

Climatic Influences on Arsenic Health Risk in the Metamorphic Precambrian Deposits of Sri Lanka: A Re-analysis-based Critical Review

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Abstract: The present study examines the arsenic associated human health risk in Sri Lanka with due consideration to age and gender vulnerability. While the existing low-grade metamorphic rock carries less possibility of arsenic contamination, the usage of old generation arsenical pesticides may increase the future possibility of a catastrophe. Through a comprehensive Health Risk Index (HRI) it could be seen that the north part of the country carries higher risk for children below eight years of age to develop cancer in long run. Interestingly rainfall pattern correlates with arsenic vulnerable zones, denoting that high monsoon led recharge may prevent rapid anthropogenic fluctuations in anoxic zones thereby preventing vigorous formation of oxy-hydroxides. However, the possibility of sea-water intrusion due to excessive groundwater abstraction may change the major-ion chemistry soon which may impact the co-occurrence scenario of arsenic and fluoride. In this regard the study identifies the wetter regions and their shift from 1981 to 2018 to propose a more stable groundwater regime in terms of sea water intrusion. There exist few scientific literatures concerning the occurrence of arsenic in the groundwater of Sri Lanka but there exists no framework to provide a meaningful interpretation by combining all the available research outputs. The present study therefore provides a basis for understanding the existing groundwater arsenic contamination of Sri Lanka by providing a scientific framework to the studies done so far by various researchers in the past 30 years.

Keywords: Arsenic; Groundwater; CHIRPS; Health risk.

Introduction

High enrichment of minerals containing arsenic (As) in the shallow and confined aquifers of Sri Lanka mediated via Precambrian era metamorphic rocks is a known challenge (Bandara et al., 2018; Bastiaanssen and Chandrapala, 2003). With more stress on groundwater abstraction to meet drinking needs, a vulnerable situation with regard to sea-water intrusion and metal-oxy-hydroxide desorption exists (Singh et

al., 2019; Singh et al., 2020; Patel et al., 2019; Kumar et al., 2007). The impact of intensive agriculture led anthropogenic forcing in the groundwater has led to erratic weatherability of rock minerals leading to further uncertainties in prediction of ion-exchange reactions (Kumar et al., 2006; Kumar et al., 2010). Further evidences of limestone (Miocene era) weathering changing under climatic extremes has limited our understanding and at the same time increased our curiosity in quantifying the role of climate change in

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causing geogenic contamination of groundwater. While the As poisoning of West Bengal and Bangladesh have been well documented, absence of scientific literature explaining the occurrence and transport mechanism of As in Sri Lanka is a cause of worry where it has been reported to be reaching to a concentration of $25 \mu\text{g L}^{-1}$ (Chandrajith et al., 2016; Chakraborti et al., 2004; McArthur et al., 2004; Nickson et al., 2007). This situation is more critical in regions where groundwater abstraction is dominantly occurring through weathered overburdens and tensional fractures.

According to Chandrajith et al. (2001), low grade metamorphic rocks usually have a very low exposure of As deposits averaging less than 5 mg/kg. While the geogenic As is limited in sedimentary deposits (1–4 mg/kg), the high concentration in agricultural soils (24 mg/kg) indicates As being introduced through phosphate fertilizers and pesticides. The vulnerability further increases in forest soil with high organic content where As has been reportedly reaching 29 mg/kg. The bio-availability of microbes further complements the release process of As in inorganic forms leaving its mineral state through either oxidative or reductive dissolution process of metal base sulphides (Kumar et al., 2019; Herath et al., 2018).

High presence of As in groundwater increases the possibility of occurrence of skin lesions, hyperpigmentation and cancer via oral and dermal uptake making As a potent mutagenic and carcinogenic threat. Recent studies have also pointed that As can aggravate into the cell cytosols by mimicking the properties of phosphates and glycerol leading to a more dangerous reality of arsenic resistant gene (ars) formation when the efflux system of the cell gets activated through ATP Binding Cassettes (ABC) and Major Intrinsic Proteins (MIP) (Hibuse et al., 2006). Another challenge with respect to As toxicity is its existence in trivalent form. Difficulty in conforming the presence of As(III) due to its high oxidation rate to As(V) outside aqueous system often leads to underreporting of the threat. Further degradation to the methylated forms subject to temperature and ion-chemistry only increases the risk.

Co-occurrence of As with fluoride on the other hand is a wider problem in Sri Lanka as compared to As occurring alone. The fluoride geochemistry is more affected by the chemistry of the major ions especially calcium (Ca^{2+}) and sodium (Na^{+}). Minerals like cryolite, hornblende and apatite release F based on the present saturation states of the minerals in alkaline conditions. The origin of F-bearing minerals traces their origin in the igneous formation resulting due to

magmatic differentiation. Presence of fluoride above 1.5 ppm has serious consequences for humans, causing bone deformities with consumption of higher fluoride contaminated drinking water (Podgorski et al., 2018; Smedley and Kinniburgh, 2002).

Rainfall led recharge is among one of the most important factors dominating the As and F mobilization process in the groundwater of Sri Lanka. The wetter region of Sri Lanka (Colombo) usually has a lower concentration of As, compared to the northern drier part, as could be seen by reported studies in the Miocene limestone formations of Mannar region where 30% of the groundwater wells exceeds the WHO permissible drinking limit. Dilution may cause a possible change in the pathway of As mobilization especially during the monsoon seasons when the dynamic equilibrium between the oxic and anoxic zones of groundwater are very much fluctuating. The mixing of high Fe bearing minerals with oxygen zones leads to formation of Fe-OOH (iron-oxyhydroxides) which are found to be great As adsorbing agents (Patel et al., 2019).

In this regard mapping and extracting precipitation information through remote sensing becomes very imperative, when the aim is to get a spatial and temporal idea at a country level and to correlate it with As vulnerable zones. A more sensitive infrared based estimation of precipitation can help build better correlation relationship. Climate Hazards Group Infrared Precipitation with Station data (CHIRPS) is a 35+-year quasi global rainfall data at a spatial resolution of $0.5^{\circ} \times 0.5^{\circ}$, incorporating both in-situ ground-based observations as well as satellite captured information. In order to quantify the impact of As on humans a more comprehensive approach in terms of human health risk is often more valuable. Based on the body weight and the ability to consume water, As has a differential impact vis-à-vis gender and age.

Daily intake via oral and dermal pathways carries equal vulnerability and hence its inclusion decides the hazard quotient in children as well as adults. The present study has therefore been done keeping following three objectives in mind: (i) to identify the temporal change in the rainfall pattern of Sri Lanka using high resolution CHIRPS data, as it has been reported that the wetter part is exposed to lesser As contamination, (ii) to examine the existing health risk due to As in Sri Lanka and its associated relationship with rainfall using secondary data through different studies and (iii) to quantify the role of sea-water intrusion in possibly changing the conditions with respect to arsenic and fluoride desorption.

Materials and Method

Study Area

Sri Lanka is located between 5°55' to 9°51' latitudes and 79°42' to 81°53' longitudes. The major Precambrian litho-tectonic units of Sri Lanka can be categorized into five classes: (i) *Miocene to Quaternary*, (ii) *Wanni Complex* – contains granitic migmatites, scattered meta-sediments with similar formation conditions as Highland Complex, (iii) *Highland Complex* – consisting of supra-crustal rocks especially marbles, calc-silicate rocks and metaquartzites. These formations usually occur under metamorphism between upper amphibolite and granulite facies condition, (iv) *Vijayan Complex* – granitic and augen gneiss and (v) *Kadugannawa Complex* – dominant hornblende and biotite formations (Cooray, 1992).

Sri Lanka has been classified under two characteristically different climatic zones defined as the “wetter” region and the “drier” region. In terms of rainfall, the southern part receives more than 3300 mm of rainfall annually and the northern drier region receives less than 900 mm. Tropical cyclones and depression dominates the monsoon season of Sri Lanka. Iron ore, zinc, manganese, nickel and cobalt dominates in terms of minerals structure in the island. Apart from arsenic, the presence of other geogenic contaminants like fluoride has also deteriorated the quality of groundwater.

Chronic Daily Intake (CDI) and Health Risk Index (HRI)

The use of HRI in evaluating the non-carcinogenic risk has been suggested by US Environmental Protection Agency (USEPA, 1989). In our study we have therefore used this approach so that we can meaningfully interpret the human health hazard based on the available literature. Chronic Daily Intake (CDI) denotes the total exposure of As through drinking and skin contact. A standard ingestion rate of 2 L and 1 L has been suggested for adults and children respectively. However, a further modification has been carried out using the guidelines of Food and Nutrition Board (2004) using its criteria of defining an Adequate Intake (AI) dosage varying with age and gender (children (1.7 L), females (2.7 L) and males (3.7 L)) (Table 1). Further calculation of Cancer Risk (CR) has been done by adjusting the Potency Factor (PF) which for the oral ingestion remains at $1.5 \text{ mg/kg/day}^{-1}$. PF ensures that in our calculation of CR the confidence interval remains at 95%. Further using Reference Dosage (R_fD), Hazard Index (HI) has been calculated (Table 2). A HQ value below 1 shows no major non-carcinogenic risk. Finally, a Cancer Incidence value between 10^{-4} and 10^{-6} validates no major risk. In the present study almost all the major scientific papers have been considered which have reported As concentration for evaluating the HRI in Sri Lanka.

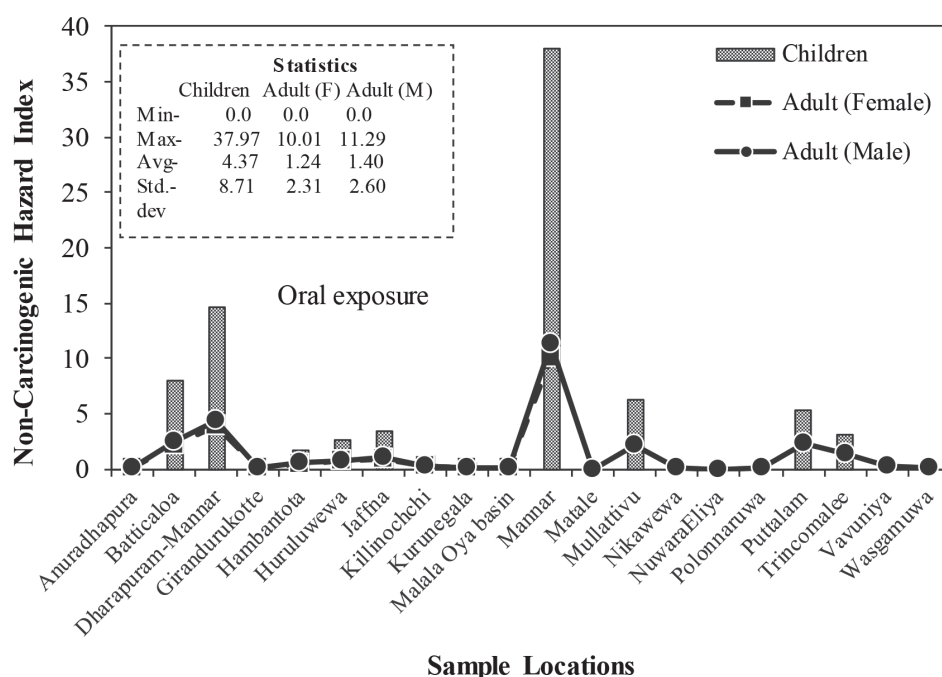


Figure 1: Non-carcinogenic risk among children, females and males in the different regions of Sri Lanka through oral ingestion of As contaminated water.

Table 1: Estimation of Hazard Quotient (HQ), Cancer Risk (CR) and Cancer Incidence (CI) for the different regions of Sri Lanka based on secondary data obtained through literature survey.

Sampling locations	HQ for oral			HQ for dermal			CR for oral			CR for dermal			CI	
	Children	Adult (Female)	Adult (Male)	Children	Adult (Female)	Adult (Male)	Children	Adult (Female)	Adult (Male)	Children	Adult (Female)	Adult (Male)	Children (Female)	Adult (Male)
Anuradhapura	1	0	0	3.98E-03	1.30E-03	1.21E-03	2.6E-04	6.9E-05	7.8E-05	1.8E-06	5.9E-07	5.5E-07	2.6E-04	7.0E-05
Batticaloa	8	2	2	3.98E-03	1.82E-02	1.70E-02	3.6E-03	9.7E-04	1.1E-03	1.8E-06	8.2E-06	7.6E-06	3.6E-03	1.1E-03
Dharapuram-Mannar	15	4	4	3.98E-03	3.32E-02	3.09E-02	6.6E-03	1.8E-03	2.0E-03	1.8E-06	1.5E-05	1.4E-05	6.6E-03	2.0E-03
Girandurukotte	1	0	0	3.98E-03	1.13E-03	1.06E-03	2.3E-04	6.1E-05	6.8E-05	1.8E-06	5.1E-07	4.8E-07	2.3E-04	6.1E-05
Hambantota	2	0	1	3.98E-03	3.90E-03	3.64E-03	7.8E-04	2.1E-04	2.3E-04	1.8E-06	1.8E-06	1.6E-06	7.8E-04	2.1E-04
Huruluwewa	3	1	1	3.98E-03	5.83E-03	5.44E-03	1.2E-03	3.1E-04	3.5E-04	1.8E-06	2.6E-06	2.4E-06	1.2E-03	3.1E-04
Jaffna	3	1	1	3.98E-03	7.80E-03	7.28E-03	1.6E-03	4.2E-04	4.7E-04	1.8E-06	3.5E-06	3.3E-06	1.6E-03	4.2E-04
Killinochchi	1	0	0	3.98E-03	2.60E-03	2.43E-03	5.2E-04	1.4E-04	1.6E-04	1.8E-06	1.2E-06	1.1E-06	5.2E-04	1.4E-04
Kurunegala	1	0	0	3.98E-03	1.30E-03	1.21E-03	2.6E-04	6.9E-05	7.8E-05	1.8E-06	5.9E-07	5.5E-07	2.6E-04	7.0E-05
Malala Oya basin	0	0	0	3.98E-03	3.25E-04	3.03E-04	6.5E-05	1.7E-05	2.0E-05	1.8E-06	1.5E-07	1.4E-07	6.7E-05	2.0E-05
Mannar	38	10	11	3.62E-03	8.44E-02	7.89E-02	1.7E-02	4.5E-03	5.1E-03	1.6E-06	3.8E-05	3.6E-05	1.7E-02	4.5E-03
Matale	0	0	0	3.32E-03	0.00E+00	0.00E+00	0.0E+00	0.0E+00	0.0E+00	1.5E-06	0.0E+00	0.0E+00	1.5E-06	0.0E+00
Mullattivu	6	2	2	3.06E-03	1.61E-02	1.51E-02	2.8E-03	8.6E-04	9.7E-04	1.4E-06	7.2E-06	6.8E-06	2.8E-03	8.7E-04
Nikawewa	0	0	0	2.85E-03	6.43E-04	6.06E-04	1.1E-04	3.4E-05	3.9E-05	1.3E-06	2.9E-07	2.7E-07	1.1E-04	3.9E-05
NuwaraEliya	0	0	0	2.66E-03	0.00E+00	0.00E+00	0.0E+00	0.0E+00	0.0E+00	1.2E-06	0.0E+00	0.0E+00	1.2E-06	0.0E+00
Polonnaruwa	0	0	0	2.49E-03	1.18E-03	1.12E-03	1.7E-04	6.3E-05	7.2E-05	1.1E-06	5.3E-07	5.0E-07	1.7E-04	6.3E-05
Puttalam	5	2	2	2.34E-03	1.74E-02	1.65E-02	2.4E-03	9.3E-04	1.1E-03	1.1E-06	7.8E-06	7.4E-06	2.4E-03	9.4E-04
Trincomalee	3	1	1	2.21E-03	1.03E-02	9.79E-03	1.4E-03	5.5E-04	6.3E-04	1.0E-06	4.6E-06	4.4E-06	1.4E-03	5.5E-04
Vavuniya	1	0	0	2.10E-03	2.25E-03	2.15E-03	2.9E-04	1.2E-04	1.4E-04	9.4E-07	1.0E-06	9.7E-07	2.9E-04	1.2E-04
Wasmamuwa	0	0	0	1.99E-03	3.99E-04	3.82E-04	4.9E-05	2.1E-05	2.5E-05	9.0E-07	1.8E-07	1.7E-07	5.0E-05	2.5E-05
Min	0.0	0.0	0.0	1.99E-03	0.00E+00	0.00E+00	0.00E+00	0.00E+00	0.00E+00	8.97E-07	0.00E+00	0.00E+00	1.20E-06	0.00E+00
Max	38.0	10.0	11.3	3.98E-03	8.44E-02	7.89E-02	1.71E-02	4.51E-03	5.08E-03	1.79E-06	3.80E-05	3.55E-05	1.71E-02	4.54E-03
Avg	4.4	1.2	1.4	3.32E-03	1.04E-02	9.75E-03	1.97E-03	5.56E-04	6.28E-04	1.50E-06	4.69E-06	4.39E-06	1.97E-03	5.60E-04
Std. Dev	8.7	2.3	2.6	7.73E-04	1.94E-02	1.82E-02	3.92E-03	1.04E-03	1.17E-03	3.48E-07	8.75E-06	8.18E-06	3.92E-03	1.18E-03

$$\text{CDI oral} = \frac{\text{CW.IR.EF.ED}}{\text{BW.AT}} \quad (1)$$

$$\text{CDI dermal} = \frac{\text{CW.SA.EF.ED.CF.ET.KP}}{\text{BW.AT}} \quad (2)$$

$$\text{HQ oral} = \frac{\text{CDI oral}}{\text{Rf D oral}} \quad (3)$$

$$\text{HQ dermal} = \frac{\text{CDI dermal}}{\text{Rf D dermal}} \quad (4)$$

$$\text{CR oral} = \text{CDI oral.SF} \quad (5)$$

$$\text{CR dermal} = \text{CDI dermal.SF} \quad (6)$$

$$\text{CI} = \text{CR oral} + \text{CR dermal} \quad (7)$$

For calculating the values of CI, maximum As data from different scientific literature has been taken.

Rainfall Data from CHIRPS

The CHIRPS data was used from the year 1981 to 2018 to map and understand the spatial and temporal variation in precipitation over the years. The data is available in NETCDF format which was scaled down to multi-dimensional raster datasets in ArcMap. Using Python model builder in ArcMap, different annual rainfall images were created using single code operation. Further cubic interpolation technique was used for resampling the data and making it smoother with less erratic changes with pixel change. In order to bring visible differences among pixels, histogram matching technique were used. Finally, specific grids were created to extract the values of precipitation for comparison with As concentration and establishing correlation.

Results and Discussions

Cancer Risk and the Cancer Incidence

Based on our analysis, done on the selected literature concerning the Arsenic concentration in Sri Lanka, it could be said that the northern region of Mannar is the most vulnerable region with respect to As groundwater toxicity (Table 3). The Hazard Quotient (HW) reaches around 15 for children in the Tharapuram-Mannar region where significant limestone (LR) deposits have been found in the fluvial geomorphology. One compelling factor for this three times higher vulnerability in children as compared to adults may be because of low ingestion rate which greatly impacts the excretion rate of As from the body. The mass of the body plays a very dominant role in determining the

intake capability of humans. Higher body weight leads to lesser aggregation of As in the body via oral uptake. The regions of Mulative and Puttalama can be identified as As vulnerable zones for children only.

Table 3: Sources of Arsenic data for estimating Health Risk

<i>Data Location</i>	<i>No. of samples</i>	<i>Source</i>
Girandurukotte	29	Nanayakkara et al., 2019
	52	Wickramarathna et al., 2017
Wilgamuwa	12	Wickramarathna et al., 2017
Nikawewa	7	Wickramarathna et al., 2017
Mannar Island	-	Herath et al., 2018
	35	Bandara et al., 2018
Tharapuram – Mannar	8	Amarathunga et al., 2019
Mulative	-	Herath et al., 2018
Puttalama	-	Herath et al., 2018
Jaffna	35	Chandrajith et al., 2016
District Specific Studies (<i>n</i> = 14)		Weragoda and Kawakami, 2017

It is important to note that these regions fall in the Quaternary deposits which are known triggers as evident from the As contamination of Mid-Gangetic Holocene Quaternary Alluviums of India. These sedimentary deposits usually are rich in organic content which leads to further oxidation under the presence of anthropogenically introduced nitrate. In case of dermal exposure, the hazard is quite low based on the frequency and amount of water being exposed onto. However, in this case body weight and skin area becomes two most important factors. In case of children an average skin area of 4900 cm² is assumed which gets increased to 16,000 cm² for females and 18,000 cm² for males. Lesser skin area decreases the chances for As excretion through skin in the form of sweating (Figure 2). As compared to adults, children are at three times higher risk of developing skin related problems due to As dermal toxicity. It could also be seen that dermal risk as compared to oral risk is significantly lower for almost entire Sri Lanka. In terms of Cancer Susceptibility (CR) which is defined as number of people susceptible per 1000, it could be seen that out of every 1000 child in Tharapuram-Mannar 15 will be susceptible to develop non-cancer symptoms ranging from skin keratosis to hyperpigmentation.

There also exists a clear demarcation of As affected areas within the country. The northern-most and the north-eastern part are susceptible to As contamination

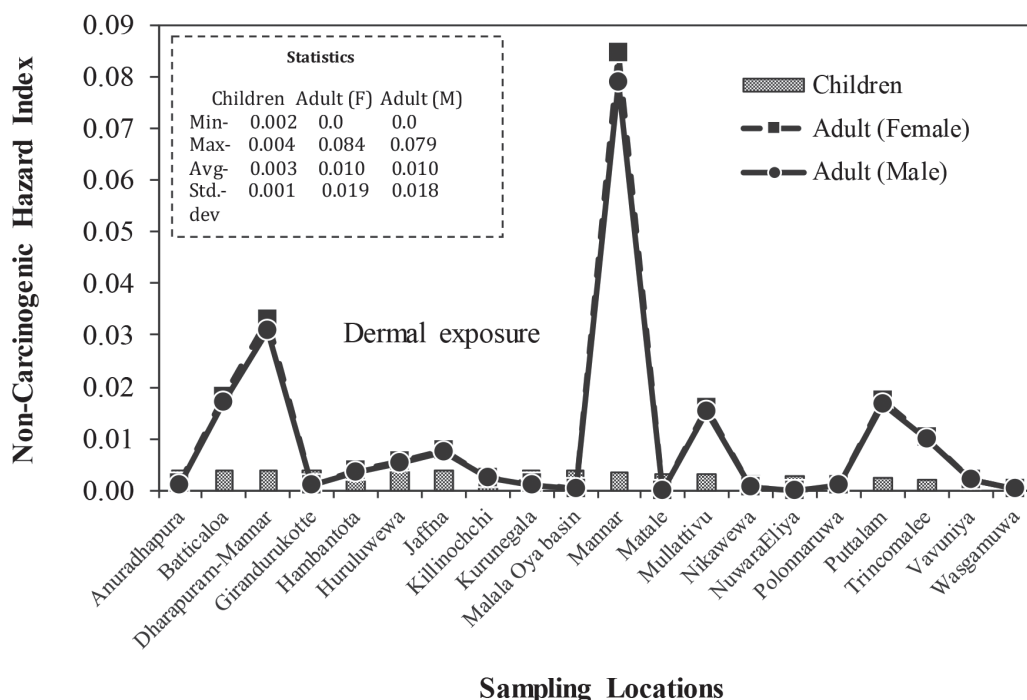


Figure 2: Non-carcinogenic risk among children, females and males in the different regions of Sri Lanka through dermal exposure of As contaminated water.

with CI reaching close to 6.6×10^{-3} . Another study by Weregoda and Kawakami et al. (2017) used 491 samples from 14 districts of Sri Lanka to estimate the groundwater arsenic concentration. According to the study the highest concentration of arsenic was found in the Mannar district As reaches to $66 \mu\text{gL}^{-1}$, followed by Batticaloa and Mullattivu where the concentration is above the drinking permissible limit of $10 \mu\text{gL}^{-1}$. In districts such as Kurunegala the amount of fluoride in groundwater has reached 5mgL^{-1} , enough to cause symptoms of fluorosis. From a As and F co-occurrence point of view, districts such as Mannar are highly vulnerable as both the toxic geogenic contaminants are crossing the permissible limit of human tolerance.

Identifying the Wetter and Drier Region of Sri Lanka through CHIRPS Data

Since, children are the most vulnerable group among all, in terms of existing arsenic risk, it is imperative to spatially demarcate the high and low zones. A high zone does not necessarily represent vulnerable zone but shows a region with comparative higher risk for future vulnerability. Through precipitation mapping from 1981 to 2018 wetter and drier regions can be identified. Except for few anomalous years in 1983 and 2016, rest all other years show a visible difference in precipitation between wetter and drier region by almost

2000 mm (Figure 3). The wetter regions also overlap with low arsenic regions signifying that more rainfall led recharge actually stabilizes the groundwater conditions from As mobilization point of view. Presence of clay minerals like montmorillonite is also a good adsorbent of arsenic under optimum cation exchange capacity of soil. In the northern drier region of Sri Lanka, the amount of clay in the soil has been reported between 3.26 to 35% (Jayasena et al., 2008). If we see in case of Cancer Risk among children, the trend is reversed. The southern region is more vulnerable to Cancer Risk through oral exposure.

Rainfall also has another role in changing the rock weatherability. It has been reported that the wetter region has more intense rock weathering as compared to the drier region based on the assumed high infiltration of rain during monsoon (Jayasena et al., 2008). High infiltration also triggers changes in the carbonate chemistry of the aquifer, which pre-dominantly is governed by the pH and mineral saturation state. High bi-carbonate leaching during monsoon forces the rate of dissolution of ions into the water system during carbonate weathering.

Usually the sandy clay mineralogy along the coastal systems force high adsorption to geogenic contaminants by providing large active sites. This causes an equal vulnerability to the sea-coastal ecosystems, especially

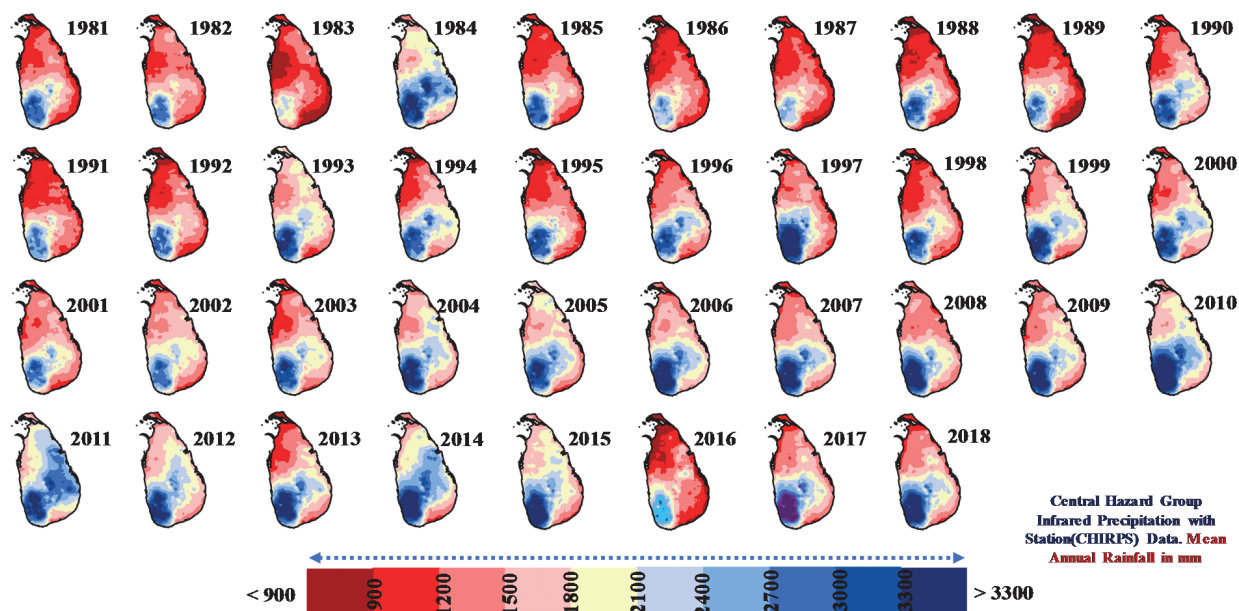


Figure 3: Pattern and distribution of rainfall over Sri Lanka mapped using data of CHIRPS.

to the phytoplankton species which are very much sensitive to any changes in their physical environment.

Role of Seawater and Groundwater Interaction in Influencing the Ion Chemistry of Geogenic Contaminants

Seawater intrusion is a known threat in coastal aquifer system. The column weight of groundwater is the only available resistive force to prevent seawater entry into the aquifer system which has increasingly being compromised owing to over-utilization of groundwater resources. At the same time projected increase in Sea Surface Temperature (SST) and Sea Level Rise (SLR) will make the situation more vulnerable leading to loss of land and agricultural productivity. Using stable isotope mass balance techniques for quantification of such changes becomes the best available option for the researchers. According to Chandrajith et al. (2014) there already exists evidences of salt-water mixing in the eastern coast of the island country. The pH of the groundwater too exhibits a peculiar existence in both the acidic and basic environment. High sodium influx may lead to over-saturation of sodium containing minerals, especially Halite which may crystallize through precipitation.

Silicate weathering have been identified as a common mechanism of arsenic desorption in the aquifer system. Usually Plagioclase minerals downgrade to clay structure leading to a buffer increase in the sodium ion concentration. Usually with rainfall-led recharge

leaching of nitrate also happens, which essentially is a very important component of fertilizers. Nitrate also has the capability to reduce the pH of the groundwater, causing changes in the arsenic speciation and increase in the toxicity. Further anthropogenic and oceanic depositions of acidic anions especially sulphate can trigger changes in the hydro-chemical facies of the groundwater. A study by Bandara et al. (2018) revealed that about 15% of sea water mixing has been happening with the groundwater samples of Mannar district.

Conclusion

The present study by using a database of the existing literature has tried understanding the prospects of health risk with respect to arsenic contamination of groundwater in Sri Lanka. Using rainfall data from 1981 to 2018, wetter and lesser vulnerable zones have been identified. The drier north region has considerable higher risk of developing future non-carcinogenic risk owing to complementing geology dominantly Quaternary sediments. The children are at almost three times higher risk compared to adults in terms of Hazard Quotient with respect to arsenic vulnerability and hence there is a dire need for employing arsenic remediation techniques in densely populated regions. Through this re-analysis review we can now specifically point out that there exists a constant threat of seawater intrusion which will only worsen with further increase in groundwater abstraction and lesser rainfall led

recharge. Therefore, there is an urgent need to formulate preventive correction plan to mitigate the challenges posed by arsenic contamination of groundwater.

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