Understanding the Contaminant Chemistry of Water Resource at Urban Solid Waste Disposal Site, Hyderabad, India

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Abstract

The Hyderabad urban solid waste dump site though located on a rocky hillock has highly weathered mantle apart from inter-connected network of fractures. These hydrological features favored massive dissipation of contaminants into the surface as well as sub-surface water resources. The surface water bodies in the vicinity turned into leachate pools and groundwater in the zone of influence is not fit for any use. It is assumed that apart from hydrological factors hydrogeochemical characteristics also contributed to contamination of pore water. Efforts were made through the Paper to underline the preferential chemistry of water resources at solid waste disposal sites that stimulated the ion enrichment. Both surface (15) and groundwater (79) samples were gathered during 2011 and 2012 hydrological cycles and tested for major parameters. Few samples (15) of 2012 were also analyzed to determine TOC, BOD, and COD. The general order of abundance of prominent ion species in groundwater was Cl^-, Ca^{2+}, Na^+, Mg^{2+}, HCO_3^- and SO_4^{2-}. Very high and similar Cl^- concentration in surface water (37 meq/l) and contaminated groundwater (40 mq/l) establish that solid waste stockpile was the point source through leachate. The presence of high (>200 mg/l) TOC, BOD and COD in many surface and groundwater samples supports the inference. Modified Piper plots display the Ca-Mg-Cl and Ca-Mg-HCO_3 were the dominant hydrochemical facies in groundwater, whereas many surface water and few groundwater samples belong to Na-Cl/SO_4 type. A good correlation (~0.90) between Cl^- - Ca^{2+}, TH, Na^+, and Mg^{2+} points out that these ion species were from the same source. Gibbs plots, positive CAI-1 and 2, together with >2 ratios of Ca^{2+}/Mg^{2+}, Cl^-/Na^+ in many samples divulge water-rock interaction and ion exchange were also controlling the hydrochemistry. The low HCO_3^-/Cl^-, TA/TH and SO_4^{2-}/HCO_3^- values indicate an insufficient influx of freshwater and non-lithogenic sources altered the groundwater chemistry. >0.50 ratio of $Na^+ + K^+/TZ^+$, Ca^{2+}/Na^+, Ca^{2+}/HCO_3^- together with cross plots of Na+K vs $Cl^- + SO_4^{2-}$, $HCO_3^- + SO_4^{2-}$ vs $Ca^{2+} + Mg^{2+}$ and Langelier and Ludwig (modified) diagram confirm the multiple ionization processes were operating in aqueous system. Favorable hydrochemistry in the form of alkaline water, high TH, Cl^- and prevailing redox conditions as well as methanogenic phase of dump yard might have spurred up ion enrichment activity within the zone of influence of point source.

Introduction

Globally growing urban population is resulting in the generation of enormous solid waste and its safe dispensation has become a challenge to municipal administrators and sanitation experts. Largely the municipal solid waste (MSW) is disposed of in an unorganized manner and is rarely processed for safe disintegration which is creating havoc on the local environment. Talalaj and Biedka (2016) shared the data from 2013 which show that in 14 countries of the European Union, the share of land-filling was over 50 %, and in 6 of these countries even over 75 % (Greece, Croatia, Cyprus, Latvia, Malta, Romania; Eurostat 2015). In the USA, about 135 million tons of solid wastes (53.8 %) were discarded in landfills in 2012 (USEPA 2012). In 2002, in China the quantity of MSW disposed of was 74.04 million tons, 89.30% of which was land-filled, 3.72% was incinerated, and 6.98% was composted (Huang et. al., 2006). India

produced 55.51 million tons of MSW in 2018-19 of which only 20.35% is treated (CPCB 2019). Dumping of solid waste either as landfill or over-ground led to the generation of harmful compounds which propagate to the regional hydrological cycle. Water has become the first victim of solid waste mismanagement, often the surface water bodies turn to cesspools of leachate and point source of aquifer contamination (Suchi et, al., 1998; Raman and Sathiya 2008; Zhiyong et, al., 2016; Nataliia and Oleg 2017; Vahabian et. al., 2019). Though many guidelines and environment protection acts exist they are more flouted than followed especially in underdeveloped countries. Land with massive rock structures or confined layers are selected as solid waste disposal sites or the sites are engineered to prevent the percolation of pollutants. But over the years maintenance and management of such structures do not receive the required attention leading to the proliferation of toxic chemicals deteriorating the local aqueous system. Ample studies were carried out on landfill sites world-over which present the prevailing poor conditions along with reasons and causes for contaminants migration (Peter Kjeldsen et. al., 2002; Kola-Olusanya 2012; Daniel et. al., 2013; Akhtar and Tang 2014; Bikash Adhikari et. al., 2014; Salar Rezapour et. al., 2018; Vongdala et. al., 2019; Dan Zeng et. al., 2021). Thus far focus of the research was mostly limited to decipher the extent and variability in contamination apart from discussing the physiological features and local hydrological properties that promote the propagation of pollutants. Innovations in analytical hydrochemistry and solute transport models helped in understanding the contaminant kinetics. Even though regional hydrological features, as well as load and age of the solid waste, determine the extent of pollution the receptive chemical characteristics of the host aqueous environment cannot be ignored. Dan Zeng et. al., (2021) concluded that the groundwater contamination near the landfill sites was influenced by leachate, soil, climate, and hydrogeology characteristics. In this context research on hydrogeochemistry of groundwater close to solid waste dump sites is gaining grounds. It is all the more important as groundwater chemistry is governed by aquifer matrix apart from anthropogenic or external inputs. Exploring the groundwater contamination from a hydrogeochemistry perspective was made by many researchers (Jorstad et. al., 2004; Reddy et. al., 2012; Selvakumar et. al., 2017; Elisabetta et. al., 2018; Emmanuel et. al., 2019). A highly contaminated hydrological environment at Jawaharnagar municipal solid waste dumpsite of Hyderabad city was studied with the hypothesis that groundwater chemistry has also influenced the absorption of contaminants and contaminant chemistry of pore water altered the ionization processes.

The Hyderabad Integrated Municipal Solid Waste (HIMSW) disposal site is located at about 35 km north of the city near Jawaharnagar. The city including urban agglomeration with a nearly 10 million population produces 5500 metric tons of solid waste a day as per the year 2020 and has the distinction of generating the highest per capita waste in the country (http://www.populationu.com/ and https://tspcb.cgg.gov.in/). The dumpsite was in operation for the past two and half decades and has gained notoriety for contaminating all elements of the environment and causing misery to local people. Many studies were carried out to chronicle the adverse impact of the huge solid waste dumps which was left unattended till recently (Vandana et. al., 2011; Sarala and Ravi 2012; Ravi Babu et. al., 2014; Rao 2015; Kurakalva et. al., 2016; Unnisa and Bi 2017; Alimuddin 2019; Venkat Charyulu 2019; Konda et. al., 2020; Soujanya 2020). Decomposition of solid waste dumped on hillocks and accumulated legacy

generates effluents that seep into foothill water bodies turning them into leachate reservoirs. Wastewater from the surface water tank and steams percolated into the underlying aquifer through preferential pathways. There is no contention on solid waste dumps being a point source of water contamination but little efforts were made to understand the underlying geochemical processes. The Paper would be a contribution to bridge the knowledge gap in this domain.

Study area

The study area comprising a solid waste dumping site and its surroundings spreads over nearly 5 sq km at Jawaharnagar village, Medchal-Malkajgiri District, Telangana State, India (Fig. 1). The area falls between 17^{o} 30' to 17^{o} 32' N latitude and 78^{o} 35' to 78^{o} 38' E longitude, its general topographic elevation ranges from 510 to 560 m. The climate in the area is semi-arid and receives a normal annual rainfall of 753 mm. The study area belongs to the Madyala stream watershed and Dammaiguda mini watershed of the Musi River which is part of the Krishna river basin. The drainage pattern is dendritic to sub-dendritic. The area is underlain by gray granite gneisses and massive granites of the Archaean age. The thickness of weathering extends down to 18m, while the fractures are recorded down to 106 m. Water table elevation contour ranges from 500 to 560 m with a gradient of 10 m/km. The groundwater flow is towards the southeast. The infiltration rate was high (29 cm/hr) at Madyala stream, low at Rajiv karmika Nagar (9.2 cm/hr) and Cherial village (9.6 cm/hr; Rao, 2015).

Method And Methodology

Hydrogeological studies were carried out in May 2011 and water sampling sources were identified. A varied sampling pattern was followed to have a wide representation of contaminated as well as normal areas. Both surface (15 nos) and groundwater (79 nos) samples were collected in pre and post-monsoon seasons of 2011 and 2012 to cover two hydrological cycles. All the samples were tested for 13 parameters and few samples (15 nos) for TOC, COD, and BOD in the chemical laboratory of Central Ground Water Board, Southern Region, Hyderabad following standard procedures of APHA (2017). The pH was measured by using the digital pH meter and EC by the EC/TDS analyzer, CM 183 model. The classical methods of analysis were applied for the estimation of Ca^{2+}, Mg^{2+}, CO_3^{2-}, HCO_3^{-} and Cl^{-}. Na^{+} and K^{+} were analyzed by flame photometry method using the CL-345 model. SO_4^{2-} was estimated by the turbidity method with the digital Nephelo-Turbidity meter 132 model. NO_3^{-} was estimated by applying the UV-Vis screen method by UV- visible spectrophotometer UV-1201 model. F^{-} was tested by the ion-selective electrode method using the Orion 290A + model. The total hardness (TH) and total dissolved solids (TDS) were estimated by calculation method (Hem, 1991). The obtained results were tested for accuracy by calculating the Normalized Inorganic Charge Balance (Huh et al., 1998). All the analyzed samples had ionic charge balances of < +/-3%.

Results And Discussion

Surface water chemistry

The results of all the tested inorganic parameters exhibit that the surface water of tanks close to Hyderabad Integrated Municipal Solid Waste Dump Site (HIMSWDS) turned into pools of leachate (Supplemental material 1). The mean of pH was about 8.00, EC 13000 m S/cm, TH 753, whereas Alkaline earths and SO_4^{2-} were normal but Na^+ was 813, K^+ 530, HCO_3^- 978, Cl^- 1304, and NO_3^- 262 were high (all in mg/l; Table 1). Many tested parameters of water samples from Irlagutta, Haridaspalli, Cherial, and Dammaiguda tanks (in order of contaminant abundance) were abnormally high and very much above the MoEF benchmark for leachate or effluent land discharge (MoEF, 2016). Mohit et. al., (2019) have also opinioned in their work on six municipal waste dumpsites (which include Hyderabad) of India that the level of almost all the physicochemical parameters concerning the regulatory threshold was found higher at all the landfill sites. Irlagutta tank located on the foothill of the dumpsite is a classic example of neighborhood water bodies turning into leachate. It recorded very high EC in all sample sessions and has reached a peak of 90560 m S/cm in pre-monsoon 2012 but in post-monsoon it receded down to 4260 m S/cm. Al-Sabahi et. al., (2009) in their research at the municipal solid waste landfill of Ibb City, Yemen observed that there would be an excessive generation of leachate during the dry season. Turbidity of 54 NTU in post-monsoon 2012 displays direct the mass flow of leachate from dump-heap by hydraulic gradient due to elevation difference of 633 to 550 m, amsl between dumpsite hillock and tank surface. In pre-monsoon 2012 these waters recorded a high amount of organic compounds like TOC (380 mg/l), COD (18000 mg/l) BOD (16000 mg/l). Except for pH, Ca^{2+}, and SO_4^{2-} all other tested parameters were abnormally high confirming the Irlagutta tank a cesspool of leachate. The Haridaspalli tank water, another closely located (1.5 km) surface water body, too had many chemical constituents in very high concentration which was reflected in EC of 12220 m S/cm. The distinction of the water was it had a higher content of Ca^{2+}, Cl^-, and SO_4^{2-} than Irlagutta water samples. Unusually high SO_4^{2-} content in Haridasappli tank (1440 mg/l) and low (5 mg/l) in Irlagutta tank which is located within proximity of a 1 Km but diverse directions was beyond comprehension. Different sources of seepage within the huge debris dumps and their outflow through separate feeder channels could be some of the causes. The tank waters at Cherial and Dammaiguda villages located at about 4 km in south and east directions respectively from dumpsite consistently had high ion strength displaying the widespread contamination (Fig. 2a and b). The mean content of Na^+ was 886, K^+ 685, Cl^- 1828, SO_4^{2-} 320 and NO_3^- 121 in Cherial tank, and in case of Dammaiguda tank Na^+ was 564, K 209^+, Cl^- 1028, SO_4^{2-} 198 and NO_3^- 72 (all in mg/l). Elevated content of Na^+, K^+, Cl^-, SO_4^{2-} and NO_3^- in both the waters confirm that the dumpsite was the source. The turbidity of about 40 NTU in these tank waters supports the contention. Chikaodili et. al., (2017) concluded that the high concentrations of potassium, nitrate, iron, lead and high occurrence of bacteria in the water resources at solid waste dumpsites in Imo State, Southeastern Nigeria indicate possible anthropogenic contamination. Cherial tank waters affected with organic contamination as displayed by the presence of TOC (395 mg/l), COD (1040 mg/l) BOD (100 mg/l) in pre-monsoon 2012. A higher rate of contamination in Cherial tank than that of Dammaiguda can be accounted for hydraulic connectivity (Rao 2015). The Stream water in Madyalavagu does not display the adverse impact of

contamination (mean TDS was 1518 mg/l) though it originates at the dumpsite and flows in the studied area. But very low HCO_3^- (9 mg/l) and high SO_4^{2-} (average 383 mg/l), as well as NO_3^- (average 93 mg/l), point out that the freshwater input is limited whereas base flow in addition to return flow from irrigation and domestic sewerage were contributing to the stream course as well as local pollution. The sampling point is about 5 km from the dumpsite in the discharge zone could also one of the factors for non-reflection of intensity of contamination in the stream. MacFarlane et. al., (1983) in a study on migration of contaminants in groundwater at a landfill reported that the plume passes beneath a small shallow stream near the landfill without significant influence on the stream. The Kundanpalli steam located at about 8 km SE of the dumpsite display signs of contamination by high TDS (about 3000 mg/l) Na^+, K^+, HCO_3^-, Cl^- and NO_3^- which can be accounted for local pollution as many unplanned new residential colonies sprouted up in the area (Supplemental material 1).

Groundwater chemistry

The mean of tested results of the entire groundwater sample belonging to four monitoring episodes (79) indicates the influence of solid waste dumps on the hydrochemistry of the area. Though concentration of few parameters (pH, Ca^{2+}, Mg^{2+}, Na^+, K^+, SO_4^{2-} and F^-) were found within the background values of granitic terrain, the EC (2062 m S/cm), HCO_3^- (268 mg/l), Cl^- (474 mg/l), NO_3^- (58 mg/l) very high (Table 1). The samples represent core area as well as periphery beyond the zone of influence of dumpsite thus the average values exhibit a moderate chemical quality scenario. Variations in water chemistry among samples of two hydrological cycles were limited, but it was distinct in Mg^{2+}, HCO_3^-, SO_4^{2-} and NO_3^- when outliers were ignored (Fig 3). Regadío et. al., (2012) stated the variation in the data are related to waste composition and local geology. Aderemi et. al., (2000) attributed the reasons for a minimal impact of the leachate from the landfill on the groundwater resource to the existing soil stratigraphy at the site consisting of clay which is deduced to have a significant influence on the natural attenuation of leachate into groundwater. To have keen insight into contaminant chemistry the samples having EC >3000 m S/cm were segregated and their chemistry was deliberated in conjunction with other samples. All the groundwater samples (79) belonging to four monitoring sessions were classified into two groups based on EC as normal groundwater (designated for discussion purpose only) with < EC 3000 m S/cm which were 62 samples and those with EC > 3000 m S/cm as contaminated groundwater those were 17 samples. The normal groundwater samples too have certain parameters in high concentration and none of them are suitable for drinking uses when compared with acceptable criteria of Indian drinking water standards. The mean concentration of all the parameters except pH and NO_3^- were very high in contaminated samples than normal ones (Supplemental material 2 and Table 1). It is obvious but most of the parameters were higher by >250% and Cl^- by 528% than normal samples. Little variation in HCO_3^- (32%) strength is noticed among two sets of samples and pH and NO_3^- were marginally lesser (-0.56 and -6.68 mg/l respectively) in contaminated samples (Fig. 4). Minimum and maximum values of pH and NO_3^- were distinctly different (low and high) in normal samples from those of contaminated. Elevated mean content of Ca^{2+} (337 mg/l), Na^+ (444 mg/l) and Cl^- (1414 mg/l) together with normal pH and low

NO_3^- in contaminated samples confirm the dumpsite as the pollution source. The mean EC was about 5000 m S/cm in contaminated samples but more than half of the samples (9nos) have it from 3000 to 4000 m S/cm range and five samples fall in 4000 to 5000 m S/cm range. Only two samples have very abnormal conductivity (>9000 m S/cm) which could be due to local contamination. In the other group of samples (normal) most of the tested wells (76%) had an EC of 1000 to 2000 m S/cm (Supplemental material 3 and Table 1). Nicholson et. al., (1983) in their study on sand aquifer at the abandoned Borden landfill observed - beneath the landfill the most highly contaminated water in the aquifer has total dissolved solids of ~4000 mg l^{-1}. Half of the (9 nos) contaminated samples have >1000 mg/l of Cl^-. Similarly, 32% of the normal samples have Cl^- >200 mg/l. The Cl^- being contributed from non-lithogenic sources, its abnormal concentration can be attributed to legacy leachate of solid waste dumps in the watershed. Chloride and sulfate in groundwater increase due to municipal and industrial waste dumped on the ground surface. Elevated concentrations of chloride (Cl^-) and sulfate (SO_4^{2-}) in surface and groundwater are common in all countries and can serve as indicators of groundwater pollution (Venkatesan and Swaminathan 2008). Decay and decompose of solid waste, the bulk of which include domestic and construction waste material, generate leachate containing Cl^-, Na^+ and Ca^{2+} in a substantial amount which joins stream flow and percolate into the sub-surface domain. Ahluwalia and Patel (2018) mentioned 50% of solid waste composed of domestic, food and organic material and 29% construction/demolition waste. Mohit Somani et. al.,(2019) reported leachate contains 9000 mg/l Cl^- in Hyderabad dumpsite and inferred - high concentration of chloride found in the leachate can be attributed to the sewage ingress, domestic effluents, fertilizers dumped at the landfill sites. Incomparable composition of Cl^-, Na^+ and NO_3^- ions, as displayed in regression analysis the R^2 for Cl:Na was 0.21 for contaminated samples and 0.49 for normal samples reaffirm that Cl^- originated from the dumpsite. The soaring mean TH (1478 mg/l) in contaminated samples was due to high bivalent cations especially Ca^{2+} which could have been sourced from the degradation of biowaste and disintegration of debris from construction activity. Najafi et. al., (2020) in their study of the landfill site, Qaem Shahr City, Iran inferred - the high concentrations of nitrate and hardness in the downstream wells (W_1–W_{11}) of the landfill were demonstrated by the results of the Kriging assay which can be correlated with the penetration of leachate into these wells. Chen et. al., (2019) reported high concentrations of total hardness, Cl^-, SO_4^{2-}, TDS from their research on Municipal Solid Waste Landfill in Lianyungang, China. Thomas Baumann et. al., (2006) opinioned that the colloids form a rather persistent coating around the aquifer matrix that reduces the hydraulic conductivity and enhances the sorption capacity of the aquifer close to the waste disposal sites. Presence of high organic compounds like TOC - 154 mg/l, COD - 176 mg/l, and BOD - 117 mg/l (average of 15 groundwater samples of pre-monsoon 2012) support the presence of biomass in waste dumps. Moderate mean HCO_3^- (332 mg/l) suggests the aquifer received limited freshwater influx but was replenished with base flow or unnatural sources which enriched the ion content of groundwater. The pH could be balanced by alkalinity as reflected by basic nature of water (pH is >7.00 in most samples). The aquatic chemistry signifies solute transport could have occurred by advection. Leachate is generated from the decay of huge and matured (aged) solid waste dumps that have reached the methanogenic

phase and located on topographic high seep into surface water bodies. Contaminant plume flow was a continuous process from pools of leachate to groundwater system supported by hydrodynamics. Adsorption of rapid ion influx could have aided by the favorable chemical makeup of groundwater and prevailing redox conditions in the host environment. Similar chemical imprints in different hydrological features of watershed support the assumption.

Hydrogeochemistry

Evaluation of hydrogeochemistry would through light on the ion enrichment activity. It is all the more necessary for contaminated water resources because apart from external infusion the natural processes also govern the groundwater chemistry (Elisabetta et, al., 2019). Sorption of contaminants depend on recipient water chemistry which would be evident in spatial hydrochemical variations thus deducing the hydrogeochemistry of local aquifers apart from contaminant chemistry is gaining grounds. Understanding the hydrochemistry of the host environment would facilitate delineating the stoichiometry involved in the mineralization of water to contamination levels. Cendón et. al., (2015) inferred the local geology favors the retention of contaminants by ion-exchange processes within the clay-rich soils and the shale layer underlying the burial site. Thomas et. al., (2000) emphasized evaluation of redox conditions in groundwater pollution plumes is often a prerequisite to perceive the behavior of the pollutants in the plume and also because redox conditions of a groundwater contaminant plume from a point source usually differ from the redox condition of the pristine aquifer. For better comprehension of contaminant chemistry, all the groundwater samples belonging to four different sampling episodes were considered for the hydrogeochemical study.

Ion dominance pattern: Cl^- was the most dominant ion in all the four sampled sessions and was followed by one of the cations Ca^{2+}, Mg^{2+}, Na^+ in second and third positions (Table 2). A similar order of cation and anion dominance in groundwaters proximal to dumpsites in Awka was reported by Egbueri (2018). In pre-monsoon 2011 the ion dominance pattern (IDP) was $Cl^- > Na^+ > Ca^{2+} > HCO_3^- > Mg^{2+} > SO_4^{2-} > NO_3^- > K^+ > F^-$, in post-monsoon it was similar except Ca^{2+} replaced Na^+ in second position and K^+ and F^- were interchanged. Joan Nyika and Ednah Onyar (2019) in their research on groundwater in Roundhill landfill vicinity of South Africa noted reverse ion exchange in BH 2 to 3 due to Na^+ and Cl^- dominance while BH 1 showed simple ion mixing and had no dominant ions. In 2012, molar content of Mg^{2+} gained significantly reaching the second position in pre-monsoon and third in post-monsoon replacing Na^+ and Ca^{2+}. Another notable change in 2012 was bicarbonate concentration reduced considerably ranking in fifth and sixth positions in pre and post-monsoon seasons respectively (Table 2). A variable number of samples and sampling points in 2011 and 2012 could be one of the reasons for changes in IDP. It also validates the assumption of ionization processes were influenced by multiple sources and external factors.

Ion strength: The percent mill-equivalent concentration of Cl^- was about 28 in the year 2011 but has risen significantly in 2012 to 35. Similar percent (25) content of Na^+ in pre-monsoon 2011 and Ca^{2+} in post-

monsoon 2011 can be accounted for ion exchange. Nicholson et. al., (1983) opinioned exchange of cations between the aquifer material and the leachate-contaminated groundwater is an apparent cause of calcium release and subsequent precipitation of calcite in the plume. Ion content of Mg^{2+} has increased to 19 % in pre-monsoon 2012 and 15 % in post-monsoon, whereas Na^{+} percent content reduced to 15 and 19 in the same periods. In post-monsoon 2012 the % meq content of SO_4^{2-} has raised to 7 from about 3 in all the past sampling sessions (Table 2). Apart from the induced ionization, natural processes can be attributed to variations in ion strength over a year of observation. Joan Nyika and Ednah Onyari (2019) noted the dominance of Ca^{2+}, Cl^{-}, Mg^{2+}, and NH_4^{+} ions in some boreholes suggested anthropogenic pollution from the landfill leachate of Roundhill Landfill, South Africa.

Water type: Water speciation was studied plotting the data in Chadha's modified Piper plot (Chadha 1999). In all the sampling episodes the groundwater samples were mostly of Ca-Mg-Cl type with permanent hardness. They were 53% in pre-monsoon 2011, 33% in post-monsoon, 60% in pre-monsoon 2012, and 68% in post-monsoon (Fig. 5a and b). Even most of the highly contaminated groundwater and some of the surface water belong to Ca-Mg-Cl facies. It indicates the mixing of high salinity water caused by surface sources, such as the liquid and solid waste discharged into the nearby land and channel (Jeyara et. al., 2016). The groundwater chemistry changes from Ca-HCO_3 type to Ca-Mg-Cl and Ca-Na-HCO_3 types due to the cations exchange reaction as well as the simple mixing with saline water (Masoud Saatsaz et. al., 2013). The water type vindicates the longer residence time of groundwater in the shallow aquifers (Prasanna et. al., 2010). Another dominant water type was Ca-Mg-HCO_3; about 30% of samples of all but the last sampling episode belong to this facies. Many surface water samples and very few groundwater samples from all the sampling sessions were of Na-Cl/SO_4 type. It confirms that surface water bodies were cesspools for groundwater contamination. Water facies further support the contention that natural ionization processes, as well as anthropogenic activity were operating in the mineralization of water. Emmanuel et. al., (2019) also made similar observations in their study of the Ga West Municipality in Ghana - the Ca^{2+} and HCO_3^{-} in these type of waters are primarily from the dissolution of carbonate minerals whilst the Na^{+} and Cl^{-} may have been input from water-rock interaction with granitic rocks, seawater intrusion, and anthropogenic activities in the watershed.

Ionization processes: Feeble seasonal, temporal, or spatial trends, together with abnormal increase and decrease of certain chemical constituents suggest that multiple processes were active in the evolution of hydrochemistry. Daniel Abiriga et. al., (2020) in a study on Revdalen Landfill sites in Norway inferred season was the least influential on concentrations of contaminants in the groundwater and observed strong attenuation of pollutants with distance. The mechanism controlling the mineralization process was studied using the Gibbs (1970) plots, in Na+K:(Na+K+Ca) vs TDS plot most of the data points of both the seasons of two years fall close to rock-dominance (Fig. 6a and b). The Cl:(Cl+HCO_3) vs TDS plots exhibit that in pre-monsoon evaporation process whereas in post-monsoon water-rock interaction was dominant in ionization of groundwater (Fig. 7a and b). It can be inferred from the Gibbs plots that apart from water-rock interaction (aquifer material) evaporation (external sources) processes controlled

the ion enrichment process. Selvakumar et. al., (2017) inferred the evaporation dominance is promoted by causes of semi-arid environmental climate conditions and surface sources. Evaporation increases salinity by increasing Na^+ and Cl^- with relation to the increase of TDS (Prasanna et. al., 2010). But the disproportionate presence of these ions, as evident in mean values of all the samples (Na^+ 184 and Cl^- 474 mg/l) and >1 Cl^-/Na^+ ratio in majority of the samples for each sampling session (mean value ranges from about 1 to 4 meq/l), confirm Cl^- was contributed through anthropogenic sources (Fig. 8a). Ca^{2+}/Mg^{2+} (meq/l) ratio of >2 (mean value vary between 1 and 14) in the majority of the samples in both the seasons (pre and post-monsoon) of 2011 (71%, 75%) and few samples in 2012 (28%, 16%) demonstrate alkali earths were contributed by silicate weathering. The high molar ratio of Cl^-/HCO_3^- (mean for contaminated groundwater samples vary 3 from to 32 and 1 to 5 meq/l in normal samples), the ratio has increased tremendously in 2012 (Table 4), it validate insufficient influx of freshwater and non-lithogenic sources altered the groundwater chemistry (Tahoora et. al., 2014). Kurakalva et. al., (2016) concluded in groundwater quality assessment studies around the Jawaharnagar municipal solid waste dumping site that the pollution source was dominated over the natural process in the vicinity of the studied area.

Chloroalkaline indices: Large variation was noticed in chloroalkaline indices-1 and 2 (CAI-1; CAI-2) among the samples of four sampled sessions. Most of the surface water samples in all the sampling sessions had positive CAI-1 and 2 values. Among the groundwater samples, 41 % in pre-monsoon and 50% in post-monsoon seasons of 2011 recoded positive indices whereas in the following year their strength has increased to 68 % in pre-monsoon and 76 % in post-monsoon and rest had negative values. This supports the assumption that direct base (cation-anion) exchange reactions were dominant, which involves the exchange of Na^+ and K^+ from the pore water with Ca^{2+} and Mg^{2+} of the aquifer material. Negative CAI ratios observed in many samples of 2011 demonstrate the exchange was of indirect base indicating chloro-alkaline disequilibrium, where replacement of Ca^{2+} and Mg^{2+} in groundwater occurs with Na^+ and K^+ of the host rock (Schoeller 1965, 1967; Al-Ahmadi 2013). The high temporal fluctuation of Na^+ and Ca^{2+} (Na^+ 11.24; 8.08; 6.32; 7.48; 7.58; Ca^{2+} 9.78; 6.60; 6.12 meq/l mean for four sampling sessions sequentially) content, as well as the abundance of certain ion concentrations and water facies, support that both direct and reverse exchange processes, though on a variable scale, were governing the cation content of the groundwater. The inference was further strengthened by about 2 (mean, meq/l) ratios of $Ca^{2+}+Mg^{2+}/Na^+ + K^+$ in both normal and contaminated groundwater samples whereas it was 0.6 in surface water samples (Table 4).

Inter-relationship of ions: Correlation (r) of all tested parameters of each sampling session shows a strong relationship exists between Ca^{2+} and Cl^- which was 0.97, 0.98, 0.86, and 0.92. Both were largely contributed from different sources, Ca^{2+} from the host environment and Cl^- from anthropogenic activity. Similarly r for TH:Cl^- was unusually high (average 0.96) in all the sampling sessions. The strong correlation of Cl^-:Na^+ in both the seasons of 2011 (>0.90) suggests halite dissolution triggered by wastewater discharge from solid waste dumps could be contributing the ions to the aquatic environment.

However, the weakening of relation (0.71) between Cl^- and Na^+ in 2012 samples was distinguishable (Table 3). It was also displayed in the cross plot of these ions in meq/l (Fig. 8a) which suggests multiple sources for these ions (Conglian et. al., 2019; Reddy 2013). The unusual relationship between Cl^- and other ions confirms the adverse impact of the solid waste dump on groundwater. In the natural environment, a negative relationship between dissolved Ca^{2+} and Na^+ was expected. In the study area good relationship exists between Ca^{2+} and Na^+ in all sampled sessions except post-monsoon 2012 (r=0. 96, 0.85, 0.65 and 0.42 sequentially) which validates anthropogenic source. Unnisa and Bi (2017) revealed that the physicochemical parameters are independent of anions and major cations in the Jawaharnagar area but some cations, anions, and physical parameters were found interrelated.

The <0.50 ratio of Ca^{2+}/Na^+, Mg^{2+}/Na^+, $Na^+ + K^+/TZ^+$, >2.0 Ca^{2+}/Mg^{2+} in many samples and the data points plotted along the 1:1 line in Fig. 8b ($Na^+ + K^+$ vs $Cl^- + SO_4^{2-}$) suggest the silicate weathering could be responsible for cation contribution in groundwater apart from dissolution of leachate. Nearly half of normal groundwater and most of the contaminated groundwater, as well as the surface water, had a Na^+/Cl^- (meq/l) ratio of <1 that emphasizes halite dissolution is responsible for sodium occurrence in these waters. The 1:2 ratio of Na^+:Cl^- in contaminated groundwater unequivocally proves that Cl^- was sourced from effluents generated from the domestic waste of MDY together with local habitations. The samples plotted close to the 1:1 line could have undergone evaporation for Na^+ enrichment. Scattered swing of data points to words x-axis (Cl^-) exhibit influence of contamination in Cl^- enrichment (Fig. 8a). The predominance of Ca^{2+} over Mg^{2+}, Na^+, HCO_3^- in many groundwater samples depicts that weathering of carbonate minerals could be responsible for the input of $Ca^{2+}$and Mg^{2+} (Table 4; Fig. 8c). The assumption was substantiated by the ($Ca^{2+} + Mg^{2+}$) versus ($HCO_3^- + SO_4^{2-}$) scatter diagram in which most of the normal groundwater and few sample points of other categories aligned along the equiline (Fig.8d). In Langelier and Ludwig (1942) plot (modified) majority of the samples fell above equiline, which proves the prevalence of water pollution in the area, and the progression of dominant ion species (Cl^-, $Ca^{2+}/Na^+/Mg^{2+}$) can be attributed to it (Fig. 9).

Contaminant chemistry

Contaminated water displays distinct chemistry, the surface water, which forms the reservoir of pollutants, has high reactive chemical characteristics like about (mean) 8.00 pH, 750 TH, and 800 mg/l Alkalinity (Table 1). These physicochemical properties promote absorption of ions from the inflow of leachate at an incipient stage which was visible in 4000 mg/l TDS. Higher molecular content of Na^+, K^+, HCO_3^- and SO_4^{2-} than groundwater suggest the influence of external influx together with the conductive hydrochemical environment on surface water quality (Fig. 10). The chemical makeup of contaminated groundwater was unique by having low pH (7.30) and high TH (1478 mg/l) in comparison with surface water and normal groundwater which confirm host water chemistry facilitated ion enrichment rather than physical features like hydraulics and thermodynamics. A mean TDS of about 3100 mg/l indicates high mineralization. The mean meq/l contents of Alkali earths, and Cl^- were higher whereas Alkali metals,

HCO_3^-, SO_4^{2-} and NO_3^- lower in contaminated groundwater than other categories of water samples (Fig. 10). Very high and near similar Cl^- concentration in surface water (37 meq/l) and contaminated groundwater (40 mq/l) indicate intermixing of these waters. The contrasting composition of cations confirms ion exchange process was active. Similarly, low Na (19 meq/l) and high Cl^- (40 meq/l) confirm they were from dissimilar sources as well as processes. In the normal groundwater the mean pH was high (7.90) and about 400 mg/l of TH was unusual for the kind of country rocks. The mean milliequivalent content of all other examined chemical constituents was much lesser than other samples of groups. It can be deduced from <5 meq/l concentrations of Ca, Mg, Na, HCO_3^- and Cl^- that unaffected water still exists in the area though the MWD has attained a stable methanogenic phase (Peter Kjeldsen et. al., 2002). Close range in the strength of SO_4^{2-} (2 to 6 meq/l) and NO_3^- (1 to 4 meq/l) in surface water, as well as both the types of groundwaters, point out the prevalence of redox and denitrification conditions. Disproportionate content of Cl^- and NO_3^- in most of the samples as evident from their ratios of 1:0.12 for surface water, 1:0.02 for contaminated samples and 1:0.16 for normal groundwater (mean; meq/l) indicate different sources and exponential strength of Cl^- in all samples can be attributed to huge solid waste dumps. More than 5 meq/l of Cl^- content and 1 to 3 meq/l of NO_3^- in half of the normal groundwater samples (largely gathered from the non-core area) could be due to multiple sources including poor disposal of sewerage from rapidly developed unplanned colonies. Sequential reduction in the content of certain ions from surface water to contaminated groundwater and normal groundwater indicates the flow path of pollutants from source to sink. The scattered distribution of contaminated water samples in cross plots could due to the unique chemical characteristics of these waters.

Conceptual mass flux model

The plume proliferation can be predicted as - leachate generated by solid-liquid reactions between decomposing solid waste and percolating water in the core area of MDY leak down into natural water tanks and get accumulated and flow into streams due to hydraulic gradient. The leachate from these surface water bodies seep into the sub-surface domain through the soil and thick weathered mantle while undergoing hydrochemical and natural attenuation processes. The fluid flow to deeper aquifer horizons occurs through a network of fracture systems where water-rock interactions and ionization occur altering the groundwater chemistry. The part of the polluted pore water under hydrostatic pressure conditions joint stream in the form of effluent seepage. The plume flow velocity and direction depend on a fluid density and hydrodynamics of receptive aquifers. The age and maturity level of the solid waste dumps also control the contaminant kinetics. Propagation of point pollution from MYD to different components of watershed occurs through hydrogeochemical cycle resulting in temporal and spatial variability in contaminant concentration. Randall (1981) inferred - contaminant transport in the groundwater environment could be in nonlinear flow systems along streamlines in a non-uniform flow field. Thomas et. al., (2000) explained contaminant transport in reactive chemistry approach as - when sufficient organic matter and other reduced components leak from a point source into an aquifer, strongly reduced redox conditions will develop close to the source and the plume will develop a redox gradient along as

well as transversal to the main groundwater flow direction. These theories were aptly reflected in the present study area.

Conclusions

Ion content of both surface and subsurface waters unambiguously establishes the municipal solid waste dumpsite as a point source for widespread contamination at Jawaharnagar dump yard. Local physiographic features, un-engineered disposal site together with favorable hydrological conditions triggered the mass movement of leachate from dumpsite to surface water bodies and in the form of the plume into aquifers though preferred pathways. It can be inferred from high molar content of Cl^-, Ca^{2+} or Na^+, dominant Ca-Mg-Cl water type, and data points confined to water-rock interaction as well as ET dominance fields in Gibbs plots that extreme ionization of water resource can be contemplated to anthropogenic source apart from geogenic processes. A favorable host water environment facilitated natural ion enrichment. Dynamic hydrochemical conditions due to continuous mass flux resulted in rapid reactions in the aquifer leading to super saturation. Unique correlation of Cl^- with Ca^{2+}, TH, and Na^+ as well as low ratios of Na^+/Cl^-, Ca^{2+}/Cl, HCO_3^-/Cl^-, TA/TH and high Ca^{2+}/Mg, Cl^-/NO_3^- values support the contention. Contamination-induced ionization led to rapid variations in the chemical makeup of pore water which created the disequilibrium resulting in ion-exchange or base-exchange. Contaminants complemented natural ionization processes and vice versa.

Declarations

-Ethical Approval	“Not applicable”
-Consent to Participate	“Not applicable”
-Consent to Publish	“Not applicable”
-Authors Contributions	"PN - analyzed and interpreted the results. SR - major contributor in writing the manuscript. RK - field work, data generation and compilation. All authors read and approved the final manuscript."
-Funding	“Not applicable”
-Competing Interests	"The authors declare that they have no competing interests".
-Availability of data and materials	All data generated or analyzed during this study are included in this published article [and its supplementary information files].

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Web sites

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Tables

Due to technical limitations, table 1, 2, 3 and 4 is only available as a download in the Supplemental Files section.

Figures

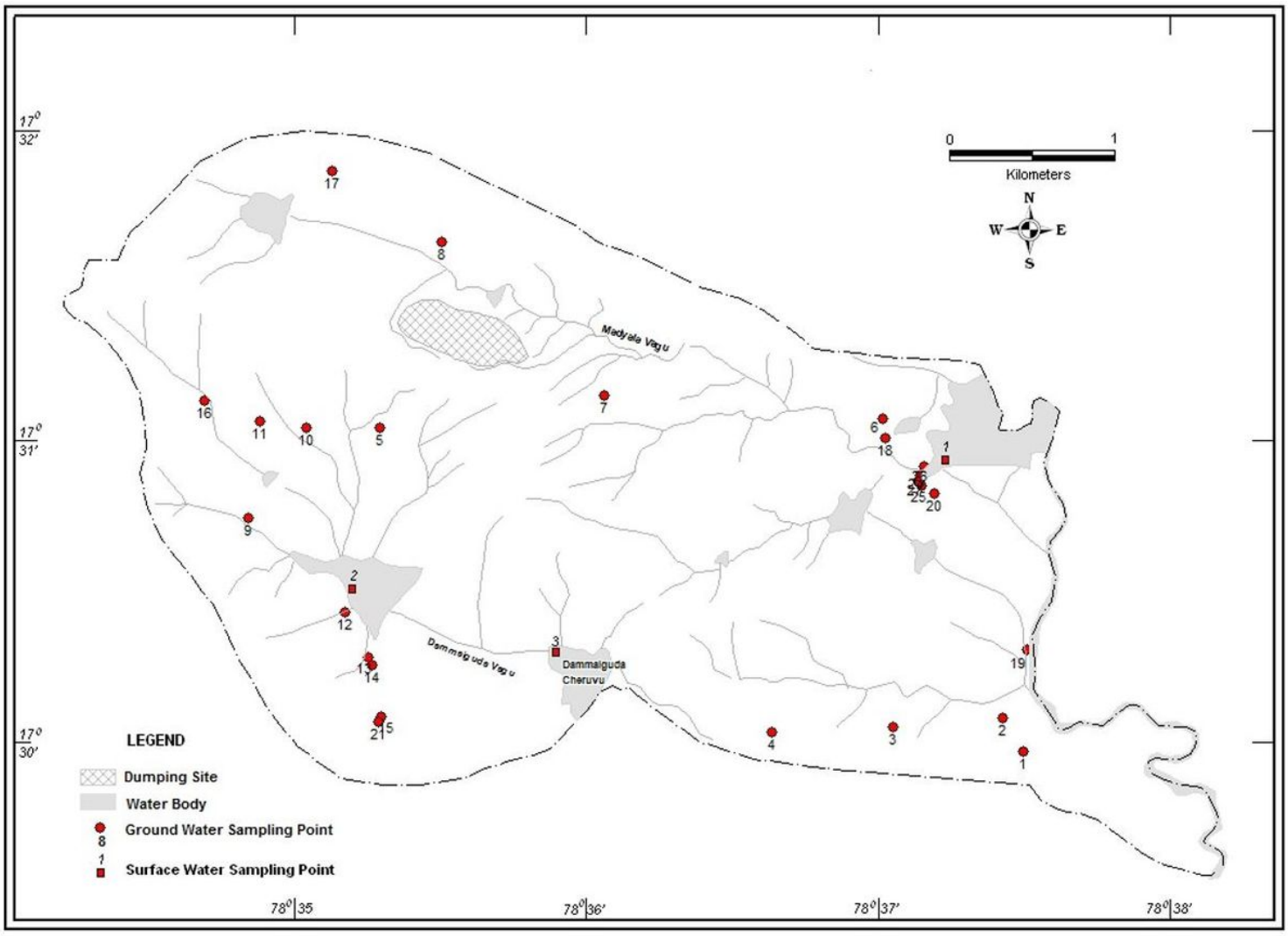


Figure 1

Study area and samples locations.

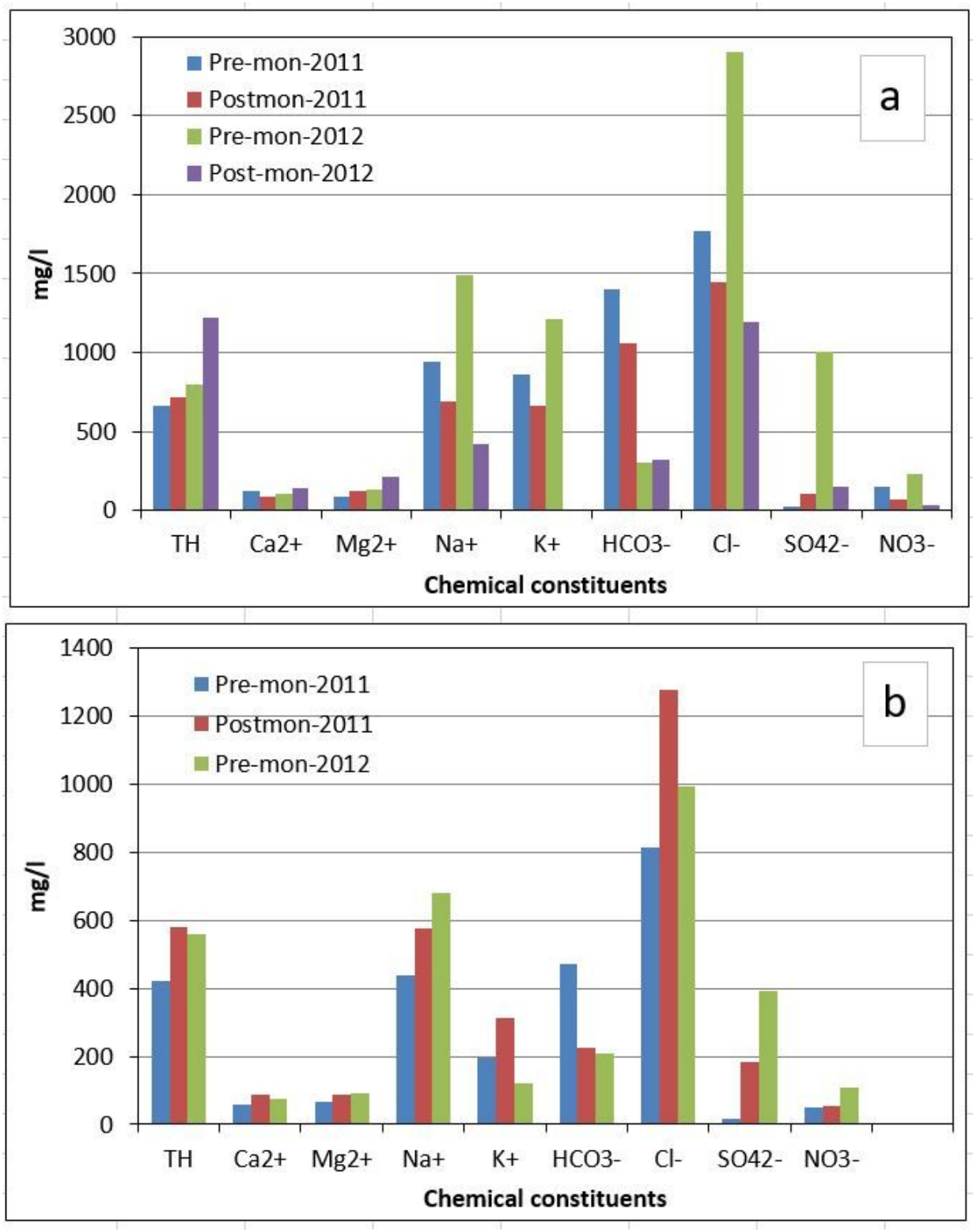


Figure 2

Variations in concentration of chemical constituents in (a) Cherial tank and (b) Dammaiguda tank (mon: monsoon)

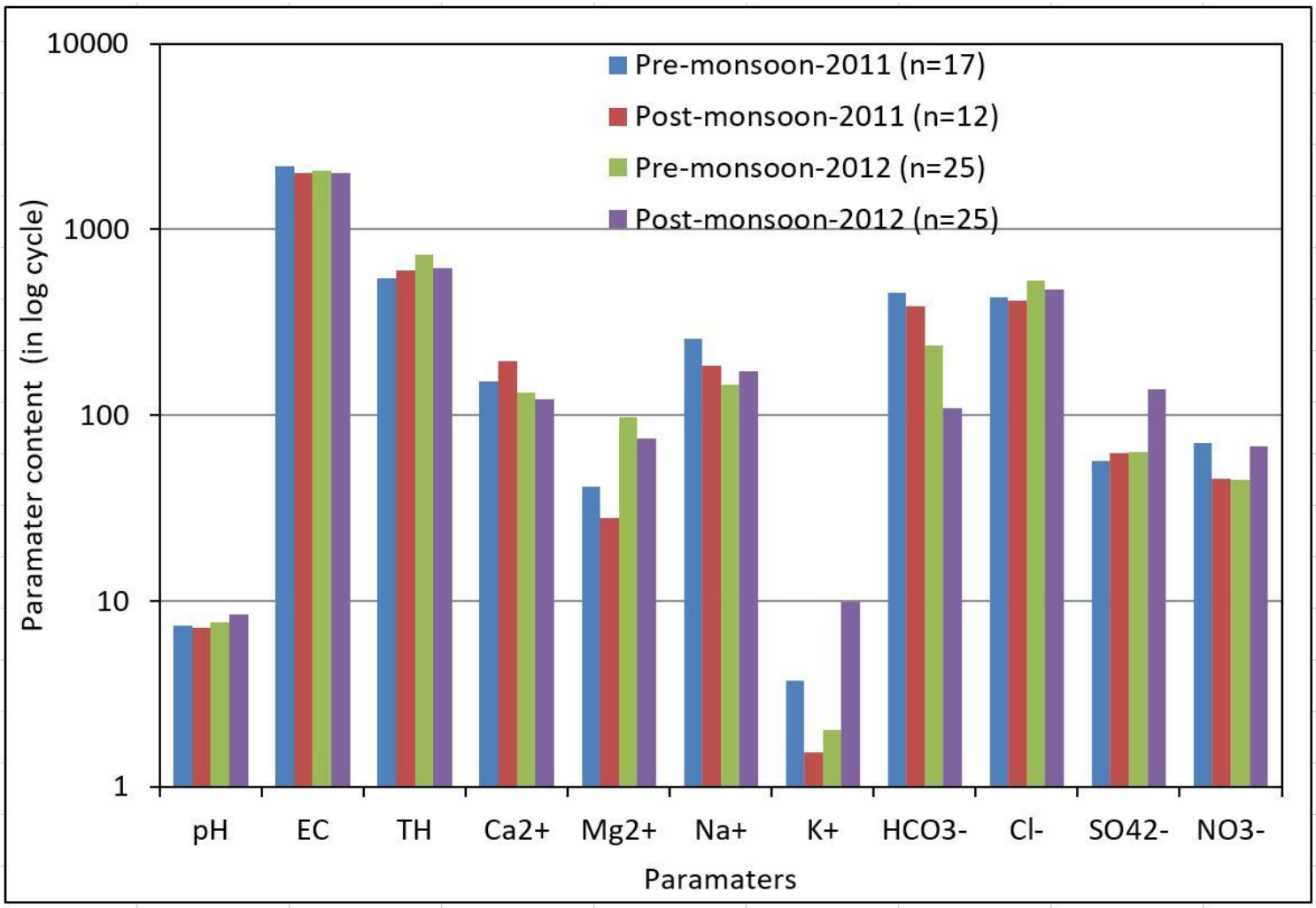


Figure 3

Varations in mean paramater content of groundwater among four sampled sessions

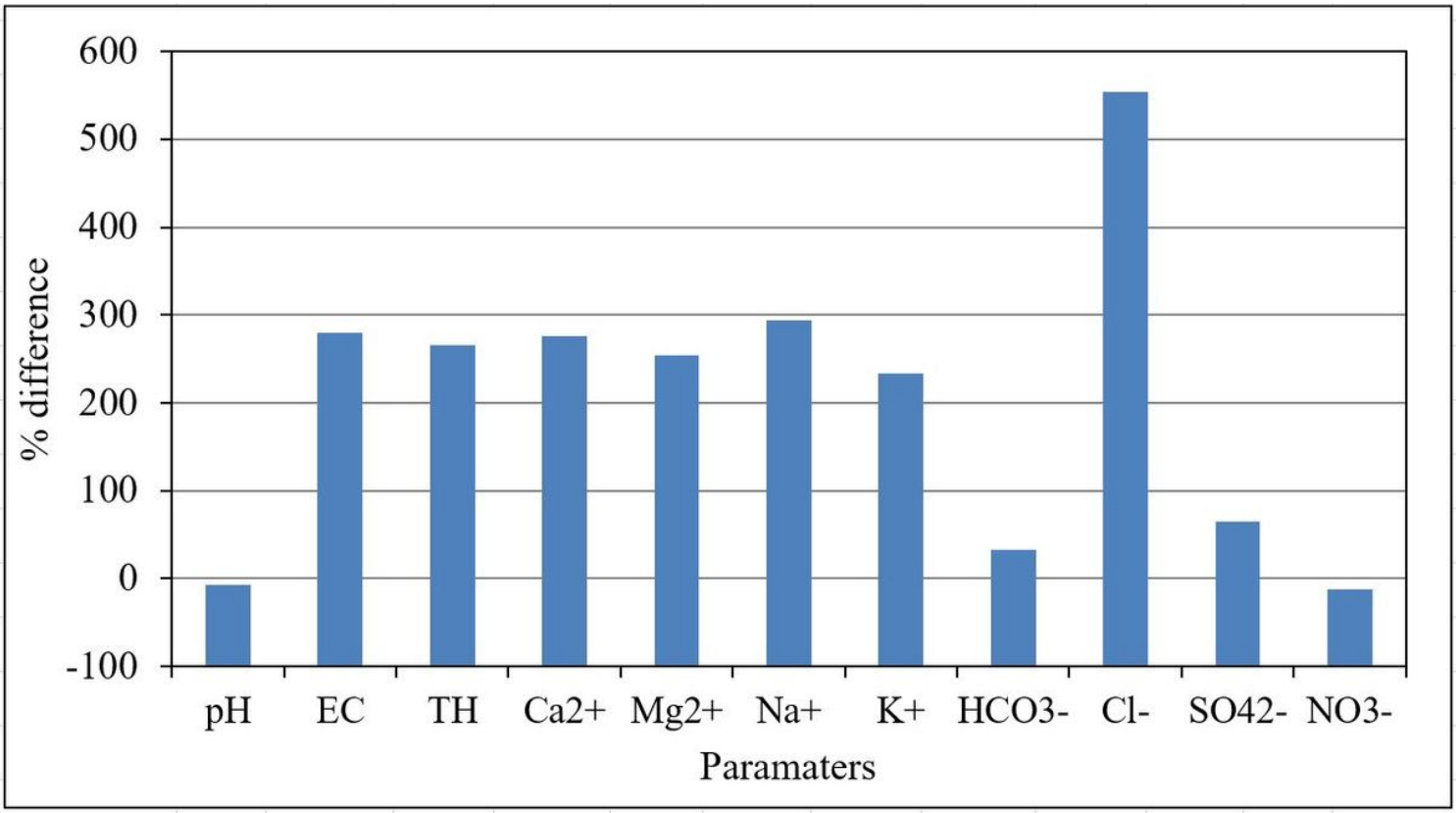
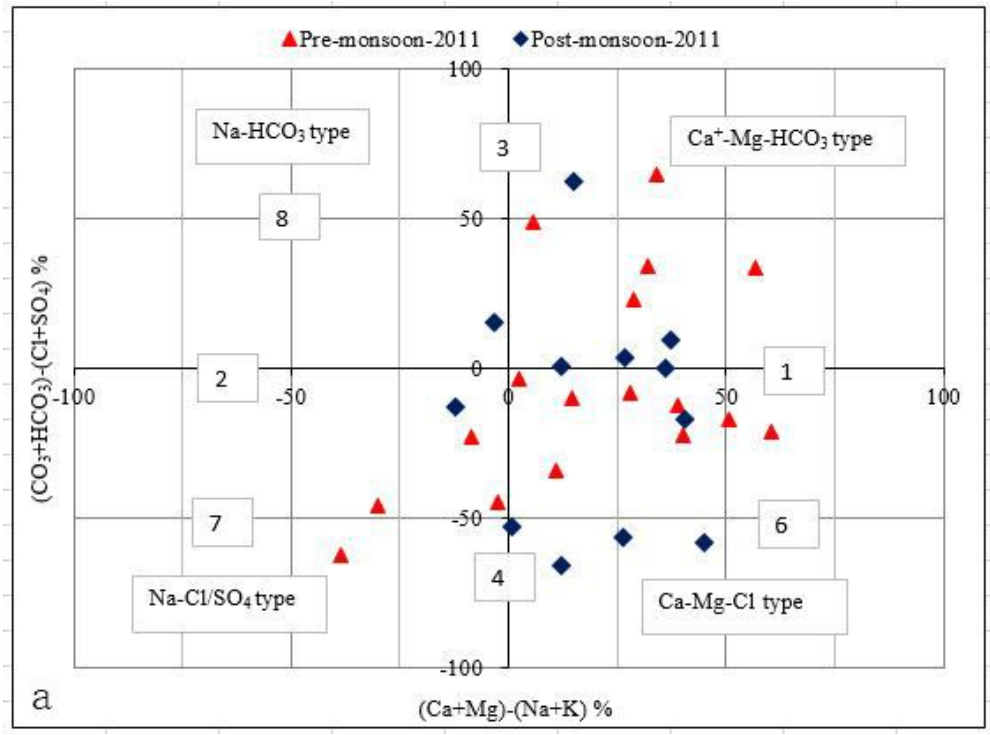


Figure 4

Variations in mean content of water chemistry among normal and contaminated groundwater samples.



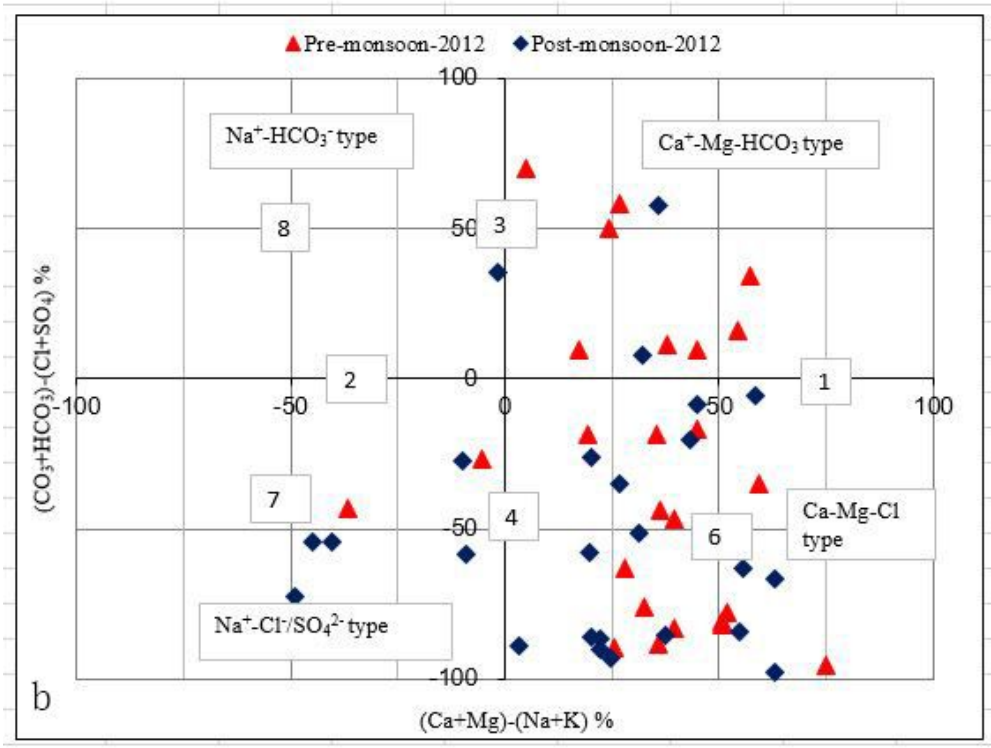


Figure 5

a. Modified Piper Diagram for pre and post-monsoon-2011 groundwater samples. b. Modified Piper Diagram for pre and post-monsoon-2012 groundwater samples.

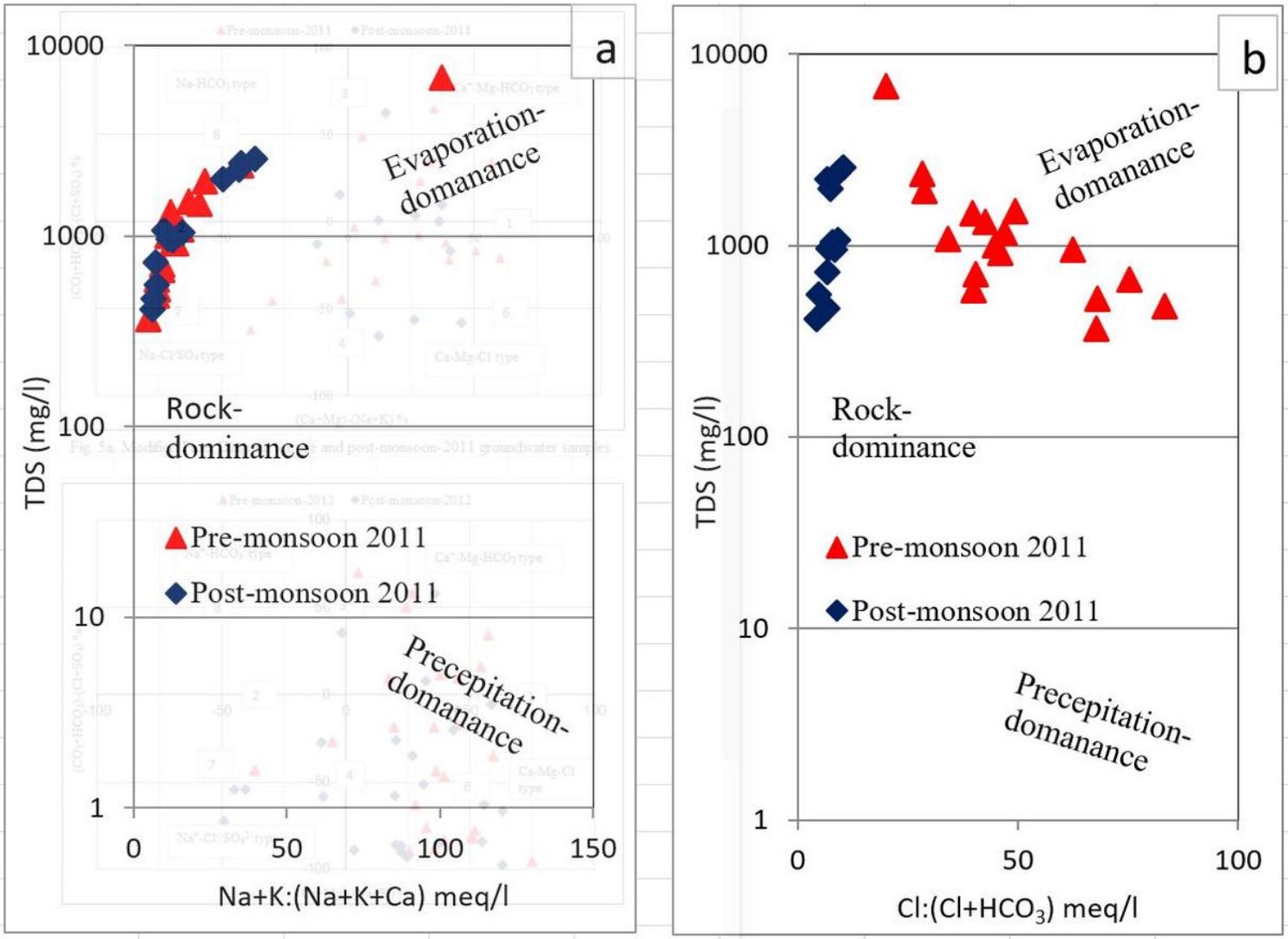


Figure 6

a and b. Gibbs plots for pre and post-monsoon-2011.

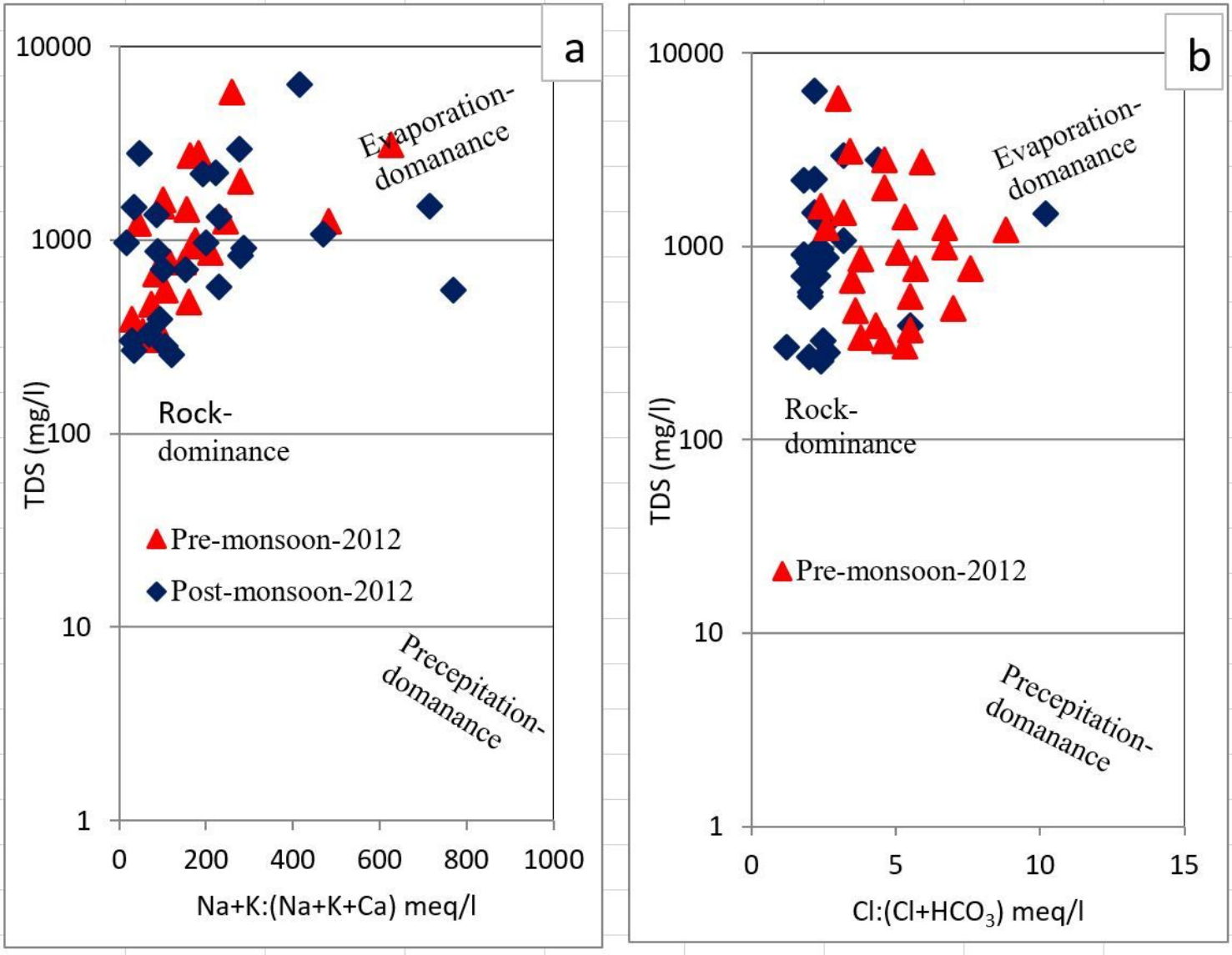


Figure 7

a and b. Gibbs plots for pre and post-monsoon-2012.

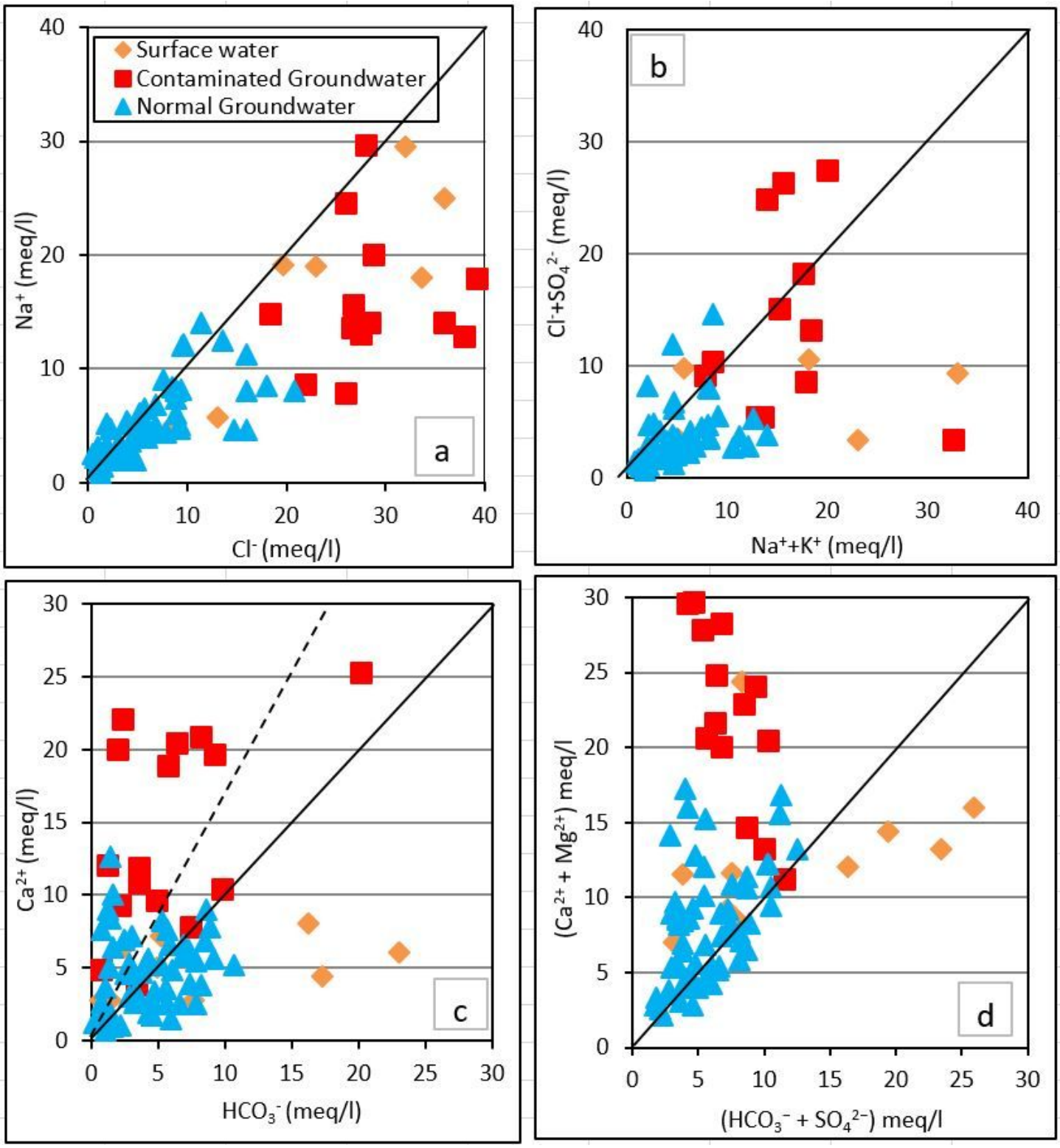


Figure 8

Cross plot of (a) Cl vs Na; (b) Na+K vs Cl+SO4; (c) HCO3 vs Ca; (d) HCO3+SO4 vs Ca+Mg.

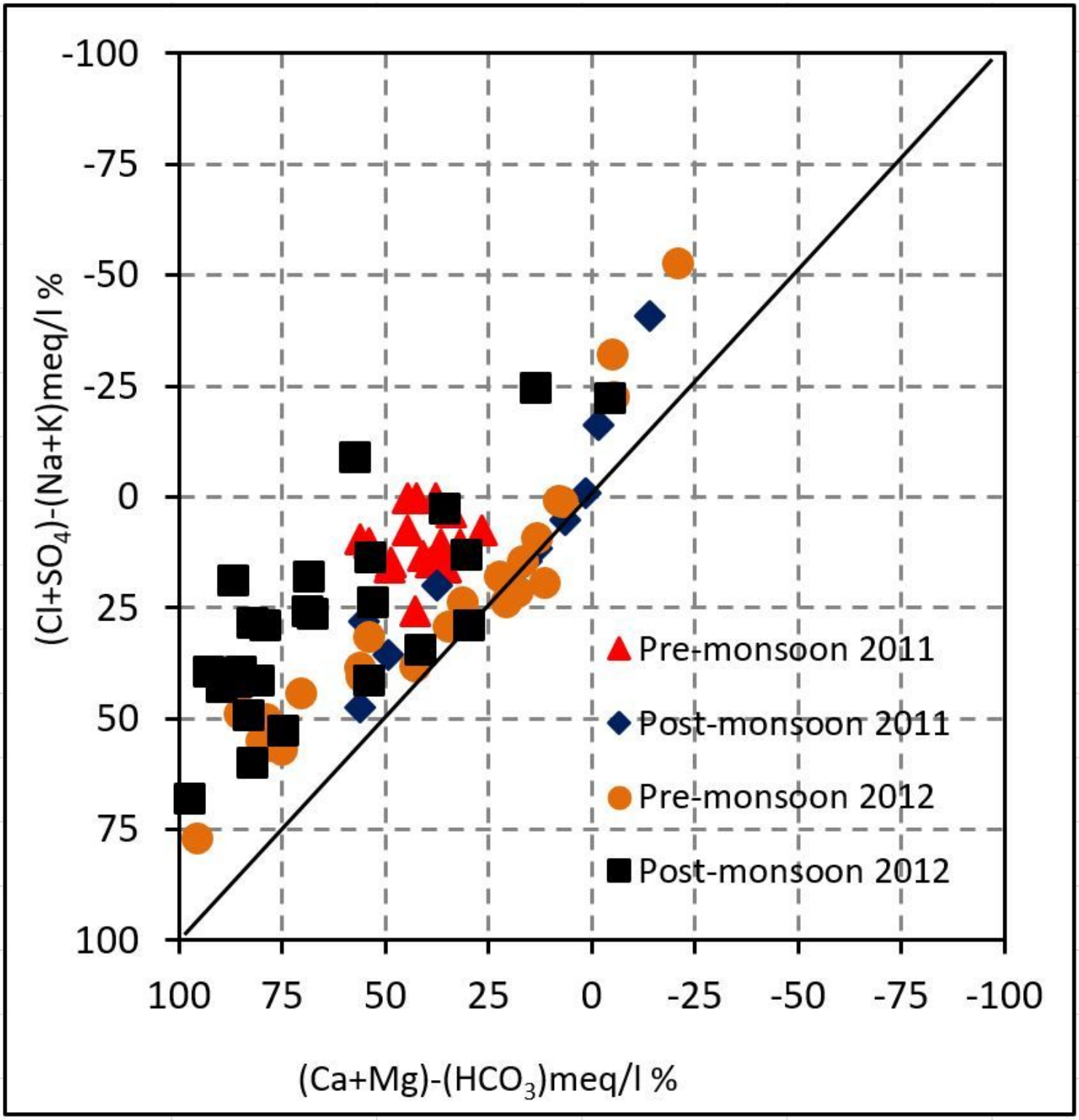


Figure 9

Langelier and Ludwig plot (modified).

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- Table1.xlsx
- Supplementalmaterial1SW.xlsx
- Supplementalmaterial2ContaminatedGW.xlsx
- Supplementalmaterial3NormalGW.xlsx
- Table2Iondominance.xlsx
- Table3CorrMatr.xlsx
- Table4aSummarryIonratios.xlsx