



Research paper

Groundwater quality in the Jaffna peninsula of Sri Lanka and a qualitative study of BTEX removal by greenly synthesized iron nanoparticles-electro-catalyst system.

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ABSTRACT

The Jaffna peninsula northern part of Sri Lanka (Longitude: 80° 06' 2.28" E and Latitude: 9° 41' 12.20" N), mainly gains water from groundwater which is continuously protected by various human activity even with reckless deterioration. The maximum nitrate concentration reached up to 51 ppm whereas the amount of phosphate found to be as 3.8 ppm. The average EC for a year is 38.1 mS/cm and corresponding salinity and TDS found to be as 20.2 and 24.3 g/L respectively. The middle region of the territory consists less saline water and 0.132 ppm of Pb and 0.092 ppm Cd presence in the groundwater. The BTEX has been completely eradicated by using the cost-effective environmentally-friendly electrochemical system. The developed system can be utilized the application areas such as service station, mechanic stores, hospitals, universities and household purpose.

1. Introduction

The water is one of the crucial natural element for the survival of the various living organism in the planet. The globe faces the serious problem that is the scarcity of the water for various purpose including household needs. This depends on economic growth, development climate change, season and human activity. Due to the human activity the vast amount of contamination discharge to the environment from various anthropogenic sources those causes detrimental impact on the environment is a growing concern (Marara and Palamuleni, 2019). Jaffna peninsula is the dry zone of Sri Lanka, mainly depend on groundwater. The groundwater consumption dramatically increased due to the increasing population, introduction of electric and other water pump and dense tube wells. The groundwater source in Jaffna is limited and the aquifers received water through seasonal rain. Reckless anthropogenic activity released, various water-soluble components to the environment and invading of seawater, affects the impinging living things directly or indirectly (Rajasooriyar et al., 2002).

The water pollution consists of dual effects such as on the environment and living organisms. In the aquatic systems the pollution affects

the productivity, diminution and elimination of environmentally sensitive organisms. In the world, nearly 14,000 people died every day due to water pollution. Various pollutants in water affect the water bodies by changing the properties like algal blooming, pH changes, turbidity, TDS, conductivity, salinity, odour, sulphate content and carbon dioxide content (Owa, 2013).

The northern part of Sri Lankan where the population heavily rely on underground water, tanks, pond and small rivers. The territory mostly covered by Miocene CaCO₃ which mainly causing water hardness that is a common issue. Recently due to the modernization, the increasing numbers of vehicles and automobile service stations, usage of electronic equipment, cracked mirrors, ceramics and deforestation have been drastically increased along with the poor wastewater management lead to a serious threat to the whole environment (Vithanage et al., 2014). The people of the Jaffna significantly utilize the groundwater for irrigation and household activities. Due to the overexploitation of the groundwater, the saline water intrusion is a crucial issue (Gunaalan et al., 2018). The heavy metals area severe hazard to the normal functioning of aquatic systems even at lower concentration. They are very toxic, non-biodegradable, and are involved in biochemical cycles. It also

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affects the human beings and causes disorders in renal and nervous system. Moreover, in the case of unmanaged disposal of lead-acid batteries, automobile oils, fuels and fertilizers relatively increase the amounts of Pb, Cd in the environment. In addition, during the war, the usage of lead, cadmium containing explosives, cracked mirrors, electronic equipment and ceramics became the potential source of heavy metal pollution. As a consequence of the above-mentioned issues, the groundwater becomes polluted with heavy metals which in turn cause adverse health effects upon ingestion (Belkhiri et al., 2018; Oyeku and Eludoyin, 2010; Momodu and Anyakora, 2010; Wu et al., 2015; Mohankumar et al., 2016; Mridul and Sarma, 2013).

Nanoscience plays a major role in waste management of the ecosystem. The nanomaterials are unique, in the sense of their extremely small size and as a result, the highly elevated surface to volume ratios compared to their bulk counterparts. The nanomaterials are being used as catalysts in many areas of science and technology. Especially, particular interest is the utilization of nanomaterials in water purification. If the nanomaterial synthesis also involves a greener approach, then the overall process of synthesis and application of nanomaterials in water purification becomes truly environmentally-friendlier (Andersen et al., 2014; Dey et al., 2012; Bhattacharya et al., 2013). The plant extracts are being used for green synthesis of nanoparticle which is stand out as an environmentally-friendly methodology for the synthesis of metallic nanoparticles. The plant extracts which are capable of reducing a variety of iron salts to generate iron nanoparticles can be replaced the inorganic reducing agents like borohydrides (Kónya et al., 2007; Nikalje, 2015; Shanmugavel and Karthikeyan, 2013).

The synthesis of iron nanoparticles by using extracts from biological sources is significantly easier when compared to conventional chemical methods. This is because the biological sources act as self-reducing agent and self-surfactant. Green synthesis of iron nanoparticles has an additional advantage that the compounds present in plant extracts can impart steric stabilization of iron nanoparticles against aggregation. This would enable to overcome the concerns related to using the reducing agents such as sodium borohydride in routine synthesis adapted so far. Recently, the successful synthesis of iron nanoparticles utilizing green tea leaf and sorghum bran extracts have been found (Kónya et al., 2007). Iron nanoparticles were readily synthesized by using green tea leaf extracts (GT-Fe NPs). Green tea is known to be contain polyphenols that act both as a reducing agent and a capping agent (Kónya et al., 2007; Nikalje, 2015; Shanmugavel and Karthikeyan, 2013; Dave and Chopda, 2014; Meyers et al., 2006). The utilization of natural or organic acids is also an alternate way for the iron nanoparticle preparation. Herrera-Becerra et al. used tannic acid and gallic acid as reducing agents to prepare iron oxide nanoparticles whereas Meeks et al. had successfully used ascorbic acid for the synthesis of iron-based nanoparticles (Devi and Singh, 2016; Sharma et al., 2012; Herrera-Becerra et al., 2010; Meeks et al., 2012).

The common organic pollutant in industrial and agricultural wastewater is one of those materials that are extremely difficult to be degraded naturally. Though various methods have been developed for the degradation of p-nitrophenol (p-NP), the utilization of catalysts to electrochemically degrade p-NP has become a novel and effective methodology where two magnetic nanoparticles (carbon-encapsulated iron, Fe/C; carbon-encapsulated cobalt, Co/C) were employed as catalysts. The above mentioned both types of modified electrodes showed favourable stability and excellent electrocatalytic activity for p-NP degradation (Dave and Chopda, 2014; Sharma et al., 2012; Herrera-Becerra et al., 2010; Meeks et al., 2012).

In this study, we carried out some basic water quality parameters such as EC, TDS, salinity, turbidity and pH for an entire year and the heavy metals such as Cd and Pb. Further, we proposed a greener method is most suitable to an aquifer contaminated by waste oil. In addition, we discover a strategy to predict the oil contamination and destroy BTEX in short period of time. It is a cheap and fruitful method for water treatment.

2. Materials and methods

2.1. Materials

Anhydrous ferric chloride (FeCl_3 -98%) was purchased from Loba-Chemie Pvt. Ltd. (Mumbai, India). Granular activated carbon was provided by Noida Chemical (Delhi, India). Sodium borohydride (NaBH_4 -99%), phosphoric acid (H_3PO_4 - wt.% 85) and sulfuric acid (H_2SO_4 -99%) were issued by Sigma-Aldrich (USA). The green tea with 100% purity was purchased from Blue field, Ceylon. BTEX standard solution with HC BTEX Mix Chromatographic with 99.9% purity was imported from Supelco Analytical (USA). The waste oil was collected from local service stations. The chemicals used were of analytical grade and used without further purification.

2.2. Sample collection

The sample collection was done in the selected places with a special instrument designed by our own. The cleaned glass bottle can be replaced instead of cylinder and immersed into the water by using thread 1 and 2. Then, thread 2 is gradually lifted below the surface of the water to open the lid. Finally, thread 2 is loosed to close the lid as soon as the cylinder gets filled. The water containing bottles are transferred in an icebox for further analysis and the analysis was finished on the same day of the sample collection. The following tests, such as electric conductivity (EC), salinity, total dissolved solids (TDS), pH and turbidity were analyzed. In one location minimum of five samples were collected for the analysis. From which the average was calculated. In addition, cadmium (Cd) and lead (Pb) were analyzed according to the manual of the autolab and samples are collected in selected dug-wells which are located near automobile service stations in Jaffna peninsula (Fig. 1, S1-S8 and Tables S1-S3).

2.3. Preparation of zero valence iron NanoParticles (ZVINP)

2.3.1. Chemical synthesis of ZVINP

Freshly prepared 5 mL of 0.001 M FeCl_3 solution was added drop by drop into the 5 mL of 0.01 M of cold NaBH_4 and then the temperature of the system increased and maintain between at 50–60 °C with continuous stirring. The pH of the system was monitored between 6 and 7 during the process.

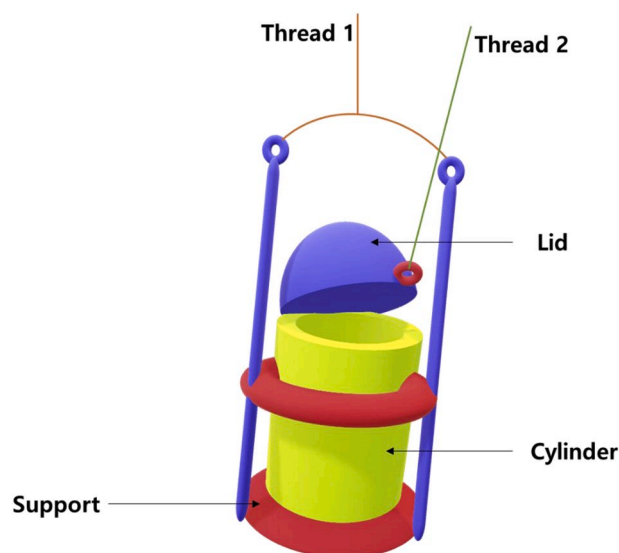


Fig. 1. Schematic representation of sample collector.

2.3.2. Green synthesis of ZVINP by using green tea extraction

The commercial green tea was washed thoroughly with distilled water for three times and dried at 101 °C. Then the green tea was ground in to finely powder by using mortar and pestle. Then, 5 g of it was heated in 50 mL distilled water at 60 °C in a 100 mL beaker for 30 min. The extract was separated by filter paper (45- μ m pore size -Whatman) and the extract was collected for ZVIN synthesis. Then, freshly prepared 5 mL of 0.001 M of FeCl₃ solution was drop wisely added into 5 mL of hot (50–60 °C) green tea extract with continuous stirring. The product was collected and utilized for deposition purpose.

2.4. Deposition of ZVINP on to activated carbon pores (ZVINPAC)

1 g of granular activated carbon was washed with diluted 50 mL of 0.1 M phosphoric acid and subsequently washed with ample amount of distilled water. Then it was heated at 110 °C for 30 min. Then the temperature of pre-treated granular activated carbon was increased up to 250 °C and remains unchanged during the treatment. The synthesized ZVINPswas added dropwise while stirring and the temperature of the system was increased up to 500 °C for 1 h. The black colour product was cooled down to room temperature in a desiccator for 24 h and it was directly utilized for oil degradation purpose.

2.5. Degradation of used oil and BTEX in water by ZVINPAC

This test was performed electrochemically by using commercially available carbon electrodes were used. The positive electrode was kept in a perforated glass vessel (with the dimension of 4 cm \times 4 cm \times 8 cm) which containing ZVINPAC. Both electrodes were kept in the 500 mL beaker containing 300 mL of used oil or BTEX aqueous solution. The electrodes were connected with 900 mA DC source. The degradation reaction was carried out at a pH of 4. During the reaction 5 mL of the solution was drawn in 10 min intervals for further analysis which was done at FROG 4000 portable gas chromatography.

2.6. Characterization

X-ray diffraction (XRD) was performed using Siemens D5000 powder X-ray diffractometer (Cu K α λ = 0.154 nm and scanning rate of 1°min⁻¹ (Rigaku, USA). The morphology was analyzed by using Hitachi SU6600 Scanning Electron Microscope (SEM) (Japan). The BTEX was examined by Frog 4000™ gas chromatography (USA). The heavy metals were analysed buy using auto lab RDE (Directive 89/336/EEC, Netherlands). The pH was measured by using Hach sensION + PH1 Basic Portable pH Meter (USA). EC, salinity and TDS were investigated by using Hach sensION + EC5 Portable Conductivity Meter (USA). The nitrate and phosphate were analyzed by using DR1900 Portable Spectrophotometer (USA). The whole analysis was done according to standard procedure recommended by instrumental manuals. The graphs were plotted using a Savitzky–Golay function (Origin 2017 graphing and analysis, Origin Lab (USA)).

3. Results

3.1. Structural analysis

3.1.1. X-ray diffraction (XRD) analysis

The XRD profile is used deduced structural features of materials and the XRD profile of iron nanoparticle deposited activated carbon shown in Fig. 2. It is obvious that two broad ((23.98,24.16) and (43.42,42.88°)) and small peaks (20.84, 65.19, 66.80 and 68.42°) indicate that the compositions are amorphous in nature. This phenomena is due to the presence of higher volume of activated carbon compare to the nanoparticles. The chemically synthesized iron nanoparticle embedded activated carbon exhibited Bragg's angles at 43.42° and 23.98° disclosed the occurrence of both zero-valent iron and iron (II, III) oxide crystalline

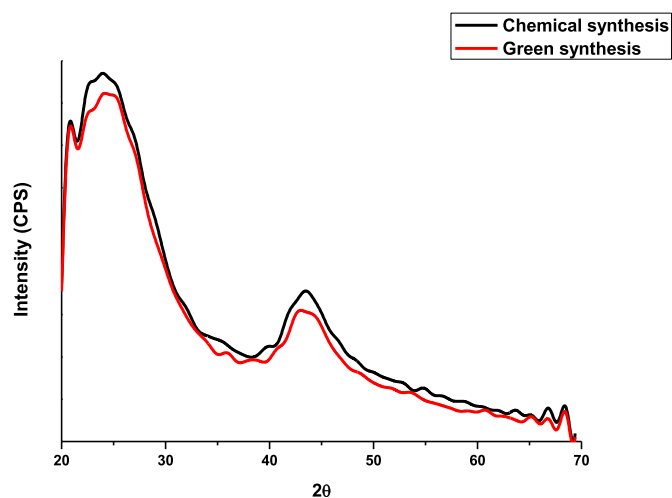


Fig. 2. XRD spectrum of iron nanoparticle prepared by chemical synthesis and green synthesis. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

phases (Fig. 2) (Rajasooriyar et al., 2002; Owa, 2013; Vithanage et al., 2014). Eventhough, peak positions of greenly synthesized slightly difere (42.88° and 24.16°) which is due to the presence of polyphenol and structural features of iron nanoparticles (Fig. 2). In addition, it is noticeable that the weak peaks at about 15–30° and 40–50° are responsible for the reflection plans of activated carbon (002) and (101), respectively. The peak at about 23° implies presence of the iron oxides whereas the peak at 44° confirms BCC zero valent iron nanoparticle (Owa, 2013; Vithanage et al., 2014). Furthermore, the characteristic small peaks are used to confirm iron nanoparticle embedded activated carbon composition. Hence, the composition consist both zero valent iron nanoparticle and iron oxide nanoparticles.

3.2. Scanning electron microscopy (SEM) analysis

The SEM images of both green-synthesized and chemically iron nanomaterials are shown in Fig. 3. The pore of the activated carbon in microscale in which nanoscale pores are present (Fig. S9). Embedding the iron nanoparticle into the nanoscale pores is a tactic of this work. The embedded particles morphologies were cone and spherical shape with size ranging from 40 nm to 110 nm and are arranged in the nanopores of carbon in an irregular fashion. The particle size of cone shape varies between 87 nm and 94 nm, while spherical shape ranged from 64 nm to 76 nm as shown in Fig. 3a–b. In addition, chemical synthesis influences the size of the particle that chemically synthesized nanoparticles were smaller than those of greenly synthesized. Further, the deposited patterns resembles internal branches within the activated carbon pores (Gunaalan et al., 2018). This indicates that the precursor solution diffused well into the entrenching branches of the pore. The maximum size of the pore in the barely activated carbon was observed is 117 nm which imply that the size of the nanoparticles is smaller than that of activated carbon pore (Fig. 3a–b, S9). Furthermore, nanoparticles well settled within the pores.

3.3. Water quality analysis of the Jaffna peninsula

The basic water quality parameters of Jaffna peninsula were tested over a year and the average was calculated for this study. According to our observation, during the dry season the salinity, EC and turbidity were high compared with the corresponding rainy season. According to the Jaffna territory the excess rainwater reached to some particular areas such as Thondaimaanaaru, Arriyalai and Araly in which the Thondaimaanaaru saline-fresh water gates are biggest in the Jaffna

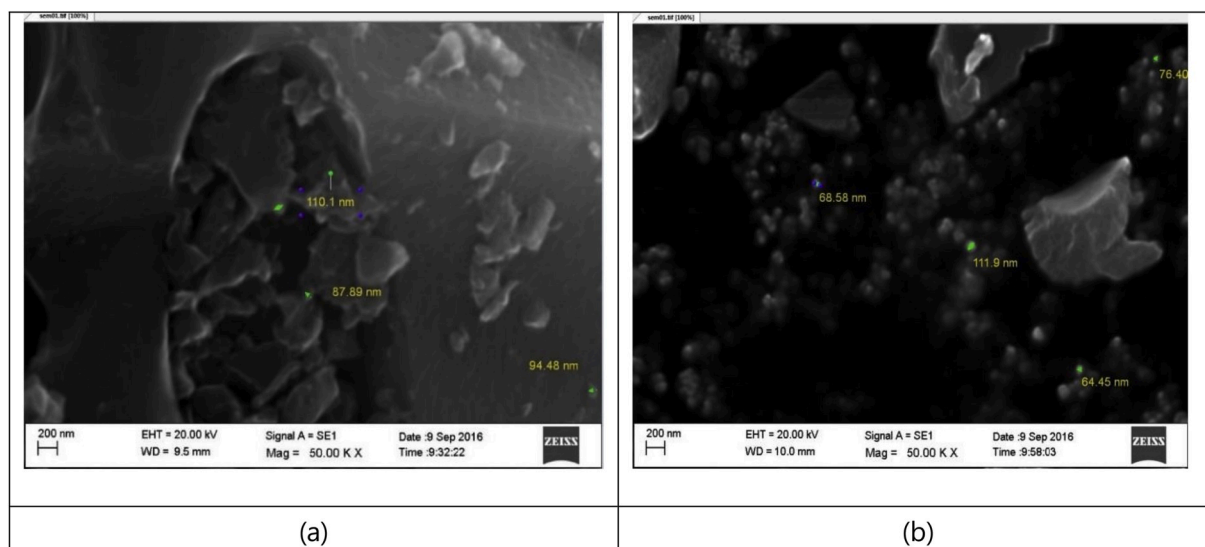


Fig. 3. SEM images of (a) iron nanomaterial synthesized using a Green tea extract. (b) Iron nanomaterial synthesized using sodium borohydride as a reducing agent. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

peninsula (Ganeshalingam et al., 2012). When the water level reached a maximum of 3.5 feet in Thondaimaanaaru, the gates are opened and excess fresh water released to sea during the rainy season. The Thondaimaanaaru barrage prevents the entering of seawater into the vadamarachi lagoon where as the Arriyalai barrage blocked the seawater entering into the Uppararu and Araly barrage allowsvallukai Aruoverflow water into the sea. The vallukaiaru is considered as the river for Jaffnaand active during the rainy season. In general, the salinity, TDS and EC are gradually reduced when move barrage to inside the land. It is remarkable in the vaddamarachi lagoon that mulli area consists dense of mangrove plants where the TDS, EC and salinity were low compared with other places. Hence, the mangrove plays a major role in balance the salinity in the land.

The groundwater of Jaffna greatly polluted by various activities such as uses of excess amount of artificial fertilizers, weedicides, insecticides, automobile waster oils, leaking of lubricant from fuel filling stations, excessive use of household cleaning chemicals (Navaratnarajah, 1994). Some fertilizers, weedicides and insecticides consist phosphate and nitrate counterparts which are the main contaminant in the Jaffna peninsula. We observed that the agricultural area (Manthigai) consist the higher amount of nitrate compare with highly populated Jaffna town area ponds such as Pullukulam, Ariyakulam and Kannathiddukulam. The phosphate content was lower in agriculture area than town area (Table 1). Jeevaratnam et al. reported that the nitrate concentration of Jaffna peninsula ranging 20–30 mg/L. But we found that the agriculture area hit up to 51 mg/L. The nitrates are readily soluble in water, cannot be retained by soil. Thus, it is easily reached groundwater and causes the various health issues. The SLS standard of phosphate is 2 mg/L. But the concentration of phosphate is higher in town area compare to agricultural area which indicates that the algal blooming prevails in town area ponds (Jeevaratnam et al., 2018). Hence, the regulations are needed about artificial fertilizers, insecticides, weedicides and etc. The government established many agricultural centres where inefficient staff fail to bring about side effect of excessive use of artificial elements in agriculture and government fails to control about imports of artificial

fertilizers. Our survey indicates that among the farmers of the peninsula don't have much knowledge about amount of the use of artificial elements in farming. Virtually they don't have the knowledge about diseases caused by the excess usage of fertilizers. The farmers purely inclined to get good yield from their product for which they unintentionally use artificial elements.

The average value of parameters of EC, salinity, turbidity and TDS are indicating that, it varies from place to place (Fig. 4a–d). The range of EC, salinity, turbidity and TDS were 0.34–18.03 mS/cm, 0.26–4.38 g/L, 0.54–5.94 NTU and 0.60–5.23 g/L, respectively. The Karaveddy, Pointpetro, Maruthankeny located near Vadamarachi lagoon whereas Chavakcheri and Kopay have a connection with uppararu. The vallukai Aru go through the area such as Thellipallai, Chankanai, sandilipay and Jaffna. Kytes, Vellanai, Delf and Karanagar mostly cover by saline water and near the sea among vellanaiarea showed higher deviation than other places. Hence, they show comparatively higher EC, TDS and salinity (Fig. 4a–d and Table 2). The water schemes like Vadamarachi lagoon, Uppararu lagoon and Valukkai Aru EC ranging 29.7–12.2, 30.5–4.5 and 38.1–2.0 mS/cm, respectively where as the corresponding salinity range is 20.2–6.4, 21.2–2.5 and 24.3–1.1 g/L, respectively. In the water schemes of Jaffna, the salinity is very high compared to the far away from the sea. There is some general question that some well near seaside has good quality water. This is because of the poor management of gates during the civil war, thus, the seawater enters into the land. Now the desalinization process is carried out by using surface water flow in the region. But it is a long-term process that the saline water bodies are being converted to freshwater lagoon which becomes surface water source for Jaffna. The pH of the groundwater seems neutral whereas Sri Lankan standard (SLS) lies between 6.5 and 8.5 (Mahagamage et al., 2019). Hence, pH of the surface water body in Jaffna peninsula is good. The EC is due to the dissolved ions such as Ca^{2+} , Na^+ , Mg^{2+} , CO_3^{2-} , NO_3^- and SO_4^{2-} and SLS is set it as 750 $\mu\text{S}/\text{cm}$ whereas WHO standard is 8.523 mS/cm (Mahagamage et al., 2019). The surface water and most of the dug wells are above the SLS standard except the Maruthankeny area and all areas found to be under WHO standard level (Fig. 4a and Table 2).

Table 1

The comparison of nitrate and phosphate in populated and agriculture area.

No	Parameter	Manthigaiv well	Pullukkulam	Ariyakulam	Kannathiddukulam	SLS Standard
1	Nitrate (mg/L)	51	0.70	0.70	0.70	10
2	Phosphate (mg/L)	1.3	2.30	3.80	3.0	2.0

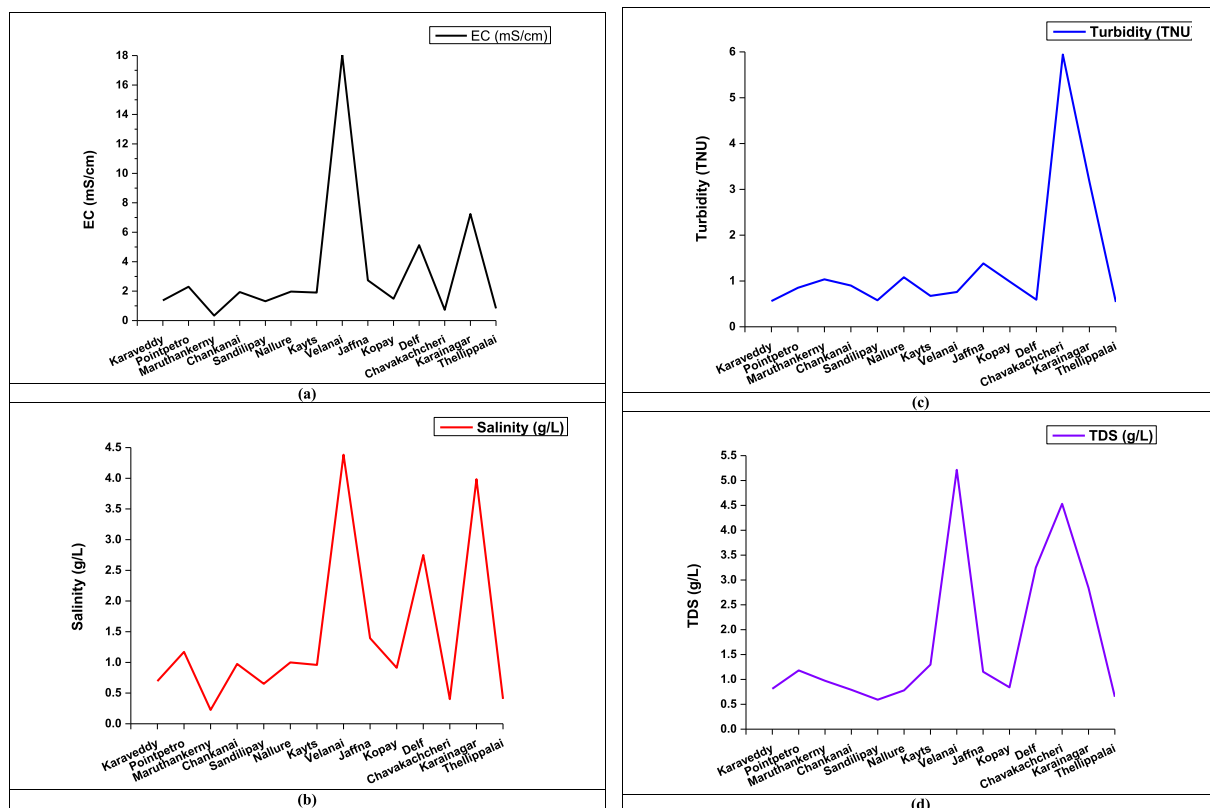


Fig. 4. The annual comparison of parameters in a different area of Jaffna peninsula (a) EC, (b) Salinity, (c) Turbidity and (d) TDS.

Table 2

Annual comparisons of different points in the water schemes of Jaffna peninsula.

Nos	Summary of Scheme	EC (mS/cm)	Salinity (g/L)	Turbidity (NTU)	TDS (g/L)	pH
1	Thondamanaru barrage	29.7	20.2	7.7	20.9	6.9
2	Vallai- Near the bridge	28.0	19.2	19.7	20.3	7.2
3	Yakkaru- Near the bridge	10.8	6.4	5.3	7.6	7.1
4	Mulli- Near the bridge	12.2	7.5	18.7	8.7	7.2
5	Ariyaalai barrage	30.5	21.2	2.7	25.2	7.0
6	A9- Road, Ariyaalai	4.5	2.5	2.4	3.1	7.2
7	Near Chemmany Road	10.1	6.1	2.2	6.8	6.7
8	Kopay- Kaithady bridge	17.5	11.1	3.1	12.4	7.2
9	Vannathypaalam	17.4	10.5	10.4	10.9	7.1
10	Surface water-Navaly	2.0	1.1	7.6	1.1	6.7
11	Kannagiamankovil	2.8	1.5	0.6	1.8	7.4
12	Navaly Madam well	2.9	1.5	1.8	2.2	7.1
13	Navaly cemetery well	9.5	5.4	3.4	6.2	7.1
14	Koddani channel	4.8	2.5	6.1	2.8	7.5
15	Valukaiaru surface water	21.2	13.9	2.0	15.8	6.9
16	Near Araly barrage cemetery well	38.1	24.3	2.1	24.1	6.7

The TDS is higher in coastal area due to presence of the sodium ion. The remarkable thing is that, the parameters like salinity, EC and TDS is lower than that of water schemes. This indicates that if the water schemes become freshwater body and the groundwater quality will be good and can be saved for future generation. The turbidity values varies place to place and that is depending on the place where the concentration of water body organisms, clouds of dust and other factors varies from place to place.

3.4. Cadmium and lead analysis in selected dug well in Jaffna

The samples were collected in the selected areas and the analysis done according to the auto lab standard procedure (Figs. S1–S8 and Tables S1–S3). The results indicates that the Pb majorly distributed than

Cd. The maximum amount of Pb concentration found to be as 0.132 mg/L and minimum concentration found to be as 0.0006 mg/L. The maximum and minimum concentration of Cd found to be as 0.092 mg/L and 0.0012 mg/L, respectively. The heavy metals cause serious health issue, as they interact with function of organs. The interaction caused malfunction of body and finally caused deadliest effect. The heavy metals enter into the body by food chains which are inevitable as heavy metals spread out by the various human activities. The Pb and Cd enter into the body by inhalation and swallowing as they are present in lead-based paints, cigarette smoke, household dust, potteries, lead-based pipe, fuels and various machines. The lead remarkably affects from fetal to adult in various level of concentration. The concentration below 50 ppb (5 µg/dL) causes decrease the intelligence quotient (IQ) level. The concentration of Pb in blood below 100 ppb (10 µg/dL) causes

reduction of hearing, IQ, postnatal growth and delayed puberty in child while in adults it causes tumour, increase blood pressure and hypertension (<https://www.niehs.nih.gov>, 2019). The Cd exposure is higher in Asian countries than Europe and United State of America. The Cd causes the chronic kidney diseases, mal-function of mitochondria, affect metabolizing of glucose and fat. Thus, this phenomenon lead to dead (Chunhabundit, 2016a). Nowadays, the commercials foods like carbonated water, coffee, rice, orange, chicken egg, Apple juice, oatmeal, corn, tomato, soya, wheat, animal products like beef and sea-foods and potato all consist particular amount of heavy metals such as Cd, Pb, Cr, As and organic substances like benzene, Diazinon and Chlorpyrifos. Therefore, modern people all around the globe, consume heavy metals and other stuff inevitably (Table 3) (Moschandreas et al., 2002, Chandorkar and Deota, 2013; Chunhabundit, 2016; Kim and Wolt, 2011; Tôth et al., 2016; Yuan et al., 2014).

3.5. Analysis of BTEX containing solution

The reaction process was carried out by using a special reactive vessel shown below (Fig. 5). The carbon electrodes were used for the electrolysis process. The anode (working electrode) was kept in modified activated carbon contained vessels which consist of many pores for the interchanging of electrolyte. The reactivity of modified activated carbon was tested by using 100 ppb BTEX standard solution with water (Fig. 5). During the reaction, the gas bubbles were observed in both electrode region. These gases were collected by using glass bottle and tested by using flame sticks. In cathode, the flame put off with pop sound, while in anode the flame burnt with shining which indicated that cathode releases the hydrogen while anode releases the oxygen. This reaction is similar to photocatalytic reaction (Raagulan et al., 2019; Gunaalan et al., 2018). The oxidation of BTEX occurred in the presence of active oxygen and iron nanoparticles which promote the oxidation via the free radical pathway and oxidation of BTEX were fastest at 900 mA. Moreover, 90% of BTEX vanished within 30 min at 900 mA. The electrolysis was done in the acidic medium (pH 5–6). Thus, cathode and anode produce hydrogen and oxygen respectively. The collision of oxygen with activated carbon surface generates reactive oxygen species because of the electron transfer from nanoparticle to oxygen. The electric field-induced this electron transfer process (Vithanage et al., 2014; Gunaalan et al., 2018). The resultant reactive oxygen species participated in the consecutive reaction.

The concentration of BTEX components gradually decrease and it is obvious that initial species converted into unknown species during the reaction. Even though, at first the BTEX species converted into at least one component of BTEX (Fig. 6 and Table 4). Ultimately, this leads to diminishing of the BTEX. Qualitatively the unknown compound can be

totally disappeared whereas similar trend showed by ethylbenzene, but, it was eradicated within 10min. In addition, benzene prevails for a longer period of time compared with other components. The xylene totally disappeared in 10 min. Hence, xylene species such as P/M Xylene and O-Xylene with an initial concentration of 13.59 and 9.21 ppb respectively are less stable under a given condition and those were removed within 10 min from the solution. The MTBE concentration remained unchanged up to 30 min and then disappeared in 40 min (Fig. 6 and Table 4). So, the persistence of the species in water can be arranged from lower to high as, following Xylene, Toluene, Ethylbenzene, MTBE and Benzene (Table 4 and Fig. 6). The schematic representation of the GC spectrum is available in the Supplementary MaterialsS1-S7. The mechanism proposed that anode produces oxygen molecules which makes the collisions with iron nanoparticle (INP) leads surface generating reactive oxygen species (radical) (Raagulan et al., 2019). The above reactive oxygen species is entered to the consecutive reaction and degraded BTEX into CO₂ and H₂O. The corresponding reactions are shown below (see Table 5).

3.6. Analysis of treated oil solution

Most of the used oil in Sri Lanka disposed directly or indirectly to the environment without following any regulation. As the rainwater or surface water seeped to underground through waste water-saturated soil. This natural process helps to reach oil content to the groundwater. As groundwater is consumed as portable water in Jaffna which affects the human dramatically. The engine oils consist inorganic elemental additives such as Fe, Pb, Cr, Sn, Cu, Mo, Ca, Mg, Ba, Al, Ti and Si. This concentration increases with the useage. This is considered due to the corrosion of engine parts. When engine under working the corrosive oxide formed in the oil. Due to the corrosive oil becomes dark in colour. Inorganic and phenolic antioxidative additives are added to improve function and life fan of the oil (Ahmad et al., 2018). Oils made up of 99.7% of aliphatic hydrocarbons, 0.27% of aromatic hydrocarbons and 0.03% olefinic hydrocarbon. The ratio between aromatic to aliphatic gets increase with the use (Kupareva et al., 2012). Further, the used oil consist carbonyl compounds and unsaturated hydrocarbons (Rudyk and Spirov, 2017). The BTEX which is naturally available in crude oil, released during volcanic eruption, forest-fire while man-made cosmetic, paints, adhesive, fuel station, petroleum refining, vehicle, cigarette and aircraft smoking also consist BTEX. World annually produce tones of petroleum products which is main cause of BTEX. The level of BTEX concentration varies based on the source and location, for instance, 4 g/L of benzene is available in crude oil whereas in 0.8 ppb of benzene found in seawater where petroleum is naturally deposited. The concentration of Benzene, Toloene, ethyl benzene, Xylene in Industrial

Table 3

Cd and Pb intake percentage of different country people.

Country	Cd (%)	Pb (%)	Ref.
USA	67.69	82.42	30
Korea	92 (Rice, fruits, vegetables, fish, meat)	*	31
Spain	74 (Fish, fruit, vegetable and cereal)	*	32
China	95.1 (Rice, meat, vegetable, flour)	*	32
Vietnam	90 (Rice)	*	32
Europe	56.1 (grain-related products, vegetables, starchy roots and tubers)	*	33
Hong Kong	88 (Vegetables, seafoods, cereals, mixed food)	88 (Vegetable, beverage, mixed dishes, cereals, seafood, fruits, meat)	34
India	79 (Fruits, vegetables, curd, cereals, roots and tubes)	91 (cereals)	35

*-considered as that there is no data for the corresponding place.

compared based on the height of the peak, varies with the time that implies the concentration changing during the treatment. The initial concentration of the benzene is 3.55 ppb which hits up to 5.96 ppb at 10 min then it gradually decreases with time and after 50 min benzene

centre with high traffic density is 349, 1310, 360 and 775 µg/m, respectively. In generally, BTEX is available in groundwater in very little percentage which is applicable to contaminated surface water where toluene is difficult to find. If the groundwater gets contaminate with

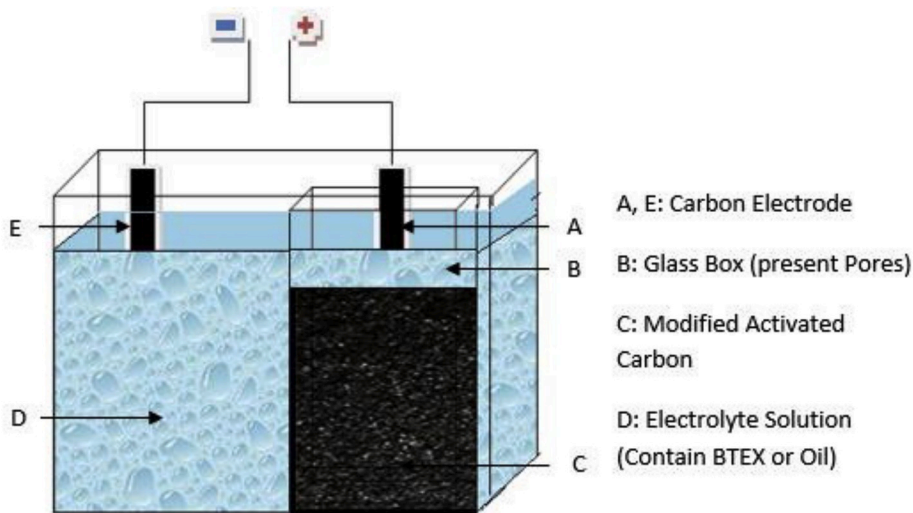


Fig. 5. Schematic representation of the electrolysis system.

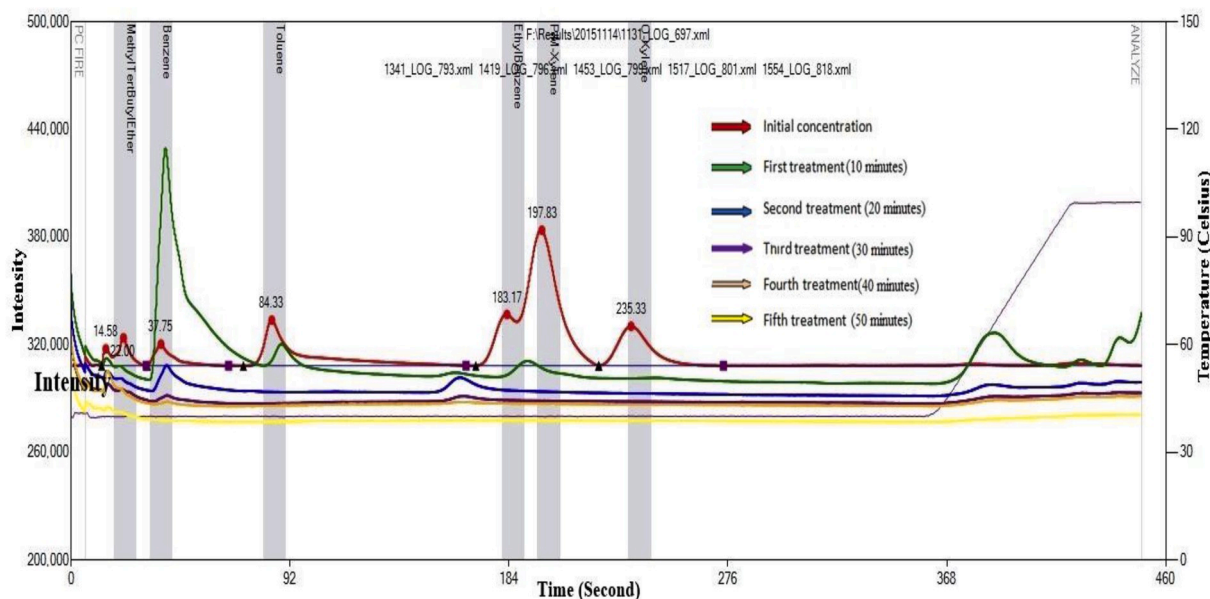


Fig. 6. Overlay of GC spectra taken after five treatments and initial solution.

Table 4

Overview of concentration of analytes from GC spectra.

Compounds	Initial concentration (ppb)	The concentration of the treated sample (ppb)				
		10 min	20 min	30 min	40 min	50 min
MTBE	0.42	0.04	0.04	0.04	Nil	Nil
Benzene	3.55	5.96	3.61	3.37	3.35	*
Toluene	1.61	1.42	*	*	*	*
Ethylbenzene	3.88	3.92	*	*	*	*
P/M Xylene	13.59	*	*	*	*	*
O-Xylene	9.21	*	*	*	*	*

“*” indicate that the corresponding component is not detected at the condition used to analysis.

BTEX, the expected concentration will be as 330, 3500, 2000 and 1340 µg/m, respectively (Leusch and Bartkow, 2010).

Gasoline consists of 11–12% of toluene, 5% of benzene, 2% of ethylene benzene and content of xylene depend on the formula of the

fuel. Exposures to BTEX for a long period of time caused high health impact in human. Among BTEX the benzene is more toxic and even higher concentration (10, 000, 000 ppb) causes death. Long term exposure with low concentration causes headache, unconsciousness, leukaemia, dizziness and tremors while when benzene is swallowed creates the symptoms like sleepiness, dizziness, vomiting, comma, rapid heart rate and death. The toluene can easily absorb by digestive system and it preferably reached kidney, brain, liver and adipose tissue which finally affect the nervous system. The ethylbenzene especially affects the hearing ability and enlargement of liver and kidney. The xylene can easily reach the body by inhalation which causes reduction of sense of balance, diminution of muscle coordination and affect the nervous system. The BTEX reaches water body by various sources like automobile, power station, fuel stations and industries. The organisms in the water bodies BTEX concentration above 1 mg/L acutely affect those organisms interact with water and BTEX stability also affect the water temperature (Leusch and Bartkow, 2010; Dórea et al., 2007; Bergerson et al., 2014). The methyl tertiary butyl ether (MTBE) is one of the contents added in

Table 5
Comparison of different BTEX standards in ppb (Dórea et al., 2007).

Nos	Standards	Benzene	Toluene	Ethylbenzene	Xylene
1	World Health Organization	10	700	300	500
2	Queenland public health regulation	1	800	300	600
3	United States National primary drinking water standard	5	1000	700	10,000 (total)
4	Australian drinking water guidelines	1	800	300	600
5	Australian and New Zealand environment conservation council environment protection guidelines	600	180	50	200
6	Queensland - environmental protection (air)	3	100	Not applicable	200
7	Queensland - Nantional environment protection (air toxic) measure	3	100	Not applicable	200

fuel to increase the fuel efficiency and its concentration varies from 84 to 234 ppm. One liter of used oil spoil 5000 L of potable water. The MTBE can easily dissolve in water than BTEX (Baker et al., 2002). Hence, disposal of waste oil in Jaffna peninsula completely destroy the underground water (Velauthamurty and Kurukulasuriya, 2016).

Initially, MTBE and BTEX is available in the sample and the peaks slightly shifted towards the left-hand side with the increasing heights along with the time. The fluctuation of the concentration of the BTEX species with time considered as, that the one species converted into another. The toluene concentration upsurge greatly whereas others significantly decline, and the concentration of the benzene almost remains constant (Fig. 7 and Table 6). After 20 min (Table 2), Benzene concentration hike drastically and others diminish which indicate that the benzene derivatives were converted into other forms and finally disappeared. Further, benzene is most stable than others (Fig. 7 and Table 6). The spectra showed that there is fluctuation in the concentration of unknown species such as MTBE, which formed during the

reaction. The benzene showed a similar trend in oil sample like in BTEX containing solution whereas MTBE exhibit strange behaviour which is considered due to presence of metals in oils and as the MTBE solubility is high it persists for a long time. The MTBE almost persisted for a longer time at the experimental condition and the Xylene compounds were absent in oil sample. Stability of compound can be arranged like Xylene, Ethylbenzene, Toluene, Benzene and MTBE. Spill of fuels on the ground and leakage of from underground fuel storage tank are the main reason for the contamination. The fuel station leak reached the dug well, which exhibit the fuel like ordos and oily layer on the top of the well. The result is miraculous only MTBE is present with the concentration of 92.08 ppb in the contaminated water. Further other parameters like EC, TDS, turbidity and fluoride were 1.297 mS/cm, 842 mg/L, 3.8 NTU and 0.2 mg/L, respectively. We consider that these factors also affect the prevailing of the BTEX in dug well. The schematic representation of the GC spectrum is available in the Supplementary Materials (Figs. S10–S23 and Tables S6–S19).

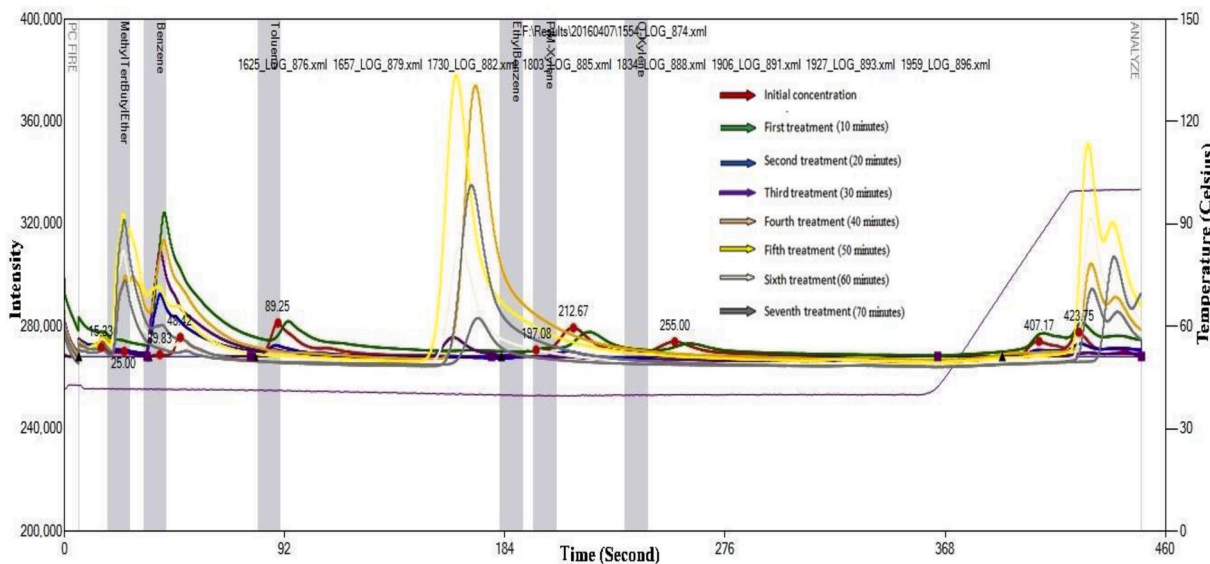


Fig. 7. Overlay of GC spectra taken after seven treatments and initial solution.

Table 6
Overview of concentration of analytes from GC spectra.

Compounds	Initial concentration (ppb)	The concentration of a treated solution (ppb)							Contaminated dug well
		10 min	20min	30min	40min	50min	60min	70min	
MTBE	0.03	0.02	0.01	Nil	0.31	0.71	1.3	0.94	92.08
Benzene	3.34	3.32	4.51	5.37	4.54	3.41	*	*	*
Toluene	2.63	1.48	1.28	1.23	*	*	*	*	*
Ethylbenzene	12.92	12.89	*	*	*	*	*	*	*
P/M Xylene	*	*	*	*	*	*	*	*	*
O-Xylene	*	*	*	*	*	*	*	*	*

“*” indicate that the corresponding component is not detected at the condition used to analysis.

4. Conclusions

The nitrate concentration ranging from 0.7 to 51 ppm whereas the phosphate content ranging from 3.8 to 1.3 ppm in Jaffna peninsula. The average value of parameters like EC, salinity, turbidity and TDS are indicating that it varies from place to place. The range of EC, salinity, turbidity and TDS were 0.34–18.03 mS/cm, 0.26–4.38 g/L, 0.54–5.94 NTU and 0.60–5.23 g/L, respectively. The water schemes behave differently, as they showed comparatively higher EC, TDS and salinity. The water schemes like Vadamarachi lagoon, Upparu lagoon and Valukkai Aru EC ranging 29.7–12.2, 30.5–4.5 and 38.1–2.0 mS/cm, respectively whereas the corresponding salinity range is 20.2–6.4, 21.2–2.5 and 24.3–1.1 g/L, respectively. The maximum amount of Pb concentration was 0.132 mg/L and minimum concentration was 0.0006 mg/L whereas the maximum and minimum concentration of Cd was 0.092 mg/L and 0.0012 mg/L, respectively. The environmentally friendly iron nanoparticle successfully deposited on activated carbon. The deposited iron nanoparticle (INP) particle size is ranging from 40 nm to 110 nm, which included zero-valent iron and iron (II, III) oxide nanoparticles. The modified activated carbon act as an electrochemical catalyst offered surface degradation of volatile organic substance like BTEX. The volatile organic substances completely eradicated by the electrochemical system in 40 min. In addition, BTEX the MTBE more stable in water than BTEX. The oil contamination of water can be confirmed by presence of MTBE in water. This method is low cost and applicable all level of the wastewater treatment in Jaffna.

Author contributions

KV and RMGR designed the research proposal, conceived the structure of the manuscript and supervised; SE and KR performed most of the experiments and analyzed all experimental data; SE, KR and KV write the manuscript; KV and ES contributed with the discussions of experimental results. All authors contributed to the preparation and review of the manuscript.

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Declaration of competing interest

The authors declare no conflict of interest.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.gsd.2020.100362>.

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