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Hydrochemical water evolution in the Aral Sea Basin. Part I: Unconfined groundwater of the Amu Darya Delta – Interactions with surface waters

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SUMMARY

The Aral Sea, which has been affected by lake level lowering of approximately 25 m and a salinity increase from 10 to >100 g/l since 1963, represents, along with the Amu Dary Delta a dynamic hydrological system under an arid climate regime. The system receives river water inflow at high seasonal and inter-annual variability from remote alpine source areas. In the Amu Darya Delta, there is a distinct salinity contrast between the low-salinity river water (~1 g/l) and the salinity of the unconfined GW (GW_{unconf}: 10–95 g/l). The GW_{unconf} levels are predominantly controlled by the seepage of the river water inflow and GW discharge into the shrinking Aral Sea.

In June 2009 and August 2009, we sampled water from various sources including surface waters, GW_{unconf}, lake water and soil leachates for chemical analyses. Evaporative enrichment, precipitation/dissolution of gypsum and precipitation of calcite drive the GW_{unconf} to an $NaCl(SO_4)$ water type presenting a positive correlation between Na and SO_4.

We model the hydrochemical evolution of the GW_{unconf} in a box model which considers the capillary rise of near-surface GW, the precipitation of minerals in the unsaturated horizon and the seasonal re-flushing of adhesive residual brines and soluble salts. The model documents a rapid increase in salinity over a few annual cycles. Furthermore, the model simulations demonstrate the importance of the aeolian redistribution of soluble salts on the hydrochemical GW evolution. In a lab experiment, halite, hexahydrite and starkeyite are precipitated during the late stages of evaporative enrichment from a representative local brine.

Processes specific to different water compartments plausibly explain the variations of selected element ratios. For example, the precipitation of low-Sr calcite in irrigation canals and natural river branches of the delta lowers Ca/Sr. The dissolution of gypsum in soils (Ca/Sr mole ratio ~ 150) and the possible precipitation of $SrSO_4$ associated with Sr-depletion in adhesive residual brines increases Ca/Sr in seepage and re-increases Ca/Sr in the unconfined GW. Aral Sea water, which receives high-Ca/Sr surface and groundwater inflow, developed due to continued precipitation of high-Ca/Sr calcite the almost lowest Ca/Sr ratio (~25) over time. We observed spatial variations in the GW_{unconf} composition: (i) ammonium levels increase strongly due to interaction with lake sediments rich in organic matter and (ii) distinct increases in levels of nitrate, U, Mo and Se locally reflect oxygenation when GW levels decrease. The Amu Darya Delta acts as a sink for boron (uptake via terrestrial vegetation) and a source for bromide (release by degradation of organically-bound Br). Our results concerning the hydrochemical evolution of the GW_{unconf} and additional data from the Aral Sea constrain the parameter 'GW discharge' in water budget models of the lake and improve the basis for palaeoclimatic interpretations of sediment records from the Aral Sea.

1. Introduction

Climatic- and anthropogenic-related variations in the river and groundwater (GW) discharge to the Aral Sea manifest in the lake level, volume and salinity of the closed lake. To date, few hydrological studies have addressed with the surface water/GW dynamics in the Amu Darya Delta (Shibuo et al., 2006; Alekseeva et al., 2009). Groundwater surface water interactions have been studied farther upstream at sites with active irrigation agriculture (Johansson et al., 2009; Scott et al., 2011). There is a high salinity contrast between saline unconfined GW (GW_{unconf}), low-salinity confined GW (Schettler et al., in press) and river water that recharges the GW_{unconf} in the delta (Johansson et al., 2009). A substantial

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increase in the salinity and lowering of the levels of the GW_{unconf} occurred after irrigation had been abandoned due to water shortage in the lower reaches of the Amu Darya (Fig. 4 in Johansson et al., 2009).

Although the Karakalpak Amelioration Administration regularly measures the levels and salinities of near-surface GW in irrigation areas, the hydrochemical evolution of the GW_{unconf} has never been investigated. For water budget models of the Aral Sea (Tshernenko, 1965, 1983; Poryadin et al., 1997; Jarsjö and Destouni, 2004; Alekseeva et al., 2009) and an improving irrigation management, knowledge on the hydrochemical evolution and composition of the GW discharging into the Aral Sea is a prerequisite.

Evaporation and seasonal dissolution/precipitation processes within the critical zone located between the GW_{unconf} level and the land surface (Feary et al., 2001; http://dels.nas.edu/Report/Conceptual-Models-Flow-Transport/10102) likely drive the salinity increase in non-irrigated areas. In addition to a simple salinity increase, we assume that these processes significantly influence the hydrochemical signatures of the GW_{unconf}, which we relate to the evolution of the GW_{unconf}. We demonstrate the importance of biogenically mediated processes (B, Br), of adsorption (Ba) and of changing redox conditions (U, Mo, Se). Our results are potentially applicable in other semiarid to arid alluvial basins that receive $Cl(SO_4,HCO_3)$-type freshwater inflow from remote (alpine) feeding regions. A study of the confined GW from Site III (Schettler et al., in press) completes the data presented here.

2. Aims, approach and assumptions

To understand the hydrochemical evolution of the GW_{unconf} in the Amu Darya Delta and the influence of the GW-inflow on the hydrochemical composition of the Aral Sea, we investigated the following compartments from Sites I to II (Fig. 1a–d): (i) surface water (river, irrigation canal, collector), (ii) GW_{unconf}, (iii) Aral Sea water, (iv) confined GW discharging on the surface, (v) water from evaporation ponds, (vi) soil/sediment samples representing the critical zone and (vii) aqueous leachates of these sediments. To characterise the critical zone, we collected additional information concerning (i) the element fractionation by leaching of soil/sediment profiles and (ii) the mineralogical and sedimentological characteristics by quantitative XRD and grain-size analysis.

Under the arid local conditions (low ratio between precipitation and potential evapotranspiration), the recharge of GW in the Amu Darya Delta must be mainly controlled by the seepage of river water. The high evaporation from the surface of the Aral Sea sustains a hydraulic gradient towards the lake. Steady-state conditions reflected by a constant lake volume become established only over long time ranges. The recent decline in river water inflow has resulted in a substantial lowering of the lake level, in a salinity increase of the lake water and in an increase in the GW discharge to the lake (Alekseeva et al., 2009). It is unclear how much inflow of river water and GW influx, have contributed to the current high solute content of the Aral Sea. It is unlikely that the hydraulic gradient towards the Aral Sea in the Amu Darya Delta can be inverted under the current hydrological conditions. The recharge of GW along the river continued even during westward directed flow (Aladin et al., 2005) of the Amu Darya in the past.

Currently, there are no back-simulations or forecasts of the effect of a decadal- or century-scale increase in river water inflow and its rapidness on the dynamics of the balance between the Aral Sea level and GW levels in the Amu Darya Delta. In the case of a high and rapid growth of river water inflow, the GW level rise may lag behind the lake level increase, which could force the infiltration of lake water into the GW_{unconf} aquifers of the delta. The solute contents of the GW aquifers in the delta may have partially originated from such former events. However, a synchronous lowering of the lake's DSC (dissolved salt content) should have restricted the potential solute export from the Aral Sea. To trace subsurface GW inflow from the Usturt Plateau into the Aral Sea (Oberhänsli et al., 2009) we studied lake water along several depth profiles and clarified the influence of lake-internal processes on the hydrochemistry of the lake.

For selected elements, we estimated the delta's role as sink or source. On-going abiotic and biotic processes in the critical zone were tracked, comparing river and canal waters with waters from collectors. Sodium and Cl, which are least influenced by adsorption, biogenic uptake and chemical precipitation, were used for normalisation purposes. We assumed that the river-suspended materials deposited in the delta have a relatively homogeneous mineralogical composition. Intercalations of coarser layers related to sand storms, strong flood events or the deflation of fine particles affects the capillary rise of water and the spatial distribution of seepage. Investigation of the soil/sediment profiles, therefore, included grain-size analyses. Capillary rise of GW leads to the dissolution/precipitation of minerals at the surface of soils and sediments or within the critical zone. Dissolved salts and saline adhesive waters are seasonally flushed into the GW_{unconf}. We used a box model to assess the significance of these processes for the hydrochemical evolution of GW_{unconf}. Data discussion further focuses on the behaviour of the large-ionic Ba, which is highly sensitive to adsorption.

In the dry season, salt particles from the soil surface, dry evaporation ponds and the margin of the shrinking Aral Sea are redistributed by wind (Ginzburg et al., 2010). The dissolution of these salts impacts the composition of the seasonal seepage and the hydrochemistry of the Aral Sea. To understand the potential salt succession we gradually evaporated brine from a salt pan at room temperature in Teflon beakers. Evaporative enrichment was accelerated with a dry nitrogen gas flow. Residual liquids were analysed at several steps of evaporation after adequate dilution of small liquid aliquots using ICP-AES. Salts were identified by XRD analysis using the ICDD (International Centre for Diffraction Data) library. For data presentation the residual volumes (V_r) were normalised to the initial volume of the brine (V_{ini}) by division and named reduction volume (V_n).

$$V_n = \frac{V_r}{V_{ini}} \tag{1}$$

Base on the chemical analyses of GW, sampled from monitoring wells along three transects south of the western basin of the Large Aral Sea (GW_{unconf} and two artesian wells), we acquired information on spatial variations in the hydrochemical composition of the GW_{unconf} in the Amu Darya Delta.

3. Geographic, geological and hydrologic settings

The Aral Sea Basin is a semi-arid to arid region west of the Tien Shan and the Pamir Mountain ranges. Deserts cover more than half of the basin. Between 1881 and 2006, the annual precipitation at the Muynak Meteorological Station ranged between 98 and 132 mm (Electronic Supplement [Suppl.], Fig. Ia and b, after Dukhovny et al., 2007 and references therein).

Two tributaries, Syr Darya and Amu Darya, discharge low-salinity water into the closed dissected Small and Large Aral Sea Basins, respectively. Over the past 50 years river inflow has decreased due to the expansion of irrigation, resulting in a lowering of the lake level by approximately 25 m (Létolle and Mainguet, 1996; Zavialov et al., 2003; Zavialov, 2005; Peneva et al., 2004). However, the discharge rates of the Amu Darya and Syr Darya already demonstrated considerable variations prior to the 1960s (Suppl., Table I, source: Asarin et al., 2010) and the salinity of the Aral Sea varied in re-