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Playas of the Thar Desert: Mineralogical and Geochemical Archives of Late Holocene Climate

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Abstract: This study presents a synthesis of the published data and summarizes new data on mineralogy and geochemistry of the late Holocene playa sediments from the Thar Desert, located in the down stream of southwest monsoon. The eastern margin of the Desert is semi-arid (>400 mm a^{-1}), whereas the western region is arid (200-300 mm a^{-1}). The negative water budget (higher evapo-transpiration/precipitation) has led to the presence of a number of playas in the region. The assemblage of clastic minerals is similar and constitutes quartz, plagioclase, K-feldspar, amphibole, mica and chlorite. The non-clastic mineral assemblage is variable and comprises of both evaporite minerals and carbonates. The geochemical proxies enabled differentiation of the shallow depth profiles at five different playas into horizons of varying chemical weathering, aeolian input and evaporation. Between ca.1.3-3.1 ka, the dominance of proto-dolomite, $[Ca_{>1}(Fe,Mg)_{<1}(CO_3)_2]$ in the eastern playas and presence of gypsum ($CaSO_4 . 2H_2O$) in the western playas suggest that the present day climatic gradient might be exiting during the late Holocene.

Key words: Evaporite minerals, geochemistry, saline lake, Thar desert, Holocene, paleoclimate

INTRODUCTION

Playas are topographically enclosed, saline shallow water bodies formed in a variety of topographical depression in arid and semi-arid regions, where evaporation exceeds precipitation (Shaw and Thomson, 1989; Briere, 2000) e.g., Northern Great Plains of Canada (Last, 1989), high plains of southwest USA (Osterkamp and Wood, 1987; Langford, 2003), northern and central Spain (Schütt, 1998, 2000), Africa (Jones *et al*., 1977) and in the Thar Desert of India (Wasson *et al*., 1984; Enzel *et al*., 1999; Roy *et al*., 2001; Sinha and Raymahashay, 2004; Roy *et al*., 2006, 2007; Roy and Smykatz-Kloss, 2007; Roy, 2007). The geochemical evolution of the playa brine and composition of minerals precipitated from the brine are controlled by composition of catchment lithology, sequential precipitation of evaporite minerals with increasing evaporation, dissolution and re-precipitation of earlier precipitated evaporites, reverse weathering of precursor clay minerals,

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seepage loss and sulphate reduction in an anoxic environment of deposition (Rosen, 1994; Rouchy *et al*., 2001; Yan *et al*., 2002; Yechieli and Wood, 2002). So the sediments deposited in the basins provide information about paleo-depositional environments, paleo-hydrology and geochemical processes responsible for the formation of economically important evaporite minerals.

Sedimentary facies, pollen assemblages, evaporite mineralogy and geochemical proxies from the sediments deposited in the Thar Desert playas have been used as tools to delineate the phases of varying southwest Indian monsoon and the paleo-hydrological condition during the late Pleistocene-Holocene (Singh *et al*., 1972, 1990; Wasson *et al*., 1984; Sundaram and Pareek, 1995; Enzel *et al*., 1999; Deotare *et al*., 2004; Sinha and Raymahashay, 2000, 2004; Sinha *et al*., 2006; Roy *et al*., 2006). Comparison and synthesis of the results suggest that the entire region experienced hyper-arid, hyper saline condition from LGM to 15 ka BP. The playa hydrology was fluctuating between hypersaline and fresh water between 15 and 7-8 ka BP. The climatic condition improved and the playas turned perennial between 7-8 and 5-6 ka BP. This improved climatic condition was relatively extensive in the eastern part of the desert compared to west. The western region is experiencing an ephemeral condition since last 6 ka BP, whereas the eastern playas are ephemeral since last 3.2 ka BP.

In this study, we summarize the comparative mineralogy and geochemistry of late Holocene sediments from ten different playas across the Thar Desert. Phases of varying chemical weathering, aeolian influx and evaporation are identified in the five different shallow depth profiles using the elemental ratios. We propose that a paleo-humidity gradient was present during late Holocene and support it by the relative abundance of evaporite mineralogy in the shallow core sediments of eastern and western playas.

Regional Setting

The Great Indian Sand Desert, better known as the Thar Desert, covers an area of ca. 4,50,000 km^2 and is divided between the state of Rajasthan (India) and the province of Sindh (Pakistan). To the east, it is bounded by the Aravalli Range of mountains, which divides Rajasthan physiographically into two parts: the semi humid eastern part and the semi arid to arid western part. The region experiences variable annual southwest rainfall and the annual average rainfall shows a gradual decrease from 500 mm in the eastern margin to 100 mm in the western margin (Fig. 1). During the summer, the temperature of the region rises up to 50°C and during the winter mornings the temperature falls to as low as 6°C. The evapo-transpiration in the region ranges from three to twenty times higher than the precipitation resulting negative water budget in the hydrologically closed, saline playas.

The playa catchments are characterized by rocks belonging to the Archaean (3.3 Ga) and Proterozoic (0.75 Ga) ages (Abu-Hamatteh, 2002). The Archaean amphibolite, granite and gneisses overlain by Proterozoic quartzite, mica-schists, gneisses (Biswas *et al*., 1982; Misra, 1982; Sen and Sen, 1983; Dassarma, 1988; Sinha and Raymahashay, 2004) are exposed along the Aravalli mountain system. The Malani basalt and rhyolites, Palaeozoic Bap boulder bed and Proterozoic sandstone are exposed in the central and western Rajasthan. The Pleistocene sand dunes are exposed in the surroundings of the eastern and western playas. Our recent work (Roy and Smykatz-Kloss, 2007) suggested that playas of the entire region receive clastics from the eastern Aravalli Mountains. Additionally, the western playas also receive sediments from the surrounding basalt and sandstone outcrops.

The tectono-geomorphic evolution of these playas has been related to excessive siltation in the river confluence, dune segmentation of former streams during the late Pleistocene climatic transition (Agarwal, 1957; Ghose, 1964; Ghose *et al*., 1977; Kar, 1990; Singhvi and Kar, 1992) and tectonic movements along the lineaments that caused formation of horst and graben structures (Sinha-Roy, 1986; Dassarma, 1988; Roy, 1999). These playas are replenished by surface runoff

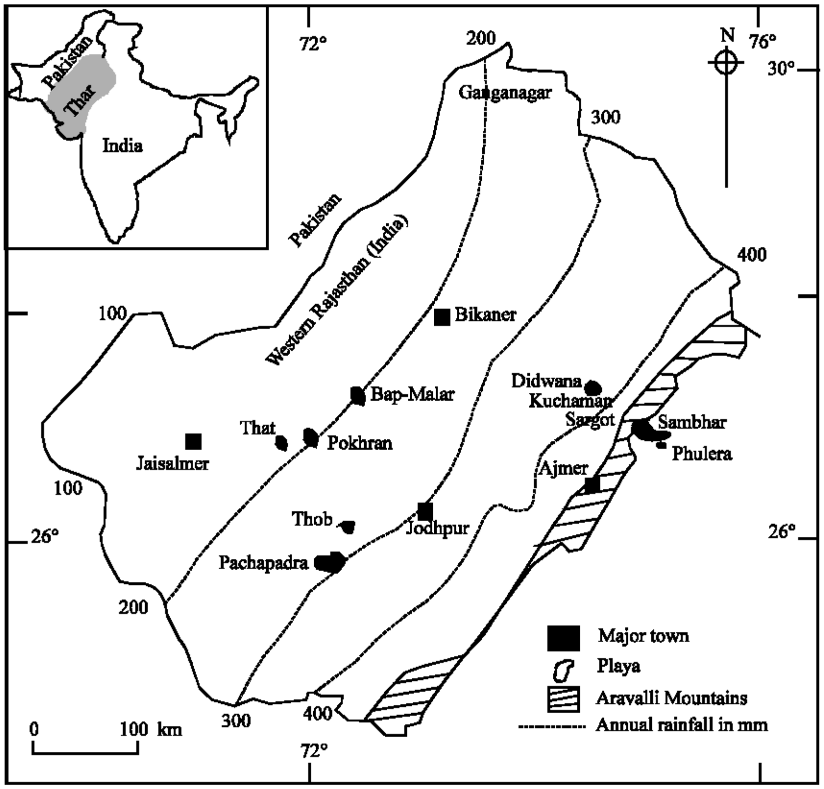

Fig. 1: Location map of the Thar desert, India. The (sampled) playas (except Phulera) are scattered in the region west of the Aravalli Mountains

during the 3-4 months of southwest monsoon and derive sediments and soluble ions from the chemical weathering of rocks from Aravalli mountains (Sinha and Raymahashay, 2004; Roy and Smykatz-Kloss, 2007). The hypersalinity of the brine is attributed to the higher evapo-transpiration and negative water budget in the region (Ramesh *et al*., 1993; Yadav, 1997).

MATERIALS AND METHODS

Sediment samples were collected from the surface of a total of ten playas namely, Phulera, Sambhar, Sargot, Kuchaman, Didwana (all sampled in May 2001), Bap-Malar, Thob, That, Pachapadra and Pokhran (all sampled in October 2002) (Fig. 1). Samples from shallow (1-1.5 m) profiles were also collected through hand augering and dug pits from the Phulera, Sambhar, Didwana, Pachapadra and Pokhran playas at an interval of 10-25 cm (Fig. 2). All the playas have an evaporite enriched crust at the surface but sampled sediments along the shallow depth profile consist of sandy silt and silty-clay (Fig. 2). The clastic and non-clastic minerals were identified with the X-ray diffraction (XRD) analysis from the dried, powdered, bulk sediments using Cu target for the 2θ range of 3° to 63° in the Siemens Diffractometer. Taking into consideration the area below the characteristic major peak as indicative of the quantity of the mineral present, the clastic and non-clastic minerals were quantified separately after recalculating the integrated XRD intensities of characteristic major peaks (area below the peaks) of all the identified minerals to 100%. In a few samples, the compositions of the clastic grains were determined with a SX50 Cameca electron microprobe analysis and the types of feldspars were

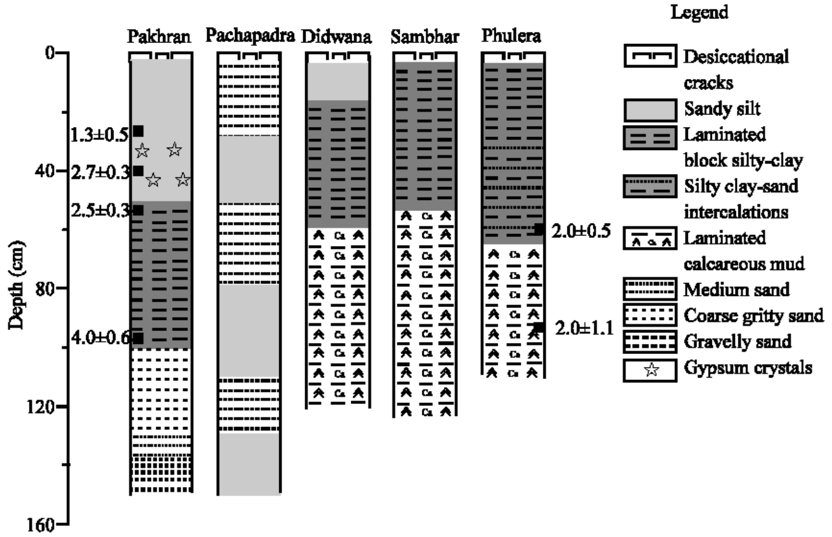

Fig. 2: Generalised lithostratigraphy of sampled shallow profiles from Thar Desert playas and OSL chronology

identified. Major elements were measured in SRS 303 AS XRF instrument and trace elements were measured by Spectrace 5000 XRF. For the major element analysis, soluble evaporite minerals were removed from the bulk sediments by washing repeatedly in double distilled water. Trace elements were measured in the total sediments. So the major element concentrations represent the composition of clastics and carbonate fractions and trace elements concentrations mirror the bulk composition of the sediments. Total amount of sulfur and carbon present in the playa sediments were measured from powdered sediments by the Leybold 5003 Carbon Sulfur Analyser (CSA). The amount of organic carbon present in the sediments was estimated by measuring the amount of inorganic carbon with the Carbon Water Analyser (CWA) and then subtracting that from the total amount of carbon.

A total of six OSL ages (2 samples from the Phulera playa and 4 samples from the Pokhran playa) from the fine grain fractions (4-11 μm) were dated using the Infrared Stimulated Luminescence (IRSL) dating technique in the Daybreak 1150 automated TL/OSL system at the Physical Research Laboratory, Ahmedabad.

RESULTS

Mineralogy and Core Lithostratigraphy

Playa sediment mineralogy allows us to reconstruct lake phases (ephemeral, perennial) as well as depositional conditions (e.g., salinity, brine composition). These sediments essentially consist of a detrital fraction, derived by chemical weathering from the playa catchment and the evaporite fraction, formed by precipitation from aqueous solution. The evaporite fraction is significantly influenced by lake water salinity and brine composition and therefore, these minerals provide very important clues for reconstruction of paleolimnic environments. Based on thermodynamic considerations, evaporites form in a predictive sequence with increasing evaporation (aridity) (Eugster and Hardie, 1978; Ingebritsen and Sanford, 1998). The first mineral to precipitate in most cases is calcite ($CaCO_3$). Subsequent precipitation of a mineral sequence of sulphates (e.g., gypsum, $CaSO_4.2H_2O$), silicates (e.g., smectite) and chlorides (e.g., halite, NaCl) is controlled by relative concentration of Ca^{+2}, Mg^{+2}, HCO_3^{-1}, SO_4^{-2} and Cl^{-1} in the brine. It follows therefore that calcite would represent the onset of salinity and aridity and halite would form in late stages of evaporation under high salinity conditions. Gypsum would represent an intermediate stage. In some instances when evaporation does not reach up to gypsum saturation, as calcite continues to be precipitated, Mg/Ca ratio in the water increases and Mg content of the subsequent carbonate precipitates rises. In other instances, however, evaporation may continue beyond the halite stage and depending upon the availability of K^+ ion, minerals like carnallite ($KMgCl_3 . 6H_2O$) and sylvite (KCl) may also precipitate. The presence of these minerals would therefore indicate extremely hypersaline conditions. However in some cases, the highly soluble chloride bearing minerals can be present through out the sediment sequence caused by their dissolution from the playa surface, percolation of Cl rich hypersaline brine from playa surface into the shallow profiles and their reprecipitation in the sediment pores. Table 1 and 2 shows comparative detrital and evaporite mineralogy of the bulk sediments of several playas of the Thar Desert.

The detrital fraction of all the playas shows very similar assemblage (Table 1, 2). They consist of quartz, feldspars, sheet silicates (micas and chlorites), amphiboles and garnets. Figure 3 shows the relative abundance of quartz, feldspars and sheet silicates in the surface and shallow core samples of the playas considering that the semi-quantitative determination from the XRD charts possibly include an error of 10-20%. Quartz is the most abundant mineral and constitutes up to 90% of the clastic fractions. Feldspars (K-feldspar and plagioclases) are the next clastic component in abundance. They are relatively higher in the surface sediments compared to the deeper samples. In bulk samples, the amount of plagioclase is much more in comparison with K-feldspars. The K-feldspars are of microcline, orthoclase and adularia types, whereas plagioclases vary between albite and CaO-rich labradorite. Sheet silicates are present in traces (up to 10%) in the surface, but it is relatively more (up to 20%) in the deeper sediments (Fig. 3). The chlorites are characterized by high intensities of the even-order reflections (002, 004) and weak intensities of the odd-order reflections (001, 003), which indicate it to be a Fe-rich type (Moore and Reynolds, 1997). Trace amounts of amphiboles and garnets are also identified in the bulk sediments.

The following sections describe the distinctive features of these playas, lithostratigraphy and evaporite mineralogy of near-surface sediments in shallow profiles. The summarized lithostratigraphy of the playas along with OSL ages are shown in Fig. 2.

Phulera

The Phulera is a small and dry playa (ca. 6 km^2) located in the east of the Aravalli mountains that form the eastern margin of the Thar Desert. The playa receives annual rainfall of ca.500 mm which dries out during the summer months forming desiccated polygonal cracks on the surface.

A shallow profile of 110 cm at Phulera shows that the near surface sediments are carbonate-rich and the concentrations of sulphate and chloride are extremely low (Table 2). The lowermost lithostratigraphic unit (110-60 cm) consists of laminated calcareous mud. In this unit, calcite ($CaCO_3$) and non-stoichiometric Fe-bearing proto-dolomite ($Ca_{>1.0}(Mg,Fe)_{<1.0}(CO_3)_2$) are the major carbonate minerals. Huntite [$CaMg_3(CO_3)_4$] and anhydrite ($CaSO_4$) are present in very minor amount. Between 70 and 80 cm, there is a distinct layer of dominant proto-dolomite with traces of halite. This horizon is also characterized by desiccation cracks filled with fine sand. Though the OSL ages are similar, an extremum age bracket is derived using (age+error) for the basal sample and (age-error) for the upper sample. This yields an age range of <3.1 ka to >1.5 ka for this litho unit (Fig. 2).

The topmost litho unit (60 cm-surface) consists of moderately laminated silty clay and medium sand intercalations capped by a thin horizon (5 cm) of well developed polygonal cracks. Between 60-20 cm, calcite is dominant compared to proto-dolomite. Trace amounts of halite (NaCl), thenardite (Na_2SO_4), anhydrite ($CaSO_4$) and proto-dolomite are present along with minor amounts of calcite ($CaCO_3$) in the surface sediments (Table 1).

Table 1: Mineralogy of surface sediments of Thar desert playas

Playa	Q	F	M	Chl	Am	Cc	Pro-dol	Anhy	Th	Gyp	Hal
Phulera	++++++	++++	++	++	+	+++	+	+			+
	++++++	++++	+	+	+	+++	+	+	+		++
	++++++	+++	+	+	+	++	+	+			+
	++++++	++++	+	+		++			+		+
Sambhar	++++	++	+	+		++	+	+	++		++++
	+++	+	++	++		++	++	+	+		+++
	++++	++	++	++	+	++	+	+	+		++++
	++++	++	++	++	+	++	+	++	+		++++
	++++	++	+	++	+	++		+	++		++++
	++++	++	++	+		++	++	++	++		++++
	+++	++	++	++		+++	+	++	+		++++
Kuchaman	++++	+++	++	+	+	++	+	+	+		+
	++++	++++	+	+	+	++	+	+	++		++++
	+++	++	++	+	+	+++	+	+	+		
Sargot	++++	++++	++	+	+	+	+	+			++++
	++++	++++	++	+	+	++		+			++++
Didwana	+++++	+++	+	+	+	+	+		+		+++++
	+++++	+++	+	+	+	+	+	+	++		++++
	++++++	+++	+	+	+	++	+	+	+		++
	++++++	+++	+	+	+	+	+	+	+		++
	++++++	+++	+	+	+	+		+	+		+++
	+++++	+++	+	+	+	++	+		++		+++
	++++++	+++	+	+	+	+	+	+	+		+++
	++++++	+++	+	+	+	+	+	+	+		++
	+++++	+++	+	+	+	+			+		+++
	++++++	+++	+	+	+	+	+		+		+++
	++++++	+++	+	+	+	+	+		+		+++
Bap-Malar	++++++	++	+	+	+	++	+	+			
	++++++	+++	+	+	+	++	+	+			
	+++	+	+	+	+	+	+	+		+	+++++
	++++++	+++	+	+	+	++	+	+			
That	+++	++				+					+++++
	+										+++++
	++++	++			+	+				+	+++++
	+									+	+++++
	++++	+				+				+	+++++
Thob	++++++	+				+		+		+	+++++
	++++++	+++	+	+	+	++		+		+	++++
Pachapadra	+++++	+++	+	+	+	++	+	+			++++
	++++++	+++	+	+	+	+++	+	+			++++
	+++++	+++	+	+	+	++	+	+			++++
	+++++	++	+	+	+	+++	+	+			+++++
	+++++	+++	+	+	+	+++	+	+			++++
	+++++	++	+	+	+	+++	+	+		+	+++++
	++++	+	+	+		++				++	+++++
	+++++	++	+	+	+	+++				+	+++++
	+++++	+	+	+	+	+++		+		+	+++++
	+++++	+	+	+	+	+++		+		+	+++++
Pokhran	++++++	++	+	+		++					+++
	+++	+	+	+		+		+			+++++
	++++++	++++	+	+	+	++					+++
	++++++	+++	+	+		++	+	+			+++
	++++++	++	+	+	+	++				+	++

Idealised compositions:

Carbonates:

Calcite, $CaCO_3$
Protodolomite, $Ca_{>1.0}(Mg,Fe)_{<1.0}(CO_3)_2$
Huntite, $CaMg_3(CO_3)_4$
Trona, $NaHCO_3 . Na_2CO_3 . 2H_2O$

Sulphates:

Gypsum, $CaSO_4 . 2H_2O$
Anhydrite, $CaSO_4$
Thenardite, Na_2SO_4
Mirabilite, $Na_2SO_4 . 10H_2O$
Glauberite, $CaSO_4 . Na_2SO_4$
Polyhalite, $K_2Ca_2Mg(SO_4)_4 . 2H_2O$

Chlorides:

Halite, NaCl
Carnallite, $KMgCl_3 . 6H_2O$
Sylvite, KCl

(Q = Quartz, F = Feldspar, M = Mica, Chl = Chlorite, Am = Amphibole, P = Palygorskite, Cc = Calcite, Pro-dol = Protodolomite, Anhy = Anhydrite, Th = Thenardite, Gyp = Gypsum, Hal = Halite, K-evap = K-bearing evaporites) (++++++ = >40%, +++++ = 30-40%, ++++ = 20-30%, +++ = 10-20%, ++ = 5-10%, + = <5%). Trace amounts of Illite-smectite and palygorskite, K-evaporite from Sambhar, trona from Kuchaman and Didwana, glauberite from Sargot are identified

Table 2: Mineralogy of shallow-surface sediments of Thar desert playas

Playa	Depth	Q	F	M	Chl	Am	Cc	Pro-dol	Huntite	Anhy	Then/Mir	Gyp	Hal	K-evap.
Phulera	20	++++++	+++	+	+	+	+++	++	+					
	30	++++++	++	+	+	+	++++	+++						
	40	+++++	++	+	+	+	++++	+++						
	50	+++++	++	+	+		++++	+++						
	60	++++++	+	+	+	+	++++	+++						
	70	++++	++	+	+		+++	+++++					+	
	80	+++++	++	+	+		++++	+++	+					
	90	+++++	+++	+	+	+	++++	+++	+	+				
	100	++++	+++	+	+	+	+++++	+++	+					
	110	++++++	+++	+	+	+	++++	++						
Sambhar (S1)	25	++++	+++	+++	++		+++	++		+	+		++	+
	50	++++	+	++	+		+++	++		+	+		++	+
	75	++++	++	++	+		++	+++					++	+
	100	++++	+++	++	+		+++	+		+	+		++	+
	125	++++	++	+	+		++++	+		+	+		++	++
Sambhar (S2)	25	++++	+++	++	++		+++	+		+	++		+++	+
	50	++++	++	++	+		++	+++		++	++		+++	+
	75	+++	++	++	+		+	++++		+	++		+++	+
	100	++++	++	++	+		++++	+++		+	++		++++	+
	125	++++	++	++	+		+++	+++		+	++		+++	+
Sambhar (S3)	25	++++	++	++	+		+++			+			++	+
	50	++++	++	++	++		+++	+		+	+		+++	+
	75	+++	++	+	+		++	++		+	++		++	+
	100	++++	++	++	+		++++	++		+	++		+++	+
	125	++++	+	+	+		+++	+		+	+		++++	+
Didwana	15	++++++	+++	++	+	+	++			+	+		+	
	30	+++++	+++	+	+	+	+++	+		+	+		+++	
	45	+++++	++++	+	+	+	+++			+	+		++	
	60	+++++	+++	+	+		+++	++		+	+		++	
	75	++++++	+++	+	+	+	+++	++		+	+		++	
	90	++++++	+++	+	+		+++	++		+	+		++	
	105	+++++	+++	+	+		+++	++			+		++	
	120	+++++	+++	+	+		+++	++			+		+++	
Pachapadra	10	++++++	++	+	+	+	+++			+		+	+++	
	20	++++++	++	+	+	+	++++					+	+++	
	30	+++++	++	+	+		++++					+	+++	
	40	+++++	++	+	+		++++					+	+++	
	50	++++++	++	+	+		+++			+		++	+++	
	60	++++++	++	+	+		+++			+		+	+++	
	70	+++++	++	+	+		++++					+	++++	
	80	+++++	+	+	+		++++					+	++++	
	90	++++++	++	+	+		+++					++	+++	
	100	+++++	++	+	+		++++					+	+++	
	110	+++++	+++	+	+		+++					+	+++	
	120	+++++	+++	+	+		+++					+	+++	
	130	++++++	+++	+	+	+	++			+		++	++	
	140	+++++	++	+	+		+++					+++++		+++
	150	+++++	++	+	+	+	+++			+		+++	+++	
Pokhran	10	++++++	+++	+			+++						+	
	20	++++++	++	+	+		+++			+		+	+	
	30	++++++	+++	+	+		+++	+		+		+	+	
	40	++++	++	+	+							++++++		+
	50	++++++	+++	+	+	+	+			+		+++	++	
	60	++++++	+++	+	+		+++	+		+		+	++	
	70	++++++	++	+	+		+++			+		+	+++	
	80	++++++	+++	+	+		+++			+		+	++	
	90	++++++	+++	+	+	+	+++	+		+		+	++	
	100	++++++	+++	+	+	+	+++	+		+			++	
	110	++++++	++	+	+		++	+		+			+	
	120	++++++	+++	+	+	+	+++	+		+			+	
	130	++++++	+++	+	+	+	++	+		+			++	
	140	++++++	++			+	++++			+			+	
	150	++++++	++				++++			+			+	

(Q = Quartz, F = Feldspar, M = Mica, Chl = Chlorite, Am = Amphibole, P = Palygorskite, Cc = Calcite, Pro-dol = Protodolomite, Anhy = Anhydrite, Th = Thenardite, Mir = Mirabilite, Gyp = Gypsum, Hal = Halite, K-evap = K bearing evaporites), (++++++ = >40%, +++++ = 30-40%, ++++ = 20-30%, +++ = 10-20%, ++ = 5-10%, + = <5%)

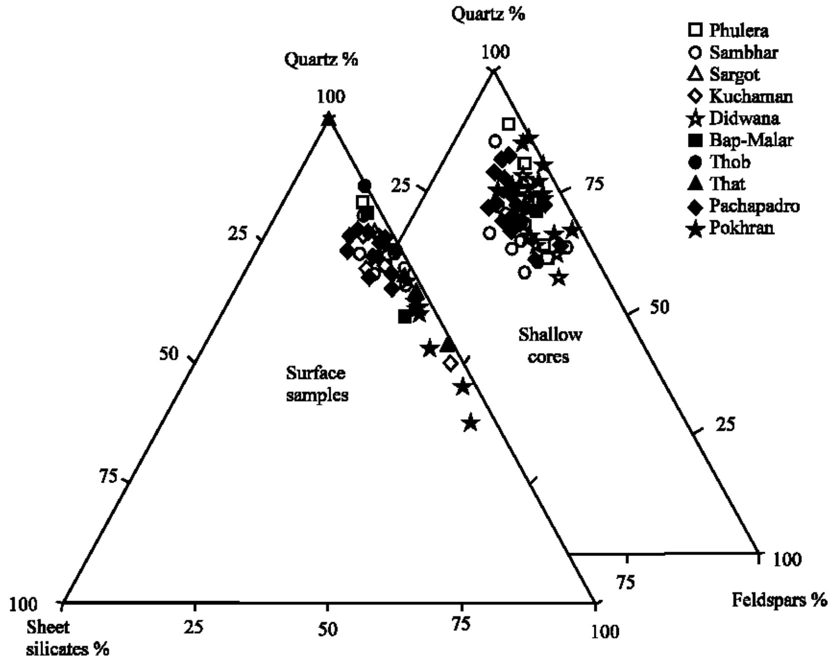

Fig. 3: Ternary diagram showing the distribution of quartz, feldspars and sheet-silicates in (a) playa surface sediments and (b) shallow core sediments

Sambhar

The Sambhar is the largest playa (ca. 225 km^2) of the Thar Desert, situated in the wind gaps of the Aravalli hills. The playa receives an annual rainfall of ca.500 mm and is fed by two ephemeral streams, the Mendha from north-east and the Rupangarh from south-west, apart from several rivulets. This playa is extensively used for commercial production of common salt, halite. The Quaternary sediments in the playa are >15 m thick spanning for more than 30 ka in age (Sinha *et al*., 2006). Much of our earlier work has been focused on this playa (Sinha and Raymahashay, 2000, 2004; Roy *et al*., 2001; Sinha and Smykatz-Kloss, 2003; Sinha *et al*., 2006; Roy *et al*., 2006, 2007; Roy, 2007) and a very comprehensive account of evaporite mineralogy is available.

Three shallow profiles correspond to three different geochemical zones in our earlier work (Roy *et al*., 2006) and show similar lithostratigraphical distribution (Fig. 2 shows the summarized log). These zones contain assemblages of carbonates, sulphates and chlorides (Table 2). But the total sulphate content is much lower in comparison to chlorides and carbonates. The bottommost litho unit (125-50 cm) consists of laminated calcareous mud. This unit shows a calcite-protodolomite-halite assemblage with minor amounts of thenardite/mirabilite and traces of anhydrite and polyhalite. A distinct enrichment of proto-dolomite is observed at 75 cm depth which might have formed due to dolomitization of a precursor calcite (Sinha and Smykatz-Kloss, 2003). This litho unit is similar to the bottommost litho unit (110-60 cm) of the Phulera playa. Considering the proximity of the Sambhar and Phulera playas, rate of sedimentation in both the playas can be assumed to be similar. This places the unit (125-50 cm) in to the age bracket of 1.5-3 ka, which is also supported by our AMS ^{14}C chronology from a deeper bore hole from Sambhar (Sinha *et al*., 2006).

The topmost litho unit (50 cm-surface) consists of moderately laminated silty-clay capped by a thin horizon of evaporite rich crust. In this unit, the abundance of proto-dolomite is much lower compared to calcite. Halite (NaCl) is the most abundant evaporite mineral in the surface sediments and thenardite is the main sulphate mineral, whereas mirabilite, polyhalite and anhydrite are present in traces (Table 1). A significant spatial variability in evaporite mineralogy exists across the playa surface and a zonal distribution as per relative solubility was reported in our earlier work (Roy *et al*., 2001). A distinctive feature of the evaporite mineralogy of the Sambhar playa is the presence of K-bearing evaporites such as polyhalite, carnallite and sylvite in the surface sediments. It is important to note that the Sambhar playa has a distinctly different mineralogy compared to the Phulera even though they are only a few kilometers apart. The Sambhar playa is enriched both in chlorides and carbonates but the Phulera is enriched only in carbonates (Table 1).

Kuchaman-Sargot

The Kuchaman and Sargot playas are located north of Sambhar playa. Both Kuchaman and Sargot receive ca. 400 mm of annual rainfall and are small playas with an area of 8.5 and 2.0 km^2, respectively. The Kuchaman-Sargot basin is fed mainly by the Palara river and some minor drainage trending east-west. In Kuchaman surface sediments, calcite is the most dominant along with traces of proto-dolomite and trona (Table 1). Thenardite occurs in significant quantities in some samples and anhydrite is recorded in traces. Kuchaman surface sediments are generally poor in halite but at least one sample recorded significant quantities of halite indicating that there may be some pockets (topographic lows) where the brine reaches halite saturation. On the other hand, halite is the major evaporite mineral present on the Sargot playa surface sediments (Table 1). Calcite is next in abundance. Anhydrite, glauberite and proto-dolomite are the other evaporites present in traces in the Sargot surface sediments. Gypsum is not present in the sediments of either of the playas. The main difference between Kuchaman and Sargot is that trona and thenardite are present only in Kuchaman but are absent in Sargot. Similarly, glauberite is present in Sargot but absent in Kuchaman.

Both these playas are completely desiccated with polygonal cracks on the surface. A hard lithified carbonate layer at 5-7 m depth in Kuchaman marks extreme aridity and a break in sedimentation (Rai and Sinha, 1990). The overlying dark colored clay with dolomite and feldspar represents the last phase of sedimentation in the playa which was eventually covered by aeolian sand.

Didwana

The oval-shaped Didwana playa lies in the semi arid (ca.330 mm of annual rainfall) part of western Rajasthan near the eastern edge of the Thar Desert dune fields, ca.50 km NW of the Sambhar playa. It is the second largest playa in the Thar Desert with an area of ca.13.5 km^2 and is commercially exploited for salt production. The playa is flanked by isolated hills of quartzite and slate to its SW margin and by longitudinal dunes to its NW margin. The playa is mainly fed by rain water during monsoons and there is no major drainage feeding the playa. The playa is filled with water during the rainy season but remains dry for most parts of the year

The litho units of the shallow depth profile (Fig. 2) correspond to the three geochemical zones of our earlier work (Roy *et al*., 2006). The bottommost litho unit (120-60 cm) consists of laminated calcareous mud and contains calcite as the major mineral (Table 2). Halite, thenardite and proto-dolomite are also present in significant quantities along with traces of anhydrite and thenardite. The intermediate litho unit (60-15 cm) consists of moderately laminated silty-clay and contains calcite and halite. Anhydrite and thenardite are present in traces and proto-dolomite is almost absent in this litho unit. The presence of anhydrite all along the profile may in fact be the dehydration product of gypsum. The uppermost litho unit (0-15 cm) consists of sandy silt and is enriched in halite. Thenardite is present in minor amounts and the presence of trona is restricted to surface sediments. The absence of K-bearing evaporites and presence of thenardite and trona in higher amount in surface sediments distinguishes the Didwana playa from the Sambhar.

A deeper profile (>6 m) at Didwana was studied by Wasson *et al.* (1984) and they clearly distinguished ‘surface evaporites’ (precipitated from shallow standing water) and ‘sub-surface evaporites’ (precipitated in pore spaces) based on their relationship to bedding and mud content. In addition, they reported a very complex evaporite, northupite ($MgCO_3.Na_2CO_3.NaCl$), from depths >3 m occurring in association with dolomite and conformable to the bedding (surface evaporite). The conventional radiocarbon chronology of Singh *et al.* (1974) and Wasson *et al.* (1984) put the sediments from the top 120 cm in the age bracket of last 3-4 ka.

Pachapadra and Thob

The Pachapadra playa lies ca.100 km NW of the city of Jodhpur with a catchment area of ca.82 km^2 (Deotare and Kajale, 1996). The playa occupies a much smaller area (ca.10 km^2) and receives an annual rainfall of 300 mm. It has been suggested that this playa formed due to disorganization of the old Luni river (Ghose *et al.*, 1966). The Thob is also a smaller playa (ca.11 km^2) lying ca.25 km NE of Pachapadra and is surrounded by the Aravalli mountains and aeolian sand dunes (Deotare and Kajale, 1996). Both Pachapadra and Thob have very similar surface mineralogy dominated by halite followed by calcite (Table 1). The playas have very low concentrations of gypsum/anhydrite and quite a few samples from Pachapadra show traces of proto-dolomite.

A shallow profile from the Pachapadra playa (Fig. 2) consist of alternate horizons of pale yellow to dark brown medium sand and dark brown sandy silt (Roy *et al.*, 2007). Minor fissures (desiccation cracks) filled with medium sand has been observed in the sandy silt horizons. Halite and calcite are present through out the shallow profile in comparable amounts (Table 2). Gypsum occurs in varying amounts in near surface sediments but its concentration increases significantly below 1.3 m with a corresponding decrease in calcite concentration. Deotare and Kajale (1996) and Kajale and Deotare (1997) also reported gypsum-rich horizons below 1 m depth at Thob playa associated with deep water facies.

Bap-Malar and That

The Bap-Malar playa is situated 140 km NW of Jodhpur, west of Aravalli ranges in the western margin of the Thar Desert. The playa occurs in the arid core of the Thar Desert, where the mean annual rainfall is 200 mm. The playa is primarily fed by the rainwater and groundwater and some minor ephemeral streams from the SW side. That is a small playa located SW of the Bap-Malar and this again has no influx from streams. It is fed mainly by summer rains (<200 mm). The surface mineralogy of both the Bap-Malar and That playas is halite-dominated followed by calcite (Table 1). That has a much lower carbonate fraction than the Bap-Malar and gypsum/anhydrite are present in traces in both the playas.

We do not have any new data from the shallow profiles at Bap-Malar but an earlier work from a 6 m thick section showed that gypsum is the major evaporite mineral below 1.2 m depth while calcite and halite occur in low amounts (Deotare *et al.*, 2004). Such sharp changes were interpreted to be climatically-induced.

Pokhran

The Pokhran is an elongated playa (ca.12 km^2), located in the western Rajasthan and receives an annual rainfall of 200 mm. The lacustrine sediments, variable in thickness from 2 to 5 m, overlie the sandstone basement, which are exposed in the north and northeastern side of the playa. In the eastern and western parts of the playa, there are outcrops of basalt and rhyolite (Rai, 1990). Similar to the Bap-Malar, the Pokhran playa is an arid core playa and halite is the most dominant evaporite mineral followed by calcite and traces of gypsum/anhydrite and proto-dolomite (Table 1).

Table 3: Concentration of CO_3, SO_4 and C_{org} (maximum in mass percent) in the Thar Desert playa sediments

Playa		CO_3	SO_4	C_{org}
Phulera	Surface	5.6	0.0	0.1
	Depth	19.4	0.1	0.7
Sambhar	Surface	5.7	0.6	1.7
	Depth	21.2	1.5	3.1
Didwana	Surface	4.1	2.6	0.1
	Depth	8.0	1.4	0.4
Pachapadra	Surface	8.2	0.2	0.2
	Depth	10.8	14.0	0.4
Pokhran	Surface	4.2	0.4	0.1
	Depth	11.9	26.3	0.1

The shallow depth profile at Pokhran (Fig. 2) is well constrained with 4 OSL ages. The bottommost litho unit (150-90 cm) consists of gritty and gravelly sand. This unit is enriched in calcite. Proto-dolomite, anhydrite and halite are present in traces. The assorted nature of the sediments suggests it to be high energy depositional event. So using linear extrapolation, this unit can be placed between 4.0 and ca.6.6 ka. The intermediate litho unit (90-50 cm) consists of moderately laminated silty-clay and is well constrained between 4.0 and 2.7 ka. It has a calcite-halite dominated assemblage with gypsum and anhydrite as traces (Table 2). The topmost unit (50 cm-surface) consists of sandy silt. A gypsum-rich lamina was recorded between 40 and 50 cm depth. It is devoid of calcite (Table 2) indicating its formation from evaporation of sulphate-rich standing water. The OSL dates place this horizon between 2.5 and 1.3 ka. The sediments younger than 1.3 ka are enriched in calcite. Anhydrite, gypsum and halite are present in traces. The surface mineralogy is halite dominated with traces of calcite, proto-dolomite, anhydrite and gypsum.

Geochemical Characteristics

Table 3 presents the summary of organic carbon (C_{org}), carbonate and sulphate contents in playa sediments. Among the eastern playas, Sambhar has the highest concentration of C_{org}, Phulera has intermediate and Didwana the lowest concentration. The surface sediments of Sambhar playa contains up to 1.7 mass percent of C_{org} and sediments in shallow profile up to 3.1 mass percent of C_{org}. The western playas contain very low contents of C_{org}. The inorganic geochemistry is very similar to that of carbonate and evaporite mineralogy. The sediments from Phulera and Sambhar exhibit 19.4 mass percent and 21.2 mass percent of CO_3, respectively. In the sediments of Pachapadra and Pokhran playas, the maximum CO_3 content are 10.8 and 11.9 mass percent, respectively. The trend is completely opposite when it comes to the sulphate contents. The sediments of western playas have higher concentration of SO_4. The shallow depth core sediments of the Pachapadra and Pokhran playas contain up to 14 and 26.3 mass percent of SO_4, respectively. In the eastern playas the SO_4 content varies between 0.1 and 1.5 mass percent.

We present here a synthesis of both the published (Roy *et al*., 2006) and new data on characteristic elemental ratios i.e. Na/Al, Sr/Ba and Zr/Al along the depth profiles to identify horizons of varying chemical weathering, aeolian activity and evaporation. Based on the elemental ratios, the shallow profiles of Phulera, Sambhar, Didwana, Pachapadra and Pokhran are divided into three different geochemical zones (I-III) (Fig. 4). As Na^+ is more soluble compared to Al^{3+} (hydrolysate), the higher values of Na/Al in the clastics indicate lower chemical weathering (sediment-water interaction) and vice versa (Mason and Moore, 1982; Nesbitt and Young, 1982; Sinha *et al*., 2006). On the other hand, the ratio of Zr/Al reflects the abundance of zircon to feldspars (Jones and Bowser, 1978; Tripathi and Rajamani, 1999). As zircon is mainly present in silt size fractions and easily transported by wind activity, the higher values of Zr/Al indicate continental input by aeolian activity in an arid environment. Similarly, Ba is associated with clastics and Sr with carbonate and sulphate minerals, so the higher values of Sr/Ba suggest higher evaporation and salinity. The geochemical zones

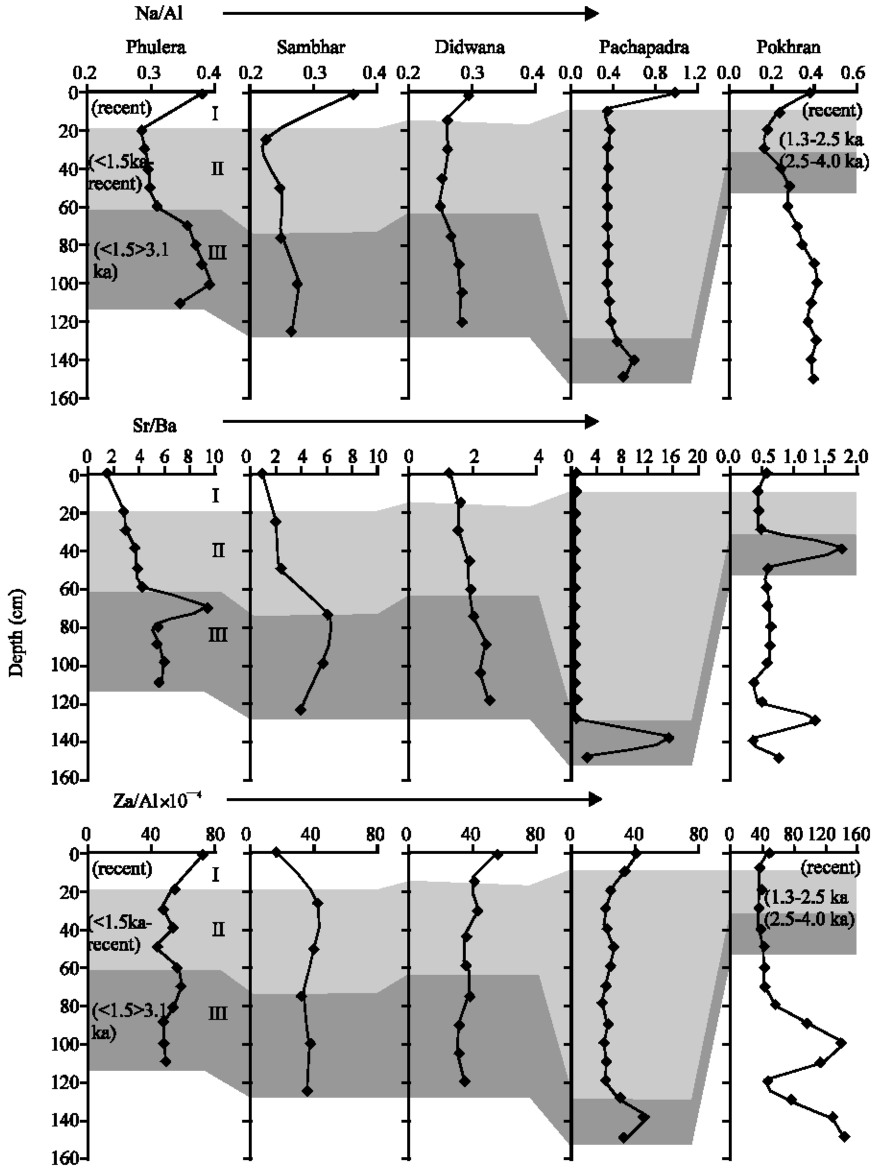

Fig. 4: Geochemical proxies (a) Na/Al (b) Sr/Ba and (c) Zr/Al×10^{-4} along the playa profiles

are comparable in the depth profiles of all the playas. The sub recent zone (I) and zone III show higher values of Na/Al compared to zone II. The ratio of Sr/Ba is highest in zone III. Except the Sambhar playa, all other profiles show high Zr/Al in zone I (Fig. 4).

DISCUSSION

Mineralogical Assemblage of Surface Sediments: Spatial Variation

The variation in the composition of evaporite mineralogy in playa sediments are controlled by the inflow composition (catchment lithology) and the sequential saturation of evaporites with the

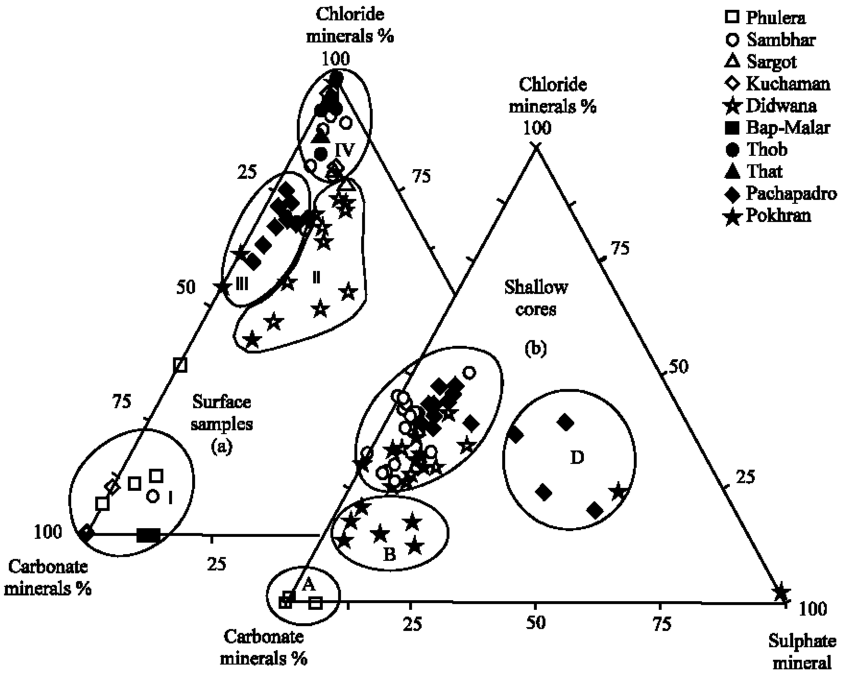

Fig. 5: (a) Distribution of carbonate, sulphate and chloride minerals in the surface sediments of the playas (Type I-Phulera; Type II-Didwana, Type III-Pachpadra, Type IV-Sambhar, That, Thob, Sargot-Kuchaman) and (b) Distribution of carbonate, sulphate and chloride minerals in the shallow core sediments (Type A-Phulera, Type B-Pokhran, Type C-Sambhar, Didwana, Pokhran, Type D-Pachpadra, Pokhran)

progressive evaporation of the brine following the principle of chemical divide (Eugster and Hardie, 1978; Ingebritsen and Sanford, 1998). The other factors which may also influence the evaporite mineralogy are sulphate reduction (Eugster and Jones, 1979; Decima *et al*., 1988; Burns *et al*., 2000; Rouchy *et al*., 2001; Yan *et al*., 2002), ground water leakage (Wood and Stanford, 1990) and clay mineral regradation i.e., reverse weathering of precursor clay minerals (MacKenzie and Garrels, 1966).

The surface sediments of the Thar Desert playas show varying assemblages and abundances of evaporite minerals. The relative concentrations of carbonate, sulphate and chloride minerals in the surface sediments have been renormalised and plotted as triangular plots in Fig. 5 (considering an error of 10-20% in the semi-quantitative data from the XRD charts). There are four distinct clusters of non-clastic mineral assemblages. Type I is the carbonate-rich assemblage represented by Phulera, Bap-Malar and Kuchaman playas. Type II is a mixed assemblage, represented by Didwana and shows the highest concentrations of sulphate minerals amongst all playas. Type III is a sulphate-poor assemblage and chlorides are relatively higher than carbonates. This assemblage is mainly represented by Pachapadra and Pokhran. Finally, the Sambhar, That, Thob and some samples from Sargot playas represent type IV assemblage characterised by dominance of chloride minerals (mainly halite) and much lower concentrations of both carbonates and sulphates.

The assemblages of clastic minerals (quartz, feldspars, amphiboles, garnets, micas and Fe-chlorites) and geological-geochemical studies suggest an igneous-metamorphic provenance for the playa sediments (Sinha and Raymahashay, 2004; Roy and Smykatz-Kloss, 2007). The exposures of silicate rocks (quartzite, granites, gneisses, schist and basalt) present in the playa catchments suggest that chemical weathering of these rocks provide HCO_3 rich inflow into the playa basins (Eugster and Hardie, 1978; Yan *et al*., 2002). We assume that the CO_2 saturated rainwater is the principal weathering agent and contributes cations i.e., Na, Ca, K, Mg and Fe, into the playa basins by the chemical weathering of the primary minerals (plagioclases, K-feldspars, amphiboles, micas and Fe-chlorites). Considering the similar assemblages and abundances of clastics present in playa sediments, it is obvious that the playa basins receive inflow with similar composition irrespective of their geographic locations. This is supported by the studies on petrography and REE geochemistry of playa sediments (Roy and Smykatz-Kloss, 2007). These rule out the effects of different catchment lithologies on the different assemblages of evaporite minerals in different playas. Apart from that the sulphate or chloride bearing rock types are absent in the playa surroundings. Both Cl and SO_4 could be contributed into the playa basins by the dissolution of the aerosol sea salts by rain water. So, we consider that the carbonates and evaporite minerals are authigenic and precipitated as a result of enhanced aridity in the region.

The relatively higher abundance of halite in the surface sediments (Table 1) can be attributed to the process of evaporative pumping of the sediment pore brine to the playa surface and its precipitation due to enhanced present day aridity (Yechieli and Ronen, 1997; Yechieli and Wood, 2002). But the relative concentration of halite with respect to other evaporites i.e., carbonates and sulphates, is extremely variable in different playas across the Thar Desert (Fig. 5). Several adjoining playas e.g., Sambhar, Phulera, Kuchaman and Sargot, show distinctly different abundances of evaporite mineralogy. Such small-scale variations, where influxes of cations, rainfall are generally similar, might be controlled by local hydrological and topographic conditions. Phulera is a much smaller playa compared to Sambhar and it receives very little runoff most of which evaporates in early summer much before the brine is concentrated to precipitate halite in significant quantities. The Kuchaman, although larger than Sargot, may drain off all its inflow through rainfall before peak evaporative conditions necessary for halite saturation. The Sargot, on the other hand, seems to be located in a topographic low where the brine stays at least in the near surface conditions to precipitate significant quantity of halite precipitation.

The presence of thenardite (Na_2SO_4) in the surface sediments of Phulera, Sambhar, Didwana and Kuchaman, glauberite ($CaSO_4.Na_2SO_4$) in Sargot and gypsum ($CaSO_4 . 2H_2O$) in Bap-Malar, That, Thob, Pachapdra and Pokhran suggest present day aridity and different mechanisms of sulphate removal from the brines of different playas (Table 1). Unlike gypsum, mirabilite ($Na_2SO_4 . 10 H_2O$), the precursor to thenardite, precipitates at a much lower temperature (4 to 6°C) (Eugster and Hardie, 1978; Cooke, 1981), suggesting that in the eastern playas a part of the SO_4 from the brine freezes out as mirabilite and later dehydrates to thenardite as a consequence of higher temperature in the region or seasonal variability in the water availability and temperature change. The minimum temperature of ca. 6°C during the winter months at the Thar Desert very well supports the mirabilite formation in the eastern region. During the monsoonal months a part of the mirabilite redissolves and reprecipitates as glauberite in the surface sediments of some of the eastern playas e.g., Sargot. The present day higher water availability in the eastern region is also reflected by the presence of H_2O-bearing evaporites, e.g., carnallite, polyhalite and trona (Table 1) in the Sambhar and Didwana playas. Compared to eastern region, the playas of western region receive less rain fall (200-300 mm a^{-1}) and do not retain inflow till the winter months. So the inflow coming into the playas dries out completely during the hot summer months forming gypsum.

Mineralogical Assemblage in Depth Profiles: Temporal Variation

A triangular plot for the shallow core sediments (Fig. 5) indicates a different clustering and a mixed assemblage of evaporite minerals. Phulera still represents a carbonate playa (Type A). A majority of samples from Pokhran playa fall in carbonate-rich type B. Type C represents a mixture of carbonates and chlorides with sulphate minerals less than 25% of the total. The playas such as Sambhar, Didwana, Pachapadra and some samples from Pokhran fall in this category. Type D represents mostly of Pachapadra samples and shows variable amounts of carbonates and chlorides with sulphates more than 25% of total. This plot clearly shows that the brine composition of all playas has undergone significant variation with time and two playas i.e., Pachapadra and Pokhran, show very large variations. Other playas have also varied in brine composition but the variation has been limited between carbonate and chloride fractions.

Along the shallow depth profiles (Table 2), halite is near-uniform and present as sub-surface precipitates. As halite is highly soluble, the occurrence of halite can be due to the percolation of Na and Cl rich hypersaline brine from playa surface into the shallow profiles and precipitation in the sediment pores. Again, non-stoichiometric Fe-bearing proto-dolomites are abundant and gypsum is absent in the shallow core sediments of eastern playas (Phulera, Sambhar and Didwana), whereas gypsum is present in variable abundances in the sediments of western (Pachapadra and Pokhran) playas. The formation of ferroan modern dolomites and absence of gypsum in Sabkha el Melah de Zarsis (Tunisia) (Perthuisot *et al*., 1990) and Lagoa Vermelha (Brazil) (Vasconcelos and Mckenzie, 1997) have been related to SO_4 reduction in an anoxic environment by microbial activity which contributes abundant HCO_3-for the precipitation of dolomites. Except Sambhar playa, the preservation of C_{org} is low in the sediments of eastern playas. This suggests an oxygenated environment of deposition in most of the Thar Desert playas. Although the geochemical process of SO_4 reduction can not be ruled out completely in Sambhar playa, the formation of proto-dolomites in the sediments of Phulera and Didwana shallow-surface sediments can not be explained by SO_4 reduction by microbial activity.

Wood and Sanford (1990) and Sanford and Wood (1991) demonstrated that the groundwater leakage (outflow to inflow ratio between 0.01-0.001) does not disturb the sequence at which dolomite, gypsum and halite are precipitated. Similarly, the process of clay mineral regradation (formation of illite and smectite at the expense of kaolinite and removal of K, Mg and Na from the brine) does not affect the precipitation of proto-dolomite and gypsum (Yan *et al*., 2002). So the presence of gypsum in shallow profiles of Pachapadra and Pokhran and its absence in eastern playas can alone be related to different extents of evaporation in two different margins of the desert. The OSL chronology of this work and comparison with radiocarbon chronology of Singh *et al*. (1974), Wasson *et al*. (1984) and Sinha *et al*. (2006) put the proto-dolomite enrich horizon of the eastern playas and gypsum enriched horizon of Pokhran playa in to the same age bracket (ca.1.3-3.1 ka). Considering that Pachapadra is located in a paleo-river bed, we assume that the sedimentation rate must have been higher at Pachapadra basin. So the gypsum horizon between 130 and 150 cm can be comparable to the proto-dolomite horizon of eastern playas.

Regional Paleoclimate and Paleolimnology

For the paleo-climatic and paleo-hydrological development of the region, mineralogy and geochemical proxies for chemical weathering, aeolian influx and evaporation are taken into consideration. During the deposition of zone III (ca.1.3-3.1), the higher values of Na/Al and higher Sr/Ba suggest low chemical weathering (sediment-water interaction) and higher evaporation in all the playas. This suggests a regional weak monsoonal rainfall. During this event, higher abundance of proto-dolomite in the eastern playas and gypsum in the western playas suggest that the eastern desert margin experienced evaporation only to the extent of carbonate precipitation, whereas the western playas experienced further evaporation till the precipitation of gypsum. But in an arid environment, proto-dolomites were formed by the dolomitisation-reprecipitation of earlier precipitated calcite and Mg enriched brine in the eastern playas. The minor SO_4 contents in eastern playa brines were removed by freezing out of mirabilite during winter months. This is reflected in the presence of minor amounts of mirabilite and its dehydration product, thenardite in the eastern profiles.

During the deposition of zone II (ca. 1.3 ka-present day), the relatively improved monsoon and humid condition is reflected by the lower Na/Al, Sr/Ba and Zr/Al and higher abundance of calcite. In the eastern playas, the abundance of proto-dolomite is low, whereas in western playas gypsum is present as traces. The modern conditions of low chemical weathering and high aeolian input are mirrored by the higher values of Na/Al and Zr/Al in the sub-recent zone I. This is also supported by the abundance of halite and presence of relatively unstable (easily weatherable) amphiboles. The absence of zircon in the modern sediments of Sambhar can be related to the location of the sampled profile in the playa centre, which might not have received any aeolian influx.

CONCLUSIONS

The playas of the Thar Desert show similar clastics, but varying assemblages of carbonate and evaporite minerals. After ruling out the effects of catchment lithology, reverse weathering, seepage loss and sulphate reduction, the varying non-clastic assemblages in the surface and shallow surface sediments is explained by the sequential precipitation of evaporite minerals as a results of topography, moisture availability, mechanisms of SO_4 removal and evaporation. The absence of gypsum ($CaSO_4 . 2H_2O$) in the late Holocene shallow core sediments of Phulera, Sambhar, Didwana playas and its presence in the sediments of Pokhran and Pachapadra suggest that during the late Holocene the eastern margin playas experienced relatively humid conditions compared to playas of western region. Between ca.1.3-3.1 ka, the geochemical proxies for chemical weathering and evaporation suggest regional weakening of monsoonal rainfall. During this arid event, proto-dolomites were precipitated as a result of dolomitisation of precursor calcite in the eastern playas and minor SO_4 were removed by precipitation of mirabilite. In the western region, the playa brines evaporated up to the precipitation of gypsum.

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