Development and application of a contaminant transport model for groundwater remediation and reservoir protection: a case study from India

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Abstract The upper Kodaganar basin, located in Dindigul district of Tamil Nadu, India, is composed of hard rock terrain. Groundwater is the major source of domestic and irrigation needs and it is being contaminated by tannery wastewater that is discharged into the nearby Sengulam Lake. The main aim of this work was to develop a contaminant transport model using the total dissolved solids (TDS) concentration of groundwater measured in the basin. The model was developed to predict the fate of contaminant in the aquifer. The TDS concentrations in the wells ranged from 249 to 20,120 mg/L, wherein extremely high values were observed in some of the severely contaminated wells. Three scenarios were proposed to predict the fate of the contaminant and to mitigate the effect of contaminant on groundwater receptors for the year 2020: scenario I: developed with the existing discharge conditions; scenario II: developed with discharge as per the standards; scenario III: developed with zero discharge. The results of this study showed that scenario III reduced the contaminated area from 12 km^2 to 6 km^2. The reduction in area for different concentration contours, namely 2000 mg/L, 5000 mg/L, 10,000 mg/L, and 15,000 mg/L, was 2 km^2, 0.5 km^2, 0.2 km^2, and 0.1 km^2, respectively, and the groundwater remediation was expected to take 2050 years. Hence, there is an urgent need for the application of clean and resource efficient technologies in process industries, and the implementation of suitable wastewater treatment technologies to prevent ground water pollution in the region.

Keywords Contaminant transport model · Model development · Total dissolved solids · Groundwater pollution · Aquifer rejuvenation

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Introduction

Water is the most essential element for the origin, survival, and sustenance of life in the world. In arid and semi-arid regions, groundwater is considered the only water resource for agricultural, industrial, and household activities/requirements because of the limited availability of surface water resources. In developing countries, the drinking water demand

in rural areas is usually met through the groundwater tapped from dug and tube wells. In India, ~ 85% of the rural population and ~ 48% of the urban population depend on groundwater for their livelihood (Khurana & Sen, 2008; WHO/UNICEF, 2012). For irrigation purposes, ~ 88% of groundwater from tube wells is presently being exploited (IDFC Rural Development Network, 2013). The escalating economy, increasing population, uncontrolled development of urban infrastructures, and improper waste disposal practices are the common challenges for the sustainable access to clean water. For example, at the global scale, the direct discharge of wastewater generated from human activities (both domestic and industrial) into rivers and the ocean is ~ 80% (UNEP, 2019).

The upper Kodaganar basin, located at Dindigul District, Tamil Nadu, India, is a hard rock terrain. The basin lies within the northern latitudes 10° 10′ 38″ to 10° 34′ 31″ and eastern longitudes 77°37′50″ to 78°10′47″. Groundwater is essential for the people in upper Kodaganar basin for their daily needs due to the scarce availability of surface water. The upper Kodaganar basin is an agricultural area that encompasses urban settlements and industrial activities that pose risk to the contamination of groundwater. The city of Dindigul is well known for its tanneries, and presently, there are 49 tanneries in operation, producing an estimated amount of 76,400 to 85,600 kg/day (Muthu, 1992). The tannery industry consumes large quantities of water and other chemicals for processing the hides and they discharge a major portion of the used water along with the dissolved chemicals as effluent. Tanning process uses chemicals such as common salt, chromium sulfate, sodium bi-carbonate, sodium sulfate, sodium carbonate, lime, liquors, vegetable oils, fat, and dyes, among others to process the hide. The total quantity of wastewater discharged for every 100 kg of skin and hide processed varies from 3000 to 3200 L (Mondal & Singh, 2010). Most of these tanneries are located along the Madurai, Battalagundu, and Ponmandurai roads (Mondal et al., 2005a). The tannery industries have been identified as the point source of contamination and they are well distributed in an area of 24 km^2 in the outskirts of Dindigul city. Most of these tanneries are present in agricultural zones, while some are also located nearby residential zones; thus, a mixed land use pattern can be observed in this region. These tanneries have been in operation for nearly four decades and

some of them have practiced uncontrolled disposal of wastewater (Mondal & Singh, 2009). Before the establishment of the common effluent treatment plant (CETP) in the year 1998, the tanneries discharged the chemical containing wastewater directly on the ground, and this has led to severe groundwater pollution and shown negative impacts on human health and the environment (Muthu, 1992). At present, the 49 tanneries discharge their wastewater through an underground conveyance pipeline to the CETP for treatment and the treated effluent is discharged into the nearby Sengulam Lake (Kanthasamy et al., 2014). Although the wastewater is treated, it is still contributing to groundwater pollution, soil quality deterioration, and aquifer damage. Hence, the wastewater discharged from CETP into the Sengulam Lake was the main source for the high total dissolved solids (TDS) concentration.

The urban settlements and the agricultural lands nearby the lake also consume large quantities of water and they depend on the same aquifer for groundwater. Therefore, there is an urgent need to manage the water resources in the region and protect the aquifer from further pollution. In this study, the persisting groundwater pollution caused by the tanneries was considered and the spatial distribution of groundwater was studied through a Geographic Information System (GIS) software (Jesudhas et al. 2021). QGIS is a free and open source software that can facilitate data preparation and provide visualization of the developed model (Park et al., 2019). The contaminant transport in groundwater was studied using the Visual MODFLOW software (Zhu et al., 2019). In practice, the complexity associated with building three-dimensional groundwater flow and contaminant transport models can be decreased using the Visual MODFLOW software (Kumar, 2013).

All the spatial and non-spatial data were collected and organized using the QGIS software. The organized data were analyzed in the QGIS by plotting the geographical coordinates (latitude and longitude) and other attribute features. The lithological data obtained from the bore holes, rainfall data, aquifer parameters, and the water table data collected from the Public Works Department (PWD) were prepared as maps using the QGIS software and were used as inputs for the model. The data prepared through the QGIS were preprocessed using the Visual MODFLOW software for model development. The results of the

model were exported to QGIS for data visualization. Groundwater head and velocity obtained through the simulated groundwater flow model was used to compute the TDS concentration in a mass transport model (Shwei et al., 2013). TDS and electrical conductivity (EC) are important water quality parameters that play a significant role in water resources management and health impact related studies (Al-Mukhtar & Al-Yaseen, 2019). To reduce the impact caused by water-related issues, assessment based on chemical, physical, and biological constituents is necessary and among these parameters, TDS is one of the most vital constituents for assessing the overall suitability and quality (Ewusi et al., 2021). Tamma Rao et al. (2013) has selected TDS as the parameter and applied flow and contaminant transport model as a management tool for the assessment of current/existing situation and forecasting the future responses of TDS concentrations. By considering all these previous literatures, TDS distribution was modeled in this study for predicting the migration behavior of the contaminant plume and this information can be used for effective management of the aquifer. Contaminant transport model scenarios were designed based on the hypothesis that the TDS concentration in the groundwater can be predicted and could support mitigation. The specific objectives of this work can be stated as follows: (i) to develop TDS-based contaminant transport model to simulate the fate of TDS in groundwater, and (ii) to apply the developed model and predict the fate of TDS under scenarios with an aim to facilitate aquifer rejuvenation.

Materials and methods

Study area and aquifer characteristics

The location of the upper Kodaganar basin is shown in Fig. 1. The geographical characteristics of the basin are presented in Table 1. The basin experiences semi-arid tropical climatic condition and is classified as an east coastal plain by the Indian Council of Agricultural Research (ICAR). The average annual rainfall in the district is 813 mm, contributing to a rise in the level of groundwater (0.02 to 0.59 m/year). The major soil types are black soil and red soil with texture ranging from clay to sand (CGWB, 2008). The upper Kodaganar basin has charnockite rock in the west

and the southern region and it is mainly composed of gneissic rock that is partially and highly weathered in the central basin that ends up in unaltered hard rocks. The composition of rocks is mainly attributed by gray and pink feldspar with quartz grains, biotite, and hornblende (Manivannan et al., 2011).

The depth of the aquifer extends up to the impermeable layer, while the weathered aquifer thickness varies from 2 to 30 m. This depth facilitates the storage and movement of water through the weathered and fractured zones of the aquifer. The shallow aquifers in the region are in phreatic condition and they are not a stable source when there is a surge in groundwater demand. Nevertheless, the deeper aquifers are partly confined, i.e., they are being recharged from the shallow unconfined aquifers through bore wells (Mondal et al., 2005b). The Kodaganar River is an ephemeral river and the flow of water in the river is mainly during the monsoon season. The surface runoffs from the west, east, and south are observed only during the rainy season. The water leaves the basin through the northern side. Aquifer recharge is mainly because of the infiltration of rainfall through the top soil layer.

Physico-chemical characteristics of groundwater and data integration

The physico-chemical characteristics of the groundwater were studied by collecting groundwater samples from the dug wells, bore wells, and dug-and-bore wells. The groundwater samples were collected from 37 wells which included dug wells, bore wells, and dug+bore wells for the year 2014. This data was helpful in assessing the spatial distribution and degree of contamination in the aquifer. Historical data were collected from PWD for a period of 13 years (2001 to 2013). During the years 2001 to 2004, there was a significant amount of missing data. As a result, this time period was excluded from consideration in our study. For this period, a number of dug wells for which the historical data of water level and water quality are available were 14 and 18, respectively. Therefore, only these wells during the period 2005 to 2014 were used for the development of the model. The bore wells and dug+bore wells were used to assess the status of contamination in the deep bore wells. Water level samples were available for every month for these years, i.e., 12 samples per year. Water quality samples were available with a frequency of 2 samples (during the
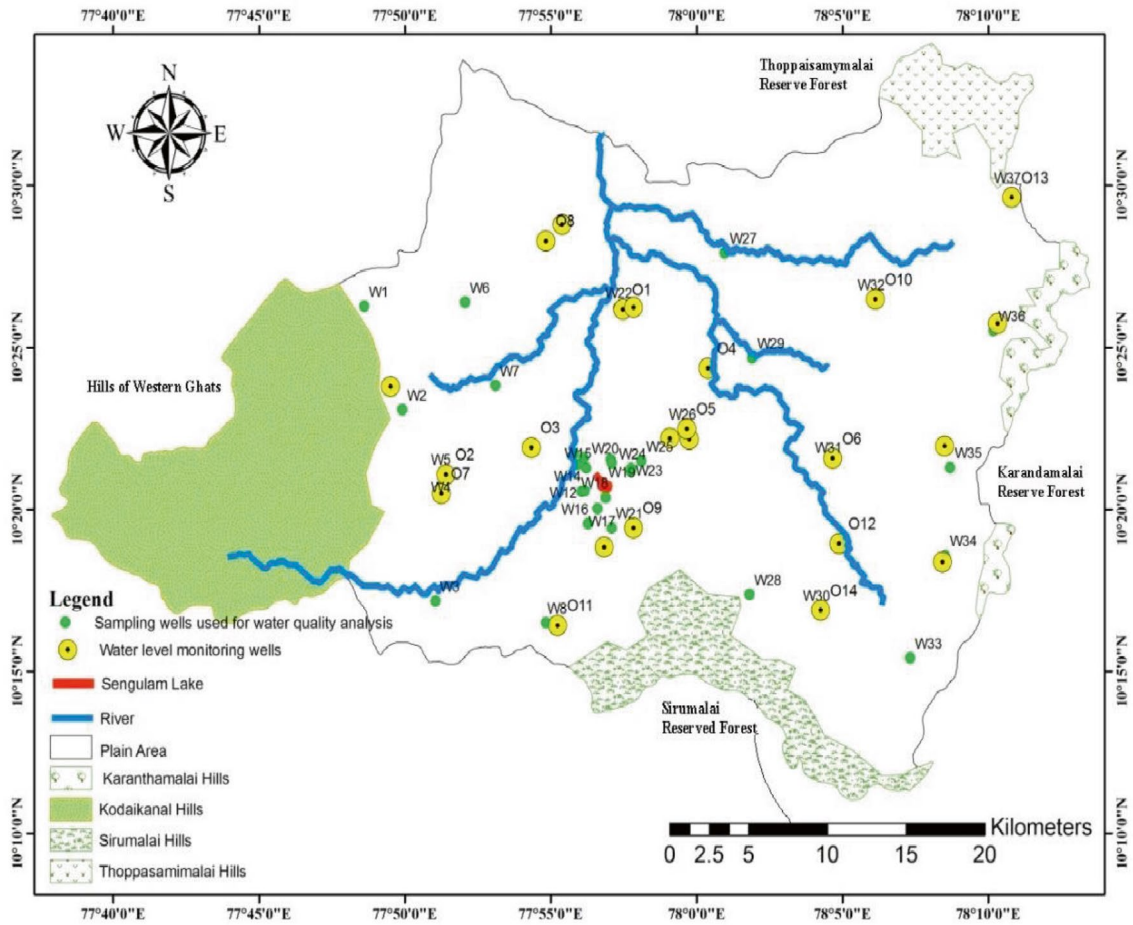
W1-Palakkanuthu	W2-Palayakannivadi W	W3-Athur	W4-Palayakannivadi S	W5-Periyakarisalpatti
W6-Gurunathanaickanur	W7-Puliarajakapatti	W8-Ambathurai	W9-Sukkanpatti	W10-Kuthathupatti
W11-PonnimanduraiSW	W12-Kuttiyapatti SW-UC	W13-Ponnimandurai	W14-Kuttiyapatti SW C	W15Kuttiyapatti W
W16-Pithalaipatti	W17-Pillayarnatham	W18-Pallapatti	W19-Muthalagupatti	W20-Paraipatti
W21- Vellodu	W22-Chettinaickanpati	W23-Begampur	W24-Mohamediapuram	W25-Dindigul
W26-Dindigul	W27-Nallamanarkottai	W28-Koovanuthu	W29-Tamaraipadi	W30-Sanarpatti
W31-Madur	32-Vadamadurai	W33-Vembarpatti	W34-Pudupatti	W35-Kambiliyanpatti
W36-Pudur	W37-Thangammapatti			
O1-Thadikombu	O2-Periyakarisalpatti	O3-Sinthalakundu	O4-Sellamanthadi	O5-Dindigul
O6-Madur	O7- Dharmathupatti	O8-Sukkanpatti	O9-Vellodu	O10-Vadamadurai
O11-Ambathurai	O12-Ragalapuram	O13-Thangammapatti	O14-Sanarpatti	O15-Silvarpatti
O16-Ammapatti	O17-Puthur	O18-NGO colony	O19-Dindigul-SW	O20-Thettupatti

W series – wells used for water quality assessment; O series – wells used for water level assessment

Fig. 1 Physiography of the upper Kodaganar basin, Kodaganar River, Sengulam Lake, and location of wells used in this study

first week of January (post-monsoon) and during the last week of May (pre-monsoon)) per year. There were few missing data and these missing data were filled by linear interpolation. The standard error was estimated

Table 1 Geographical characteristics of the upper Kodaganar basin

Sl. no	Characteristics	Details
1	Blocks in the Kodaganar basin	Dindigul
		Sanarpatti
		Athur
		Reddiyarchatram
2	Latitudes	10° 10′ 38″ to 10° 34′ 31″ N
3	Longitudes	77°37′50″ to 78°10′47″ E
4	Area	1168 km^2
5	Elevation	207 to 608 m AMSL
6	Average annual rainfall	813 mm
7	Major river	Kodaganar
8	Predominant Soil type	Red sandy soil and black cotton soil
9	Predominant rock type	Charnockite and granite gneiss
10	Weathered formation	2 to 30 m
11	Depth to water table	1 to 12 m
12	Hydraulic conductivity	0.05–6 m/day

for all the observations of water level in each well. The observations of all the wells were uniformly distributed and were within the standard error and at a significance level of 5%. The water in the Sengulam Lake was also sampled from different locations, i.e., from the north, east, west, and south directions. The exact location of the sampling wells was determined in the field with the help of a hand-held GPS instrument. The latitude and longitude information of the wells acquired in the field was imported into the QGIS software for spatial visualization and analysis. The location of the sampling wells used for hydro-chemical analysis is shown in Fig. 1. The methods of collection and analysis of water samples were done according to the standard protocol described by the American Public Health Association standards (APHA, 2012).

Development of the TDS fate transport model

The 3D modular finite difference groundwater flow model was constructed using the Visual MODFLOW software. MODFLOW can be used to solve the equations of groundwater flow in any aquifer-based system (Tavakoli-Kivi et al., 2019; Kirubakaran et al., 2018). This software can simulate both steady-state and transient-state flows of groundwater for different hydrological systems. It has been suggested that groundwater flow calibration has to be done with historical data of the water level (Sashikkumar et al., 2017). In this study, the flow model was developed for unconfined condition by following the standard modeling procedures commonly used for groundwater studies (Lachaal et al., 2012). The quality of water in the aquifer can be assessed by the anions and cations present in the groundwater. The effluents from the tannery industries usually contain a mixture of salts such as calcium, sodium, chloride, and sulfate, among others. To model the transport of these ions in the groundwater, every ion has to be treated as a species. However, the transport modeling of individual species is practically difficult because of problems arising in handling the data. Therefore, one representative parameter that could generalize these ions and provide a good representation of the quality of groundwater should be identified. The TDS concentrations reflect the anions and cations present in the water sample and therefore, this parameter can generalize the ions and represent the status of water quality in the study area. Besides, TDS concentration is a good representation of the salts filtering from the soil and other environmental pollutants being contaminated by the leachate (Wagh et al., 2016). Leaching of ions leads to an increase in the TDS content of groundwater and hence TDS is a good, representative indicator of the pollution level (Sharma et al., 2020). Therefore, in this study, TDS was selected for the contaminant transport model. The higher concentration of TDS in groundwater will influence the density and viscosity of the water (Ophori et al., 1998). For example, density and viscosity play a major role in sea water intrusion studies (Lee et al., 2019). The presence of these salts can vary the density and

viscosity of the contaminated groundwater. However, most of the studies conducted in the freshwater aquifer did not consider the density and viscosity in the contaminant transport model (Shwei et al., 2013; Tamma Rao et al., 2013). A model developed for the same area by Mondal and Singh (2009) has also assumed that TDS has no influence over the density and viscosity. The wells used for the calibration of the model had TDS concentration not exceeding 2000 mg/L. Therefore, the influence of density and viscosity was not considered in the study. The remediation in the aquifer contaminated by the miscible contaminants was done through the careful design and evaluation of different alternatives of remedial strategy. This model has the following assumptions: (i) the hydraulic characteristics of a grid cell does not vary within itself, (ii) the density and viscosity of water was not affected by the TDS concentration, (iii) the discrete measurements of parameters are representative values over a region and hence may vary from cell to cell, and (iv) a hard rock terrain advection is the most dominant than dispersion. Due to the poor network of the water quality monitoring wells, the historical data was not available for all the 37 wells, especially for the region in the vicinity of the Sengulam Lake where the primary data was collected. Therefore, the TDS concentration of the Sengulam Lake and observation wells measured for the year 2014 was used as the initial TDS concentration. Thus, for the development of the TDS fate transport model, only the wells with historical data were used. For the calibration of the model, the parameters such as specific yield and the hydraulic conductivity were used. For the additional inputs, the average conditions of parameters such as water recharge and pumping rate were assigned. Advection and dispersion were considered in this study, while the velocity vectors, i.e., magnitude and direction of groundwater flow, were used by the model for prognostics.

Model conceptualization

The conceptual model designed for groundwater flow modeling includes the model grid design, defined boundary condition, defined hydraulic parameters, assigned wells, sources and sinks, and the determination of flow directions and estimation of components of the groundwater budget (Anderson et al., 2015). The conceptual model involves a careful survey and interpretation of the data available over the previous years (Lasagna et al., 2020). The shape and size of the basin were used for the model grid design, while hydrogeologic setting was used for defining the hydraulic parameters, geologic setting, and recharge. The discharge sources were used for defining the boundary conditions, and the observation wells and pumping wells were used for the development of the conceptual model. The physical system was conceptually translated into a mathematical system using the Visual MODFLOW software. The governing equation for steady-state flow condition is presented in Eq. (1), while the equation for transient-state condition is presented in Eq. (2) (Harbaugh, 2010):

$$\partial/\partial x(K_x \partial h/\partial x) + \partial/\partial y(K_y \partial h/\partial y) + \partial/\partial z(K_z \partial h/\partial z) = 0 \tag{1}$$

$$\begin{aligned} \partial/\partial x(K_x \partial h/\partial x) &+ \partial/\partial y(K_y \partial h/\partial y) + \partial/\partial z \\ (K_z \partial h/\partial z) &\pm Q = S_s \partial h/\partial t \end{aligned} \tag{2}$$

where K_x, K_y, and K_z are values of hydraulic conductivity along x, y, and z coordinate axes, h is the hydraulic head, Q is a flux term accounting for recharge, pumping, and other forms of source/sinks, S_s is the specific storage, and t is the time. The WHS Solver that uses the Bi-Conjugate Gradient Stabilized (Bi-CGSTAB) acceleration routine was used to solve the differential equations in the MODFLOW environment.

Model grid design

The model grid design involved dividing the area into regular grids of appropriate size and shape. The average length and width of the basin were 42 km and 28 km, respectively. The model domain was divided into 80 columns and 60 rows with a grid spacing of 500×500 m. At the basin level, the plain considered in the study area was assigned as an active layer, while the rest of the regions were assigned as inactive. A two layer aquifer model was developed to simulate groundwater flow in the basin. Block-centered approach was adopted where the water table head was solved for the center of the grid cell. The Shuttle Radar Topographic Mission (SRTM) data (Fredrick et al., 2007) was imported as ground surface elevation to the model. The highly weathered layer with a thickness of 30 m was imported as the first layer into the model, i.e., from the bottom of the ground surface. On

the other hand, unaltered bed rock with mild fractures that extended up to a thickness of 200 m was assigned as the second layer. The equivalent porous medium (EPM) method was applied to model groundwater flow and weathered hard rock formation (Surinaidu et al., 2013).

Boundary conditions

The boundary conditions were fixed based on the physical boundaries envisaged in the study area. Charnockite was present in the western and southern sides of the basin. The eastern side of the basin was composed of quartzite rocks that ran from the north–south direction. All these three sides were covered by the igneous rock type and they were found to be elevated. Therefore, the Neumann's boundary with zero flux (no-flow boundary condition) was assigned to the south, west, and discontinuously at the eastern side of the basin. The regional slope of the topography was towards the northern side of the basin. The River Kodaganar flows through the northern side of the basin that was found to be a low-lying region, and therefore, the Cauchy or head-dependent boundary condition was assigned to the northern side of the basin. A numerical parameter conductance was used in the river boundary condition to simulate the resistance to flow from the surface water body to the groundwater. No-flow boundaries were established at the impermeable bed rocks located at the bottom of the aquifer's subsurface layer.

Hydraulic parameters

Hydraulic parameters were determined through pumping tests in few wells and for few wells, this data was obtained from the Public Works Department. These parameters are point estimates and they were used as the initial inputs to the model. The aquifer parameters, namely, the hydraulic conductivity, thickness, and connectivity, vary spatially and they were mostly unknown (Haitjema, 1992). The hydraulic parameters that varied over a region induced uncertainty in the model. It was therefore assumed that the discrete measurements were representative over a region and they were almost constant over the grid. The model was parameterized by interpolating the point estimates to represent the hydrological system. Hirata et al. (2020) used the parameters such as transmissivity, hydraulic conductivity, flow velocity, and

specific yield in the MODFLOW-based contaminant transport model. The hydrological parameters such as transmissivity (T), hydraulic conductivity (K), and specific yield (S_y) were assigned as discrete values in the model. These values were distributed in the basin after interpolation using the inverse distance weighted (IDW) method. As the IDW is a statistical approximation method, these parameters were also used for model calibration.

Wells, sources, and sinks

The wells for which the historical data of water level and water quality are available were used in this study. These data were available for the period ranging from 2005 to 2014 and therefore, this period was selected for model development. The number of wells that has the historical data of water level for this period was 14. The water level observation wells and the pumping wells were imported into the model. The initial head of the wells was assigned to the model domain based on the water table data collected from the PWD (i.e., during 2005). The extinction depth was assigned as 3 m, wherein the water table is below this depth in most of the months of a year. All the surface water features other than Sengulam Lake are hydraulically separated from the groundwater system by an unsaturated zone and have little or no direct influence on the groundwater flow. Through the unsaturated zone at Sengulam Lake, there was a significant interaction between the surface water and the underlying groundwater in the lake. Head-dependent boundaries were assigned to the lake as the treated water is discharged into the lake. The interaction between the surface and groundwater was characterized by leakance. After the input of all the required well data, such as well screen and water level, was obtained, the model was run to compute the head (h_c) under steady-state condition. The computed head (h_c) should agree with the observed head (h_o). The difference between the heads is termed as the residuals, i.e., $\Delta h = h_o - h_c$. Since the values of residuals were high, steady-state calibration was attempted to minimize the residuals and to optimize the parameters (Hill & Tiedeman, 2007).

Calibration of the model

The parameter values such as hydraulic conductivity, storage, and effective porosity were used as the initial

parameter estimates during the start of model calibration. The hydraulic boundaries delineated by the contour lines and the stream lines were used for steady-state calibration. The output from the steady-state calibration was used as the initial condition for the transient-state calibration. The steady-state model was extended for the transient-state conditions by incorporating the historical data of water level for a period of 10 years, i.e., from the year 2005 to 2014. The period of study was stratified into stress period (~3 months) that resulted in 40 time steps. The pumping well locations were introduced in the model based on the position of the well in a projected coordinate system. The pumping wells were introduced based on the consumptive requirement of urban and irrigation areas. The pumping rate (m^3 per day) was used for representing the pumping scenario in the model domain. The domestic water consumption through the pumping wells in the urban area was estimated by the product of the population of the village and their per capita demand. Irrigation water demand was estimated by the product of crop water requirement and the area irrigated. Recharge to the aquifer was estimated based on the Soil Conservation Service–Curve Number (SCS–CN) method (Satheeshkumar et al., 2017). The model was executed to compute the water level for a period of 10 years (i.e., from 2005 to 2014). This data was used for the calibration of the transient-state model. The convergence criterion for the solution was based on the changes in the head and the residual criteria. In this study, PEST, a non-linear least squares inverse model calibration, was used (Ezzy et al., 2006). The PEST optimization program can be utilized in a zone-based approach for calibrating the hydraulic conductivity (Teimoori et al., 2021). The sensitivity analysis can be carried out using different parameters such as hydraulic conductivity, vertical anisotropy, specific yield, specific storage, and recharge (Tolley et al., 2019). The maximum number of outer iterations used for the convergence was 25. The maximum number of inner iterations used for the convergence was 10. The head change and the residual criteria used for the convergence were 0.01 m. The residual calculation and optimization procedure of transient-state calibration were similar to those of the steady-state calibration.

Development of the groundwater quality model

There were 18 wells with data on water quality during this time period, and these wells were utilized to create the model that was used to predict water quality. The information about the water quality observation wells were imported into the model. The initial concentration was assigned to the model domain based on the collected TDS concentration data, i.e., from the year 2005. After the input of all the required parameters, the model was run to compute the TDS concentration (TDS_c) under steady-state condition. The computed TDS concentration is expected to agree with the observed TDS concentration (TDS_o). The difference between the concentrations was termed as the residuals, i.e., $\Delta TDS = TDS_c - TDS_o$. Whenever the values of the residuals were high, the steady-state calibration was carried out to minimize the residuals and to optimize the parameters. PEST optimization function was used to minimize the Δh and ΔTDS values by adjusting the hydraulic parameters. The 3D Mass Transport in three-Dimension (MT3D) package was used to simulate the advection and dispersion in groundwater flow systems. The TDS introduced as a contaminant in the Sengulam Lake was treated as a point source. To track the path followed by the contaminant, the particles were introduced in the lake and the MODPATH package was used to simulate the path traveled by the particle in the groundwater system.

Simulation for prognostics and mitigation: scenario development

The developed TDS transport model was used to simulate different scenarios in an attempt to protect the receptor systems, i.e., the water resources (the aquifer and the Kodaganar River) and water source (pumping wells) in the basin. In order to protect the receptor systems that could be possibly contaminated by the pollutants from the tanneries, three scenarios were developed to simulate and predict the fate of the TDS in the groundwater system. Scenario I predicted the fate of TDS in the groundwater receptor system when the same TDS concentration was allowed to continue. Scenario II predicted the fate of TDS in the groundwater receptor system, when the TDS concentration was maintained as per the standard. Scenario III predicted the fate of TDS in the groundwater receptor system when the TDS concentration was maintained at zero. All the prognostics were attempted with an assumption of the existence of average conditions of natural events, i.e., rainfall, evapotranspiration, and pumping. A regression model was developed to

predict the area affected by different concentration distribution for the mentioned discharge condition in a view to protect the receptor systems in the basin.

Results and discussion

Physico-chemical characterization

The results of physico-chemical parameters measured from the different groundwater samples during the pre-monsoon and post-monsoon seasons of the year 2014 are listed in Table 1. These parameters were useful in determining the quality of the groundwater in the aquifer beneath the basin. The major parameters such as TDS, EC, pH, calcium, magnesium, sodium, chloride, sulfate, carbonate, bicarbonate, nitrate, and potassium were measured. These parameters were also considered for determining the ionic balance, i.e., the cations and anions, in the water samples. Every groundwater sample was subjected to an estimation of the error of ionic balance, and the error was found to be within acceptable limits (< 5%). Electrical conductivity, an indirect measure of ionic strength, was found to be in the range of 149–28,300 μS/cm during the post-monsoon measurements and 402–28,570 μS/cm during the pre-monsoon season that indicated the presence of higher concentration of ions in some samples (Table 2). The pH in the basin ranged from 7.0 to 8.8 during the post-monsoon season and from 7.8 to 8.7 during the pre-monsoon season that indicated the alkaline nature of groundwater. The TDS concentrations were extremely high and it ranged from 249 to 19,810 mg/L post-monsoon and 283 to 20,120 mg/L pre-monsoon that showed the presence of higher dissolved salts. These values were > 500 mg/L in some samples, the prescribed limit by the Bureau of Indian Standards (BIS, 2012). This indicated that some wells were extremely contaminated

in the basin. The TDS concentration during the pre-monsoon season was higher than those observed during the post-monsoon season. The presence of higher concentration of TDS in some of the regions in the basin was classified as brackish water (Mondal & Singh, 2011). TDS is directly related to the electrical conductivity (EC) and has a strong correlation with the anions and cations present in the groundwater (Jesudhas et al., 2017). Therefore, several previous studies (Shwei et al., 2013; Al-Mukhtar & Al-Yaseen, 2019; Ewusi et al., 2021; Tamma Rao et al., 2013) have considered TDS as the main parameter for modeling the contaminant transport in groundwater.

It is noteworthy to recall that the Sengulam Lake, located in the Dindigul-Batlagundu highway, is also presently being used as a storage tank for collecting the treated water discharged from the CETP. The continuous discharge of treated wastewater has completely changed the color, odor, and quality of the lake water. The average TDS concentration in the Sengulam Lake was found to be 14,000 mg/L during the pre-monsoon season and 13,530 mg/L during the post-monsoon season, indicating the influence of dilution caused by precipitation. The lake was found to be devoid of aquatic organisms (e.g., fish, frogs, snails) and its water quality did not appear to be a favorable ecosystem/habitat for its growth. Besides, the water in the Sengulam Lake was also not suitable for agricultural purpose or for surface irrigation (Kanthasamy et al., 2014). The TDS in the soil was found to be ~ 16,000 mg/L, and the pH was found to be 7.9. This condition of alkaline soil may be due to the presence of high chloride concentrations (Murali & Rajan, 2012). These chemical contaminants had also infiltrated into the subsurface and contaminated the groundwater in the aquifer. However, during model prediction, some of the wells with high TDS concentrations were also included in the model. This

Table 2 Statistical summary of groundwater quality parameters and comparison with BIS drinking water standards (IS-10500)

| Parameter (unit)	IS-10500 Desirable limit	Post-monsoon				Pre-monsoon			
		min	max	mean	sd	min	max	mean	sd
TDS (mg/L)	500	249	19,810	2281	4027	283	20,120	2467	4143
EC (μS/cm)	300	149	28,300	2154	5677	402	28,570	3504	5884
pH	6.5–8.0	7.0	8.0	7.8	3.1	7.7	8.7	8.1	3.2

Note: *TDS* total dissolved solids, *BIS* Bureau of Indian Standards, *IS* Indian Standards, *EC* electrical conductivity, *min* minimum, *max* maximum

might have influenced the density and viscosity of the contaminant plume. This inclusion affected the predictive ability of the model developed in this study. The results from physico-chemical analysis of water samples clearly show that the freshwater source has been severely mismanaged, leading to loss of natural habitat and biodiversity. Therefore, it is important to protect and restore the water quality of Sengulam Lake and the aquifer. The sustainable development goals (SDG-6.6) also reinstate to protect and restore the water-related ecosystems, such as wetlands, rivers, aquifers, and lakes (UNEP, 2013).

Calibration for head and concentration

A model should be able to achieve the minimum objective function before it is applied for prediction purposes in real field conditions. In this study, a sensitivity analysis was done on the parameters such as hydraulic conductivity and specific yield to provide a reasonable range of possible outcomes. The PEST optimization model in Visual MODFLOW was first run in estimation mode to achieve the minimum objective function. Once the minimum objective function was achieved, it was used to identify the sensitive parameter. The

Fig. 2 Pathway followed by the contaminant in the aquifer from Sengulam Lake
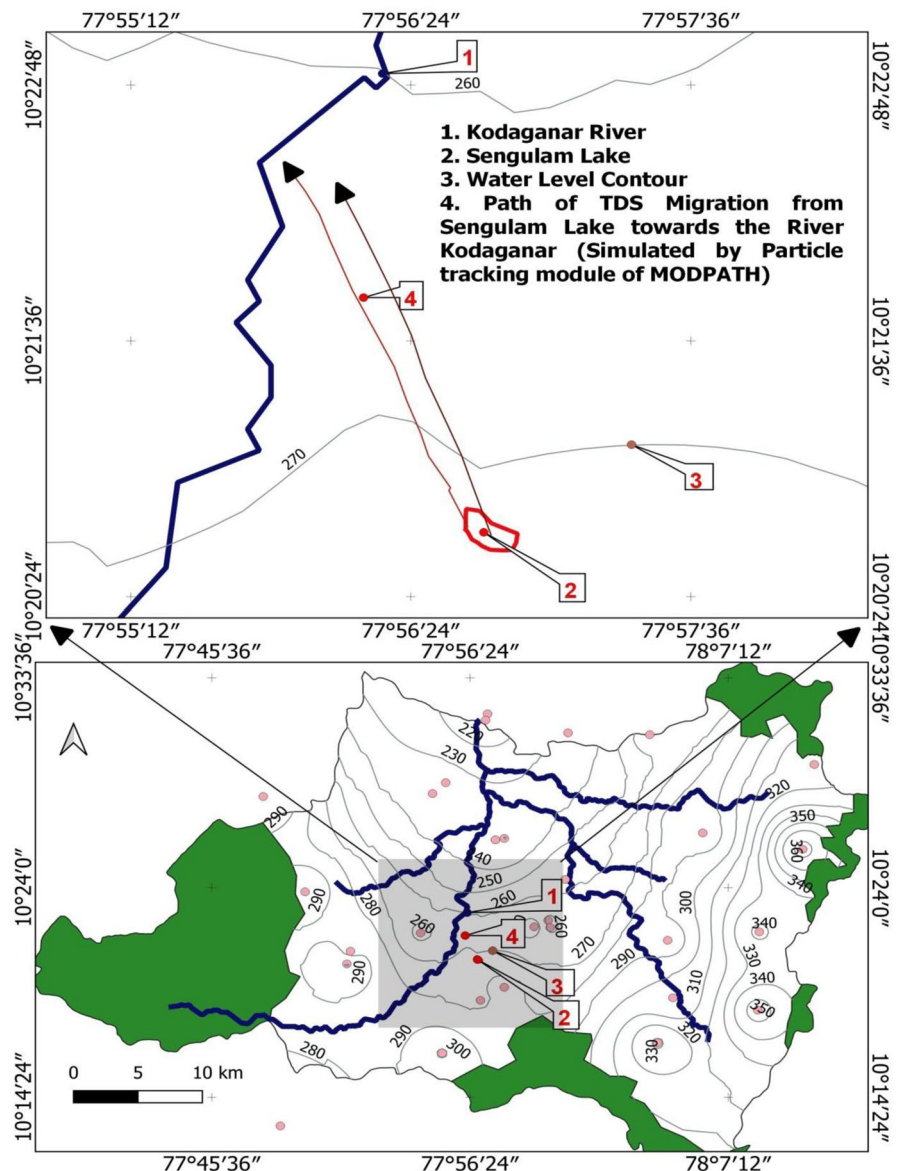
model identified hydraulic conductivity as the most sensitive parameter when compared to specific yield. The study conducted in the same region showed that advection, and not dispersion, played the dominant role in the contaminant transport process (Mondal & Singh, 2009). Therefore, it was assumed that the migration of TDS in the aquifer was predominantly controlled by advection and hence this aspect was incorporated in the model. The model was simulated for transient-state condition to predict the fate of TDS in the groundwater regime. The direction and distance of migration of the contaminant, i.e., TDS in the aquifer from the Sengulam Lake, are shown in Fig. 2. The contaminants were introduced along the circumference of the lake and the forward tracking module was used to compute the location of particle at different time steps. The direction and distance were computed from the velocity vectors generated by the groundwater flow model. The velocity of groundwater and TDS, i.e., as the pollutant load (Sundararajan & Sankaran, 2020), was used for the prognosis. The contaminant from the lake got infiltrated into the aquifer. It has started moving towards the Kodaganar River and contaminated the groundwater in the aquifer along its direction of travel. The average distance traveled by the contaminant in the aquifer was estimated to be 1.2 km/year. The mechanism of dispersion in the water table was modeled by the MT3DMS package. A correlation coefficient of 0.98 was achieved, indicating that the calibrated steady-state condition had been finalized.

The graphical representation of the observed and computed TDS concentration under steady-state condition is shown in Fig. 3. The results obtained after steady-state calibration are presented in Table 3 in which the standard error of the estimate was found to be 26.714 mg/L, minimum residual concentration=70 mg/L (WQ5), maximum residual concentration=63 mg/L (WQ35), mean residual concentration=16.333 mg/L, and absolute residual mean=39.333 mg/L. The root mean square error (RMSE) was used for the steady-state calibration of the model that yielded a value of 73.01 mg/L. There were no observation wells in the PWD that had historical data near the tannery industries; therefore, only the wells with historical data were used for simulating the transient-state model. The calibrated transient-state condition was finalized when a correlation coefficient of 0.94 was reached. The graphical representation of observed and computed TDS concentration under transient-state condition is shown in Fig. 4. The results obtained after transient-state

Fig. 3 Comparison of observed TDS concentration and calculated TDS concentration calibrated for the steady-state condition
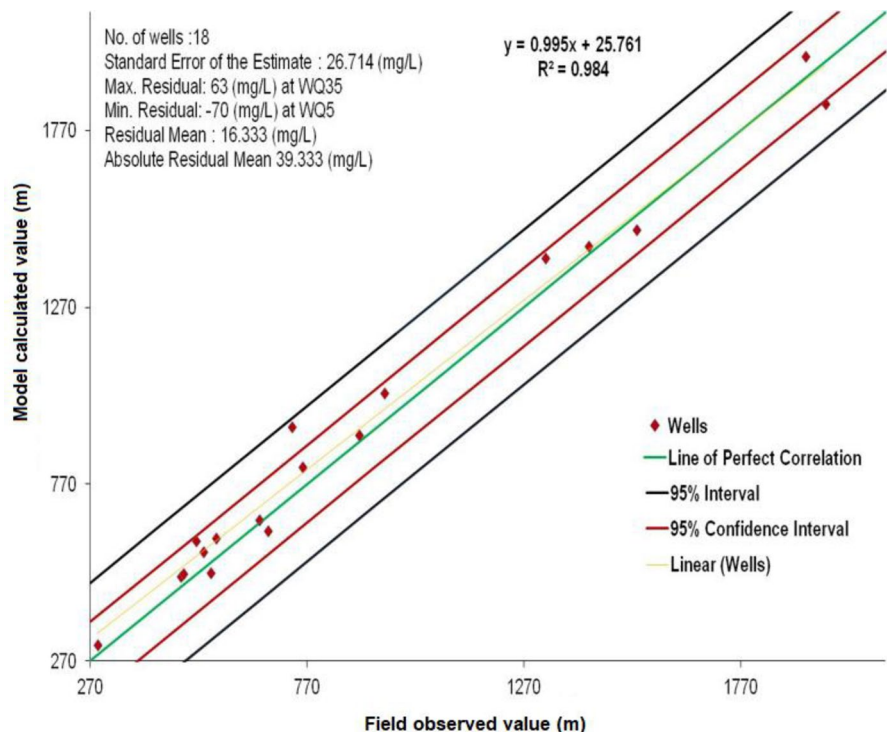

Plot details:
No. of wells :18
Standard Error of the Estimate : 26.714 (mg/L)
Max. Residual: 63 (mg/L) at WQ35
Min. Residual: -70 (mg/L) at WQ5
Residual Mean : 16.333 (mg/L)
Absolute Residual Mean 39.333 (mg/L)
$y = 0.995x + 25.761$
$R^2 = 0.984$

Y-axis: Model calculated value (m)
X-axis: Field observed value (m)

Legend:
♦ Wells
— Line of Perfect Correlation
— 95% Interval
— 95% Confidence Interval
— Linear (Wells)

Table 3 RMSE validation chart used for calibration of steady-state and transient-state conditions

No	Well no	Steady-state condition				Transient-state condition			
		O [mg/L]	E [mg/L]	$(E-O)^2$	$(E-O)^2/E$	O [mg/L]	E [mg/L]	$(E-O)^2$	$(E-O)^2/E$
1	WQ1	1212	1150	3844	3.34	1320	1360	1600	1.18
2	WQ2	495	512	289	0.56	515	566	2601	4.60
3	WQ3	283	316	1089	3.45	288	316	784	2.48
4	WQ4	500	530	900	1.70	549	520	841	1.62
5	WQ5	1380	1310	4900	3.74	1420	1445	625	0.43
6	WQ6	1240	1210	900	0.74	660	670	100	0.15
7	WQ7	496	530	1156	2.18	532	580	2304	3.97
8	WQ8	1435	1480	2025	1.37	1530	1490	1600	1.07
9	WQ9	450	480	900	1.88	480	510	900	1.76
10	W21	1800	1860	3600	1.94	1920	1980	3600	1.82
11	W22	460	480	400	0.83	485	519	1156	2.23
12	W31	714	752	1444	1.92	760	783	529	0.68
13	W32	1766	1820	2916	1.60	1966	2010	1936	0.96
14	W33	871	826	2025	2.45	890	910	400	0.44
15	W34	524	553	841	1.52	560	620	3600	5.81
16	W35	711	774	3969	5.13	736	776	1600	2.06
17	W36	905	930	625	0.67	948	997	2401	2.41
18	W37	611	634	529	0.83	680	640	1600	2.50
		RMSE = 42.39		$\chi^2 = 32.52$		RMSE = 39.56		$\chi^2 = 34.99$	

Fig. 4 Comparison of observed TDS concentration and calculated TDS concentration calibrated for the transient-state condition
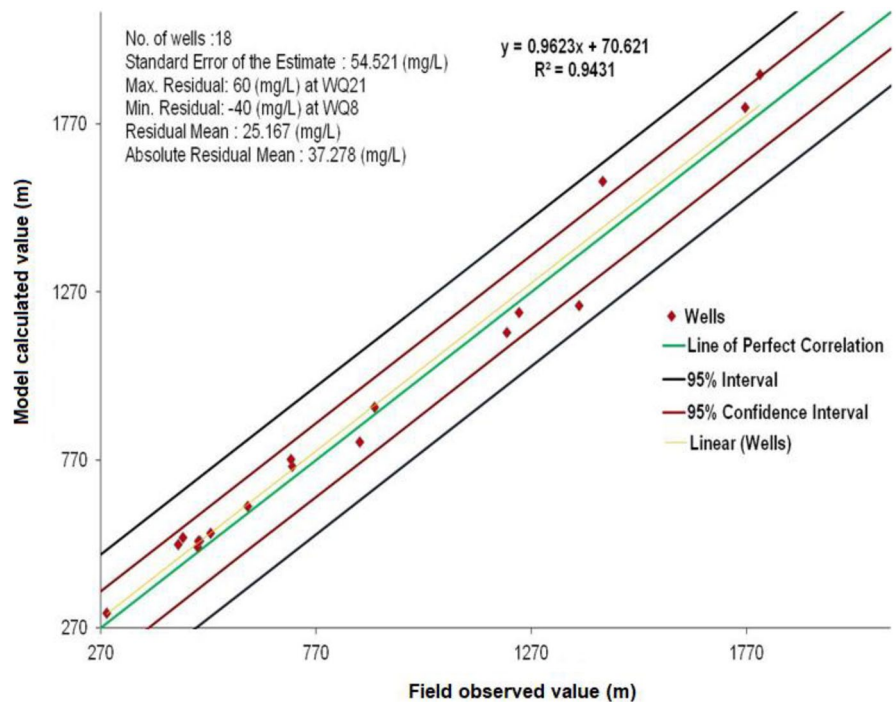
calibration are presented in Table 3 in which the standard error of the estimate was found to be 54.52 mg/L, minimum residual concentration = 40 mg/L (WQ6), maximum residual concentration = 60 mg/L (WQ18), mean residual concentration = 25.167 mg/L, and absolute residual mean = 37.278 mg/L. A test for accuracy of fit was done to ascertain whether the computed values support the observed values by considering the level of significance as 5%. A χ^2 hypothesis test was also performed to compare the observed and the computed TDS concentration for the steady-state and transient-state models. The data from a total of 18 observation wells were used as inputs to the model; therefore, the degree of freedom (n-1) was 17. For a 5% level of significance, the tabulated $\chi^2 = 35.72$, which was more than the calculated $\chi^2 = 32.5$ for steady-state condition and $\chi^2 = 34.98$ for transient-state conditions. Hence, there was no significant difference between the observed TDS concentration and the computed TDS concentration.

Thus, based on these results, it is evident that the contaminant that reached the water table gets dispersed along the transverse direction. The ratio of hydrodynamic dispersion coefficient along the transverse to the longitudinal direction was generally found to be 0.1 (Gelhar et al., 1992). Nevertheless, it was observed that the TDS concentration in the wells exceeded the computed TDS concentration indicating the larger influence of dispersion. Presumably, this was due to the uncontrolled extraction of groundwater for agricultural and domestic requirements of water.

Fate of TDS and concentration distribution

The contour map representing the distribution of TDS concentration near the tannery industries is shown in Fig. 5. The well in the upstream side of the Sengulam Lake (W18) had higher concentration of TDS. This well was not energized and remained non-utilized due to the contaminated water. The wastewater from the tannery industries, located in the upstream side, was carried by surface runoff during the rainfall season. It was reported that the flora and fauna around a radius of 6 km were affected by pollution caused by the nearby tanneries (Kanthasamy et al., 2014). The groundwater quality in the central plains of the basin,
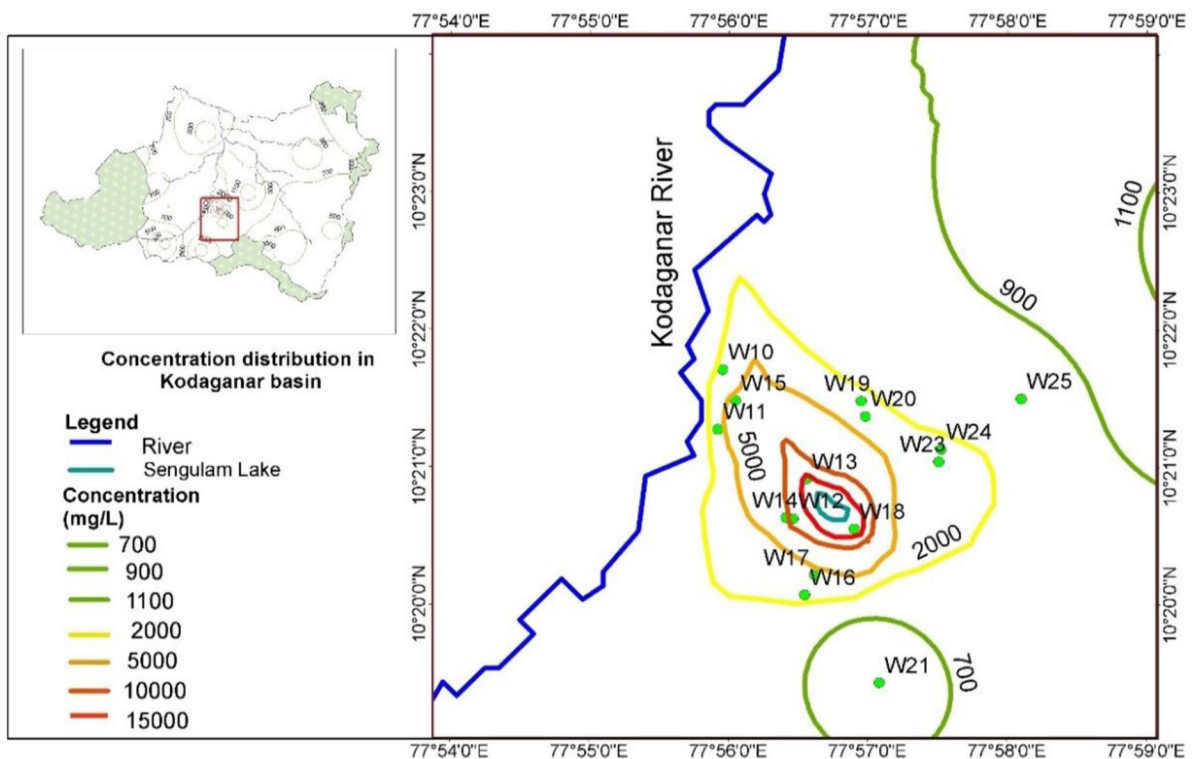

Fig. 5 Contour map representing the distribution of TDS concentration near the tannery industries (year: 2014)

i.e., in the south west of Dindigul city, was also highly contaminated. The concentration of TDS in groundwater in this region was > 2000 mg/L. The contaminated regions have low nutrients in the soil, leading to uncultivable land (Vasanthavigar et al., 2009). Notably, the fertility of an area of ∼100 km² has been lost due to severe pollution in the region (Mondal & Singh, 2005). In this study, it was estimated that 12 km² of aquifer was affected due to pollution caused by the tanneries, although the quality of groundwater in the peripheral region of the basin was found to be moderately good. The villages such as Kuttiyapatti, Pallapatti, Ponnimandurai, Athur, Pillayarnatham, Muthalagupatti, Paraipatti, Begampur, and Mohamadiapuram were found to be contaminated. Meanwhile, according to the results, the contamination was also extended into Dindigul city, up to ∼2 km². Water resource management could be significantly impacted by the switching of interactions between surface water and groundwater, as well as by anthropogenic activities such as pumping, wastewater discharge, and water transfer (Zhu et al.,

2019). The wastewater in the lake can be monitored online with the help of sensors (Capodaglio, 2016) to avoid serious any future damage to the aquifer. The sustainable development goals (SDG-6.3) reinstate to improve water quality by reducing pollution caused by untreated wastewater, i.e. by enforcing the companies to treat their wastewater at site, follow the strict discharge guidelines stipulated by the Pollution Control Board (PCB) and adopting cleaner production measures within their facilities.

Prognostics for different source concentrations

The developed model was used to simulate different scenarios in an attempt to protect and rejuvenate the aquifer, the Kodaganar River, and the pumping wells located in the basin. The model was used to predict the status of TDS concentration in groundwater by considering three different scenarios for different input conditions (mean recharge condition, average evapotranspiration, etc.).
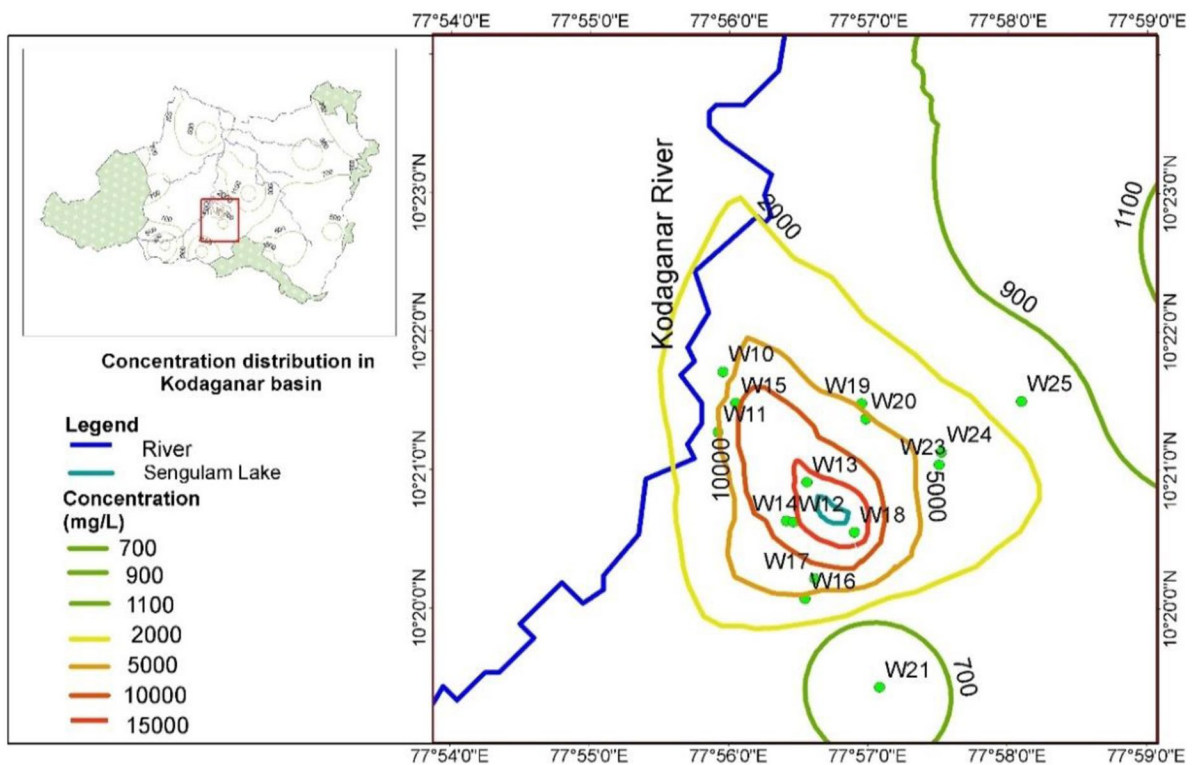

Fig. 6 Contour map representing the distribution of TDS concentration near the tannery industries (year: 2020, estimated by allowing the existing TDS concentration in the Sengulam Lake)

Scenario I

In the first scenario, the TDS concentration in the Sengulam Lake was maintained constant, and the concentration was predicted for the year 2020. The distribution of TDS concentration is shown in Fig. 6. This scenario was observed to be mainly controlled by advective transport. The dispersion in the transverse direction was found to be higher than expected due to the average pumping conditions that were exerted for the domestic (household) and irrigational requirements. The distribution of TDS concentration was found to have been increased over the years. In this scenario, the distribution of TDS in groundwater has also crossed the Kodaganar River. This condition could severely pollute the water that flows into the river during the rainy season. The estimated areal extent of aquifer affected by tannery industries was estimated to be 18 km^2. According to the results of scenario I, (i) the contamination has extended in the aquifer and could contaminate 3.5 km^2 of the residential area in the south-west urban settlements of Dindigul city, and (ii) an increase in the TDS concentration

has negatively affected the aquifer and both the Kodaganar River and the pumping wells are at high risk.

Scenario II

In the second scenario, the TDS concentration in Sengulam Lake was maintained as per the permissible discharge standard (2100 mg/L) and the concentration was predicted for the year 2020. The distribution of TDS concentration is shown in Fig. 7. This scenario was also observed to be mainly controlled by advective transport. Because of the pumping activities, it was discovered that the dispersion in the transverse direction was more than planned and/or expected. However, in this scenario, the spatial distribution of TDS concentration in groundwater was found to have decreased although it did not significantly contribute to the improvement in the water quality of the contaminated aquifer. The estimated areal extent of aquifer affected by the tannery industries was estimated to be 17 km^2, while the contamination has extended in the aquifer and has contaminated 3.25 km^2 of residential
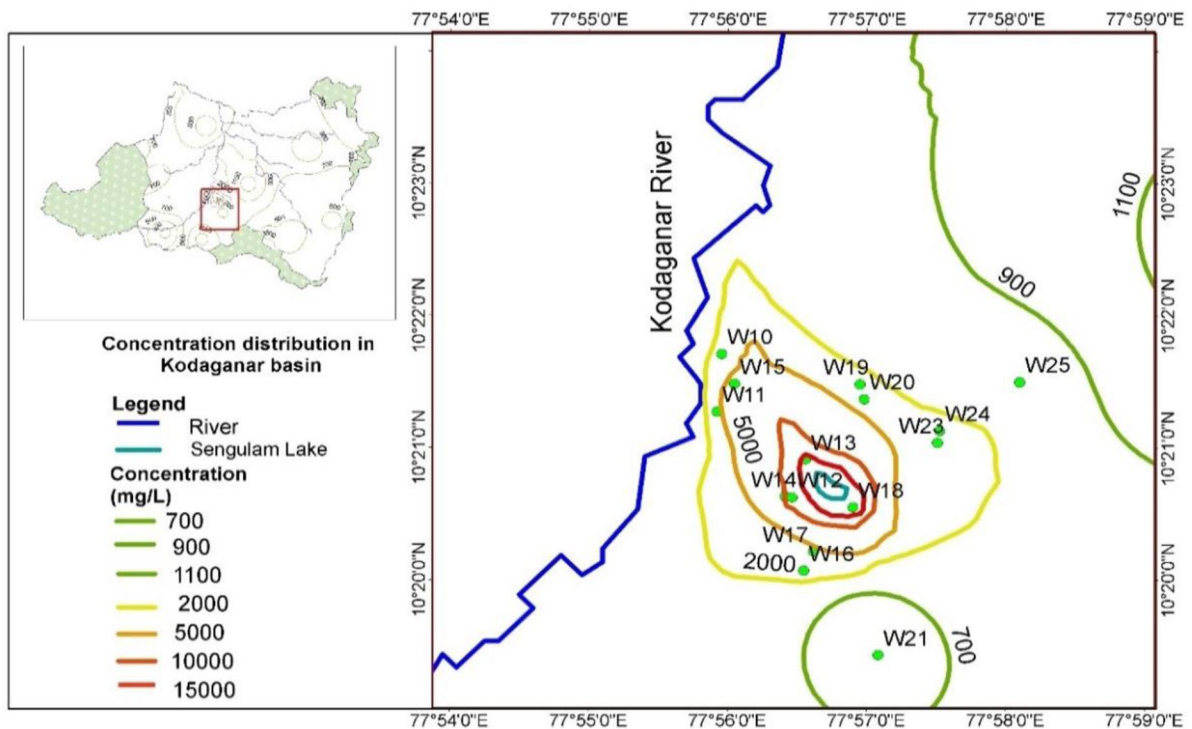

Fig. 7 Contour map representing the distribution of TDS concentration near the tannery industries (year: 2020, estimated by allowing the permissible discharge limit concentration in the Sengulam Lake)

area in the south west of Dindigul city. According to the results of scenario II, (i) the contamination has not reached the regime of Kodaganar River and (ii) maintaining the TDS levels within the permissible discharge standards has prevented the pumping wells from being contaminated, and (iii) TDS progression has been controlled/contained along the direction of flow of groundwater in the aquifer.

Scenario III

In the third scenario, the TDS concentration in the Sengulam Lake was considered to be zero (0 mg/L), and the concentration was predicted for the year 2020. The distribution of TDS concentration is shown in Fig. 8. The dispersion in the transverse direction due to advective transport was found to be higher than expected due to the pumping activities. In this scenario, it was observed that the decrease in TDS concentration was slow due to the influence of other contaminants present in the soil particles and the aquifer matrix. The estimated areal extent of aquifer affected by tannery industries was estimated to be 6 km^2 and the contamination has extended in the aquifer and

affected up to 1.4 km^2 of the residential area in the south west of Dindigul city. Compared to scenarios I and II, the condition of scenario III has significantly improved the status of groundwater in the region. According to the results of scenario III, (i) the contamination has not reached the regime of Kodaganar River, (ii) the villages such as Kuthathupatti, Athur, Begampur, and Mohamadiapuram could be protected from contamination, and (iii) the pumping wells were also preserved from contamination; because of this, they were not regarded to be in danger or at risk.

Mitigation of receptor contamination

In this study, the natural resistance offered by the hydro-geological setting to conserve the groundwater from contamination was studied using the TDS transport model. The model computed the transit time of the contaminants from the Sengulam Lake. Sengulam Lake falls in a region of moderately sloped, highly weathered gneissic rock that is overburdened with silty soil deposits. The hydraulic conductivity in this region was low (0.5 m/day) that contributed to moderate recharge of the aquifer. The moderately shallow
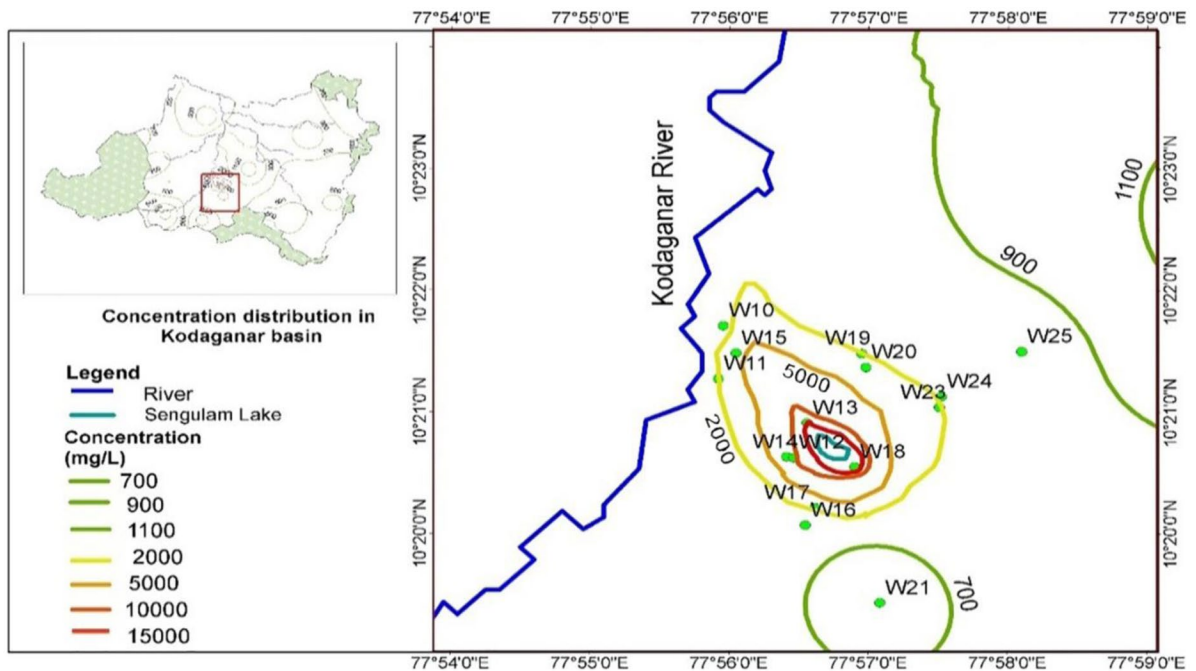

Fig. 8 Contour map representing the distribution of TDS concentration near the tannery industries (year: 2020, estimated by allowing the zero discharge concentration in Sengulam Lake)

Fig. 9 Predicted TDS
concentration and the
associated areal extent of
contaminated aquifer for
different concentrations: **a**
2000 mg/L, **b** 5000 mg/L,
c 10,000 mg/L, and **d**
15,000 mg/L

◆ Existing Discharge ◇ Permissible Discharge ◆ Zero Discharge
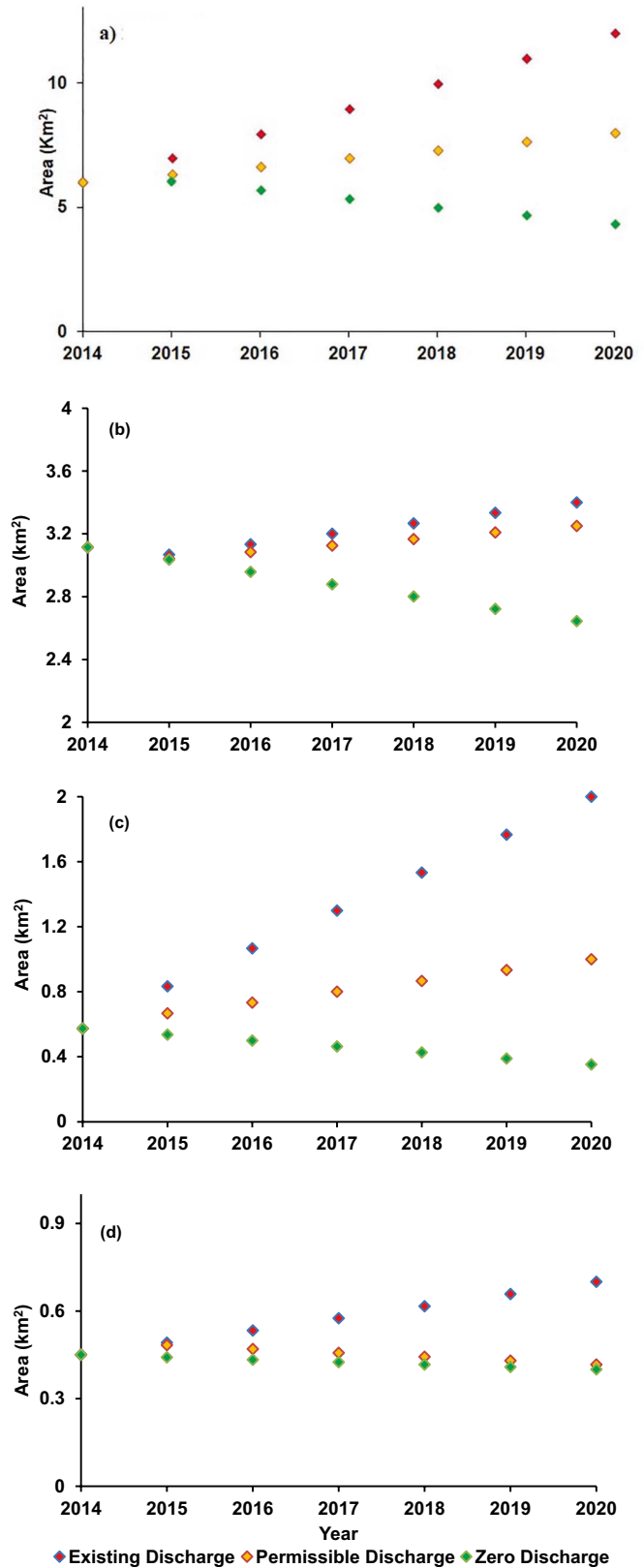
water table was attributed to interconnected pores and passages between the aquifer and the lake bed, and there was continuous interaction/flow of water between the Sengulam Lake and the aquifer. Even though the Sengulam Lake and the aquifer are situated in a low and medium vulnerable region mainly because of poor conductivity, the aquifers in these regions are highly contaminated by TDS (Jesudhas et al., 2016). This may be due to the overexposure of contaminants to the aquifer because of the perennial availability of water in the lake and the improper disposal of wastewater in the nearby area. In an attempt to mitigate the contamination, three scenarios were simulated in the TDS fate transport model. The predicted results of the TDS fate transport model for the three scenarios are shown in Fig. 9. The influence of a particular concentration and its areal distribution has been listed separately for every TDS concentration chosen, i.e., (a) 2000 mg/L, (b) 5000 mg/L, (c) 10,000 mg/L, and (d) 15,000 mg/L, respectively.

For example, at a TDS concentration of 2000 mg/L, the following observations were made: (a) the areal distribution of TDS was found to increase over the years when the same discharge was allowed to continue, (b) the spatial growth of this concentration was controlled when the discharge was according to the permissible discharge standard, (c) the spatial growth decreased when the discharge was maintained as zero, (d) the affected area was forecasted to be ~2 km^2. Similar interpretations could also be done for TDS concentrations of 5000, 10,000, and 15,000 mg/L. A linear regression model was developed for the zero-discharge condition to predict the rate of decrease of areal extent and the model equations are presented in the respective figures. This model was used to predict the time taken for the shrinkage under natural attenuation. Under linear conditions, the model predicted that it would take 2050 years to reduce the concentration of 15,000 mg/L to the permissible discharge limit, whereas it predicted that it would take 2030 and 2020 years for 10,000 mg/L and 5000 mg/L concentrations, respectively. Among the three scenarios, the third scenario, i.e., by stopping the discharge of treated water into the Sengulam Lake, the quality of groundwater in the nearby village can be improved significantly. The mobility, bioavailability, and toxicity of certain toxic metals (e.g., chromium) depend on its oxidation states (Beretta et al., 2020). Thus, as expected, the recovery time for the groundwater

under natural attenuation was too high and it might not be able to restore the receptors. Therefore, in situ groundwater remediation technologies such as permeable reactive barriers, pump and treat technologies, biostimulation, bioaugmentation, chemical oxidation/reduction, and bio-electoremediation or bioelectrochemical systems (Cecconet et al., 2020) could be used to enhance and accelerate the remediation process.

Conclusions

Groundwater in the upper Kodaganar basin (Tamilnadu, India) is contaminated with high TDS concentration and other toxic chemicals due to the discharge of treated tannery wastewater to the Sengulam Lake. The absence of water quality monitoring wells near the vicinity of the tannery industries is a major challenge in obtaining proper spatial water quality data. The application of the contaminant transport model to predict the transport of TDS in the Sengulam Lake revealed information on the possibility of risk/danger to the nearby Kodaganar River and the aquifers of the basin. The results from this study underline the urgent need to implement tailor-made/customized and knowledge-based in situ treatment systems to protect the depleting water resources in the region.

Author contribution Conceptualization and writing: Colins Johnny Jesudhas; supervisor: Madurai Chidambaram Sashikkumar; proof reading and suggestion: Rajesh Banu Jeyakumar, Eldon R. Rene.

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Declarations

Ethics approval and consent to participate Not applicable

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