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Groundwater Memories of Past Climate Change—Examples from India and the Nordic Countries

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Abstract: The last glacial period can be identified in groundwater globally in hydrochemistry and groundwater turnover. To illustrate this, three examples representing very different conditions are presented here, two from India and one from the Nordic countries. The last glacial period resulted in a 125 m lowering of the sea level below present level and the return to the same level within a relatively short geological time span. The low sea water level at Last Glacial Maximum (LGM) induced, in combination with variations in the SW monsoon, recharge of coastal aquifers here exemplified by the Tertiary aquifers in Kerala. The sea level lowering before LGM and its rapid subsequent recovery after LGM has caused different sedimentation conditions resulting in more oxidised Pleistocene sediments compared to Holocene sediments. This has affected the redox conditions and resulted in mobilisation of arsenic in groundwater in the Holocene strata notably in the Bengal delta and upstreams in the Ganga valley.

In the Nordic countries there was a 2.0-2.5 km high load of ice on the land. The result of the melting is seen in land uplift, which is still active to this day. The connections between the Baltic Sea and the ocean via the North Sea has varied during the postglacial period resulting in brackish and fresh water conditions making their imprint in the hydrochemistry and turnover of the groundwater. A common feature is seen from both regions in the form of the Na-HCO_3 type of groundwater formed during fresh water flushing of a formerly saline aquifer. Along some shorelines there are reducing environment similar to those in India but the main manifestation is acid drainage as a result of slow land uplift and drainage for agriculture.

Keywords: Groundwater; Climate change; Recharge; Hydrochemistry.

Introduction

Groundwater ages can be calculated by considering stationary conditions (Post et al., 2013). However, this is not always a realistic assumption when considering past climate change events surrounding the last glaciation and the large changes in sea level. Traces from the Pleistocene and Holocene periods can be identified in hydrochemistry and groundwater turnover globally. In particular the low sea level at and around the Last Glacial Maximum (LGM) has left imprints. At LGM the sea level was approximately 125 m below the present level (Poore et al., 2000). The effects of that phenomenon are especially seen in coastal aquifers where saline-freshwater exchanges have taken place (Ericson et al., 2006).This study will use various coastal and near-coastal aquifers to illustrate the potential consequences of using aquifers as water supplies and why changes in water quality may be of health concern.

Coastal aquifers are important and subject to considerable stress as the population density in coastal areas is usually high. Even outside urban centres the

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population density is in the order of 1000-1500 per km^2 along coastal tracts in South Asia. In addition, coastal areas have good agricultural land with need for irrigation water. In this study, two different areas with very different Pleistocene-Holocene history are investigated, one in India and the other in the Nordic countries. The Indian peninsula has experienced a low sea level promoting recharge. In spite of the different environments in the Nordic countries and in India there are some clear hydrochemical similarities observed, notably the effect of the postglacial recharge in terms of the formation of the Na-HCO_3 type of groundwater. Finland and Sweden had a complicated Holocene history after the slow retreat of the inland glacier which had a height of 2.0-2.5 km. The land uplift during the Holocene, when the load of the ice was released, resulted in several saline and freshwater periods in the Baltic which forms the shoreline of these two countries.

Kerala Tertiary Aquifers and Aquifers on the Indian Southeast Coast

Kerala has a narrow coastal plain with recent deposits underlain by laterite (Jayalakshmi et al., 2004). In the southern part from Kochi to Thiruvananthapuram these shallow deposits are underlain by Tertiary sediments divided into four beds: the Warkali, the Quilon, the Vaikom and the Alleppey beds. The Warkali and Vaikom beds are good aquifers while the Quilon beds—largely consisting of limestone and clay—are less permeable. The Tertiary beds are deposited in a graben formed by faults in the underlying Precambrian rocks (Soman, 2002; Dhiman and Thambi, 2012).The topography of the underlying Precambrian is formed by faults in the SSE-NNW direction. This has facilitated the recharge in the southern part of the area where the overlying recent beds are thinner and thus guided the recharge in the SSE-NNW direction. This is mirrored in the distribution of the groundwater types with the flushing completed in the south and brackish-saline groundwater still left in the north.

The Warkali and Vaikom beds are currently used as water supply for the larger towns and urban centres while most of the water supply to the scattered settlements comes from the recent strata.

The Warkali and Vaikom beds have been subject to a large number of investigations by Central Ground Water of India (CGWB, 2014; Thambi, 2012). The hydrochemistry shows a typical zonation characterising the flushing of formerly saline aquifers by fresh water (Mercado, 1985) (Figure 1). In this case the flushing is completed in the south resulting in Ca-HCO_3 type of water. This is less northwards with a zone of Na-HCO_3 water before the water quality becomes brackish towards Kochi (Table 1).

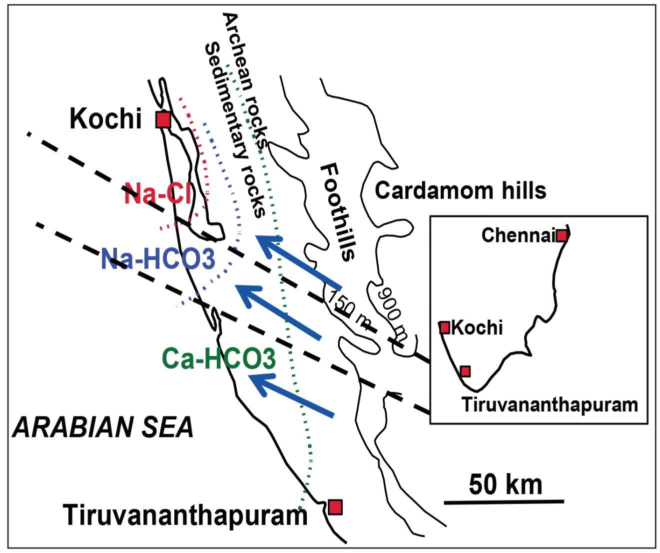

Figure 1: The Kerala coast and the groundwater movements with flushing of the Tertiary aquifers with freshwater resulting in a zonation of the water quality The faults on the figure are found in the Precambrian underlying the Tertiary aquifers. The graben formed by the faulted Precambrian has guided the groundwater flow direction.

The Na-HCO_3 groundwater is typically enriched with fluoride (Jacks et al., 2005). This is also seen here in the Warkali beds in the Na-HCO_3 zone (Dhanya Raj and Shaji, 2016). This zonation might have been guided by the thicker and more permeable beds in the south, north of Thiruvananthapuram.The fresh water flushing is well illustrated by plotting the Na/Cl ratio versus chloride (Figure 3). The Na/Cl ratio in the groundwater has a typical peak just before the exchangeable sodium from the saline period is depleted (Figure 2).

Similar zonation is seen in other deep aquifers in southern India. On the east coast in southern Tamil Nadu Sukhija et al. (1996) have investigated a sequence of aquifers from Holocene to Tertiary. Their work shows a similar zonation of groundwater types from Ca-HCO_3 via Na-HCO_3 to brackish groundwater. Still another sedimentary aquifer at Pondicherry is investigated by Thilagavathi et al. (2012). Here the sedimentary sequence stretches from recent via Tertiary to Cretaceous age. Similarly in Kerala the aquifers often have higher mineralisation than the underlying Tertiary beds. The presence of Na-HCO_3-type of groundwater can be inferred from the correlation analysis (Thilagavathi et al., 2012).

Table 1: Selected analyses from the Kerala Tertiary beds. The order of analyses are from south to north

Site	*Aquifer*	*pH*	*EC mS/ cm*	*Ca^{2+} mg/l*	*Mg^{2+} mg/l*	*Na^{+} mg/l*	*K mg/l*	*HCO_3^- mg/l*	*SO_4^{2-} mg/l*	*Cl^- mg/l*	*F^- mg/l*
Karthikapalli	W	7.45	460	52	12	23	4.0	244	Trace	40	1.00
Karthikapalli	Q	7.52	560	38	13	69	8.9	372	Trace	18	0.74
Karthikapalli	V	7.37	520	46	8.5	53	9.6	305	Trace	33	0.09
Muttom	W	6.95	710	64	16	53	3.9	159	7.5	154	0.48
Muttom	V	6.50	560	56	11	54	7.7	348	Trace	27	0.63
Milk Plant Alleppey	W	7.21	419	14	12	65	11	262	Trace	11	1.6
TDMC Alleeppey	W	7.24	450	16	9.7	69	11	235	Trace	20	1.5
Mannanchery	W	7.36	3700	84	85	480	33	220	Trace	1050	0.6
Kuttamangalam	V	6.52	3540	54	19	630	20	421	35	914	1.3

W = Warkali, Q = Quilon, V = Vaikom. Values in mg/l

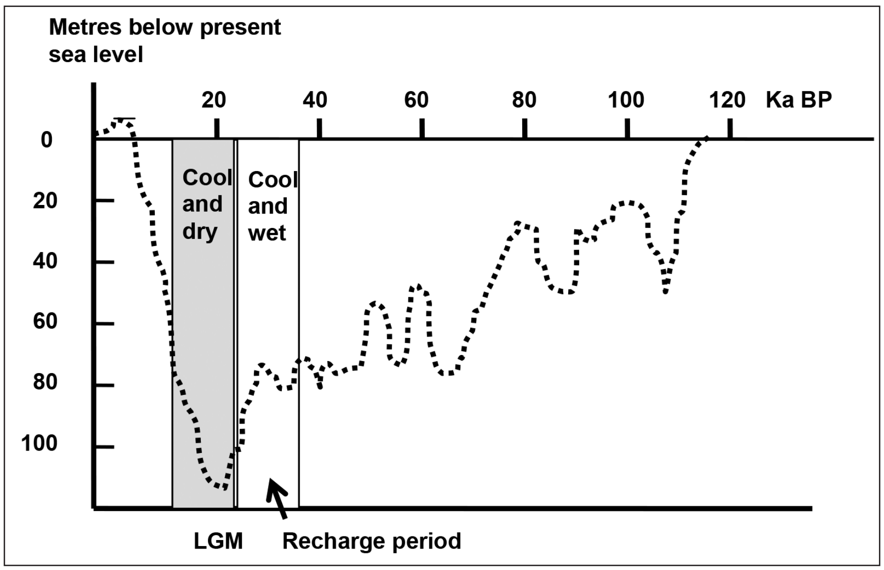

Figure 2: Sea level variations in Pleistocene and Holocene times and the period of recharge. The variation of the monsoonal strength is indicated.

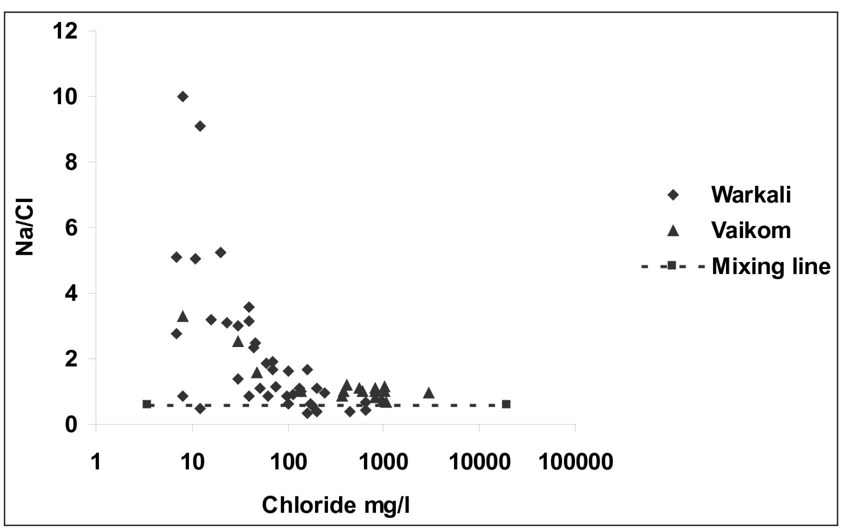

Figure 3: Na/Cl versus chloride concentrations for groundwater from the Kerala Tertiary beds. The peak in Na/Cl at low chloride levels is typical for fresh water flushing.

The Tertiary groundwater has been dated by ^{14}C-dating in nine samples from the Warkali and Vaikom beds and the range of ages varied from 23 to 34 Ka BP (Figure 3). The methodology for the age determination followed that given by Pearson and Hanshaw (1970). This is similar to the ages of groundwater in the Karaikal and Tanjavur area where ages between 20 ka BP to 32 ka BP were recorded (Sukhija et al., 1996). In a Cretaceous confined aquifer in Ramanathapuram District in Tamil Nadu, Saravana Kumar et al. (2009) found groundwater ages of 20-40 ka BP. The strength of the Indian Summer Monsoon (ISM) is one factor that could be considered when it comes to explain why the three aquifers show more or less the same interval for recharge.

There is an abundance of palaeoclimatological data regarding the Indian Summer Monsoon (ISM) and regarding the recharge period of the three aquifers mentioned above. There is data for the ISM that quite well describes the period of recharge for the three aquifers. The strength of the ISM has been assessed by several means from terrestrial archives as well as marine sediments. One of the tools used is the relative abundance of C3 and C4 plants.Sediments in the Darjeeling foothills in the eastern Himalayas indicate a wetter period with a higher abundance of C3 plants during the period 31-22 ka BP and a drier period 22-18 ka BP, thus during the LGM (Ghosh et al., 2015). From the Thar desert in western India, sediments indicate a wetter period 60-30 ka BP and a drier period 20-11 ka BP, the latter during LGM (Juyal et al., 2006).

The low rainfall during LGM is also noted by Kumaran et al. (2013) in terrestrial archives in the Konkan region in the Western Ghats of India. Sediment cores with data for the relative abundance of C3 to C4 plants from the Ganga Plain indicate periods of increased rainfall at 40 ka BP and 25 ka BP (Agrawal et al., 2012). Another sediment investigation from the Ganga Valley (Roy et al., 2012) shows increased aggradation for the period 30-23 ka BP, mirroring a more active ISM but also noting a lower monsoonal activity during LGM. Prabhu et al. (2004) have studied marine sediments from the eastern Arabian Sea and noted a drier climate for 24-15 ka BP and a wetter climate for 50-24 ka BP. A weak ISM for 24-15 ka BP is found by Saraswat et al. (2014) from a Holocene record. A peat deposit in southwest China covers the period 32.7-11.4 ka BP (Huang et al., 2016). Their data based on mineral weathering among other things, mirror a wetter period 33-23 ka BP and drier conditions for 23-18 ka BP. A low monsoonal activity during LGM, is also recorded by Juyal et al. (2009). After 18 ka BP there is a gradual increase to wetter conditions. The review of climatological data largely show a wetter climate in the past before around 23 ka BP and a dry period from 23 ka BP during the LGM. In spite of that the head for recharge was the largest during LGM; it seems that the good conditions for recharge were interrupted by the onset of the dry conditions during LGM.

Over the last three decades there has been little or no change in the total dissolved contents in the Tertiary groundwater in Kerala despite a considerable pumping rate that has lowered the groundwater level below the present sea level. Elevated chloride contents are as per $\delta^{18}O$ analyses not to due mixing in of sea water but due to diffusion of chloride from intercalated clay layers (Figure 4). The groundwater in recent sediments show an increasing $\delta^{18}O$ ratio and chloride concentration presumably due to evaporation but at higher chloride level there is new trend towards the $\delta^{18}O$ for sea water. A sample of pore water from an intercalated clay layer in the Warkali section of sediments showed 2000 mg/l while the over- and underlaying sand and gravel sections had a groundwater with only 15 mg/l. The groundwater in recent shallow aquifers show considerable change over time due to tides which have flooded the coastal plain in part (Shaji et al., 2009; Prasanth et al., 2012). The connection to the Tertiary beds is poor protecting

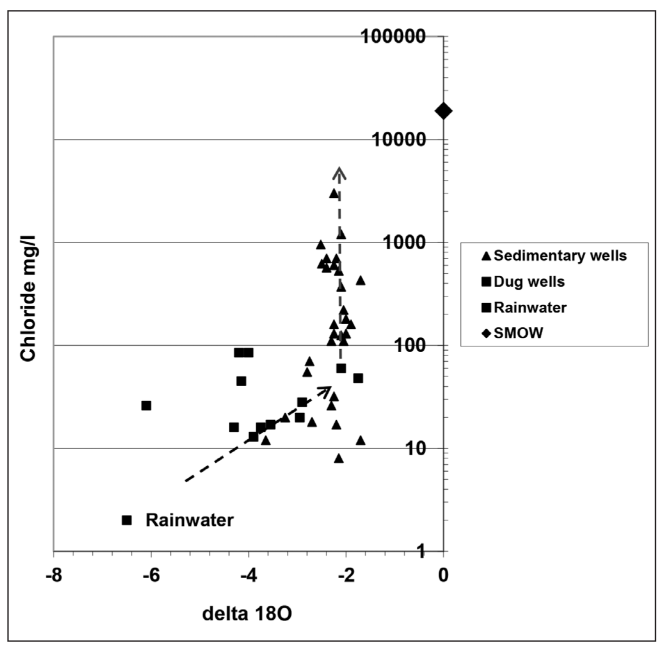

Figure 4: $\delta^{18}O$ ratios versus chloride concentrations. Wells from recent sediments and the Tertiary beds indicating that chloride comes from diffusion of intercalated clay beds.

them from salinization while simultaneously hindering recharge (CGWB, 1992; Dhanya Raj and Shaji, 2016). Tertiary beds in southern India do often have freshwater while the overlaying recent sediments are more affected by tides or by industrial activities (Sonkamble et al., 2014). In the case with the Tertiary beds in Kerala there might be fresh groundwater offshore, a common feature globally (Post et al., 2013). These authors list approximately 30 basins globally that contain fresh or brackish groundwater offshore, among them several in south and southeast Asia. However, there are no offshore drillings available on the Kerala coast to confirm the presence here. Indirectly offshore freshwater discharge has been detected by ^{222}Rn measurement as radon is enriched in groundwater compared to surface and seawater (Gopal Krishan et al., 2015). In testing hypotheses for the formation of fluid mud along the Kerala coast Jacob et al. (2015) have by tritium analyses and ^{222}Rn measurements concluded that there is no connection between the groundwater in the Warkali beds and the offshore water.

Arsenic Mobilisation in the Bengal Basin and the Middle Gangetic Alluvial Plain

Another memory of past climate is the often seen difference between Holocene and Pleistocene sediments in the Bengal delta and areas upstream along the Ganga and Brahmaputra rivers. A major problem here is the presence of elevated arsenic in groundwater (Mukherjee et al., 2015). The Holocene sediments deposited under more anaerobic conditions have more reactive organic matter, while the Pleistocene sediments deposited in better drained sites during late Pleistocene have less organic matter (Acharyya et al., 2000). In a later review of the sedimentary characteristics in the Bengal basin and the the Middle Gangetic Alluvial Plain, Acharyya (2005) has concluded that the arsenic affected groundwater is occurring in the Holocene sediments and close to the present draining streams and rivers.The bacterial diversity is larger in the Holocene sediments where more labile organic matter is still preserved (Al Lawati et al., 2012; Ghosh et al., 2015). This also has the secondary effect that the Holocene sediments are more prone to develop anoxic conditions and mobilise arsenic while Pleistocene sediments are often brownish, oxidised, and have groundwater with low arsenic (Kumar et al., 2016; Kulkarni et al., 2016).

The colour of the sediments do mirror the redox conditions very well and can be used to locate low arsenic as well as low manganese aquifers (Hossain et al., 2014). Shamsudduha and Uddin (2007) found a connection between Holocene sediments formed during the rapid sea level rise after LGM and aquifers with elevated arsenic groundwater. Wetland formation and accumulation of organic matter is likely to be promoted during the sea level rise (Umitsu, 1993), while before LGM, with a sinking sea level, the drainage was better and aerobic degradation of organic matter was favourable. The sinking sea level also promoted erosion and re-sedimentation of the sediments (Umitsu, 1993). Pleistocene elevated terraces are free from groundwater arsenic pollution (Acharyya, 2005). In general this pattern seems to be the case in many coastal sites in South and Southeast Asia (Stuckey et al., 2016). Besides the significance of the organic matter content, the bacterial diversity is also important for the degradation of organic matter and the development of reducing conditions (Sutton et al., 2009).

Groundwater in the Nordic Countries

In a very different environment, i.e. the Nordic countries, the effects of climate variations during glaciation and deglaciation in the Holocene and Pleistocene are clearly visible in hydrochemistry. Much of the land, especially in Finland and Sweden, was under water after the last glaciation. Land uplift, amounting to as much as 285 m around the Bothnian Bay caused various connections to the North Sea, which created brackish periods in the Baltic sandwiched with fresh water periods. After melting of the ice the land rose and is still slowly rising at a maximum rate of 9 mm/year in the Bothnian Bay between Sweden and Finland. As the current sea level increases at a rate of approximately 2-3 mm/year, there is still a shift of the shoreline offshore in the Bothnian Bay allowing freshwater to replace the brackish water resulting in a zonation of the groundwater from a mixed brackish water via a Na-HCO_3-water to a Ca-HCO_3-water, similar to what is found in Tertiary aquifers on the Indian coast.

Table 2. Saline and freshwater periods during Holocene in the Baltic Sea

Location	*Period/Age interval ka BP*	*Salinity*
Baltic Sea	Present-3.0	Brackish
Litorina Sea	8.0-3.0	Brackish
Ancylus Lake	9.5-8.0	Fresh
Yoldia Sea	!0.3-9.5	Brackish
Baltic Ice Lake	12.8-10.3	Fresh

Source: Björk, 1995; Tikkanen and Oksanen, 2002; Berglund et al., 2005

The main period when brackish water was introduced into aquifers accessible for water supply, usually down to about 100 m depth, is the Litorina period with chloride contents of about 6500-7000 mg/l (Nurmi et al., 1988). This is considered to have happened about 7 ka BP (Laaksoharju et al., 1999). The brackish Yoldia sea period was short and the period of elevated salinity shorter than indicated in Table 2. The interplay between the brackish and fresh periods and the postglacial land uplift has created a complex pattern of water qualities in Sweden and Finland in which the flushing of saline aquifers is often seen in terms of a groundwater zonation. An important imprint is the Na-HCO_3 type of groundwater having elevated fluoride levels. More than half of the area of Finland lay below the highest shoreline and about 1/3 of the coastal and central areas of Sweden. The highest shoreline is situated at 285 m above the present sea level in northern Sweden while it lies at around 50 m in southern Sweden.

During the periods with considerable inflow of water from the North Sea the aquifers were flooded with brackish water. After being raised above the sea the groundwater was flushed by fresh water. This result of fresh water flushing is still seen in terms of Na-HCO_3 type of groundwater, notably in drilled wells reaching down to about 100 m and in areas with a rather flat topography. Where the land has a higher more pronounced topography the washing out of brackish water is completed to a larger depth. However, even at sites where investigations to deeper levels have been done a similar zonation is seen (Laaksoharju et al., 2008). A secondary effect is elevated fluoride levels occasionally coming up to 8.5 mg/l (Berger et al., 2016). SW in Table 2 is a typical analysis from a bedrock well in western Sweden, an effect of freshwater flushing of a formerly saline aquifer. M1 is another bedrock well where the flushing has gone to a typical Na-HCO_3 type of water also enriched in fluoride while M2 is a bedrock well situated within a few hundred metres from M1 and where the flushing is complete resulting in a Ca-HCO_3 type of water.

In the Litorina period (Table 2) sulphidic clay sediments were deposited (Åström and Björklund, 1995). During the isostatic uplift and agricultural drainage they are subject to oxidation. The main effect is acidic runoff in streams.

In Finland similar types of groundwater occur especially in so-called rapakivi granites (Lahermo and Backman, 2000). These granites are hornblende-biotite granites with rounded crystals of orthoclase mantled by oligoclase and enriched in fluorine. The fluoride concentrations in groundwater come up to similar values as in Table 2. There is a local risk of dental fluorosis in Sweden and Finland in areas where people have their own private wells (Augustsson and Berger, 2014).

The Ramloesa aquifer in the southern-most part of Sweden is a sedimentary aquifer known for its bottled water (Ramloesa water). This aquifer has a history of flushing of a formerly saline aquifer with fresh water after the land has been uplifted (Agerstrand et al., 1981; Frengstad et al., 2010). A test drilling from the Ramloesa aquifer shows a zonation from a typical Ca-HCO_3 type to a Na-HCO_3type below an aquitard (Figure 5).The fluoride is elevated and in the bottled water it contains 2.4 mg/l. Ramloesa water has been produced for decades without much change in composition as the sediments have a large ion exchange capacity. Similarly very extensive extraction of groundwater from the Tertiary aquifers in Kerala has not appreciably changed the water quality. In fractured hard rocks pumping can induce rapid changes (Agerstrand et al., 1981).

Groundwater in the deeper sections of the Precambrian shield has been investigated in connection with the search for safe burial sites for spent nuclear waste (Gimeno et al., 2014). In general the age of the groundwater at around 500 m depth is the range of tenths of thousands of years and below 1000 m the residence time is in the order of 1.5 M years and bearing a reduced character (Louvat et al., 1999). The latter is considered as favourable for the immobilization of radioactive isotopes. The groundwater in hard rock

Table 3. Fluoride-rich groundwater resulting from freshwater flushing of a formerly saline aquifer in SW Sweden, values in mg/l

Site	*pH*	*Na^+*	*Ca^{2+}*	*Mg^{2+}*	*K^+*	*HCO_3^-*	*Cl^-*	*SO_4^{2+}*	*F^-*
L	8.9	190	2.0	1.3	2.0	300	93	15	8.5
M1	8.3	185	13	3.0	3.1	325	55	55	3.5
M2	7.4	22	89	15.1	2.5	290	51	51	0.7

L is a hard rock well in SW Sweden and M1 and M2 are hard rock wells from the Stockholm region, both situated in close vicinity to each other. Values in mg/l.

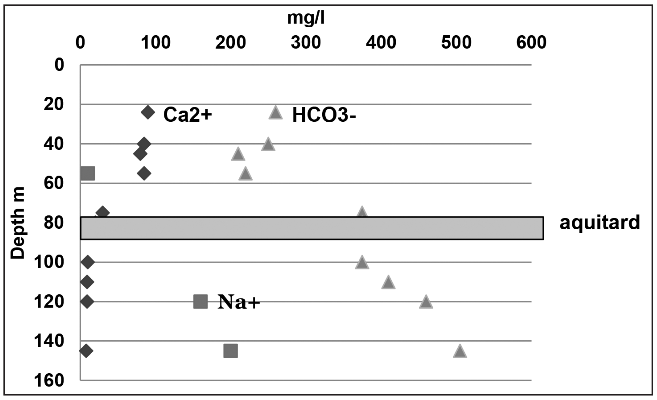

Figure 5: Test boring at Ramloesa site in southern Sweden showing the change from a Ca-HCO$_3$ type of water to a Na-HCO$_3$-type below an aquitard. The sediments are sandy-gravelly with a clayey aquitard at 80-90 m depth.

aquifers show a complex mixing pattern of meteoric, past and present Baltic seawater which has been modelled in connection for planning for repositories for spent nuclear fuel (Laaksoharju et al., 1999).

Summary

Groundwater has globally a very varying residence time, from less than a year to millions of years. The last glaciation is an important event that is traceable in the turnover rate and hydrochemistry in many aquifers. Along the coast of the Indian peninsula larger aquifers bear memories from the low sea level at Last Glacial Maximum(LGM). Before LGM there was a cool and wet period which promoted recharge by the SW monsoon. At LGM and later a cool and dry period interrupted this recharge. Age determinations from three deep aquifers, in Kerala (CGWB, 1992) and two sites on the south-eastern coast (Sukhija et al., 1996; Saravana Kumar et al., 2009) indicate a recharge period during late Pleistocene in a time span approximately between 40 to 20 ka BP. This recharge period as well as the dry period during and after LGM is verified by data mirroring the strength of the southwest monsoon. The flushing with fresh water during the pre-LGM period was not complete and thus a sequence of water types is seen from the Ca-HCO$_3$ type of water via Na-HCO$_3$ type to a brackish water. Fresh groundwater may exist offshore, a phenomenon which has been observed in a large number of sites globally. However, this has not been proved in India. The slow recharge rate in the deeper aquifers should be considered to avoid overdraft and saline intrusion. In Kerala larger urban centres are switching over to treated surface water.

The different sedimentation conditions during Pleistocene compared to Holocene has resulted in differing redox conditions. The rapid lowering of the sea level before LGM resulted in sediments deposited under more oxidising condition while the rapid rise of the sea level in Holocene resulted in more wetland and more accumulation of organic matter. This has resulted in moderately reducing conditions in the shallower sediments in the Bengal Delta and the Middle Gangetic Alluvial Plain and mobilisation of arsenic into the groundwater.

In the Nordic countries, notably Sweden and Finland, the Baltic Sea has varied from fresh water to brackish conditions which, in combination with a rapid land uplift after the release of the ice cap, has resulted in rather complicated groundwater turnover and hydrochemical conditions. The same flushing pattern discovered along the coast of the Indian peninsula can also be seen in Sweden and Finland. However, the varying hydraulic conductivity in predominantly hard rock areas in these countries has resulted in more of mixing and a less clear pattern of water types. In sedimentary aquifers, the flushing pattern is more visible. In the coastal areas there are redox phenomena in the form of sulphidic soils which after uplift and drainage produce acidic discharge (Nystrand and Österholm 2013). In the Bengal delta the redox level is higher and found at the ferric reduction level resulting in arsenic mobilization.

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