Sustainable Treatment of Emerging Pollutants in the Context of India

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***Abstract*: Emerging pollutants have been increasingly studied over the past decade to improve our understanding of their fate, occurrence and toxicological effects on the environment and human health. The aim of this research is to develop a model that calculates the removal of emerging pollutants in India using different treatment unit processes. Different wastewater treatment scenarios based in India were defined considering several variables and factors including: influent water quality, intended use of effluents, available resources, operational envelop and treatment efficiency of technologies. WiSDOM Tool was used to find optimal wastewater trains/packages for treatment keeping in view technical, environmental, social and economic aspects. The tool also evaluates the performance of each optimal solution in terms of removal of current pollutants (such as BOD, COD, TN, TP, FC etc.) using multi-objective genetic algorithms and multi-criteria decision analysis. An Excel spreadsheet model was developed, where the treatment trains (generated/selected by the WiSDOM tool) were passed through to determine the removal efficiency of emerging pollutants. Each emerging pollutant has different physical and chemical properties and therefore, each compound should be monitored separately to generate the optimum removal. Further research is required to bridge the knowledge gap regarding emerging pollutants and their removal during treatment.**

***Keywords*: Emerging pollutants, pharmaceuticals, wastewater treatment, WiSDOM.**

I. INTRODUCTION

Emerging pollutants are also known as micropollutants, emerging organic contaminants, contaminants of emerging concern, and emerging contaminants [1], [2], [3]. The Environment Protection Agency (EPA) of the US defines emerging pollutants as 'new compounds without regulatory status and which impact on environment and human health is poorly understood' [4]. Other definitions highlight the lack of monitoring of these substances and the unknown toxicity effects that they may have. One of the earliest sightings of emerging pollutants was recorded in 1965 [5] which focused on steroid hormones in the aquatic environment. Between 1965 and the 90's further publications appeared regarding pharmaceuticals, and hormones as pollutants in the water [6], [7], [8]. Gavrilescu et al. (2015) identified that between the 1930's to 2000's an increase occurred globally in the production of anthropogenic chemicals from around 1 million to 4 million tons per year.

Pharmaceuticals, personal care products (PCPs) and endocrine disrupting chemicals (EDCs) are the most common classed emerging pollutants posted in the literature. However, there are other emerging pollutants researched that are less mentioned e.g. steroid hormones, surfactants, perfluorinated compounds (PFCs), flame retardants, industrial additives and agents (herbicides and insect repellents), gasoline additives, illicit drugs, UV filters, and nanomaterials [9], [10], [11], [12], [13], [14], [15], [16], [17], [18], [19], [20], [21], [22], [23], [24], [25]. Wastewater and water treatment is necessary to eliminate these products from the aquatic environment, however as the existing treatment plants were not originally designed for the removal of these compounds the current recorded removal rates are extremely low [10], [26].

This paper sets out to provide an overview of different sustainable treatment solutions, and their ability to effectively remove a list of chosen emerging pollutants within developing countries. Due to the ubiquitous types of emerging pollutants, and time limitations, thirty-nine were chosen, with a focus on those found within India. The following sections summarise the information which was collected during an extensive literature review, used to help create the Excel spreadsheet to calculate the most suitable treatment option.

II. CURRENT POLICIES, LEGISLATIONS AND RESEARCH

There are many generic water quality policies put in place which focus on current listed pollutants, however, there are no global worldwide policies regarding emerging pollutants [27]. As the topic of emerging pollutants is a growing field there is presently sparse information regarding them. Therefore, different projects have been launched which are currently looking into a broad range of issues such as fate and occurrence in water resources, potential health effects to human health and the environment, control and removal and policy approaches or development. UNESCO, funded by the Swedish International and Development Cooperation Agency (Sida) is covering case studies in 20 different countries such as: Australia, Brazil, China, Ethiopia, India, Kenya, Kuwait, Mexico, Mongolia, Nigeria, Norway, Rwanda, Saint Lucia, Thailand, Tunisia, Ukraine and Vietnam [3]. The EAWAG Institute in Switzerland has proposed environmental criteria for several emerging compounds including pesticides [28], [22]. The SOLUTIONS project set up by the EU integrates

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modelling and monitoring to improve the relationship between water quality regulations (Water Framework Directive and Drinking Water Directive) with authorisations such as REACH (Registration, Evaluation, Authorisation and Restriction of Chemicals). The REACH regulation was set up in 2006 and its aim was to identify dangerous chemicals and find less dangerous substitutes [26], [29].

India:

Currently no official legislation revolves around Emerging pollutants in India, however there has been a total of 42 published papers regarding concentrations of these pollutants in India. A paper written by Gani and Kazmi (2017) provided the first review of all the contaminants present in aquatic sources of India. Out of all the published data they explored it was discovered that 57% of the contaminants were pesticides, 17% were pharmaceuticals, 15% were surfactants, 7% were PCPs and 5% were phthalates. Moving forward the aim of the review was to act as a framework for any future research or regulatory initiatives regarding monitoring of Emerging pollutants in India. No data was reported on endocrine disruptors in the published literature, therefore further research into other classes of pollutants should be carried out [30].

III. SELECTION OF EMERGING POLLUTANTS FOR INVESTIGATION

A list of 39 emerging pollutants were selected for further analysis to review their removal via different treatment options. Therefore, the main pollutants which were chosen are included in journals based within India. Five emerging pollutants were published within Gani and Kazmi (2017): Acetylsalicylic Acid (ASA), Ampicillin (AMP), bis (2-ethylhexyl) phthalate (DEHP), di-n-butyl phthalate (DNBP), and Trimethoprim (TMP). Thirteen emerging pollutants occurred within the paper published by Gani and Kazmi (2017) but were also present within other countries: Amoxillin (AMX), Bisphenol A (BPA), Carbamazepine (CBZ), Ciprofloxacin (CIP), Dichlorodiphenyltrichloroethane (DDT), Diclofenac (DCF), Dimethyl Phthalate (DMP), Endosulfan (END), Naproxen (NPX), Nonylphenol (NP), Norfloxacin (NOR), Ofloxacin (OFL), and Triclosan (TCS). Lastly, twenty-one emerging pollutants were not present within the paper focussing its research in India but were strongly mentioned in other published literature regarding emerging pollutants. These are 3-benzopheone (BP3), Atenolol (ATN), Atrazine (ATZ), Caffeine (CAF), N, N-Diethyl-meta-toluamide (DEET), Diazinon (DZN), Diuron (DIU), Erythromycin (ERY), Estradiol (E2), Estrone (E1), Galaxolide (GAL), Gemfibrozil (GFZ), Ibuprofen (IBP), Methylparaben (MP), Perfluorooctanesulfonic Acid (PFOS), Perfluorooctanoic Acid (PFOA), Roxithromycin (ROX), Sucralose (SUC), Sulfamethoxazole (SMZ), Tetracycline (TCN), and Tonalide (TON).

IV. SOURCES AND PATHWAYS OF EMERGING POLLUTANTS

Sources of emerging pollutants can include pesticide application on agricultural land, parks and gardens, urban infrastructure, as well as domestic, hospital and industrial waste and wastewater; which can contain pollutants such as pharmaceuticals and PCPs [27], [15], [31], [19], [32], [33]. Point source locations, consist of those containing a single point as the source such as: industrial effluents, wastewater and water treatment plants, and landfill sites. On the other hand, non-point source/diffuse source [35] locations will cover a broader geographic range for example agricultural land [31], [33], where the specific pollution point cannot be identified. Non-point sources can have a larger impact on groundwater quality [31]. However, point source pollution results in higher concentrations entering the environment, therefore making it easier for detection [19].

The pathway which the emerging pollutants take is dependent on their physiochemical properties [14]. For example, the chemicals solubility in water; those chemicals which have a lower solubility are more likely to be found within the sediments [26], [33]. Direct pathways routes for emerging pollutants are listed by Stuart et al. (2012) as ‘leaking sewers, discharge of wastewater treatment effluents, landfill leachate, leaking storage tanks and other discharges to the ground that bypass the soil zone such as septic tanks’. If groundwater tables are low, septic tanks act as an important source [19]. On the other hand, agricultural pesticides and sewage sludge will transport through the soil zone [33].

V. OCCURRENCE OF EMERGING POLLUTANTS IN WASTEWATER TREATMENT PLANTS

Along with spatial and temporal variations of emerging pollutants, Luo et al. (2014) highlighted other influences which can cause changes in concentrations of the influent and effluent of wastewater. Contributing factors include rate of production, sales of emerging pollutants, metabolism, water consumption per person per day, size of treatment plants, and the elimination efficiency of wastewater treatment processes [21]. These factors also play a part in the occurrence of emerging pollutants in drinking water, along with the lack of removal treatment options in wastewater treatment plants. A study conducted by Kleywegt et al (2011) identified that carbamazepine and caffeine were found at high concentrations exceeding 600 ng/L [34]. Further studies are required into the safety of drinking water and any effects which may be posed by parent emerging pollutant compounds or by their transformation by products [32].

Pharmaceuticals are metabolized within the human body and excreted within urine and faeces which therefore, results in the compounds ending up in wastewater treatment plants [21]. Pal et al. (2010) focussed on the pharmaceutical occurrence on studies in the literature from Europe, North America, Australia and Asia. It was noted that the metabolite of erythromycin was found in high concentrations than the parent compound in Asia [2]. In Europe, the recorded values were higher than the predicted no effect concentration (PNEC) for ibuprofen, whereas in Asia and North America ibuprofen concentrations were lower. It should be noted that ibuprofen, carbamazepine, sulfamethoxazole, diclofenac and primidione have low excretion rates. However, these compounds are found in high concentrations in wastewater treatment plants due to the products frequent and high usage [21].

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It was also found that estrogenic compounds exceeded their PNEC values and a major source for this was veterinary excreta. Li et al. (2013) reported on hormones mostly occurring at concentrations lower than 100 ng/L with the exception of hospital effluents. For example, antibiotics such as lincomycin were detected in concentrations up to 56,7000 ng/L in Taiwan. High levels of tramadol, codeine, gabapentin, and atenolol were detected at high levels in the raw wastewater in a treatment plant in Wales, UK. Areas where high levels of pharmaceutical products are detected can been seen to match locations which have high quantities of pharmaceuticals [34]. The concentrations of PCPs entering a treatment plant can be affected by climatic condition. Kasprzyk-Hordern et al. (2009) found that in dry weather conditions the concentration of PCPs entering the treatment plant would double. This is because in wet weather conditions the rainwater would act as a dilution, therefore reducing the concentration entering the plant. Luo et al (2014) also stated that in warm weather conditions, temperature can impact the concentration of pollutants entering the plant.

VI. FATE OF EMERGING POLLUTANTS

The transformation of active pharmaceutical ingredients (APIs) occurs in wastewater treatment plants dependent on the composition of sewage, weather conditions, and treatment options available [23]. The different fate of APIs and their metabolites within treatment plants could be due to mineralization to carbon dioxide and water, adsorption onto solids, or release within the effluent [35]. Some pharmaceuticals will adsorb onto sludge within a treatment plant, therefore, when this sludge is later used for agricultural purposes (fertilizer) the pharmaceutical products can enter the environment [23]. Pharmaceuticals are ubiquitous in the environment as rates of release are greater than the rate of removal and transformation [29]. Nikolaou et al. (2007) identifies that the most persistent pharmaceuticals are sulfonamides and fluoroquinolones, and the most easily adsorbed by soils and sediments are (listed in order): tetracyclines, fluroquinolones, macrolides, sulfonamides, aminoglycosides, ß-lactams.

Research regarding pharmaceuticals and their presence in food webs is limited. However, Nikolaou et al. (2007) identified that diclofenac was reported in the prey of vultures, such as the liver and kidneys of fish. Other chemicals also detected in fish were fluoxetine, sertraline, norfluoxetine, and desmethylsetraline [36].

VII. METHODS

A. *Excel Programme Development*

The Excel model was developed using a spreadsheet to calculate the removal efficiencies of the chosen emerging pollutants. The treatment options (unit processes as well as treatment trains) used were taken from the WiSDOM tool, to allow for a clear comparison and analysis against current Indian Water Standards, and the removal efficiencies of emerging pollutants.

First a table was produced regarding concentrations of the chosen emerging pollutants which included the name of the emerging pollutant, abbreviations, Chemical Abstracts Service number, surface water concentrations, ground water concentrations, untreated wastewater concentrations, drinking water concentrations and treated wastewater concentrations. This information was populated to provide a range of minimum and maximum values regarding the concentrations recorded of the contaminants and was found within a range of sources. Next the minimum and maximum removal efficiencies (%) were looked at for all thirty-nine emerging pollutants when treated through different unit processes. The different unit processes used were from the WiSDOM tool which was adapted from Joksimovic (2007). A surface literature search was carried out using a variation of different input words for each pollutant. For example, 'removal of ibuprofen', 'treatment of ibuprofen' and 'removal of ibuprofen from wastewater'.

Large amounts of research which is published, looks at the overall removal rate of the different pollutants in different treatment plants. However, not all the information recorded lists the different unit processes used, nor does it contain a breakdown of the removal efficiencies within the effluent after each process. Due to a lack of research within this area, there is currently insufficient data recorded regarding the removal rate for each of the unit processes and emerging pollutants mentioned. Therefore, assumptions were made to allow a large enough data-set to be produced to run the calculations (Table 1).

Table 1 - Assumptions that were made to allow for the creation of the Excel programme regarding the removal rates of emerging pollutants.

	Assumption
1	All unit processes involving activated sludge will have the same removal rate.
2	P-Precipitation and EBPR are focussed on the removal of phosphorous only, therefore unless any specific information is found regarding their removal rate, they will not be included. The values will be inputted at 0% removal.
3	The following unit processes will not be considered and their removal rates will be 0%: DAF and EBPR.
4	If no information is found for a unit processes regarding an EPs removal the removal rate will automatically be placed at 0%.
5	If a value is found for anaerobic conditions than all other treatment processes will pose similar removal rates.
6	The overall removal rate for stabilization ponds will be split equally between the different pond stages.
7	Regarding Caffeine, all disinfection stages will have similar removal rates unless stated otherwise in the literature.
8	If information is only found regarding one certain type of pond i.e. algal ponds, then the removal rate will be present for all different pond types.
9	When only one overall value is given for more than one treatment option, this percentage will be split between the processes included.
10	That Chlorine Dioxide and Chlorine Gas will have the same removal rates (%) unless otherwise specified.

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To create the spreadsheet, the list of emerging pollutants chosen for this study were populated into separate rows in a column, and the minimum and maximum recorded concentrations were displayed in a separated column alongside. Using a separate sheet, the possible unit processes from the WiSDOM tool [37] were listed and a drop down facility was created in the main sheet to allow the user to change the required treatment train options. A formula was used to calculate the water quality after each stage/unit process. Each cell used the same formula, however when references to cells were made within, the formula number changed to correlate to the relevant cell in question.

A. *WiSDOM Tool*

WiSDOM is a decision support tool designed for the optimisation and selection of wastewater treatment trains/technologies in the context of India. The tool contains a user friend graphical interface which consists of both Genetic Algorithm Based Many-Objective Optimisation and Multi Criteria Decision Anaylsis. This tool is used to calculate the removal of COD, BOD, Suspended Solids, TN, Phosphorous, Faecal-coliform, Turbidity and Intestinal Nematode Eggs to meet Indian Standards. A user manual is available for an in-depth user friendly guide and further explanation of the optimisation processes used [38], [39].

B. *Combination of WiSDOM Tool and Excel Programme*

Different scenarios were developed which consisted of different influent water quality. The different scenarios were processed through the WiSDOM tool, to determine the best treatment removal options for current Indian Water Standards depending on the constraints inputted. The eight best solutions from WiSDOM were run through the Excel programme to determine which had the better option for the removal of emerging pollutants. The results were then collaborated to find the overall best solution for both the removal of emerging pollutants and the removal of current water pollutants which are monitored by India.

VIII. RESULTS

The top eight treatment train solutions from the MCDA were taken for each scenario and a scatter graph was produced from the Multi-Objective Genetic Algorithm (MOGA) optimisation of the best solutions. These solutions were then entered into the Excel programme to determine the best treatment train solution for the removal of the emerging pollutants. Different scenarios were created to test the Excel spreadsheet: Scenario 1 - Treatment Options Suited to a Varied Population, Scenario 2 - Treatment Options Suited for Industrial and Hospital Wastewater.

A. Scenario 1 – Treatment Options Suited to a Varied Population

The first of the two scenarios explored focussed on the impact of public festivals and holidays such as Diwali and Ganesh Chaturthi, within Goa. The second scenario which was tested look at the removal of emerging pollutants in a tourist location, Jaipur. In both scenarios, it was expected that higher levels of PCPs and pharmaceuticals would be found in the locations chosen.

The most optimum treatment train for the first scenario (Fig 1.) which focussed on occurrence events consisted of a fine screen, RBC, SAT, constructed wetlands – polishing, and chlorine gas. The removal rates of AMP, CBZ, CIP, DEET, DDT, DMP, DnBP, DIU, ERY, NOR, SMZ and TMP were due to the use of chlorine gas only (1-100%). Whereas, the removal of DIA, NPX, ROX, SUC occurred during just the use of wetlands (12-100%). E1, E2, OFL and TCN used both the wetland and chlorine gas for the removal of emerging pollutants. Constructed wetlands – polishing contributed to 90-100% removal for hormones (E1 and E2), whereas the use of disinfection only reached removal rates of 33%. For OFL the removal percentages for both tertiary treatment and disinfection were around 70% and 80%, respectively. The removal of TCN during wetlands had removal rates of 17-100%, whereas the use of chlorine gas only removed 29% of the remaining pollutant. The use of RBC contributed to the removal of BPA (74%), CAF (78-97%), CBZ (1-5%), DCF (13-66%), GAL (81-90%), IBP (77-97%), MP (49-69%), NP (46-88%), TON (83%) and TCS (4-80%).

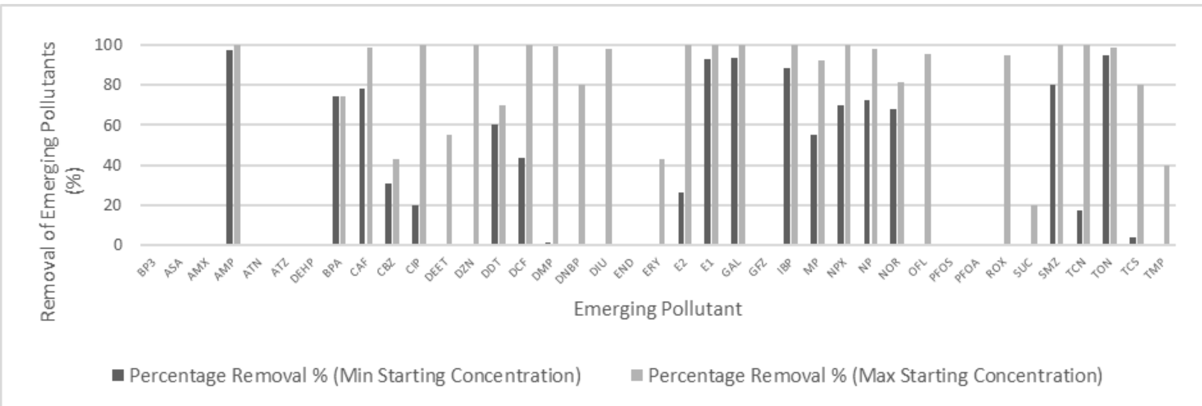

Fig. 1 - Graph showing the removal percentages of different emerging pollutants for the optimum solution for Scenario 1a.

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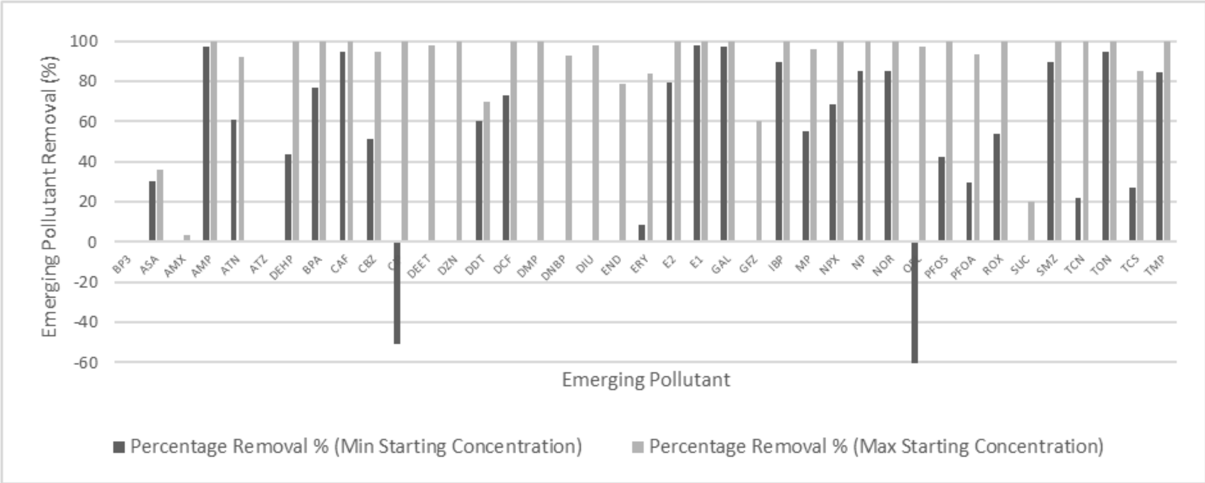

Fig. 2 – Graph showing the removal of different emerging pollutants for the optimum solution for Scenario 1a. Not to scale.

The most optimum treatment train for the second scenario involved bar screen, sedimentation w/o coagulant, low loaded activated sludge and secondary sediment, RB, UF, constructed wetlands-polishing and chlorine gas. Although this solution was effect at removing a large proportion of emerging pollutants, CIP and OFL had negative removal rates when minimum removal efficiencies were used. When activated sludge has been used within a treatment train, it has been found to have negative removal rates of -124.2% when trying to remove OFL, and a removal rate of -88.6% when removing CIP (Fig.2). Both emerging pollutants saw no removal during tertiary and disinfection stages.

B. Scenario 2 - Treatment Options Suited for Industrial and Hospital Wastewater

The first type of wastewater explored was hospital effluent, therefore a focus was placed on the ability of the treatment train to remove UV filters, PCPs and pharmaceuticals which were included. The most optimum solution included a coarse screen, sedimentation w/o coagulant, fine screen, advanced oxidation (UV/H2O2), SAT and chlorine gas. The use of an AOP led to the removal of BP3 with removal rates ranging from 50-100%. The unit processes used within this treatment train were ineffective at removing ASA and NPX.

The second example used industrial wastewater, resulting in ensuring that the treatment train was suited to remove emerging pollutants which are found or used in industrial products. The optimum solution involved sedimentation w/o coagulant, DAF w/ coagulant, stabilisation pond: aerated ponds, constructed wetlands – polishing, micro filtration and chlorine dioxide.

I. CONCLUSIONS

Emerging pollutants were previously not listed as a cause for concern, therefore water treatment plants were not (purposely) designed to remove them. This in turn has allowed for emerging pollutants to access our water systems leading them to enter freshwater and drinking water systems. The most common types of emerging pollutants explored amongst the literature were pharmaceuticals, personal care products and endocrine disrupting chemicals. However, there are many other different types of emerging pollutants, which

still need to be further explored e.g. surfactants, flame retardants, industrial additives and agents and UV filters. Further research is needed on the concentration of these new emerging pollutants in different water sources worldwide.

The overall aim of this study was to analyse sustainable treatment options for the removal of emerging pollutants within India. Natural processes such as wetlands and ponds are a more sustainable treatment option to remove current and emerging pollutants. However, the land requirement for these options is not always suited in urban areas. Equally, more energy intensive options such as AOPs are not suited in areas such as Dharavi where steady electricity sources are not viable. This research has provided a way to assess the removal of emerging pollutants within separate unit processes. The main conclusion which can be taken from this study is that

each emerging pollutant needs to be treated and monitored irrespective to any other. Each emerging pollutant has its own physical and chemical components resulting in the compound to be broken down or removed in its own unique way. Therefore, in order to allow for the effective removal of emerging pollutants it is important to study each compound separately including their transformations during unit processes. This study has provided the basis for further research concerning the removal of emerging pollutants.

RECOMMENDATIONS

The research surrounding emerging pollutants is increasing globally, however, the main limitation with this research occurred due to the lack of data regarding their removal, fate and occurrence within India. Further investigations are needed to fill the current gap within the published literature. Advances are required regarding the funding availability and access to equipment within India, to allow for a better understanding of emerging pollutants.

This study provided the basis for further investigations regarding the removal and treatment of emerging pollutants. Primary data collection would allow for more accurate removal rates during different treatment stages, therefore

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allowing for the creation of a more accurate model. Monitoring and testing of emerging pollutants would allow for a list of emerging pollutants which occur within India to be produced. This in turn would help to create a model that would be more specific to those pollutants found in India to provide an effective treatment option. In addition, separate unit processes should be tested along with stages of wastewater treatment to allow for a better understanding of the removal rates involved at individual unit processes. Currently, large proportions of the data collected provide an overall value for removal given from the concentration in the effluent. However, removal rates more specific to each unit process would help to provide a better understanding of the fate of emerging pollutants during wastewater treatment.

Time constraints of this study resulted in an additional Excel spreadsheet model being created to work alongside the WiSDOM tool. However, future work would involve updating the tool to apply the ideas behind the Excel spreadsheet. By updating and changing the hard code implemented within the WiSDOM tool it can be changed to generate suitable treatment train removal solutions for both current listed and emerging pollutants.

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