



Hydrogeochemical classification of deep formation waters



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ARTICLE INFO

Article history:

Available online 5 November 2014

Editorial handling by M. Kersten

ABSTRACT

Deep formation waters from Western Europe, Russia and North America are classified by the major water components (Ca^{2+} , Cl^- , Mg^{2+} , Na^+) and barium/sulphate ratios. The data are used to identify important hydrogeochemical processes (e.g., halite dissolution and albitisation) that lead to the different composition of formation waters. Two significant water types are identified: Na–Cl water and Na–Ca–Cl water. Furthermore, differences in formation water according to stratigraphical units are shown for deep reservoirs in the North German Basin and the North Sea. Based on the collected hydrogeochemical data, development trends are stated for the formation waters, and albitisation is favoured as the main process causing Ca enrichment.

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1. Introduction

There are several terms to describe deep fluids of the Earth's crust. Kharaka and Hanor (2007) list the following descriptions: oil-field brine, basinal brine, basinal water, and formation water. In our paper we decided to use the term “deep formation water”, because we focus on groundwater from deep formations in all kinds of geological units. The term brine refers to water salinity higher than 5%, but not all formation waters reach this salinity level.

Anthropogenic activities (e.g., shale gas extraction, CO_2 sequestration, geothermal energy production and deep waste disposal) can change the hydrogeochemical characteristics of deep formation water. To predict such changes, the natural processes in deep reservoirs should be known and the influence of anthropogenic impact should be evaluated.

Formation water in deep sedimentary basins can be classified according to depth, temperature, and salinity (e.g., Graf et al., 1966; Kharaka and Hanor, 2007). Most of the deep formation waters contain more dissolved solids than seawater and are unsuitable as human drinking water. The hydrogeochemical development of formation water has been discussed for a long time. It is widely accepted that deep aquifers are influenced by the meteoric water cycle and geochemical processes within the crust (Hebig et al., 2012). Similar hydrogeochemical signatures are found in deep formation waters on all continents and can be explained by

general geochemical processes within the deep reservoirs (Land, 1995). The main hydrogeochemical processes that are thought to be responsible for the hydrochemical composition of deep formation waters are:

1. Seawater evaporation

This process has been thoroughly investigated (e.g., Usiglio, 1849; Clarke, 1924). The steps in evaporation that lead to different brine compositions and mineral precipitations are understood and can be retraced through modelling (e.g., Eugster et al., 1980). Recently formed brines resulting from seawater evaporation can be observed in dry areas (e.g., Dead Sea, Kara Bogas Gol) and in salt mines (Fontes and Mantray, 1993a). Several authors have discussed seawater evaporation as a process leading to the composition of deep formation water (e.g., Lowenstein et al., 2003; Lowenstein and Timofeeff, 2008; Houston et al., 2011). Lowenstein and Timofeeff (2008) favour residual brines as the main contributor to deep formation water and also suggest that changes in the chemistry of ocean water (especially $\text{Ca}^{2+}/\text{SO}_4^{2-}$ ratios) can be responsible for the development of brines dominated by calcium chloride. Residual evaporation brines dominated by magnesium, sulphate and chloride seem to be only occasionally present in deep reservoirs (Tesmer et al., 2007).

2. Salt dissolution

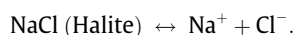
The dissolution of salt layers and salt domes will increase the salt concentration of meteoric and surface groundwater that is in contact with the salt deposits. Halite is the dominant precipitated

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salt in most geological units, and therefore halite will be dissolved, and halite dissolution leads mainly to the enrichment of sodium and chloride in deep formation waters. Mass balance scenarios considering the interaction of formation water with crustal rocks (Land, 1995) validate the conclusion that evaporites must have been deposited and destroyed at much greater rates during geological history than existing salt inventories suggest.

The dissociation of halite is chemically described as:

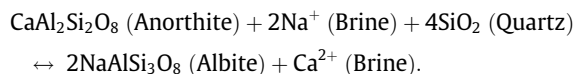


The amount of dissolved halite increases with rising temperature.

Bromide is another indicator for the dissolution of halite deposits (e.g., Land and Prezbindowski, 1981; Walter et al., 1990). Bromide cannot be included in the crystal lattice of halite, because of its larger ionic radius, therefore it enriches the residual solution during seawater evaporation. Formation water derived from the dissolution of halite will have the chloride/bromide ratio of halite. This leaching induces low bromide concentration, and the chloride/bromide ratio resulting from halite dissolution is much higher than the ratio developed during seawater evaporation.

3. Feldspar transformation

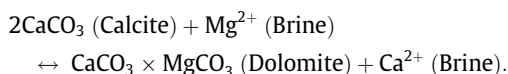
During this process Ca^{2+} ions are added to deep formation waters. The transformation of different feldspar minerals has been discussed already over many years (e.g., Davisson and Criss, 1996; Houston et al., 2011; Land, 1995; Pinneker, 1966; Spencer, 1987; Tesmer et al., 2007). Special focus has been given to the albitisation of anorthite in Na–Cl dominated brines summarised by the following reaction:



According to thermodynamic equilibrium conditions, this reaction favours Ca enrichment of formation waters.

4. Dolomitisation

The term dolomitisation refers to the replacement of calcite by dolomite. Dolomitisation is often explained as a reaction between brines and calcite. It leads to a loss of Mg as dolomite is formed and is summarised by the following chemical reaction:



The opposite process, dedolomitisation, caused by Ca-rich brines can also change the formation water composition and lead to an enrichment of Mg (Land and Prezbindowski, 1981; Stoessell et al., 1987).

There are several conceptual models for dolomitisation that Gasparrini (2003) has split into three main groups: evaporate and seepage-reflux models, fresh and seawater mixing models, and burial (subsurface) models. Machel (2004) argues for the use of the term dolomitisation only when $\text{CaMg}(\text{CO}_3)_2$ replaces CaCO_3 , and not for the primary precipitation of dolomite.

Further processes that are thought to influence deep formation water are membrane filtration or osmosis in clay layers, shales and siltstones (e.g., Graf, 1982; Hitchon et al., 1971; Kharaka and Hanor, 2007), diffusion (e.g., Ranganathan and Hanor, 1987), and freezing of saline water (e.g., Herut et al., 1990). The solubility of calcite at different temperatures and pressures has been well investigated (e.g., Coto et al., 2012; Duan and Li, 2008). Even considering the high CO_2 pressures within deep formations, calcite solubility does not induce the high Ca concentrations found in most formation waters, whereas the portion of HCO_3^- concentrations can be attributed to calcite solubility (Lehmann, 1974). Often the origin of the salt contents in groundwater cannot be explained by just one process. The saline groundwater of the Canadian Shield, for example, where no salt layers are found, is attributed to the inflow of brines from other strata, the leaching of salt from fluid inclusions and special mineral reactions (Frape and Fritz, 1987).

Table 1
Short characteristics of regions from which geochemical data were used for this study (occurrence of salt layers: from large deposits “+++” up to minor deposits “+” and no salt deposits “–”).

Region	Geological units from which water was mainly sampled	Presence of evaporite layers	Reference
<i>Western and Middle Europe</i>			
North German Basin	Carboniferous – Tertiary	+++ (Permian, Triassic) Permian salt domes often reach up to the surface.	Hesshaus et al. (2013) Kühn et al. (1998) Lüders et al. (2010) Regensburg et al. (2010) Schulz and GeotIS-Team (2009) Warren and Smalley (1994)
North Sea	Devonian – Eocene	+++ (Permian, Triassic) Permian salt domes often reach up to the surface.	
Paris Basin	Triassic – Tertiary	++ (Triassic: Muschelkalk and Keuper)	Fontes and Mantray (1993b)
<i>Russia</i>			
Southwest Ural	Cambrian – Permian	++ (Cambrian, Devonian, Permian)	Nosareva (2007)
Siberia	1. Precambrian – Ordovician	1. ++ (Cambrian)	Shouakar-Stash et al. (2007)
	2. Precambrian – Permian	2. ++	
	3. Cambrian – Tertiary	3. ++	
	4. Precambrian – Silurian	4. –	
Angara Lena Basin ^a	Precambrian – Ordovician	++ (Cambrian halite–carbonate deposits: 3000–4000 m thickness)	Pinneker (1966)
<i>America</i>			
Canadian Shield	Precambrian (crystalline rocks)	–	Frape and Fritz (1987)
Western Canada (Alberta)	Devonian – Cretaceous	++ (Devonian)	Hitchon et al. (1971)
Illinois Basin	Silurian – Devonian	+ (Gypsum and anhydrite)	Stueber and Walter (1991)
Palo Duro Basin	Carboniferous – Triassic	+++	Knauth (1988)
Mississippi Salt Dome Basin	Jurassic – Cretaceous	+++	Kharaka et al. (1987)
Texas Gulf Coast	Jurassic – Miocene	+++	Kharaka et al. (1977)
Gulf of Mexico	Jurassic – Miocene	+++ (Jurassic)	Houston et al. (2011)

^a The dataset of Pinneker differs from the data given in Shouakar-Stash et al. (2007).

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