



Chemical weathering and atmospheric CO₂ consumption in the semi-arid Swarnamukhi basin (Peninsular India) estimated from river water geochemistry

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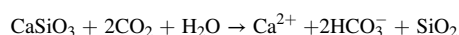
ABSTRACT

A small ephemeral Swarnamukhi River (hard rock) basin, more sensitive to climate change, was sampled and analyzed for various parameters. A total of 66 river water samples (22 of each season) were collected from different sampling stations covering the whole Swarnamukhi River basin in pre- and post- and monsoon seasons. Chemical weathering rate data of the Swarnamukhi River basin is integrated with the available data of other large perennial (Godavari and Krishna) and ephemeral (Kaveri and Periyar) hard rock river basins of Peninsular India. The Peninsular river basins, including Swarnamukhi River basin, are mainly dominated by granitic gneissic terrain. Contribution of inputs through forward modelling shows: silicate > evaporite > carbonate > atmosphere. The mean silicate and carbonate weathering rates in the river basin observed are 27.15 tonnes.km⁻².yr⁻¹ and 9.45 tonnes.km⁻².yr⁻¹ in pre-monsoon season, whereas 32.73 tonnes.km⁻².yr⁻¹ and 45.90 tonnes.km⁻².yr⁻¹ in post-monsoon season, respectively. The Swarnamukhi river has lower silicate weathering rate (annual average: 30.57 tonnes.km⁻².yr⁻¹) than other published river basins data (Narmada: 33.9 tonnes.km⁻².yr⁻¹; Nethravati: 42 tonnes.km⁻².yr⁻¹; Godavari: 45.6 tonnes.km⁻².yr⁻¹) due to different localized geological setting. Mean annual CO₂ consumption for the Swarnamukhi River is 6.9 × 10⁵ mol km⁻².yr⁻¹ which is comparable to other Peninsular rivers such as the Godavari (6 × 10⁵ mol km⁻².yr⁻¹) and Gad (5.7 × 10⁵ mol km⁻².yr⁻¹) rivers. The study further provides the inventory for CO₂ consumption on river basin scale, which is an important consideration from the global warming point of view.

1. Introduction

Chemical weathering is an important process for understanding the global carbon cycle and climatic evolution which is dependent on many factors such as kinetics of mineral dissolution, physical erosion, geology, meteorological conditions and hydrology (Dessert et al., 2001; Gaillardet and Galy, 2008; Donnini et al., 2016). Chemical weathering of silicate rocks is of major significance in the long-term (10⁶ yrs) global climate change rather than carbonate weathering or evaporite dissolution, because every mole of silicate weathering utilizes an equal amount of atmospheric CO₂ (Berner, 1991). Conversely, in carbonates, the amount of CO₂ utilized in weathering will get further released to the atmosphere during its precipitation in ocean. Evaporite dissolution process does not consume CO₂ in semi-arid region but has a significant role in the contribution of chemical composition (K, Na, Ca, Mg, Cl and

SO₄) of river waters. Up and down arrows in the equations represent the sources and sinks of CO₂ respectively (Berner, 1991).



Solute load generated by continental chemical weathering is carried to the ocean by inland rivers, thus its composition and CO₂ drawdown is a good indicator of chemical weathering activities that link continent-ocean-atmosphere systems (Navarre-Sitchler and Thyne, 2007). Many studies related to CO₂ drawdown by a river basin have been reported such as Amazon (Mortatti and Probst, 2003), Orinoco (Mora et al., 2010), Alpine rivers (Donnini et al., 2016), some Asian rivers, such as Narayani (Galy and France-Lanord, 1999), Han (Li et al., 2009), Hong (Moon et al., 2007), Pearl (Xu and Liu, 2010), Xishui (Wu, 2016),

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Mekong (Li et al., 2014), Sanchahe (An et al., 2015), Himalayan rivers such as Indus (Krishnaswami and Singh, 1998; Karim and Veizer, 2000), Yamuna (Dalai et al., 2002), Ganges–Brahmaputra (Tripathy and Singh, 2010) and Indian peninsular rivers such as Narmada–Tapti–Wainganga (Dessert et al., 2001); Krishna (Das et al., 2005), Narmada–Tapti (Sharma and Subramanian, 2008), Godavari (Jha et al., 2009), Narmada (Gupta et al., 2011) and Nethravati (Gurumurthy et al., 2012) are well documented. The aforementioned studies of various authors reflect the broad interests in understanding the controlling processes on river basin geochemistry; hence, already published data of these basins are

used here for comparison. In this study, a small ephemeral Swarnamukhi River basin was sampled in detail to know river geochemistry and CO₂ consumption rates. All the large rivers mentioned above have a mixture of hard rocks and soft rocks whereas an ephemeral Swarnamukhi river is entirely in hard rock region. Among these river systems, riverine chemical load with spatio-temporal variations in the Swarnamukhi river basin is the least studied so far. An attempt is made to fill the gap by studying many temporal and spatial river water samples along the river course to understand potential future CO₂ change with reference to hard rocks. Integrated analysis of weathering processes and associated CO₂

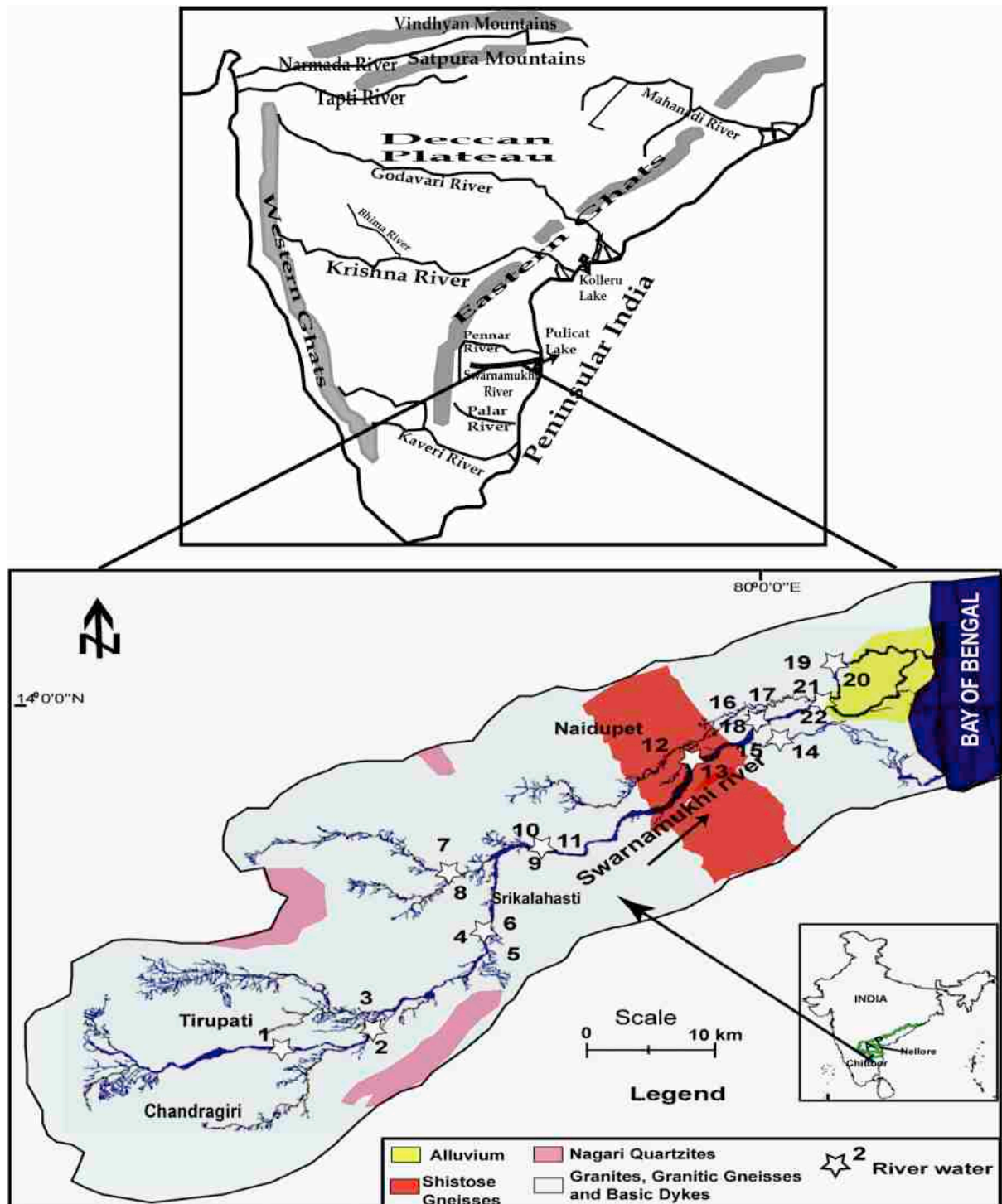


Fig. 1. Physiography of the Peninsular India region and Geological map of Swarnamukhi River basin with sampling locations (Source: Reddy 1981).

consumption has been carried out in granulite and granitic gneisses of Kaveri (Pattanaik et al., 2013), gneiss/amphibolite in the subtropical monsoon climate zone of Xishui (Wu et al., 2013) and granitoid and gneissic rocks of Mucone river basin (Borrelli et al., 2014). However, for Swarnamukhi River, it is the first attempt to quantify the CO₂ consumption in similar geological and climatic conditions.

2. Study area and geology

Swarnamukhi river basin, located in the Andhra Pradesh State of India, lies in between latitudes 13°25'30"-14°08'30"N and longitudes 79°07'39"-80°11'0"E (Fig. 1). It rises at an elevation of 300 m in the Eastern Ghats ranges passing through the Tirumala-Tirupati hills flowing in N-E to S-E directions, finally meeting the Bay of Bengal. The Swarnamukhi is an ephemeral east flowing river with a total drainage area of 3225 km² and length of about 192 kms. The river gets run-off flow during monsoon and is almost non-existent during summers. The Swarnamukhi, Independent River, has no major tributaries and therefore its flow mainly depends on rainfall in its upper catchment and subsurface water inflow (Patel et al., 2016). The river has dendritic to sub-dendritic drainage pattern that is the characteristic feature of granitic terrain. The study area is mainly dominated by agricultural farmlands with no major industrial activities except a few small scale factories producing bricks, ceramic, metal plating and cement. The major soil types are red loamy, red sandy, black clay, black loamy and alluvial soils. The river basin has sub-tropical climatic conditions with summer temperature touching nearly 38–46 °C and winters 12–18 °C. The average annual rainfall in the Swarnamukhi basin decreases from 1270 mm at the eastern extremity to 762 mm at the western extremity of the basin with 47% by S–W monsoon and 42% by N–E monsoon. The rainfall distribution of SW monsoon shows a decreasing trend eastward, while NE monsoon shows decreasing trend westward. The period between January to May, being the main dry season, accounts for only 10–15% of the annual rainfall. The mean annual evaporation recorded through pan evaporation is about 860 mm. The potential evapotranspiration calculated using thornthwaite's formula is 2049 mm for a normal year. The bulk of the annual rainfall occurs usually in a period of 3–4 months and hence it is heavily lost as surface runoff. The dominant aspect of the climate is general aridity, but for the rain during the monsoon for a short period of the year imparting humidity to the atmosphere and moisture to the soil. The Swarnamukhi river basin has only one GDQ (Gauge, Discharge and Water Quality Site) hydrological observation site at Naidupet terminal point with an observed average annual runoff of 60 MCM discharge having a catchment area of 2650 Km² (Hydrological Data Directorate Report, 2012). The peak water level at the Naidupet hydrological observation site has been observed 44.34 m in the year 1979. Monthly average flow (during 1978–2009) per unit drainage area of minimum 0.0 mm (April) and maximum 56.64 mm (Nov.) is observed at Naidupet site of the river basin. The number of sub-basins present in the Swarnamukhi catchment area is 18. The total dependable yield at 75% confidence limit is 13,740 Mcft. The utilization of the yield in the basin is around 10,663 Mcft and in some of the sub-basins it substantially exceeds the yield of the 75% confidence limit. However, after compensating the deficits in some sub-basins with the surplus yield of the other sub-basins, there is a cumulative balance of 3077 Mcft available (as base flow) in this river basin.

Peninsular India is mainly composed of the western and eastern Ghats, Vindhyas and Satpuras (Fig. 1). The northern part is composed of the Deccan traps, while the southern part consists of Archaean gneisses, schists and charnockites. More resistant flows of Deccan traps form a series of step-like terraces in which Godavari and Krishna Rivers originate. A large number of rivers such as Godavari, Krishna, Cauvery and several smaller rivers such as Periyar, Pennar, Bhima and Swarnamukhi flow from west to east (Fig. 1). Most of the Peninsular Rivers have reached a mature state of development, particularly in the lower portions of their valleys. The Godavari basin consists heterogeneous

geology i.e. Deccan trap (48%), Dharwars (phyllites, schists and amphibolites) and Archaean (granites and gneisses) rocks (39%), Precambrian and Gondwana sedimentary rock (11%) and recent alluvial cover (2%) (Biksham and Subramanian, 1988; Raman et al., 1995). Jha et al. (2009) studied Peninsular India to calculate the weathering rate of the Deccan trap using major ion chemistry of the water of the Godavari River, which is the third largest river in India. The Swarnamukhi basin is underlain by geological formations of Archaean, Proterozoic and Quaternary ages. Granitic rocks are intercepted by innumerable dyke rocks of narrow width. Large alluvium tracts are present along the major stream of the river which belongs to a recent age with most spreading in coastal part up to a thickness of 3–7.5 m underlain by weathered granitic terrain. The famous Tirumala-Tirupati hills (Eparchaeon Unconformity) of Precambrian period are composed of Nagari quartzites and granites (Fig. 1).

3. Methodology

The available published data from the literature for large river basins such as Godavari, Narmada, Krishna, Kaveri etc. used for comparison with the data generated for small Swarnamukhi River basin, but not for recalculations. Analytical and sampling methodology is the same for the published data and freshly sampled river basin for the better interpretation and comparison. A total of 66 river water samples (22 of each season) were collected for the Swarnamukhi river basin during January 2014 (post-), June 2014 (pre-) and October 2015 (monsoon). River water was collected from the middle and both sides of the river bank to get a holistic picture. Rain water sample was collected on the roof of the Department of Geology, Sri Venkateswara University with the help of polypropylene funnel and bottle in March 2015. During normal monsoon (June–September), the river basin usually does not get significant rain (Indian Meteorological Department, 2015). Grab river water samples were collected in pre-cleaned polyethylene bottles after filtered by axiva 45 µm Millipore filter paper. Cleaning of all the bottles was carried out by soaking in 5% nitric acid for 24 h and then thoroughly rinsing with doubled distilled water. All the sample bottles were stored in a refrigerator at approximately 4 °C till analysis. General parameters such as pH, electrical conductivity (EC) and total dissolved solids (TDS) were measured at the time of sampling using a multi parameter ion meter (pH/Cond 340i SET 1). Dissolved silica was measured by ammonium molybdate method that actually gives H₄SiO₄⁰. Hence Si mentioned in the text actually refers to dissolved silica. HCO₃ analysis was performed by potentiometric titration (end point pH 4.5) of samples. Other major ions analysis was done as per the standard protocol of American Public Health Association (APHA, 2005): Ca, Mg, Na and K were analyzed by AAS (Model: Thermo Scientific M series). Anions were determined by spectrophotometry method (UV 3200 double beam spectrophotometer, Lab India). The precision of the derived dataset has been estimated using normalized inorganic charge balance. All the samples are within ±5% errors and are acceptable for further interpretations.

4. Results

4.1. General observation for present sampling periods

4.1.1. River water chemistry

The physio-chemical parameters of the Swarnamukhi River water for pre-, post- and monsoon seasons are presented in the supplementary material (Appendix Table) and ranges of all seasons are shown in Table 1. The pH is relatively low in the monsoon river water due to supply of low pH water from rainfall. Higher EC values noticed in Munagapalem and Sodavaram locations indicating the anthropogenic source i.e. mainly from municipal and agricultural wastes. The lower values of EC in the monsoon samples are due to dilution resulting from high rainfall events. The high TDS observed in post-monsoon is due to

Table 1

Statistical summary of various physio-chemical parameters of river water collected in pre-, post- and monsoon seasons.

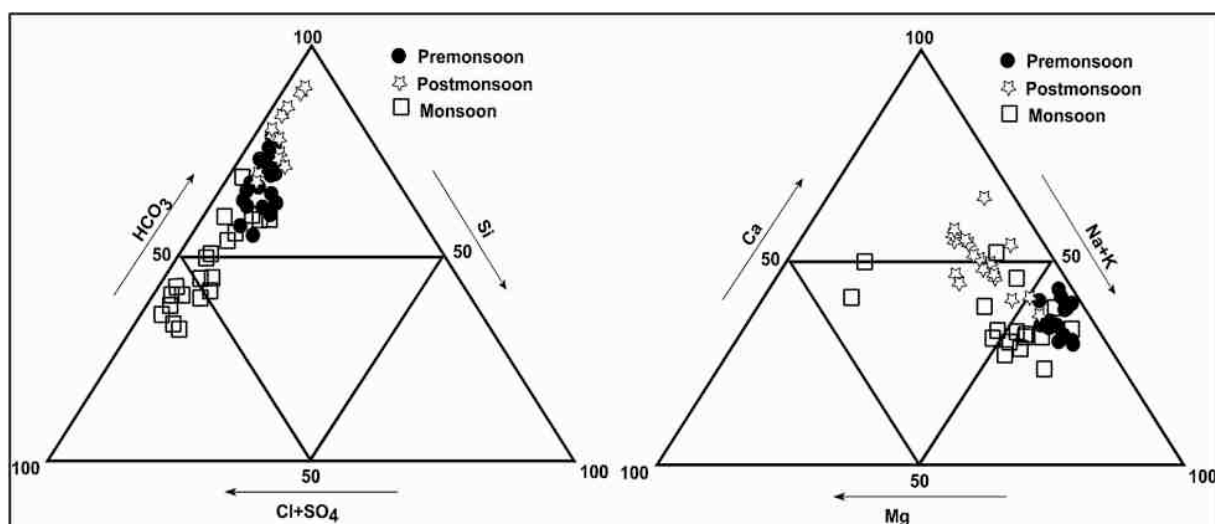
Season	Pre-monsoon		Post-monsoon		Monsoon	
Parameter	Range	Mean	Range	Mean	Range	Mean
TDS	161.56–346.44	206.90	157.79–333.22	219.91	53.01–150.21	114.27
EC	230.80–494.91	295.57	225.42–476.02	314.15	75.72–214.58	163.24
pH	7.51–8.51	8.05	7.30–8.55	8.10	7.00–7.80	7.47
Ca ²⁺	400.70–1071.16	643.62	701.23–1347.36	968.73	149.96–460.81	319.55
Mg ²⁺	66.69–343.64	196.95	123.22–750.10	333.69	100.33–381.25	236.17
Na ⁺	1458.70–3497.83	1920.34	899.55–2178.91	1307.87	111.74–1273.96	771.59
K ⁺	20.00–105.50	60.10	20.00–130.83	45.01	13.33–104.86	67.74
CO ₃ ²⁻	0.00–58.20	11.73	0.00–50.01	16.65	–	–
HCO ₃ ⁻	1400.29–4500.95	2248.83	1606.56–4500.95	3109.03	343.43–1489.31	758.01
SO ₄ ²⁻	111.34–309.88	221.69	10.35–499.80	194.51	125.81–649.74	308.58
Cl ⁻	588.35–1297.60	820.61	309.99–619.97	473.32	100.00–689.97	483.68
F ⁻	0.00–61.05	18.31	9.47–79.53	23.95	7.52–90	29.04
NO ₃ ⁻	0.16–157.26	44.06	0.00–35.00	3.93	0.00–40.00	6.93
PO ₄ ³⁻	0.00–11.13	3.13	0.16–7.63	2.09	0.00–12.21	4.66
H ₄ SiO ₄ ⁰	122.05–407.49	234.46	110.58–503.83	183.20	37.94–245.14	135.55
TZ ⁺	2.82–6.25	3.66	2.81–5.92	3.95	0.93–2.58	1.95
TZ ⁻	2.69–6.29	3.60	2.69–6.19	4.04	0.89–2.47	1.91
NICB	(-2.54)–4.56	1.68	(-5.01)–4.23	-1.87	(-4.40)–5.00	2.09
SO ₄ /Cl	0.12–0.45	0.29	0.02–0.81	0.39	0.20–6.50	0.90
Na/Cl	1.60–3.04	2.37	1.21–5.44	2.80	0.48–12.07	2.07
NO ₃ /Na	0–0.11	0.02	0–0.03	0	0–0.17	0.02
NO ₃ /Cl	0–0.24	0.06	0–0.06	0.01	0–0.08	0.02
NO ₃ /K	0–2.07	0.69	0–0.73	0.07	0–1.25	0.16
SWR	12.72–51.50	27.15	13.56–70.04	32.73	1.34–100	31.85
CWR	0.32–42.80	9.45	16.66–98.52	45.90	1.12–90	17.92
CO ₂ sil	0.85–20.43	7.87	0.76–30.81	9.75	0.20–61.02	8.70
CO ₂ carb	0.04–4.42	1.07	1.43–10.53	5.19	0.24–43.08	8.60

All ions, SO₄/Cl, Na/Cl, NO₃/Na, NO₃/Cl, NO₃/K in μM , TDS in mg/l, EC in $\mu\text{S}/\text{cm}$, TZ⁺ is total cations and TZ⁻ is total anion in meq/l, NICB is normalized inorganic charge balance in %, SWR is silicate weathering rate and CWR is carbonate weathering rate in $\text{ton.km}^{-2}.\text{yr}^{-1}$, CO₂sil is CO₂ drawdown by silicate weathering and CO₂carb is CO₂ drawdown by carbonate weathering in ($\times 10^5 \text{ mol km}^{-2}.\text{yr}^{-1}$).

dissolution of evaporites and municipal wastes and in pre-monsoon due to diffusion solute load increases in the river water. However, we have not quantified contribution from each source. Average TDS of the Swarnamukhi River (mean: 180.36) is below 283 mg/l of world's major rivers (Gaillardet et al., 1999) but higher than other rivers like Amazon (44), Orinoco (82) and Brahmaputra (71) (Galyand France-Lanord, 1999). Na is the dominant cation that contributed (μM) 60–72% in pre-, 40–63% in post- and 19–67% in monsoon of total cations (TZ⁺) followed by Ca, Mg and K. Ca is attributed from carbonate and silicate weathering apart from atmospheric contribution. High Mg has been observed in some samples (13-Naidupeta, 14, 15-Pulikalva and 16, 17, 18-Gunupadu) of monsoon which indicates high dissolution rate of

Mg-containing minerals in the basin. TZ⁺ of the Swarnamukhi River (mean: 3.66 in pre-, 3.95 in post- and 1.95 meq/l in monsoon) is considerably higher than the mean value of world's rivers (1.25 meq/l) imparting its higher solute load (Meybeck, 1987). Ternary diagram (Ca–Mg–Na + K) indicates the dominance of Na + K among total cations in all the seasons except two samples of monsoon (sample 2, 3: Saraswatikandriga) which shows the Ca and Mg dominance (Fig. 2). The dominance of Ca and Mg in monsoon season could be due to high rate of both silicate and carbonate weathering.

HCO₃ is the dominant anion contributing (μM) 53–74% in pre-, 67–91% in post- and 36–65% in monsoon of total anions (TZ⁻) followed by Cl, SO₄, F, NO₃ and PO₄. HCO₃ in river water is contributed from

**Fig. 2.** Ternary diagrams (meq/l).

mineral weathering and organic matter decomposition in the basin. Slightly higher values of Cl and SO_4 in monsoon samples (sample no. 9–11–Pulluru bridge and 13–Naidupeta high bridge) can be explained by higher dissolution of evaporites existing in the semiarid basin. High NO_3 during pre-monsoon season could be due to the contribution of nitrate rich groundwater which recharges during lean river flow conditions as groundwater studies show its contamination with nitrate (Raju et al., 2015; Patel et al., 2016) apart from indiscriminate municipal discharge into the nearby flowing river stretch. Raju et al. (2016) published Swarnamukhi basin groundwater nitrate range values of pre-monsoon season (0–143 mg/l) which suggest that pre-monsoon river water nitrate is due to groundwater contribution whereas lower nitrate levels in the monsoon flow are due to dilution effect. However, relatively low NO_3 in other seasons is a result of high runoff and its consumption in biological processes. Some of the river water samples are inconsistent with the NO_3 content in rain water suggesting its atmospheric contribution as well. The ternary plot of HCO_3 –Si–Cl + SO_4 shows the clear dominance of HCO_3 in the anion budget (Fig. 2) (Raju et al., 2011). Si is calculated from $\text{H}_4\text{SiO}_4^\circ$ values. It is also observed that some monsoon samples also lie along the mixing line of HCO_3 and (Cl + SO_4) species indicating the contribution from (Cl + SO_4). The relationship between (Ca + Mg) vs. HCO_3 and (Ca + Mg) vs. (HCO_3 + SO_4) plot suggest less contribution of evaporites in enriching the river water with Ca + Mg as it seems insignificant variability in the data points of two plots (Fig. 3). (Ca + Mg) vs. (HCO_3 + SO_4) plot also illustrates the excess HCO_3 which could be derived from silicate weathering. $\text{H}_4\text{SiO}_4^\circ$ average value (184.40 μM) of the Swarnamukhi River water is above the global river average of 145 μM (Meybeck, 2003), Yamuna (67–378; Dalai et al., 2002), Amazon (60–160; Gaillardet et al., 1997) and Congo (140–210; Dupre et al., 1996) due to presence of silicate minerals. The mean values of all analyzed ions in the river basin are depicted in Fig. 4.

4.1.2. Rainwater chemistry

In order to calculate the CO_2 drawdown from the Swarnamukhi River basin, local rainwater chemical composition needed for atmospheric corrections. Thus, various physio-chemical parameters of local

rainwater (collected in March 2015) were analyzed and compared with average Indian rainwater chemistry (Table 2). Rain water has a pH of 6.37 indicating its slight acidic nature. Na and HCO_3 (μM) are dominating ions in the rainwater sample. Na/Cl molar ratio of 1.40 in rain water is imparting continental dust deposition (dust storm, traffic activities and constructions) rather than marine aerosols contribution. It is found that the solute load of the basin's rain water is higher than the Indian scenario due to its closeness to the Bay of Bengal or semi-arid climatic conditions with less rainfall events which could not be able to wash out solute load from the atmosphere.

5. Discussion and comparison with published data for other rivers

5.1. Source of major ions in riverine water

5.1.1. Atmospheric contribution

The sources influencing rainwater chemistry are mainly local dust and sea salt aerosol. In semi-arid climate, dust storms play a significant role in the atmospheric dust contribution. In general, the Swarnamukhi River joins the Bay of Bengal (saline) indicating the influence of marine aerosol in solute load. So, marine aerosol contribution to riverine solute load should probably increase close to the mouth of the river. Cl concentration levels along the river course do not significantly vary; hence, marine contribution may not be important factor in the river water chemistry (Li et al., 2014). Two samples (4&5, Srialahasti) in pre-monsoon show the highest values of Cl due to urban municipal wastewater discharge directly into river water. The Na/Cl molar ratio (1.40) of rainwater indicates the atmospheric dust-derived Na to rainwater. High HCO_3 of rainwater is due to high particulate matter in the atmosphere such as dust storm, construction and traffic activities which are prominent in most of the Indian semi-arid climatic environments (Gupta et al., 2011). The major ionic contribution from atmospheric rain to the Swarnamukhi River water was evaluated and found that mean TZ^+ derived from atmospheric contribution to the river water is 8.98% for pre-, 9.38% for post- and 11.41% for monsoon seasons. The percentage TZ^+ values suggest higher contribution of rain water cations in monsoon season to the riverine solute load due to heavy rainfall events as compared to pre- and post-monsoon seasons.

5.1.2. Anthropogenic contribution

A significant level of riverine solute load contribution is due to anthropogenic point sources (municipal and industrial wastewaters) and non-point sources (atmospheric inputs, fertilizers and irrigation runoff). The high TDS values are observed in sample 1 (Munagapalem), 7 and 8 (Sodavaram) (Appendix Table) showing high Ca, Na, SO_4 and Cl and negligible NO_3 concentration values, which can be attributed to evaporation during hot summer months and less anthropogenic influence. Less NO_3 concentrations in river water samples indicates minimal impact from agricultural or other anthropogenic activities. In order to have a better insight into anthropogenically driven ions, scatter plot between Na/Cl vs. Cl, SO_4 /Na and Na/Cl vs. NO_3 /Na have been considered (Fig. 5). Cl input from anthropogenic activities in river water could result in elevated Cl concentration with decrease in Na/Cl ratio, which is not observed in the basin suggesting the evaporite and atmospheric sources of Cl (Gurumurthy et al., 2012). Scatter plot between SO_4 /Na and Na/Cl with NO_3 /Na shows the poor relation with each other which suggests different origins of ions to the river solute load. Except two samples (2 and 3 Saraswikandriga) which are falling on one end of the line (Fig. 5), bulk of the samples are falling in a cluster and hence the indicated R^2 values in the plot may not truly reflect the real variability of the concerned values. Since most of the basin area is agriculture dominated, chemical signatures of agriculture end-member have been estimated. Na/Cl, NO_3 /Na, NO_3 /Cl and NO_3 /K ratio of 5, 10, ≥ 1 and 3–4, respectively shows the impact of agricultural activities and fertilizers on river water chemistry (Zeng and Sun, 1999; Liu et al., 2006; Chetelat

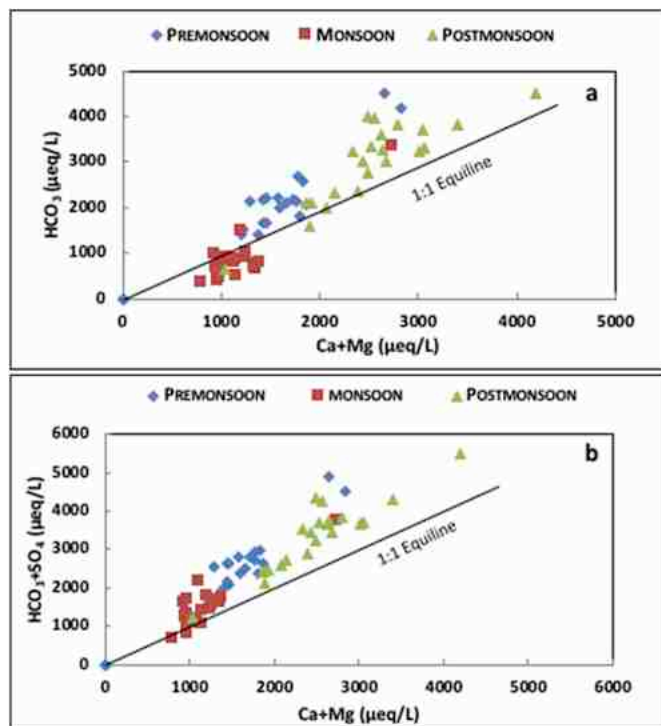


Fig. 3. Relation between different ions [a. (Ca + Mg) vs HCO_3 , b. (Ca + Mg) vs (HCO_3 + SO_4)].

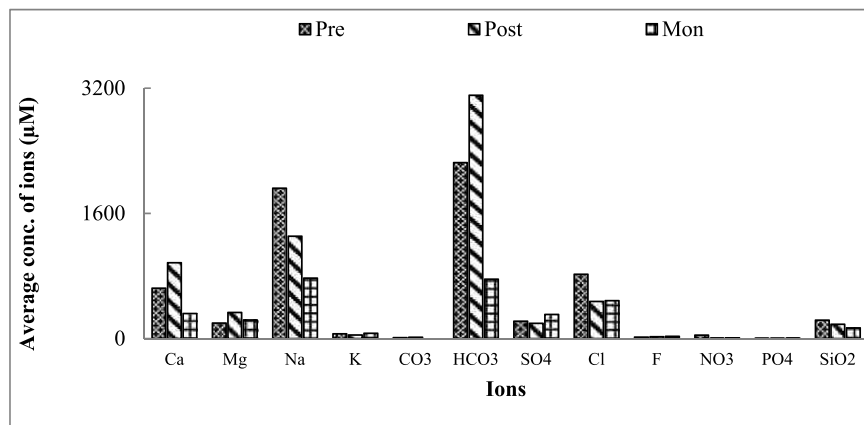


Fig. 4. Histogram showing temporal changes in ionic content in river water.

Table 2

Rain water composition of the Swarnamukhi River basin collected in March 2015 and comparison to average Indian rain water chemistry.

Parameters	Concentration values	Indian average values
Ca ²⁺	44.89	37.41
Mg ²⁺	12.35	6.36
Na ⁺	82.61	29
K ⁺	5.12	6.65
HCO ₃ ⁻	81.97	20
Cl ⁻	59.15	34
F ⁻	BDL	5.17
PO ₄ ³⁻	BDL	BDL
SO ₄ ²⁻	23.93	14.80
NO ₃ ⁻	6.45	20.23
pH	6.37	6.22
EC	109.20	–
TDS	60.67	–

All ions in µM, pH - unitless, EC - µS/cm and TDS - mg/l.

BDL = Below Detection Limit.

et al., 2008) (Table 1). Comparison of these agricultural end-members with the Swarnamukhi River data indicates that there is no impact of agriculture onto the river solute load. Anion ratios can be used for quantifying percentage of pollution in the river water (Pacheco and Van der Weijden, 1996).

Samples with <50% pollution are considered as limit of chemical weathering whereas >50% pollution depicts polluted water. The mean

values (38% in pre-, 22% in post- and 49% in monsoon) of river water samples confirm lesser impact of anthropogenic activities and significant role of mineral weathering onto the river solute load. River water samples of the earlier studied ephemeral Kaveri river basin (Table 3) also shows lower pollution values due to dilution effect (Rai et al., 2010; Pattanaik et al., 2013). In this study, few samples in monsoon (Saraswatikandriga and Srikalahasti locations) show high percentage of pollution which could be due to the recent heavy flash flooding event of 2015 which substantially contributed nearby wastes. Overall, based on ionic ratios and % pollution calculated values, it can be concluded that anthropogenic input in the river solute load is minimal.

5.1.3. Weathering contribution in hard rock terrain

Chloride (Cl) correction is made to quantify the atmospheric contribution. This method is based on assumptions that a conservative element Cl results from the atmosphere due to its non involvement in biogeochemical cycling and very minimal content in carbonate and silicate rocks (Fan et al., 2014). This correction accounts for dilution and evaporation processes, but neglects element fluxes due to biomass removal or dry deposition. Atmospheric corrected Ca–Mg plot reveals that majority of the samples are clustered towards Ca and only few monsoon samples falls over equiline indicating dolomite weathering in the basin (Fig. 6a). Excess Ca over Mg can be explained by extra supply from silicate weathering and release of Ca by calcretes found along the river banks. (Na + K) and Cl plot shows that majority of the samples lie towards (Na + K) axis which suggest the extra supply of Na from silicate weathering apart from evaporite weathering (Fig. 6b). Scatter plot of

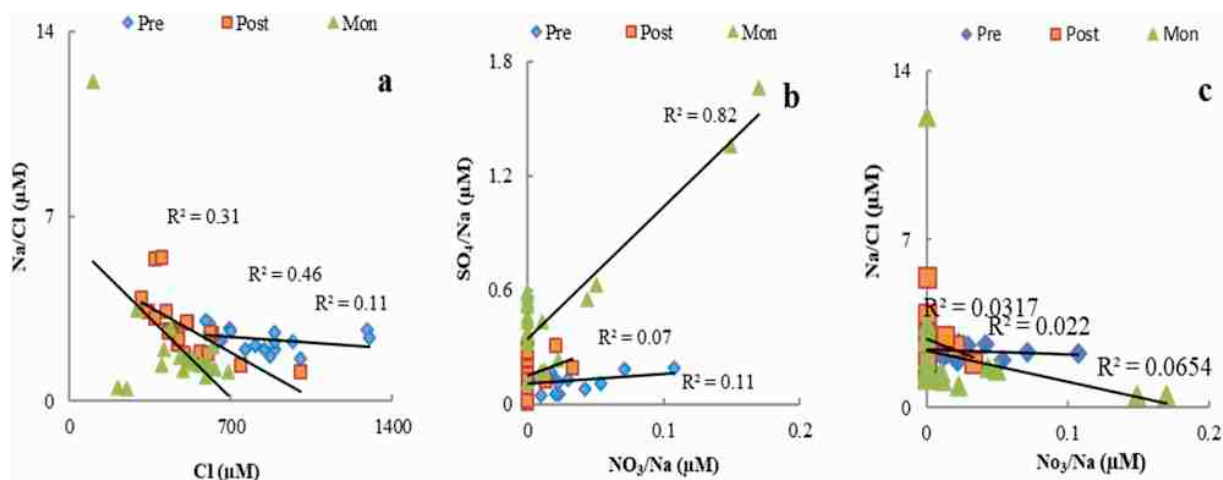


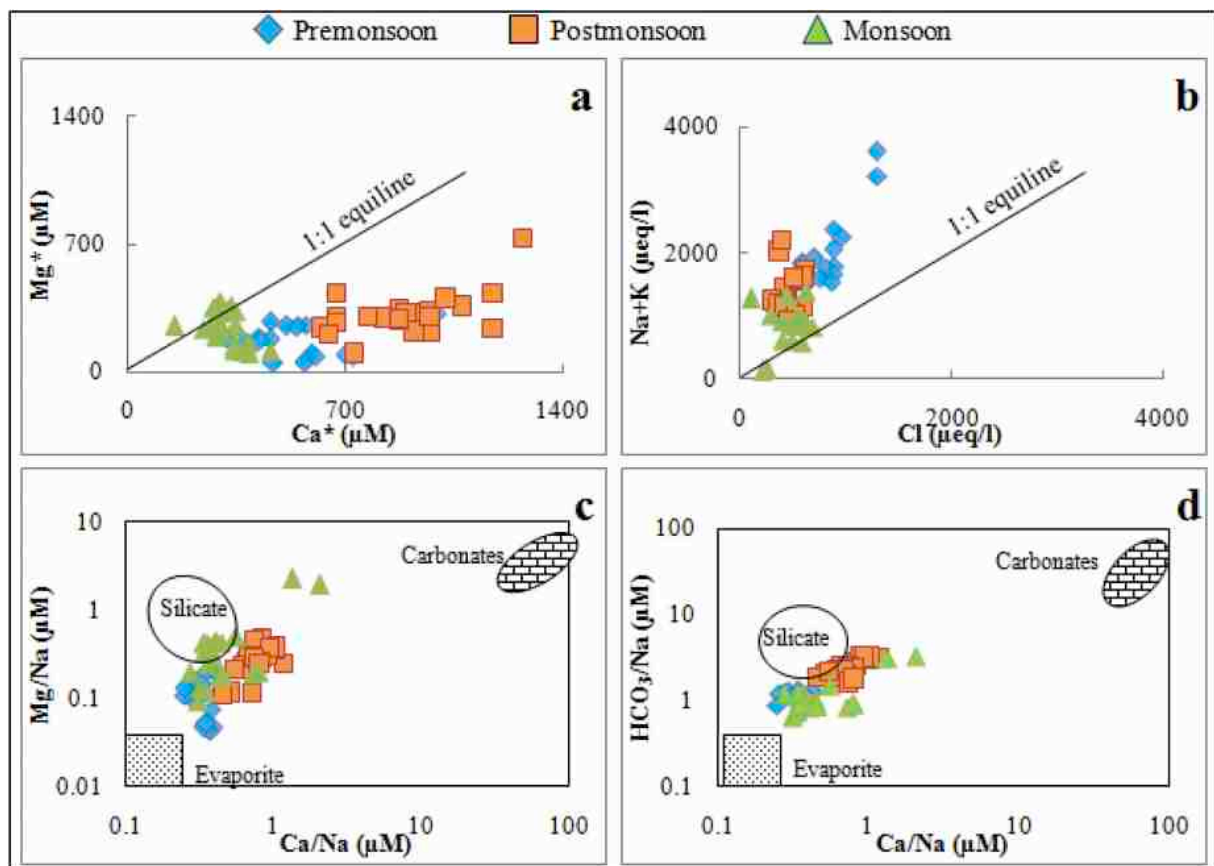
Fig. 5. Relation between different ionic ratios.

[(a) Na/Cl vs. Cl; (b) SO₄/Na vs. NO₃/Na (c) Na/Cl vs. NO₃/Na];

Table 3Comparison of chemical weathering rate and CO₂ consumption of the Swarnamukhi River with other rivers.

River Basin	Discharge Km ³ /yr	Area 10 ³ km ²	SWR ton.km ⁻² .yr ⁻¹	CWR ton.km ⁻² .yr ⁻¹	CO ₂ sil 10 ⁵ mol km ⁻² .yr ⁻¹	CO ₂ carb 10 ⁵ mol km ⁻² .yr ⁻¹	References
Amazon	6590	6112	13	11.1	0.52	1.05	Gaillardet et al. (1999)
Bhima	–	33.9	12	–	3.3	–	Das et al. (2005)
Brahmaputra	510	580	10.3	35.4	1.50	3.40	Gaillardet et al. (1999)
Chambal	30	139	5.5	–	1.1	–	Rengarajan et al. (2009)
Gad	–	0.98	40	–	5.7	–	Das et al. (2005)
Ganga, Rishikesh	22.4	19.6	12.9	51.7	3.8	–	Krishnaswami and Singh, 1998
Ganges	493	1050	14	28	4.50	2.40	Gaillardet et al. (1999)
Godavari	110	310	–	–	6.4, 7.8, 3.3 ^a	–	Jha et al. (2009)
Huanghe	28.3	752	3.23	30.97	0.35	1.34	Fan et al. (2014)
Indus	90	916	3.8	13.8	0.60	0.90	Gaillardet et al. (1999)
Kaveri, Musiri	8.5	66.24	7.9	–	2.6–3.0	–	Pattanaik et al. (2013)
Krishna	–	36.3	19	–	4.2	–	Das et al. (2005)
Mackenzie	308	1787	1.8	14.9	0.34	1.35	Gaillardet et al. (1999)
Mekong	470	795	10.2	27.5	1.91	2.86	Li et al. (2014)
Mississippi	580	2980	3.8	16.1	0.70	1.46	Gaillardet et al. (1999)
Narmada	–	89	33.9	12.7	21.2	3.6	Sharma and Subramanian (2008)
Nethravati	12	3.7	42	–	2.8–2.9	–	Gurumurthy et al. (2012)
Red	123	120	27.5	31.7	6.83	3	Moon et al. (2007)
Swarnamukhi	5.39	3.225	30.57 (27.15, 32.73, 31.85) ^a	24.42 (9.45, 45.90, 17.92) ^a	8.77 (7.87, 9.75, 8.70) ^a	4.96 (1.07, 5.19, 8.60) ^a	Present study
Tapti	–	61.1	33.6	7.32	18.1	2.2	Sharma and Subramanian (2008)
Upper Han	41.1	95.2	5.6	47.5	1.01	6.80	Li et al. (2009)
Yamuna	10.8	9.6	28	115	7	–	Dalai et al. (2002)
Yangtze	928	1808	5.5	55.9	0.60	5.51	Gaillardet et al. (1999)
Yili	–	–	1.30	7.39	–	–	Wu (2016)

Where, a denotes the mean values of pre-, post- and monsoon seasons.

**Fig. 6.** Scatter plot of ionic constituents[a. Ca²⁺ vs. Mg²⁺, b. (Na + K) vs. Cl, c. Mg/Na vs. Ca/Na, d. HCO₃⁻/Na vs. Ca/Na].

molar ratios of Mg/Na , HCO_3/Na and Ca/Na indicate the presence of silicates and evaporite end member (Fig. 6c and d) (Gaillardet et al., 1999). The river basin area is geologically dominated by granitic gneisses, quartzite rocks and quartz, biotite, feldspars minerals; therefore, silicate weathering has important contribution to the hydrochemistry of the area. Ions such as Na, Cl, Ca and SO_4 are generally used to recognize the process of evaporation in any riverine or groundwater system (Meybeck, 1987). Na in the river water is also contributed from evaporite dissolution. The study area is a semi-arid region where evaporation plays an important role in controlling the hydrochemistry of the river system. High rate of evaporation eventually forms halite which on dissolution during heavy rainfall events dissociates into Na and Cl and releases ions in adjacent flowing river water. In Na + K and Cl plot, sample falls below the equiline towards Na indicating the excess Na which could be supplied from Na-feldspars, shales or Na-silicate clays of the basin (Fig. 6b). From the scatter plot of SO_4 -Cl, it is observed that clustering of majority of data points are around the equiline which supports the dissolution of CaSO_4 and NaCl in the river basin. Nevertheless, some data points of all seasons are also shifting towards Cl axis indicating Cl enrichment by evaporation prevalence in the river basin (Fig. 7). Some monsoon samples also show high concentration of SO_4 than Cl, which is an indication of anhydride (i.e. evaporite mineral).

5.2. Ionic mass budget: Forward model (this model has not been used in earlier publications for other rivers in peninsular India)

Dissolved major ions in riverine system are primarily derived from chemical weathering (silicates, carbonates and evaporites) of various lithologies, biomass and groundwater contribution. It is essential to know the lithologies source contribution by deriving chemical weathering rate and CO_2 consumption flux. The role of chemical weathering contribution to the major ion solute load of the Swarnamukhi River is discussed in the following sections. Forward method is employed in this study to know the ionic mass budget (Zhu et al., 2013) and the calculation is based on some straightforward assumptions. K, Ca, P and N are the major nutrients for the biomass which are accumulated in different parts of their body. Nevertheless, decay of organic matter releases back these elements in the river. At a steady state, plant uptake and decay do not change the ionic budget of the river water significantly and thus, biomass contribution has not been included (Drever, 1997). The contribution of groundwater to river waters in this basin is considered negligible as the Swarnamukhi is an ephemeral river, thus the main contribution in river water flow is from rainfall.

5.2.1. Silicate weathering

Atmospheric corrected Cl is applied as an index of evaporite dissociation to understand the Na silicate contribution to the river water. It is assumed that all Cl is from rainwater and evaporation processes. The molar Na/Cl ratio in rain water is 1.40 and Na/Cl for halite dissolution is 1. K is mainly from dissolution of K-feldspars and biotite because carbonates and evaporites do not supply K in appreciable amount. The K contribution is around 3–5% of total cations in the river water and reveals insignificant contribution from the silicates and atmosphere. It is difficult to estimate K quantitatively from evaporite dissolution as well. An appreciable amount of Ca and Mg is originated from silicate weathering. The molar ratio of Ca/Na (0.35) and Mg/Na (0.24) is released to river drainage system from silicate rocks (granitic gneiss bedrocks) during chemical weathering process (Gaillardet et al., 1999). The major mean cationic contribution from silicate weathering percentage (%) is 60, 46 and 26, atmospheric input is 9, 9 and 11, carbonate weathering is 8, 32 and 20 and from evaporite dissolution varied 22, 12 and 42 in pre-, post- and monsoon seasons, respectively, which shows that silicate weathering contribution had declined in monsoon due to cationic contribution from evaporites dissolution (42%).

5.2.2. Carbonate weathering

Carbonate mineral weathering is very important in controlling river water solute load irrespective of local geological setup. After atmospheric input correction, it is assumed that SO_4 contribution is mainly from evaporation process and Ca, Mg contributions are from evaporite, silicate and carbonate weathering processes (Gaillardet 377 et al., 1999). Mean cation contributions from the carbonate weathering are 8% in pre-, 32% in post- and 20% in monsoon seasons, indicating the high rate of carbonate weathering in post-monsoon is related to heavy rainfall and runoff in the basin.

5.2.3. Evaporite weathering

The semi-arid nature of the study area does not have evaporite lithology but the material loss due to evaporation is often considered for the weathering processes (Gaillardet et al., 1999). Mean evaporite dissolution varied 22% in pre-, 12% in post- and 42% in monsoon. Any remaining cations which are not accounted for by atmospheric, silicates and carbonates derive from evaporites dissolution. The Forward model calculation of different sources of major cation contribution by weathering process in the Swarnamukhi River basin is shown in Fig. 8. The order of contribution of various inputs are Silicate > Evaporite > Carbonate > Atmosphere. Silicate weathering is the dominant contributor of cation in the basin due to the granitic gneissic terrain which is

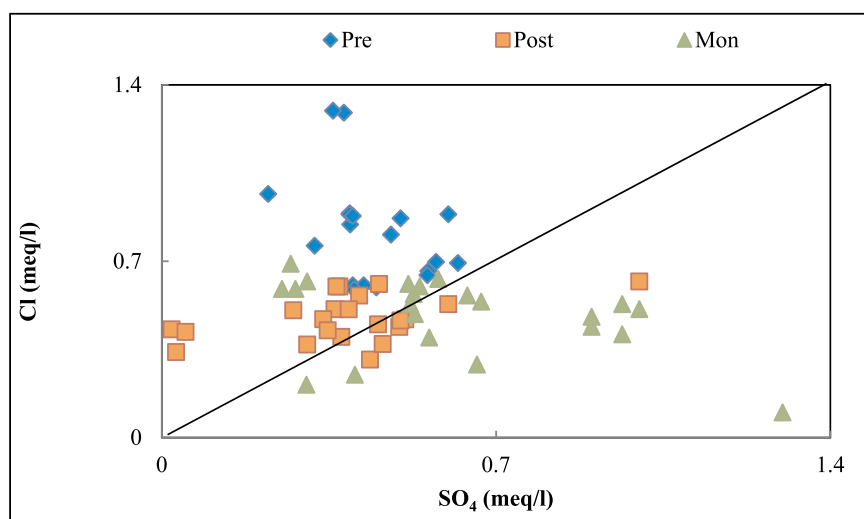


Fig. 7. Scatter plot of SO_4 vs. Cl.

followed by evaporite weathering due to semi-arid conditions with irregular rainfall events. During evaporation, water evaporates leaving behind salts such as halite which dissolve during wet events and enrich the water with lots of Na and Cl.

5.3. Chemical weathering rate

In order to estimate chemical weathering processes, silicate weathering rate (SWR) and carbonate weathering rate (CWR) ($\text{tonnes.km}^{-2}.\text{yr}^{-1}$) are calculated (Pattanaik et al., 2013). Average density of silicate (2.7 g/cm^3) and carbonate (2.4 g/cm^3) is used to convert the rate ($\text{tonnes.km}^{-2}.\text{yr}^{-1}$) into mm/ky (millimeter/1000 years) by dividing with density (Galy and France-Lanord, 1999). Mean silicate weathering rate calculated ($\text{tonnes.km}^{-2}.\text{yr}^{-1}$) is 27.15 (10.06 mm/ky) in pre-, 32.73 (12.12 mm/ky) in post- and 31.85 (11.80 mm/ky) in monsoon and mean carbonate weathering rate ($\text{tonnes.km}^{-2}.\text{yr}^{-1}$) is 9.45 (4.72 mm/ky) in pre-, 45.90 (22.95 mm/ky) in post- and 17.92 (8.96 mm/ky) in monsoon seasons. Silicate weathering rate in pre- and monsoon is higher than carbonate weathering rate which indicates the dominance of silicate weathering in both the seasons. Whereas, carbonate weathering rate is higher in post-monsoon due to rise in groundwater levels which would dissolve the calcretes/Ca precipitates occur around the water wells and supply these carbonated waters to the adjacent streams or the main river course. This would be the reason for high CWR in post-monsoon season in the Swarnamukhi River basin.

5.4. CO_2 consumption by chemical weathering

Based on different stoichiometric coefficients, major cations released during silicate and carbonate weathering are mainly driven by carbonic acid. CO_2 consumption rates (ΦCO_2 : $\text{mol.km}^{-2}.\text{yr}^{-1}$) are calculated (Pattanaik et al., 2013) using the following equations:

$$\Phi\text{CO}_{2\text{sil}} = [2\text{Ca} + 2\text{Mg} + \text{Na} + \text{K}]_{\text{sil}} \times \frac{\text{Discharge}}{\text{Drainage area}}$$

$$\Phi\text{CO}_{2\text{carb}} = [\text{Ca} + \text{Mg}]_{\text{carb}} \times \frac{\text{Discharge}}{\text{Drainage area}}$$

where, cations are in μM , discharge in km^3/year and drainage area in km^2 .

Mean CO_2 consumption rate for silicate weathering ($\times 10^5 \text{ mol km}^{-2}.\text{yr}^{-1}$) is 7.87 in pre-, 9.75 in post- and 8.70 in monsoon seasons. Mean CO_2 consumption rate for carbonate weathering ($\times 10^5 \text{ mol km}^{-2}.\text{yr}^{-1}$) is 1.07 in pre-, 5.19 in post- and 8.60 in monsoon (Table 3). The CO_2 consumption is higher in silicate weathering than the carbonate weathering in the river basin. High CO_2 consumption rate is observed in post-monsoon for silicate weathering whereas in the case of carbonate weathering high value is observed in monsoon samples. Prominent CO_2 consumption in post- and monsoon seasons can be due to heavy rainfall which results in high level of discharge in the Swarnamukhi River basin which has a direct relation with carbon sequestration.

5.5. Comparison of weathering rates and CO_2 consumption rate with global rivers

Silicate weathering rate of Peninsular Swarnamukhi River ($30.57 \text{ tonnes.km}^{-2}.\text{yr}^{-1}$) is higher than Amazon, Huanghe, Mekong, Himalayan Ganga and Brahmaputra and lower than Peninsular Indian rivers Gad, Narmada and Nethravati (Table 3). West flowing rivers draining Deccan basalt show high SWR ($33.9 \text{ tonnes.km}^{-2}.\text{yr}^{-1}$) due to their high runoff, drainage area and easy weatherability of basaltic terrain as compared to east flowing rivers of the Peninsular India which are mainly dominated by granitic gneissic topography (Das et al., 2005; Sharma and Subramanian, 2008). Carbonate weathering rate of the Swarnamukhi River ($24.42 \text{ tonnes.km}^{-2}.\text{yr}^{-1}$) is lower than Huanghe, Ganga, Yamuna and comparatively higher than Mississippi, Amazon and Mackenzie. The final chemical erosion rate (CER) calculated for three seasons of the Swarnamukhi River ($36.60 \text{ tonnes.km}^{-2}.\text{yr}^{-1}$ in pre-, $78.63 \text{ tonnes.km}^{-2}.\text{yr}^{-1}$ in post- and $49.78 \text{ tonnes.km}^{-2}.\text{yr}^{-1}$ in monsoon) is higher than global mean values of $26 \text{ tonnes.km}^{-2}.\text{yr}^{-1}$ (Meybeck, 1979), $21 \text{ tonnes.km}^{-2}.\text{yr}^{-1}$ (Berner and Berner, 1996) and $24 \text{ tonnes.km}^{-2}.\text{yr}^{-1}$ (Gaillardet et al., 1999). This weathering variability could be due to difference in degree of rock weathering in different geological settings and climatic scenarios. High silica flux value in the Swarnamukhi river waters indicates the raised degree of chemical erosion rate of the basin. The Godavari River flows through different lithologies of Deccan trap, Dharwars and Archaean rocks and has chemical weathering rate higher than Swarnamukhi River which consists of mainly single lithology of granitic gneissic terrain. This shows that a river flowing through multiple lithologies has more weathering rate than mono-lithological terrain (Jha et al., 2009).

The CO_2 consumption rate of the Swarnamukhi River ($13.73 \times 10^5 \text{ mol km}^{-2}.\text{yr}^{-1}$) is higher than the Kaveri river (Das et al., 2005), Nethravati river (Gurumurthy et al., 2012), Changjiang river (Chetelat et al., 2008), Upper Han river (Li et al., 2009) and Amazon and Congo-Zaire (Gaillardet et al., 1999). The western flowing Narmada ($24.8 \times 10^5 \text{ mol km}^{-2}.\text{yr}^{-1}$) and Tapi ($20.3 \times 10^5 \text{ mol km}^{-2}.\text{yr}^{-1}$) have higher CO_2 consumption rate (Dessert et al., 2001) than the Swarnamukhi River (Table 3). The annual consumption of CO_2 for the Swarnamukhi River basin accounts for 0.021% and 0.004% in pre-, 0.04% and 0.02% in post- and 0.02% and 0.03% in monsoon by silicate and

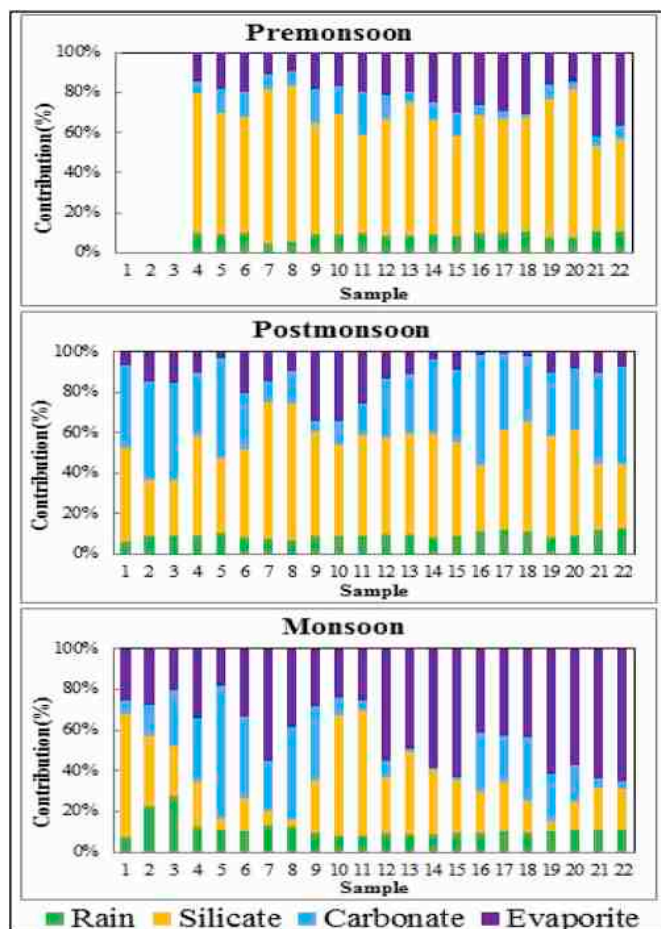


Fig. 8. Contribution of different sources to major cations in pre-, post- and monsoon seasons of the Swarnamukhi River water.

carbonate weathering, respectively of the global CO₂ consumption fluxes by silicate rocks ($12,300 \times 10^9 \text{ mol yr}^{-1}$) and carbonate rocks ($8700 \times 10^9 \text{ mol yr}^{-1}$). The total drainage area of the Swarnamukhi River accounts for 0.0022% of the global continental area of $150 \times 10^6 \text{ km}^2$. The annual CO₂ consumption in the Swarnamukhi River is less mainly due to its relatively smaller drainage area, semi-arid climatic conditions, low discharge and granite gneissic rocks which are highly resistant to chemical weathering (White and Blum, 1995; Ibarra et al., 2016). Total CO₂ consumption rate of the Swarnamukhi River per km² ($13.73 \times 10^5 \text{ mol km}^{-2} \cdot \text{yr}^{-1}$) is around 5.7 times lower than the global average rate of $237 \times 10^5 \text{ mol km}^{-2} \cdot \text{yr}^{-1}$ indicating less CO₂ consumption in the basin.

6. Conclusion

The Swarnamukhi River shows spatio-temporal variation in its physio-chemical composition. The mean total dissolved solid (TDS) of the studied river basin (278.96 mg/l) is similar to mean value of global rivers. Na and HCO₃ are the dominant ions (μM) in all studied seasons with Na–HCO₃ water type. The decreasing order of contribution of various inputs in the whole Swarnamukhi river basin is: silicate > evaporite > carbonate > atmosphere. Silicate and carbonate weathering rates were calculated and mean SWR (tonnes.km⁻².yr⁻¹) is 27.15 in pre-, 32.73 in post- and 31.85 in monsoon and mean CWR (tonnes.km⁻².yr⁻¹) is 9.45 (4.72 mm/ky) in pre-, 45.90 (22.95 mm/ky) in post- and 17.92 (8.96 mm/ky) in monsoon seasons. The Swarnamukhi River has mean net CO₂ consumption rate of $13.73 \times 10^5 \text{ mol km}^{-2} \cdot \text{yr}^{-1}$. The comparison of silicate weathering CO₂ consumption rate of the Swarnamukhi River to other Peninsular rivers of India indicates that the CO₂ consumption rate in the Swarnamukhi is lower than the Narmada–Tapti (Dessert et al., 2001) rivers draining into the Arabian Sea and higher than Kaveri and Krishna river system (Das et al., 2005), Nethravati (Gurumurthy et al., 2012) rivers draining into the Bay of Bengal. The annual average CO₂ consumption rate due to silicate weathering in the Godavari river basin was 2–4 times lower than Narmada and Tapti river basin (Sharma and Subramanian, 2008), which may be due to spatial variability in the weathering rate within the Deccan trap. When compared Godavari basin with the larger size/discharge Himalayan river basin (the Ganges and the Brahmaputra), the CO₂ consumption rate due to silicate weathering in the Godavari river basin was found higher than that of the river Ganges ($0.45 \text{ mol km}^{-2} \cdot \text{y}^{-1}$) and Brahmaputra ($0.15 \text{ mol km}^{-2} \cdot \text{y}^{-1}$) (Gaillardet et al., 1999). This is attributed to the presence of basalt lithology in the Godavari basin. The annual drawdown of CO₂ by silicate and carbonate weathering for the whole Swarnamukhi river basin accounts for 0.021% and 0.004% in pre-, 0.04% and 0.02% in post- and 0.02% and 0.03% in monsoon seasons of the global CO₂ consumption fluxes by silicate and carbonate rocks, respectively. The annual CO₂ drawdown in the Swarnamukhi River is less mainly due to its relatively smaller drainage area, and discharges as compared to other tropical rivers as well as the predominance of granite gneissic rocks which are highly resistant to chemical weathering processes.

Declaration of competing interest

There is no conflict of interest of the study.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.apgeochem.2020.104520>.

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