

# Occurrence of Perchlorate in Drinking, Surface, Ground and Effluent Water from Various Parts of South India

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**Abstract-** In spite of widespread usage of perchlorate ( $\text{ClO}_4^-$ ) for different applications in India, the  $\text{ClO}_4^-$  being an environmental pollutant a complete survey study has not been reported yet. Present study investigates the occurrences of  $\text{ClO}_4^-$  in the drinking water, ground water, surface water and effluent water from various parts of south India. Water samples were collected from Ambattur, Bangalore, Chennai, Coimbatore, Ranipet, Tirupattur, Tirupati and Vellore and analyzed for  $\text{ClO}_4^-$  concentration using ion selective electrode method. Water samples further characterized by pH, salinity, oxidation-reduction potential, conductivity and total dissolved solids. The overall  $\text{ClO}_4^-$  concentrations ranges from 0 to  $93.5 \times 10^3 \mu\text{g l}^{-1}$  and the highest concentration was detected in real effluent samples from Vellore. Out of 130 samples analyzed, 46.92% water samples contain perchlorate level above  $0.10 \times 10^3 \mu\text{g l}^{-1}$  and 24 samples showed  $\text{ClO}_4^-$  quantity above  $0.70 \times 10^3 \mu\text{g l}^{-1}$ . Mean concentrations of  $\text{ClO}_4^-$  in drinking water, ground water, surface water and effluent water found to be 126.08, 830.61, 1110.0 and 15754.4  $\mu\text{g l}^{-1}$  respectively. The permitted level of  $\text{ClO}_4^-$  concentration in drinking water by US environmental protection agency is 4  $\mu\text{g l}^{-1}$  and irrespective of different sampling points chosen, reconnaissance data reveals that there is no remarkable difference in perchlorate contamination in drinking water samples.

**Keywords –** Perchlorate, pH, Oxidation-reduction potential, Salinity, Reconnaissance

## I. INTRODUCTION

Perchlorate ( $\text{ClO}_4^-$ ) is an inorganic chemical which exist naturally as well as manmade. Ammonium perchlorate is the most prevalent form and widely used in solid propellants for rockets, missiles, fireworks and explosives [1]. Perchlorate compounds are also used in air bag initiators for vehicles, chemical laboratories in analytical testing, production of matches etc [2]. Improper disposal of  $\text{ClO}_4^-$  containing chemicals are increasingly being discovered in soil and water. In 2005, United States Environmental Protection Agency (USEPA) reported that  $\text{ClO}_4^-$  contamination of soil and groundwater prevails at more than 150 separate locations in 25 different states in US [3]. It has been found in ground and surface water in number of states in US and in fact, many wells in California closed because of perchlorate contamination [1]. Due to the high mobility of  $\text{ClO}_4^-$  in water and soil, it is readily absorbed by agricultural produce and cattle exposed to  $\text{ClO}_4^-$ . As a result, significant levels of  $\text{ClO}_4^-$  have been detected in dairy products, vegetables and in breast milk [4-5]. The primary route of  $\text{ClO}_4^-$  exposure is through ingestion of water or food contaminated with  $\text{ClO}_4^-$  and besides skin absorption, direct inhalation can also be considered as a secondary route of exposure [6]. The presence of  $\text{ClO}_4^-$  ion interferes with iodine transport which in turn reduces thyroid hormone production and thus affects the normal metabolism in human body [7]. The adverse effects are predominant in pregnant women in developing fetuses (cretinism), infants and children which result in behavioural changes, delayed development and decreased learning capabilities in new born [8].

With increasing levels of  $\text{ClO}_4^-$  in soil and water, there is a need of well defined  $\text{ClO}_4^-$  standards around the world. As such, in India there is no such recommended standard for perchlorate present in the contaminated water or soil. The reference dose according to national academy of science review is that, daily ingestion of  $\text{ClO}_4^-$  up to 0.07  $\mu\text{g kg}^{-1}$  of body weight can occur without adversely affecting the thyroid and iodide uptake [9]. In 2006, the New Jersey department of environmental protection recommended that drinking water with  $\text{ClO}_4^-$  concentration above 5  $\mu\text{g l}^{-1}$  should be avoided [9]. Although, it is considered to be strategic chemicals, around this time the usage capacity in India (ammonium perchlorate experimental plant, Kerala) is approximately close to 300-500 tons per year [10].

To meet the rising demand and lack of paucity in identifying an alternative substance, it is challenging us to treat the contaminated sites and waste water polluted with perchlorate. Although the existence of more number of armament, cracker manufacture and perchlorate production facilities in India, the environmental contamination by perchlorate in water or soil has been received little attention by researchers. Very few reports show the information on presence of  $\text{ClO}_4^-$  in India, in which Kannan and his workers showed that the occurrence of  $\text{ClO}_4^-$  in water and human saliva samples collected from urban and rural locations (Tamil Nadu, West Bengal, Bihar, Maharashtra, Karnataka and Pondicherry) [11]. Another study states that the occurrence of perchlorate contamination in groundwater from 6 cities (Sivakasi, Chennai, Trichy, Mumbai, Madurai and Kolkata) in India [12].

The objective of the work described was to ascertain  $\text{ClO}_4^-$  concentrations in various water samples collected from various locations in south India. The study samples consist of drinking water, ground water, surface water and industrial effluent water. The water samples were filtered,  $\text{ClO}_4^-$  quantity measured and further characterized by pH, salinity, oxidation-reduction potential (ORP), total dissolved solids (TDS) and conductivity level.

## II. MATERIALS AND METHODS

### A. Study area and sample collection–

Water samples were collected from Tamilnadu, Karnataka and Andhra Pradesh during February-May 2011. Sampling location covers Ambattur, Bangalore, Chennai, Coimbatore, Tirupattur, Tirupati, Ranipet and Vellore. The water specimens were collected in sample bottles (100 ml volume) with no head space volume and stored at 4°C. The land area marked with the help of online map as shown in Figure 1. Overall 130 specimens were collected from 14 different locations and it comprise of drinking water-56.9%, ground water-21.5%, surface water-3.1% and effluent water-18.5%. The drinking water samples consists of bottled water collected from public places (road side shops, restaurants, bus stand, railway station, temple, church etc.), well water and bore well water from houses and apartments. In addition to that it also includes the drinking water distributed through municipality after water treatment. Ground water samples were collected from bore wells, public wells and tube wells. Surface water samples were collected from natural sources like rivers, lakes, local water deposits and ponds. The industrial effluent samples were collected from outskirts of various industries (explosive, steel, leather, tannery and textiles manufacturing) located in Ambattur, Ranipet, Tirupattur, and Vellore. Standard sampling procedure was followed during sample collection [13] and effluent samples observed to be brownish-black in colour with pungent smell.

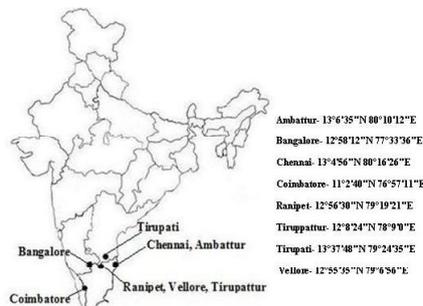


Figure 1. Map of India showing water sampling locations chosen for reconnaissance study during February-May 2011

### B. Sample preparation and parametric measurement–

The pH and ORP of the samples were measured at the sampling site by using a pH meter (Hanna digital pH meter, model-671p) and an ORP tester (Eutech, range: -999 to +1000 mV). Further the samples were withdrawn by using a sterile syringe and measured the salinity, conductivity and TDS by using a salinity tester (Eutech, range: 0-999  $\text{mg l}^{-1}$ ), conductivity tester (Eutech, range: 0-199.9  $\mu\text{S cm}^{-1}$ ) and a TDS tester (Eutech, range: 0-99  $\text{mg l}^{-1}$ ) respectively. Instruments were calibrated, and the sensors were washed thoroughly with deionized water and placed in the respective recommended dipping solutions at room temperature.

### C. Perchlorate quantification–

A standard operating procedure was used for  $\text{ClO}_4^-$  quantification in water samples by using perchlorate ion selective electrode method [14]. The water samples (75 ml) were filtered by vacuum filtration (vacuum pump: oil free mini 15-75 LPM, Riviera) using 0.45  $\mu\text{m}$  cellulose nitrate membrane filters (47 mm D) and the  $\text{ClO}_4^-$  quantity measured by using an Orion ion analyzer (Thermo Fisher Scientific, range: 0-19,900  $\text{mg l}^{-1}$ , accuracy  $\pm 0.001$ , Orion, USA). Ion analyzer consists of a perchlorate ion selective electrode (Thermo Fisher Scientific, cat.no.938101, range:

0.70-99,500 mg l<sup>-1</sup>, Orion, USA) and a double junction reference electrode (Thermo Fisher Scientific, cat.no.900200, Orion, USA). Before sample analysis, calibration was carried out by using known perchlorate standards (0.1, 1.0, 10 and 100 mg l<sup>-1</sup>) added with perchlorate ionic strength adjuster (cat.no.930711, Orion, USA). Fresh standards were prepared for each sample analysis. A continuing calibration verification standard was analyzed after every 10 samples. The analytical method should exhibit minimal interferences for the species examined, sample analysis was carried out in triplicates and the mean value reported.

### III. EXPERIMENT AND RESULT

The water samples were characterized by quantifying the pH, salinity, ORP, TDS, conductivity and perchlorate level. The obtained values were compared with the respective standards and discussed below.

#### A. Effect of pH-

Since the pH is a major factor which affects the bio-degradation of perchlorate under real field conditions. Therefore, the pH of all samples checked and showed in Table 1. Most of the industrial effluent and contaminated sites contains number of toxic chemicals which makes a long variation in the pH. Studies have been proved that perchlorate degradation occurs throughout the pH from 5.0 to 9.0 and the optimum reduction observed under alkaline range (7.5-8.5) [15]. The reconnaissance data reveals that the effluent samples found to be highly acidic in nature compared to other water samples. Overall, it varies from 4.1-8.96 as shown in Table 1. The recommended pH level in surface water and ground water is 6.5-8.5 [16]. Apart from effluent water, the pH of other samples found to be near neutral (7.0) and observed to be within the recommended limit.

Table -1 Summary of various parametric characteristics of water samples collected from Tamil Nadu, Karnataka and Andhra Pradesh in India

Samples	pH	Salinity (g l <sup>-1</sup> )	ORP (mV)	TDS (g l <sup>-1</sup> )	Conductivity (mS cm <sup>-1</sup> )
DW	6.93-8.46	0.214-0.87	28-146	0.264-1.23	0.236-1.733
GW	7.13-8.3	0.234-0.801	37-143	0.367-1.14	0.489-1.561
SW	7.1-8.96	0.258-2.81	22-102	0.567-3.68	1.037-5.230
EW	4.1-8.32	0.337-6.11	-12-158	0.483-2.89	0.760-37.6

DW-drinking water, GW-ground water, SW-surface water, EW-effluent water

#### B. Effect of salinity-

The salinity of the water samples were measured and found to be in the range of 0.214 to 6.11 g l<sup>-1</sup>. Studies shows that the ability of perchlorate respiring microorganisms to grow under high salinity (>4% w v<sup>-1</sup> NaCl) conditions appears to be limited [17]. The salinity variations observed in drinking water, ground water, surface water and effluent water samples were summarized in Table 1. The bacteria responsible for reductive respiration of perchlorate are active below 2% w v<sup>-1</sup> NaCl and saline tolerance is found to be a limiting factor in the biodegradation mechanism. The effluent samples were found to be more saline (6.11 g l<sup>-1</sup>) and it may raises the groundwater levels in the ecosystem. The perchlorate contamination from industrial washout brings up the dissolved salts level in water sources and gets stored in the ground water. As a result, the pH tends to be more acidic [18] and it will increase the hardness of ground or surface water. When the salinity increases the oxygen solubility decreases, thereby it adversely affects the solutes and other aquatic organisms present in the environment. We speculate that, the rise in salinity level in the water bodies is mainly due to the exposure of industrial effluent washout into various water sources. This will incur significant impacts such as reduced crop yields and at higher concentrations over long periods, can harmful to plants/microorganisms which may helps to degrade perchlorate in the environment. The present report concludes that the effluent water needs to be treated further before it reaches to the environment.

#### C. Oxidation-reduction potential-

The perchlorate biodegradation predominant under oxygen free environment, therefore, the redox potential was analyzed. ORP evaluates the effectiveness of water sanitation and the true measure of safe water. In this study, ORP was found to be within the range of -12 mV to +158 mV (Table 1). The ORP value may vary slightly when it gets exposed to the atmosphere. Therefore, to avoid the erroneous results, the ORP measurement was carried out at the sampling site. ORP represents the ability of water samples to either accept or donate electrons and expressed in millivolt. Results showed that apart from effluent water, other water samples exhibited positive voltage. The effluent water samples were negative (-12 mV) which indicates that the water molecules were more reductive. It may be due to lack of aeration in the industrial effluent. Therefore, sufficient aeration is required for effluent washout before it get exposed to the environment. The ORP values of drinking water samples were in the range of 28 to 146 mV (Figure 2)

and it reveals that samples were bacterial infected [19]. Previous studies showed that *E.coli* were inactive at reduction potential of 650 mV and yeasts and molds may require around 750 mV in order to be inactive in drinking water [19].

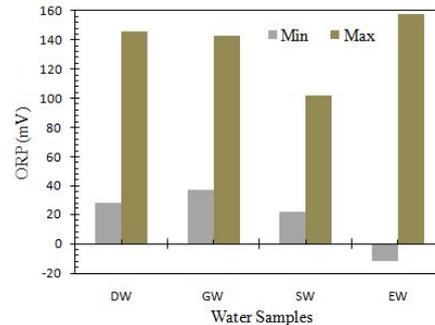


Figure 2. The deviations in oxidation-reduction potential observed in drinking water (DW), ground water (GW), surface water (SW) and effluent water (EW) samples. Min-minimum, Max-maximum

#### D. Total dissolved solids in water samples–

TDS is not considered as a primary pollutant, it is an indication of aesthetic characteristics of drinking water. The variation in TDS was found to be within the range of 0.264 to 3.68 g l<sup>-1</sup> (Table 1). The maximum contamination level set by USEPA for palatable drinking water is 0.5 g l<sup>-1</sup> TDS and in fresh water it should be less than 1.5 g l<sup>-1</sup> [20]. The survey data contemplate that the drinking water and ground water samples were fresh and observed no remarkable variation in TDS. However, surface water and effluent water samples showed an increase in TDS level and it was more predominant in surface water (3.68 g l<sup>-1</sup>) as depicted in Figure 3. This is may be due to leaching of soil contaminant and effluent discharge from sewage treatment plants to nearby water bodies. In fact, the effluent samples showed comparatively higher TDS (2.89 g l<sup>-1</sup>) reason being, the presence of various organic and inorganic substances present in the effluent and also may be due to the presence of potassium, chlorides, sodium, lead, arsenic, cadmium and nitrate dissolved in water.

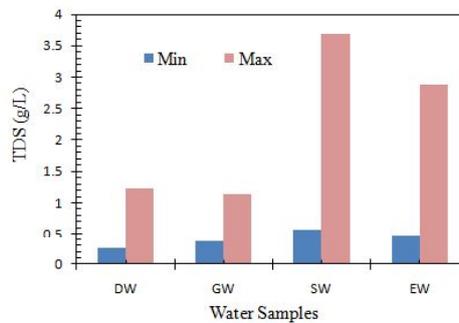


Figure 3. The variations in total dissolved solids observed in drinking water (DW), ground water (GW), surface water (SW) and effluent water (EW) samples. Min-minimum, Max-maximum

#### E. Conductivity level–

Conductivity depends upon the presence of total ionic concentration, mobility, valence and temperature measurement. The difference in conductivity level observed in all water samples as showed in Figure 4, however, it was found to be in the range of 0.236 to 37.6 mS cm<sup>-1</sup>. The effluent water had the maximum conductivity (37.6 mS cm<sup>-1</sup>) as compared to other water samples, reveals that may be the presence of toxic chemicals, heavy metals or other non-biodegradable substances present in the effluent rises the conductivity. The least conductivity was observed in bottled drinking water (0.236 mS cm<sup>-1</sup>) followed by in ground water and then in surface water (Figure 4). The potable water recommended with a conductivity level lie within the range of 0.05 to 1.5 mS cm<sup>-1</sup> and distilled water appears in the range of 0.0005 to 0.003 mS cm<sup>-1</sup> [21]. It was observed that the analyzed drinking water and ground water samples could be used for human consumption; however, the surface water samples need to be treated.

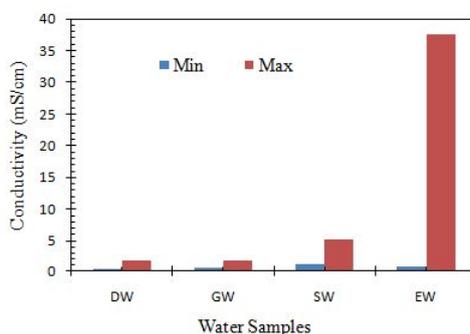


Figure 4. The difference in conductivity level observed in drinking water (DW), ground water (GW), surface water (SW) and effluent water (EW) samples. Min-minimum, Max-maximum

#### F. Perchlorate contamination in water samples–

The  $\text{ClO}_4^-$  concentration in water samples collected from various parts of Tamil Nadu, Karnataka and Andhra Pradesh is summarized in Table 2. The effluent samples showed the maximum  $\text{ClO}_4^-$  contamination and least observed in drinking water with mean value of  $126.08 \mu\text{g l}^{-1}$ . Among the 130 samples were analyzed, the mean perchlorate concentration found in drinking water, ground water, surface water and effluent water as follows: 126.08, 830.61, 1110.0 and  $15754.4 \mu\text{g l}^{-1}$  respectively as depicted in Figure 5. The effluent samples collected from various places, the maximum contamination was detected in Vellore ( $93.5 \times 10^3 \mu\text{g l}^{-1}$ ), and this may be due to improper disposal of industrial waste and effluent discharge in Vellore district, Tamil Nadu. Further, the effluent specimens from Tirupattur showed considerable level of perchlorate contamination ( $22.38 \times 10^3 - 1.7 \times 10^3 \mu\text{g l}^{-1}$ ), followed by in Ranipet:  $10.0 \times 10^3 \mu\text{g l}^{-1}$  and Ambattur:  $8.79 \times 10^3 \mu\text{g l}^{-1}$  (Table 2). The reason may be due to the source contamination of these watersheds in the tannery industries, high density livestock, elevated use of ammonium perchlorate as fertilizer in crop cultivation and unusual crop farming practices. Apparently, leading to bioaccumulation in plant species and indirectly it reaches to the human as showed in Figure 6. The decrease in perchlorate level in surface, ground and drinking water may be due to various factors such as simple dilution, adsorption to plant or microbes and microbial degradation mechanism [4, 17].

Table -2 Overall perchlorate concentrations in drinking water, ground water, surface water and effluent water samples from India as stratified by state and city

State/City	Perchlorate concentration ( $\mu\text{g l}^{-1}$ )				
	N	Min ( $\mu\text{g l}^{-1} \times 10^3$ )	Mean	SD	Max ( $\mu\text{g l}^{-1} \times 10^3$ )
<b>Tamil Nadu</b>					
Vellore (south)	9	0.0	32.08	42.8492	93.5
Brahmapuram (VIT campus)	20	0.0	0.04975	0.1084	0.65
Katpadi	5	0.027	0.8294	0.7845	2.1
Muthamil Nagar	3	0.17	0.2533	0.0737	0.31
Thorapadi (CMC campus)	3	0.13	0.41	0.2426	0.56
Muthyl Pettai	2	0.0	0.125	0.1767	0.25
Ranipet	10	0.12	2.958	3.1802	10.0
Vellore (north)	4	0.0	0.3318	0.4122	0.9
Chennai	11	0.0	0.08118	0.1448	0.43
Coimbatore	12	0.0	0.07408	0.0822	0.23
Tirupattur	12	0.0	5.7903	13.0055	41.7
Ambattur	7	0.0	1.9414	3.2896	8.79
<b>Karnataka</b>					
Bangalore	28	0.0	0.10957	0.1902	0.68
<b>Andhra Pradesh</b>					
Tirupati	4	0.0	0.0325	0.0275	0.06
<b>Overall</b>	<b>130</b>	<b>0.0</b>	<b>3.1933</b>	<b>14.2873</b>	<b>93.5</b>

N-Number of samples, Min-Minimum, Max-Maximum

Amongst the analysed 130 samples, the bottled drinking water showed the least perchlorate contamination with mean value less than  $2.0 \mu\text{g l}^{-1}$  (below the detection limit) which is recommended for human consumption. However, Perciasepe's experiment has reported that drinking water with  $> 5 \mu\text{g l}^{-1}$  perchlorate should be avoided [9]. It is noteworthy that 55.4% of the analyzed drinking water samples showed perchlorate contamination above the permitted level ( $5 \mu\text{g l}^{-1}$ ) and it suggests that further epidemiological investigation on human health effects is required. In fact,

long term consumption of low level  $\text{ClO}_4^-$  could causes serious health implications to infants and adults. Reports from India has shown that perchlorate concentrations in water samples are one to two orders of magnitude lower (drinking water- $0.1 \mu\text{g l}^{-1}$ , ground water- $1.0 \mu\text{g l}^{-1}$ , bottled water-  $0.02 \mu\text{g l}^{-1}$ , surface water- $0.05 \mu\text{g l}^{-1}$  and rain water-  $0.02 \mu\text{g l}^{-1}$ ) than the concentrations reported in the US (bottled water- $0.07$  to  $0.74 \mu\text{g l}^{-1}$ , ground water-  $0.04$  to  $0.12 \mu\text{g l}^{-1}$  and rain water-  $0.005$  to  $0.102 \mu\text{g l}^{-1}$ ) [11]. The bottled drinking water results were equivalent to that of the reported data and variations were observed in ground and surface water samples. This may be due to the difference in selection of sampling points, sampling time, weathering and influence of industrial waste discharge in India. Another study reported that perchlorate contamination was detected in groundwater samples ( $<0.005$ – $7700 \mu\text{g l}^{-1}$ ) in India and 41% of the samples contained perchlorate level above  $6 \mu\text{g l}^{-1}$  [12]. The maximum contamination was detected in Sivakasi ( $0.009$ – $7700 \mu\text{g l}^{-1}$ ) followed by Chennai ( $0.005$ – $24 \mu\text{g l}^{-1}$ ), Trichy ( $<0.005$ – $46 \mu\text{g l}^{-1}$ ), Mumbai ( $0.029$ – $0.09 \mu\text{g l}^{-1}$ ), Madurai ( $<0.005$ – $3.5 \mu\text{g l}^{-1}$ ) and Kolkata ( $<0.005$ – $0.09 \mu\text{g l}^{-1}$ ).

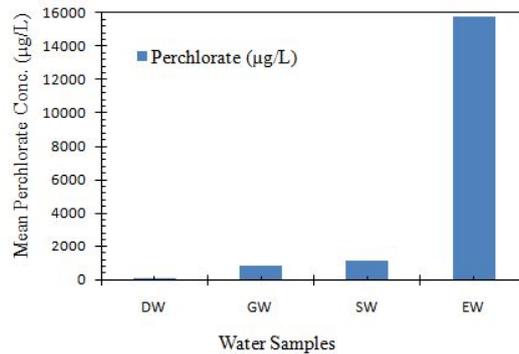


Figure 5. The overall variations in mean perchlorate concentration observed in drinking water (DW), ground water (GW), surface water (SW) and effluent water (EW) samples from India

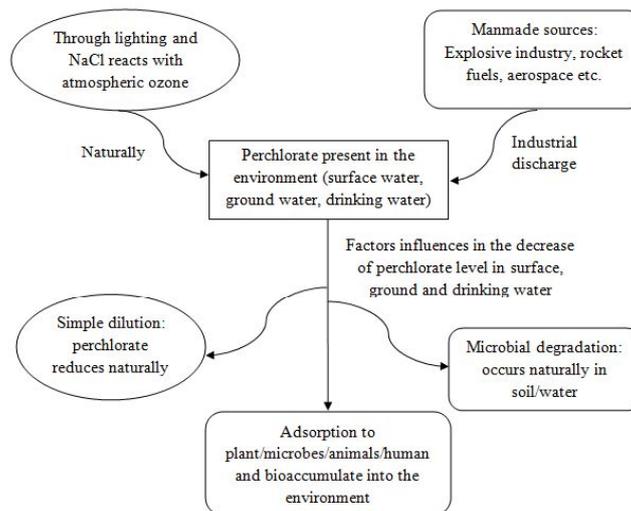


Figure 6. Generalized flow diagram representing the natural distribution of perchlorate present in the environment

#### IV. CONCLUSION

The work conducted for reconnaissance has culminated new information to understand the low level of  $\text{ClO}_4^-$  present in south India and the major source of contamination was industrial effluent discharge. The high density livestock and emerging industrial exposure in India makes the effluent water to become more saline ( $6.11 \text{ g l}^{-1} \text{ NaCl}$ ) and acidic (pH 4.1) in nature. Overall, there was no significant difference in  $\text{ClO}_4^-$  contamination observed in drinking water samples analyzed from all the sites. The increasing population has put a lot of stress in our natural resources and the incomplete combustion of various forms of perchlorate salt and the usage of other toxic chemicals lead to bioaccumulation of these contaminants into the environment. Wastes from the production, storage and improper

disposal of perchlorate containing chemicals has resulted in the contamination of drinking water, ground water and surface water at various places in south India. However, the biological degradation does occur under certain environmental conditions by natural microbes present in the soil and water. Therefore, it is necessary to create a passive remediation system at the contaminated site or we have to find an alternate substance for perchlorate in the nearby future.

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