that the integrated water reuse system would be the most sustainable option, offering an overall eco-efficiency improvement of 12.6% in comparison. By adopting this approach it is therefore possible to gain a greater insight into the relative sustainability of each GBM format and the reference technology.

This work therefore aims to provide not only the first economic assessment of EC removal by alternative GBM adsorption formats, but also the first assessment of their relative sustainability in terms of eco-efficiency and in relation to an energy-centric alternative. In addition, the most influential aspects of the life cycle for each GBM filter type are identified by way of sensitivity analysis while the model uncertainty is also quantified for each technology.

2. Materials and Methods

2.1. Goal and Scope Descriptions

The goal of the present study was to identify and compare the economic costs of the four novel EC removal technologies, before incorporating them with environmental impact scores to evaluate and compare the eco-efficiency of each. At the time of investigation, the breakthrough time of the GBM filters was not yet well understood when treating ECs in tertiary municipal wastewater. For this reason and for comparability with other water or absorbent types, the life cycle costing of the GBM filters was performed at a range of different breakthrough times in line with the previous LCIA [10]. This was necessary as the work mentioned highlighted the significance of breakthrough time on resource use which is likely to strongly influence the LCCA. Cost inventories for each technology can be found in Tables S1–S8 of the Supplementary Materials.

With regards to the scope of the LCCA, Figure 1 displays the costs and system boundary considered in the analysis. These include consideration of the materials required for the initial and ongoing GBM synthesis, process energy in production, other structural materials, operational energy, maintenance energy and materials (for backwash/desorption), and hazardous waste disposal. Filter material containers are doubled to account for reuse following refill. Due to the novelty of these materials, GBM synthesis was costed in respect to laboratory-scale batch sizes (0.1–0.503 kg). A service life of 15 years for each technology is considered, each operating at a treatment capacity of 1 m^3 per day.
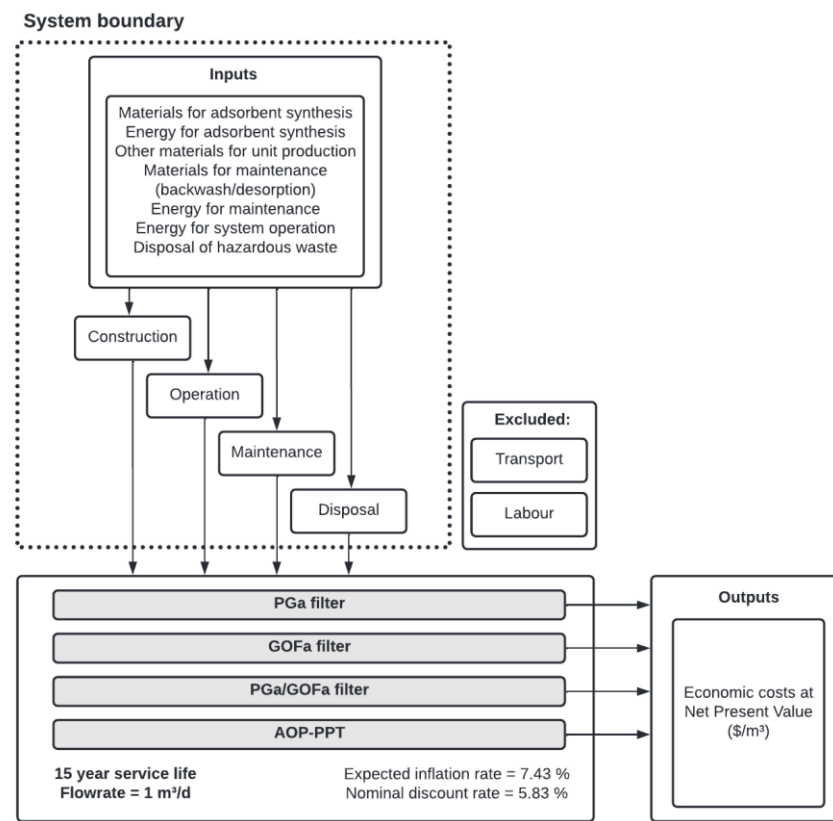
Figure 1. System boundary of the comparative LCCA.

2.2. Life Cycle Cost Model Development

LCC values were calculated using Equation (1). This is based on a model used by Younis et al. [55] to compare the economic performance of structural concrete material scenarios. For both the costs of operation and maintenance, $C_{O\&M}$, and disposal, C_D, the net present value (NPV) is calculated to incorporate the influence of inflation and interest rates on future worth into present value [55,56]. Parameters are included as necessary, i.e., C_D would only be considered in analyses that consider the final year.

$$\text{Life cycle cost, LCC} \left(\$/m^3\right) = C_{CAP} + \left[\sum_{t=0}^{T} \frac{C_{O\&M}}{(1+r)^t}\right] + \left[\sum_{t=0}^{T} \frac{C_D}{(1+r)^T}\right] \quad (1)$$

To consider the individual components, the capital costs, C_{CAP}, are considered as represented by Equation (2):

$$C_{CAP}\left(\$/m^3\right) = C_{SyMi} + C_{SyEi} + C_{StMi} \quad (2)$$

where the C_{CAP} includes the costs of materials for the initial adsorbent synthesis, C_{SyMi}, as well as the initial structure materials ($\$/m^3$), C_{StMi}, that considers other filter materials ($\$/m^3$) and the cost of process energy required for the initial adsorbent synthesis, C_{SyE} ($\$/m^3$).

Equation (3) represents the annual operation and maintenance costs, $C_{O\&M}$:

$$C_{O\&M}\left(\$/m^3\right) = C_{SyMo} + C_{SyEo} + C_{StMo} + C_{Mm} + C_{Eo} + C_{Em} \quad (3)$$

This equation accounts for the costs of materials for ongoing adsorbent synthesis, C_{SyMo} ($\$/m^3$), the cost of process energy used in the ongoing material synthesis, C_{SyEo} ($\$/m^3$), the costs of ongoing structure materials, C_{StMo} ($\$/m^3$), the cost of mainte-

nance materials such as for backwashing/desorption and regeneration, C_{Mm} ($/m^3), the cost of energy required for operation, C_{Eo} ($/m^3), and the cost of energy required for maintenance, C_{Em} ($/m^3).

The real discount rate, r, is calculated according to Equation (4) [56]:

$$\text{Real discount rate, } r = \frac{i - d}{1 + d} \tag{4}$$

where the expected inflation and nominal interest (discount) rates are represented by i and d, respectively. In the current work, i was provided as 7.43% based on an average consumer price index (CPI) inflation rate between years 1960–2020 [57]. A nominal interest rate of 5.83% was applied based on an average from years 1978–2020 [58].

In addition, the operational longevity of the tertiary technologies is considered as 15 years and represented by T, while t indicates each year of the service life.

Finally the disposal costs, C_D, are considered to account for the hazardous waste that is generated through EC adsorption and calculated as demonstrated in Equation (5):

$$\text{Disposal costs, } C_D(\$/m^3) = C_{HW} \tag{5}$$

where hazardous waste for disposal is considered, C_{HW} ($/m^3). Non-hazardous waste was considered to be negligible for the present study, based on the use of only two filter columns (recyclable stainless steel) per life cycle. This aspect will need to be better represented in commercial scaling considerations.

2.3. Eco-Efficiency Assessment

The eco-efficiency assessment (EEA) is based on a binomial impact-cost assessment of any given product and can be used as a means to select a more favourable technology from alternatives in terms of economic and environmental performance [2]. While the analysis would allow for the weighting of these factors to be distributed disproportionally, equal weighting has been given to each aspect in the present study and is expressed as follows [2,59].

$$EEI = EEF_E + EEF_C; \ 0 \ \leq \ EEI \ \leq \ 1 \tag{6}$$

where EEF_E is the impact aspect of EEI, while EEF_C accounts for the economic consideration. These can be determined as according to Equations (7) and (8), where the relative weighting of each factor can also be assigned:

$$EEF_E = \frac{LCIA_{REL}}{2} \tag{7}$$

$$EEF_C = \frac{LCC_{REL}}{2} \tag{8}$$

Both $LCIA_{REL}$ and LCC_{REL} are expressions of their respective impact and cost efficiencies relative to the least efficient alternative. These are expressed as follows:

$$LCIA_{REL} = \frac{LCIA}{LCIA_{MAX}} \tag{9}$$

$$LCC_{REL} = \frac{LCC}{LCC_{MAX}} \tag{10}$$

where LCIA is the environmental impact score across the technologies' life cycle, measured as Pt/m^3 treated wastewater in the present study. $LCIA_{MAX}$ is therefore the alternative technology that derives the highest impact score. LCC represents the economic costs incurred across the technologies' life cycle measured as $/m^3 of treated wastewater and, consequently, LCC_{MAX} is the highest reported life cycle cost of the compared technologies.

Once the EEI score is calculated, a final step is taken in the EEA to provide the results in a more intuitive format as a percentage increase of eco-efficiency relative to the lowest performing technology. This is achieved by way of the following equation:

$$\text{EEI } (\%) = (1 - \text{EEI})100 \qquad (11)$$

Calculation of the EEI for each material can be found in Table S9 of the Supplementary Materials, while the environmental impact scores are provided by previous work [10].

2.4. Life Cycle Impact Assessment

In order to calculate the EEI indices for each of the four technologies, the life cycle impacts incurred by each was required. This information was taken from our previous work where full details can be found [10]. In short, a life cycle inventory (LCI) was first developed for each technology to derive associated environmental impacts from the Ecoinvent database version 3.4. The IMPACT 2002+ method was then used to first categorise each impact into 14 midpoint categories including human toxicity, respiratory effects, ionizing radiation, ozone layer depletion, photochemical oxidation, aquatic ecotoxicity, terrestrial ecotoxicity, aquatic acidification, aquatic eutrophication, terrestrial acid/nutr, land occupation, global warming, non-renewable energy and mineral extraction [60]. The relative scores in each midpoint category were then fed into four endpoint categories including human health, ecosystem quality, climate change and resources [60]. Scores were given in points (pt) values as the normalized unit of measure that represents the average impact caused by a person during one year in Europe for all categories except human health, which is instead given as the average impact on a person in one year [60]. No changes were made to the LCIA data for the present study.

2.5. Sensitivity Analysis

Sensitivity analyses provide a way to investigate the relative influence of model parameters on output [61]. In this way efforts can be allocated accordingly in determining each of the parameters and importance can be assigned to parameters that are based on assumption. In the present work, a local sensitivity analysis (LSA) was used for this purpose. The LSA works by deviating each investigated model parameter one at a time by a standard set amount. In this work a deviation of 10% was applied. Arguments have been made against the use of LSA such as its limited investigation of the parameter search space, no detection of interactions between parameters and lack of self-verification [62]. However, these concerns are less warranted in the present model due to the linear relationship of input parameters and model outputs with little interaction expected between the small number of parameters [62]. In fact, in this case the LSA is found to be preferable due to its ease of implementation and interpretation, and reduced computational demand [63].

The sensitivity coefficients, S_C, are calculated according to the following formula:

$$S_C = \left[\frac{O_2 - O_1}{P_2 - P_1} \right] \frac{P_d}{O_d} \qquad (12)$$

where P_d is the default input parameter before deviation and P_1 and P_2 are the -10% and $+10\%$ derivative values, respectively. O_1, O_2 and O_d are the respective model outputs for each P_{value}.

2.6. Uncertainty Analysis

An uncertainty analysis is often performed to estimate the reliability of results and thus install confidence in a model [64]. While it can be used to add validity to a model, the uncertainty analysis can also be used to ensure that there is little uncertainty in the parameters that may jeopardise the effectiveness of the LSA [62].

For each of the investigated parameters, a uniform probability distribution was assumed. This was because there was no reason to believe that there was a bias towards

a central value or the range limits for material/energy costs, while also being the most conservative distribution [65]. In this case the standard deviation for each of the input parameters, σ, with a rectangular (uniform) distribution is calculated as follows [65]:

$$\text{Standard deviation, } \sigma = \frac{a}{\sqrt{3}} \tag{13}$$

where a is the distribution boundary, i.e., true parameter values must fall within ±a and without bias in the case of the uniform distribution. Range limits were considered as ±10% as this was enough to envelop all prices found during the costing exercise. No uncertainty was considered for the service life due to being a design parameter.

Values were then taken at random from the distribution of each parameter providing 10,000 sets of values. Each value set was processed through the model and the output recorded. The standard uncertainty, U_C, could then be calculated as the standard deviation of all 10,000 model outputs produced by way of the STDEV.P function in Microsoft Excel$^{\text{TM}}$. Finally, U_C was multiplied by a coverage factor, k, of 1.96 to provide the expanded uncertainty of 95% confidence which is ultimately reported [65]. Results of the uncertainty analysis can be found in the Supplementary Material (Figures S1–S4).

3. Results and Discussion

3.1. Life Cycle Costs

At an assumed breakthrough time of 5 days, the PGa filter was observed to cost $2.97 ± 0.15/m^3 as shown in Figure 2. The GOFa filter incurred the lowest cost of $1.73 ± 0.09/m^3, while the hybrid filter incurred a cost of $2.12 ± 0.11/m^3. The elevated costs incurred during material synthesis of the PGa can be attributed to the greater demand of hydrazine (0.588 kg) required per replacement event compared to 0.126 kg and 0.319 kg for the GOFa and hybrid adsorbent, respectively. Hydrazine was found to account for 63.7% of the total material synthesis costs for PGa per replacement at $103.07 and 56.5% for the GOFa at $22.09. The next greatest material costs during synthesis were $13.22 for the PGa which was substantially less, while for the GOFa filter the next greatest cost was $5.53. The higher asymmetry observed between hydrazine and the other materials for the PGa compared to GOFa highlights the added vulnerability of the PGa filter to possible price hikes of this single material. Hydrazine is a notoriously expensive material to produce [66] and, as Leader et al. [67] showed, technologies can see substantial price increases when underlying material costs rise. Conversely, material costs of the GOFa filter are more broadly distributed and would therefore be less impacted by single material cost rises. This suggests a greater resilience of the GOFa filter to the PGa filter in terms of supply and demand imbalances as well as the reduced costs of production.

In contrast to the GBM filters, Figure 2 also shows that the AOP-PPT incurred a significantly higher cost ($25.02 ± 1.72/m^3), due primarily to its high demand on energy supply. While it is not so prone to material price hikes, its heavy reliance on energy for effective operation is arguably more at risk. Electricity prices are influenced by many factors including privatization of electricity companies [68], regulation and liberalization [69,70], renewable energy source adoption [71,72], clean energy initiatives [73], fossil fuel price fluctuation [74], market integration and competition [75], commodity price imbalances [76], and geopolitics [77–80], to name a few.

Besides the environmental benefits that increased uptake of renewable energy affords [81], reduced costs are also achievable that may favour energy-centric technologies like the AOP-PPT technology over adsorption [82,83]. While the economic gains of renewable uptake are rarely realised in the short term [66], substantial cost reductions are achievable [72]. Such gains are facilitated by the development of increasingly efficient renewable technology and cost degression [83–85] as well as macroeconomic conditions and experience effects [86]. Child et al. [87] suggested that levelized electricity costs could be reduced by as much as 26% in Europe by 2050 following a full switch to renewable energy. However, despite this, in order to achieve economic advantage we calculated that

electricity costs would need to be reduced ~13 times for the AOP-PPT to offer competitive pricing to the hybrid filter at a 5-day breakthrough and ~3.5 times if a breakthrough time of 1 day is considered.
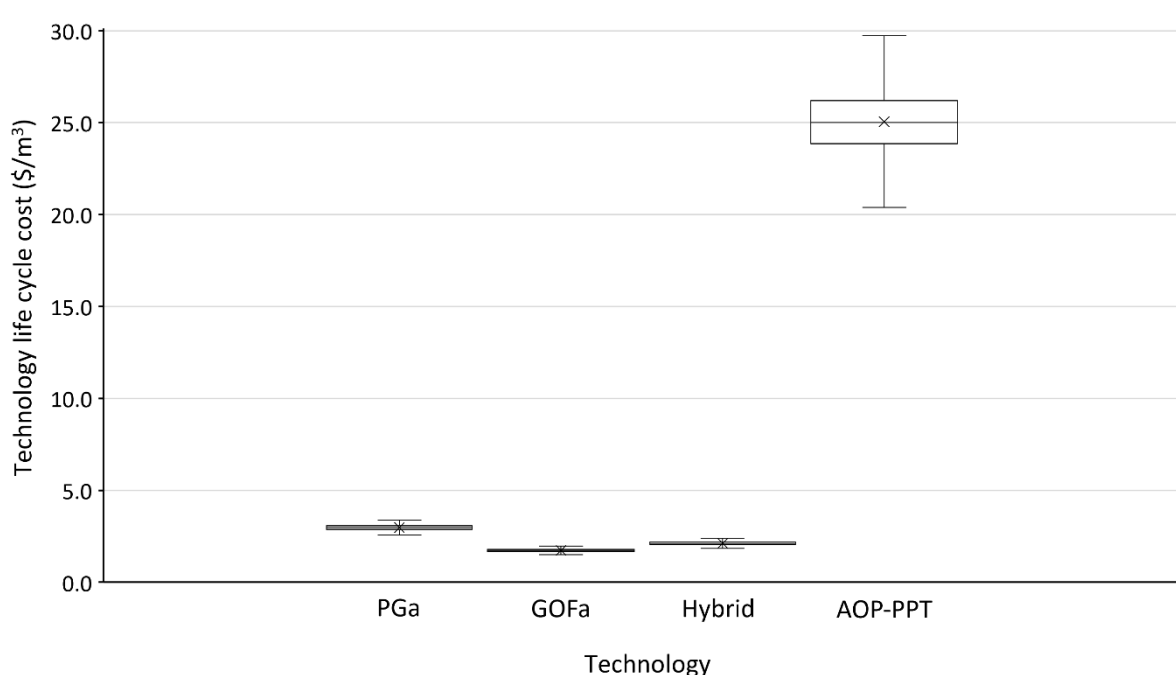

Figure 2. Life cycle costs ($/m^3) for each of the technologies with limits of uncertainty.

The results of this work are in contrast to results reported by Tarpani and Azapagic [88] who investigated the life cycle costs of several advanced wastewater treatment types. In their study, ozonation was found to be the most affordable option when compared to another adsorbent, granular activated carbon (GAC). Despite the GBM filters incurring greater costs per kg (PGa = $799/kg, GOFa = $70/kg) compared to the $1.83/kg assumed for GAC in their study [88], it is likely that the GBM adsorbents would offer a significantly higher breakthrough time for the studied pollutants than the 11–33 days assumed. This is due to the sizeable difference in theoretical surface area afforded by GO of 2630 m^2/g when compared to the 580 m^2/g surface area of GAC [31,32,89,90]. More critically, the ozonation process investigated by Tarpani and Azapagic [88] only incurred costs of approximately $0.009–/m^3 assuming the same electricity costs of the present study ($0.077/kWh), while the AOP-PPT demands 22.4 kWh/m^3, which incurs a considerably higher cost of $1.72/m^3 [91].

3.2. Sensitivity Analysis

Parameters found to be most influential on the model were *n* with a coefficient of 0.89 and *i* with −0.45, as displayed in Figure 3. It is unsurprising that the model should be most sensitive to the longevity of the technology, as this will largely govern the quantity of costly materials required for regeneration or synthesis throughout its service life [10]. In fact, every extra year would incur an additional cost of $551.75 for the GOFa filter, $949.26 for the PGa filter and $679.37 for the hybrid filter assuming a breakthrough time of 5 days. These annual costs account for 5.84–5.89% of the total life costs and are therefore substantial. Similarly, the expected inflation rate (*i*) that was given at 6.7% [57] will be expected to have a profound effect on annual and overall costs. This is more concerning, as inflation is prone to rapid fluctuations due to macroeconomic events that will clearly have a knock-on effect on ongoing costs.
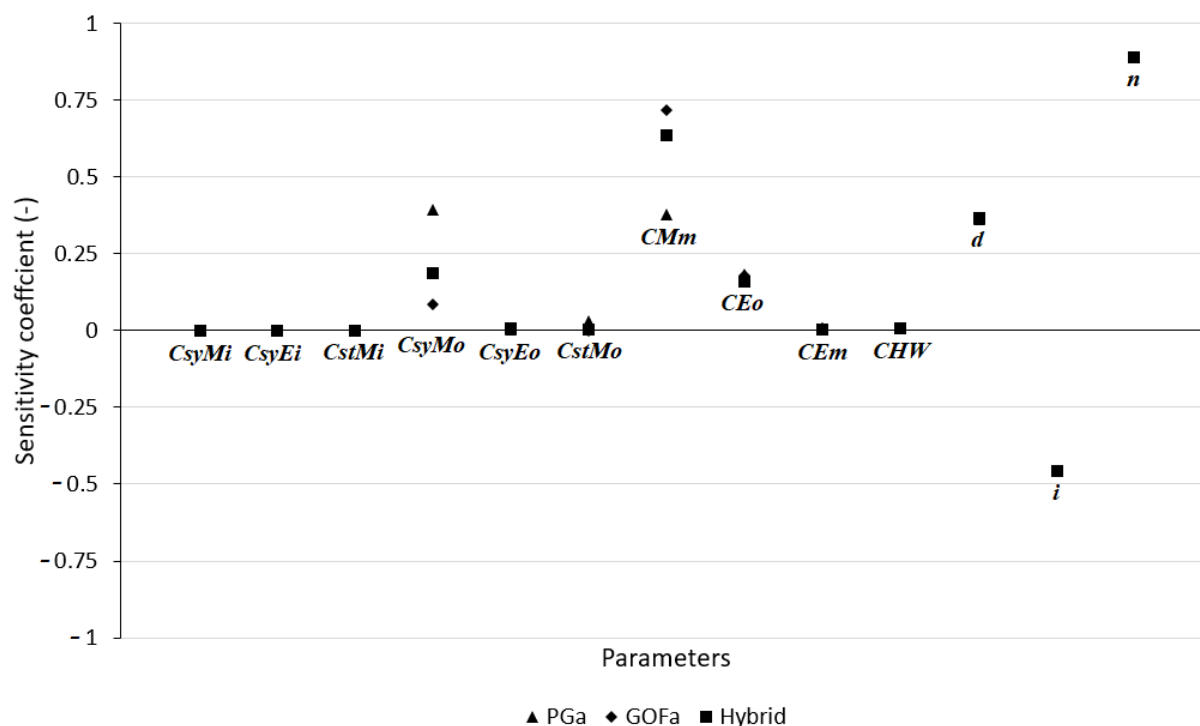
Figure 3. Relative sensitivities of parameters within each of the GBM life cycle costing models.

From the cost parameters, the model was found to be most sensitive to the maintenance materials, *CMm,* with coefficients of 0.50, 0.41 and 0.38 for GOFa, Hybrid and PGa filters respectively. While the PGa filter incurs greater annual costs for backwash/desorption events than the other filter types at \$358.08 compared to \$278.61 for the GOFa, this is a significantly lesser portion of its overall costs (36.8%). In contrast, the annual *CMm* costs of the GOFa filter account for 49.1% of total life cycle costs assuming a 5-day breakthrough. This observation is further supported by the greater relative coefficient of the PGa filter for ongoing synthesis materials (0.39) when compared to the GOFa (0.15) and Hybrid filters (0.31), while reinforcing its greater vulnerability to price fluctuations of the synthesis materials (i.e., hydrazine). Only negligible differences were observed in sensitivities of each filter type for other cost factors.

Other work has also shown the material synthesis costs of adsorbents to be less influential on total life cycle costs. When analysing the sensitivity of a GAC LCC model, Tarpani and Azapagic [88] made a similar inference. This was due to the material costs of GAC only accounting for 25% of the total life cycle costs in their study. While both the total cost portions and the relative sensitivities of the GBM filters are higher than the GAC, a similar assessment can be made due to the reducing total cost portions of each filter types in the present work (PGa > hybrid > GOFa).

3.3. Breakthrough Times

Each of the GBM filters demonstrated a lower life cycle cost when compared to the AOP-PPT. Each calculated trendline (power) was given a backwards extension of 0.5 days which indicates that, at all breakthrough periods above, at least twice daily would favour the GBM filters over the AOP-PPT from an economic perspective. Figure 4 also shows negligible difference in economic costs across the higher breakthrough times for each of the filters, but at the lower breakthrough times differences were observed, with the PGa filter demonstrating the greater cost compared to each of the alternatives. This suggests that, at breakthrough times below 5 days, the PGa filter will be the less economically preferable of the filter types, while the GOFa filter offers the greatest cost efficiency. This increasing asymmetry at lower breakthrough times highlights the additional costs needed by the PGa

filter in both material synthesis and backwash/regeneration when compared to the other filter types. For example, the maintenance events of the PGa filter incurred a cost of $5.08 each time, while the GOFa and hybrid filters both incurred only $3.95 for each event. A similar trend was observed for the costs associated with material synthesis to facilitate the replacement events, whereby the PGa filter cost $161.90 per replacement compared to $39.09 and $92.81 for the GOFa and hybrid filters, respectively.
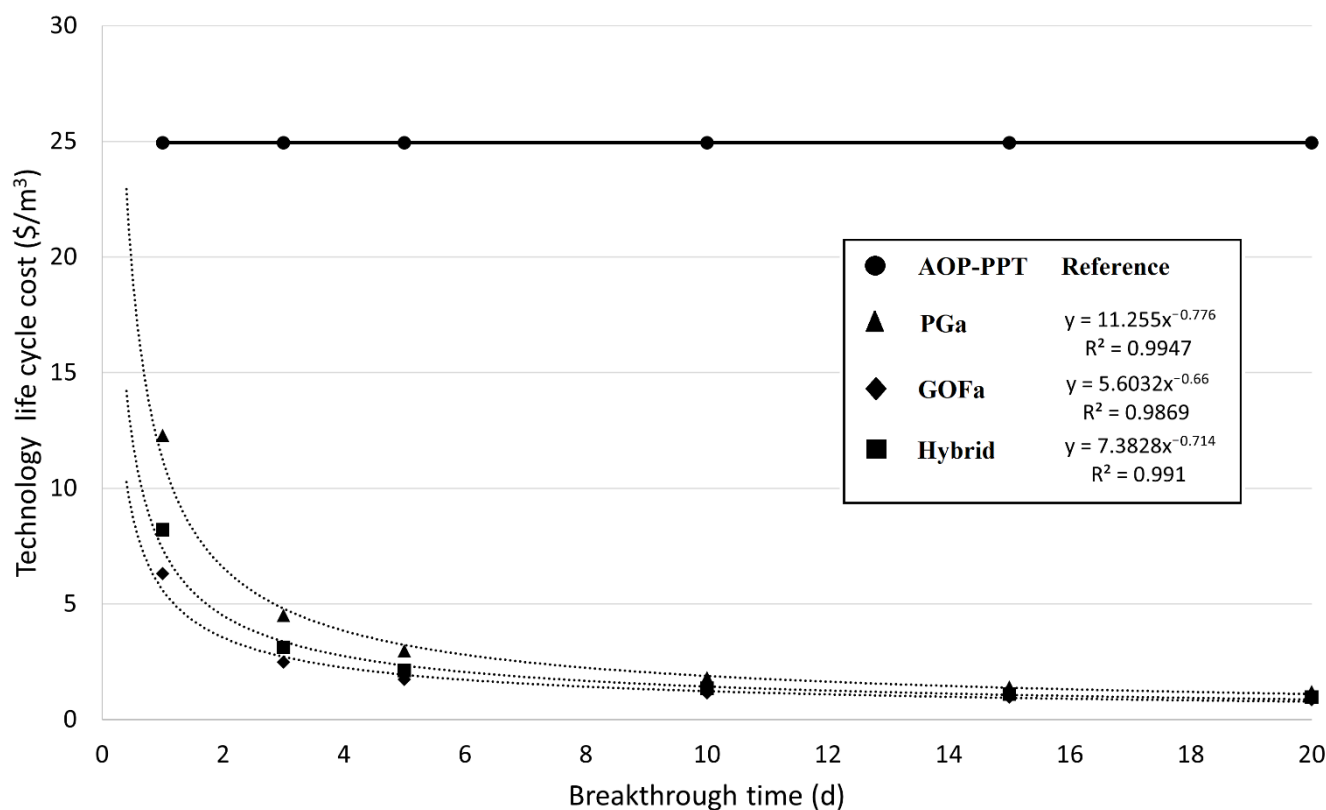

Figure 4. Compared life cycle costs of each technology at different breakthrough times.

At higher breakthrough times, it is the operational costs that account for the largest portion of the total life cycle costs. Figure 5 shows that electricity costs incurred during operation become the dominant expenditure for the hybrid filter when the breakthrough time is greater than 10 days. At similar breakthrough times (11–33 days), Tarpani and Azapagic [88] found electricity costs during operation of the GAC treatment to account for less than 10% of the total life cycle costs. In their study, however, enhanced coagulation as a pre-treatment step was required that incurred as much as 45% of the total costs which would otherwise lead to comparable results.

While much of the life cycle costs are accounted for by the material synthesis costs for the GBMs, it is worth noting that these are costed per lab-sized batch according to present protocols [42,92]. Due to economies of scale, these costs are likely to be greatly reduced as the rate of production increases [93]. Furthermore, the protocols for GO synthesis are still relatively novel and better economy may be achieved as the synthesis methods are further developed and optimized for efficiency. The development of new exfoliation and reduction processes for graphite has already reduced costs by over an order of magnitude [90]. While validating the removal performance of PGa, Khalil et al. [90] highlighted several ways in which the process could be streamlined for better efficiency. Similarly, efficiency gains are likely to be realised as the maintenance events are developed [94]. For example, Kawachale and Kumar [94] showed that the costs of lab-scale adsorbents used for the removal of isoflavones could be reduced during scale-up by recycling the waste solvent streams. This

may have implications for the methanol usage in the backwash/desorption events of the GBM filters that incur the greatest overall costs at the lower breakthrough times.
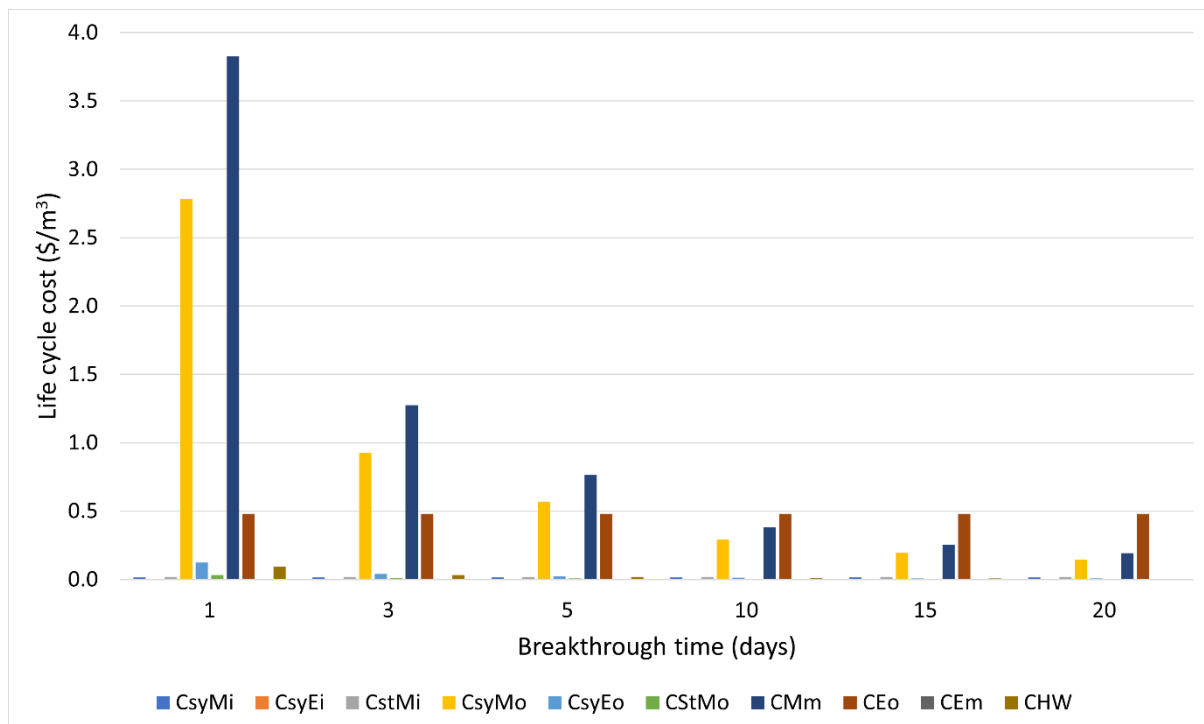

Figure 5. Life cycle costs of the hybrid filter, uncorrected for NPV.

3.4. Eco-Efficiency Assessment

The results of the LCCA were found to be succinct with the findings from the LCIA for the investigated technologies [10] and, as such, the EEA did not demonstrate any deviation of the prevailing costs trend formed during each of the individual analyses (GOFa > hybrid > PGa > AOP-PPT). Relative to the least efficient technology (AOP-PPT), Figure 6 shows that even at a breakthrough time of 1 day the GOFa filter would offer a 63.7% improvement in eco-efficiency, while the PGa filter and hybrid filter afford a 49.0% and 59.4% improvement, respectively.

In terms of breakthrough time, Figure 6 also shows that this strongly influenced the eco-efficiency of each filter with diminishing returns as it increased. Above 5 days the differences in eco-efficiency between filters were negligible, while the greatest differences were observed between 1 and 3 days, which reflects the exponential increase in demand for resources at lower breakthrough times. While the relative order of the GBM filters remained the same at each breakthrough time, the asymmetry between each filter type increased at the lower breakthrough times. This would be expected, with increasing demand of material synthesis exacerbating subtle economic and environmental cost differences incurred during the production process. This is further supported by the greater disparity between the PA filter at the lower breakthrough times compared to the alternative filter types. For instance, the eco-efficiency of the GOFa filter and hybrid filter increased only 22.6% and 25.5%, respectively, when the breakthrough time was increased from 1 to 3 days, while the PGa filter increased by 32.3%.

These results suggest that, of the investigated technologies, the GOFa filter offers the greatest advantage from both an economic and environmental perspective. Furthermore, as adsorbents are known to offer different adsorption capacities when treating different absorbates [46], it is note-worthy that the overall efficiency of the GOFa-based filter has here been shown to remain less affected at lower breakthrough times. In other words, the

GOFa filter is expected to incur less environmental impact and financial expenditure than the investigated alternatives, even as a lower breakthrough time is realised.
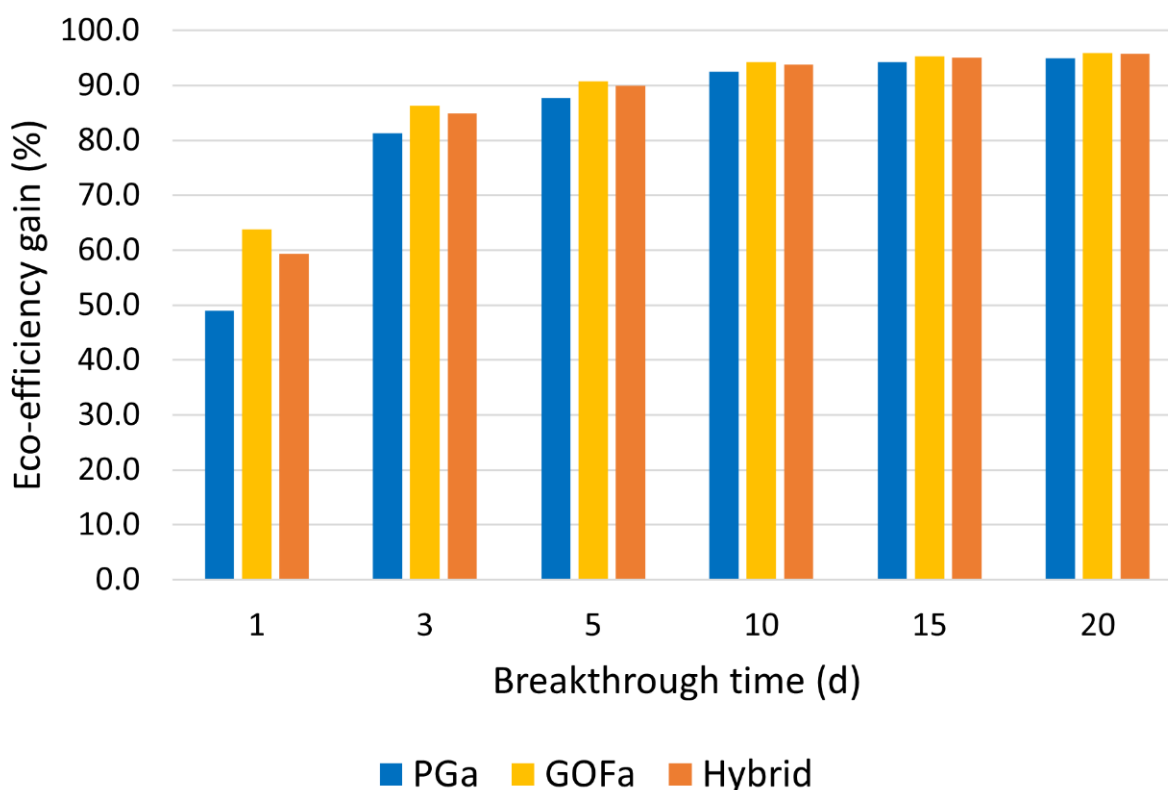

Figure 6. Eco-efficiency scores across technologies and breakthrough times shown as % improvement compared to the least eco-efficient technology (AOP-PPT).

While the EEA provides a better proxy for sustainability than either the LCCA or LCIA alone [48], it still fails to incorporate any measure of performance. Although each of these technologies have demonstrated the capacity to remove ECs from wastewater [31,32,91,92,95,96], direct comparisons are limited due to their novelty and the testing of different contaminants. Furthermore, in the case of the GBM filters, experiments investigating the adsorption capacity have shown good results only in treating trace amounts in distilled water, while showing their performance to be negatively affected by interference, due to the presence of multiple ECs and other present ions as would be expected in municipal wastewater [31,32]. Khalil et al. [31] suggested that this may be overcome by increasing the GBM:EC ratio, but this will affect the life cycle costs of the GBM filter, which will need to be reassessed. Recent work has also shown that GOFa adsorption performance can be improved following prior treatment by UV radiation [92] but this will again incur additional sustainability costs that will need to be accounted for. In contrast, the AOP-PPT has shown strong removal performance without such limitations [91,96]. In fact the AOP-PPT has shown to offer additional services including the removal of COD and BOD ($91 \pm 1\%$) as well as other pollutants that will be expected to pay additional dividends in a more comprehensive analysis [91].

A further consideration should be given to the electricity source used in each of the analyses. The inventories of both the LCIA and LCCA for each of the four technologies were considered in an Indian context, therefore an Indian country mix was assumed. India still generates as much as 59.7% of its electricity from fossil fuels [97], and this is known to incur significant environmental damage during production [10,98]. Ahmadi et al. [82] found that environmental costs could be reduced five-fold under a scenario of maximum renewable energy use in a water treatment plant utilizing energy-intensive systems. The rate of

renewable energy uptake differs substantially between global regions [99]. In countries such as Iceland that now draws 100% of its energy from renewable sources [100], energy-centric technologies like the AOP-PPT will incur relatively little environmental damage compared to the GBM filters. Likewise, with energy prices known to differ substantially across global regions [101], similar shifts in bias will be expected for the AOP-PPT within the LCCA. However, with relatively low electricity prices in India and relatively high electricity prices in Iceland, testament must be given to the more holistic perspective of the EEA that would overcome the conflicting results of the LCIA and LCCA alone for these countries.

It would therefore be premature to conclude at this point that the AOP-PPT is a less sustainable solution than the GBM filters in light of these points. What can be stated is that, at the present time, GBM filters have demonstrated a greater eco-efficiency in an Indian context and may offer the greatest overall sustainability should the ambitious assumption of equal EC-removal performance by each technology be validated. Of the GBM filters, the GOFa variant appears to offer the least economic and environmental costs over the life cycle, but this again will need to be further evaluated in light of relative performance compared to the alternatives. Further work should now incorporate the relative performance of each technology to provide a more comprehensive index by which to compare the sustainability of each technology.

4. Synthesis

While the costs for EC removal determined in the present work may be considered unviable as a solution within typical wastewater treatment budgets, several reasons to expect significant cost reductions have been discussed with particular reference to the upscaling of the GBM synthesis processes. More importantly it is the net value of this treatment that now needs to be assessed through cost-benefit analysis in light of opportune costs and willingness of consumers to pay for it. However, costing the environmental and public health benefits of EC removal remains challenging despite the mounting evidence of their risk [1]. Furthermore, consumers are unlikely to accept higher water bills for EC removal when the direct links between their unhindered release into the environment and the consumer's own detriment is still heavily understated in public perception [102]. Further work is therefore suggested to gain a better understanding of the causal link between EC release and public and environmental health, as well as to explore potential ways of quantifying the benefits of their abatement in economic value. One possible avenue for achieving this may be the use of shadow prices as a measure of the undesirable outputs of EC release, this method having been developed for the purpose of quantifying net benefits of wastewater treatment in previous work [103].

5. Conclusions

Emerging contaminants continue to persist in urban water systems, but novel approaches are being considered as viable solutions. The present work has shown that GBMs can offer economic advantage over energy-intensive solutions due to the relatively high costs of electricity compared to the costs of materials required for adsorbent replacement and maintenance. The GOFa filter was shown to incur the least expenditure throughout the life cycle at $1.73 \pm 0.09/m^3$ compared to $2.97 \pm 0.15/m^3$ and $2.12 \pm 0.11/m^3$ for the PGa and hybrid filters, respectively, while the AOP-PPT was more expensive at $25.02 \pm 1.72/m^3$. When environmental impacts were also incorporated into the analysis, the order of efficiency was unchanged (GOFa > hybrid > PGa > AOP-PPT). It was also calculated that the eco-efficiency of EC removal could be increased by at least 49.0% by employing a GBM filter instead of the AOP-PPT, and that further efficiency gains would be made under longer breakthrough times. Sensitivity analysis showed that service life and expected rate of inflation were the most influential parameters on the life cycle costs, followed by the cost of maintenance materials. Further work has been suggested to provide a more comprehensive evaluation of the sustainability of each technology for comparison

as well as the necessary steps required to gauge the net value of EC removal in the light of environmental and public health benefits and consumer contribution.

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