The importance of river water inflow and shelf sediment-sea water exchange in the East Siberian Sea for the Nd isotopic composition of Arctic Ocean water

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With data generated from cruises to the Canada Basin in 2000, to the Eurasian and Central Arctic Ocean basins in 2001, to the Fram Strait in 2002 and to the Chuckchi Sea in 2005 we now have a good general view of the distribution and isotopic composition of Nd (ε_{Nd}) in the Arctic Ocean [1, 2]. The restricted Arctic Ocean basin is surrounded by large continental shelves, covering more than 50% of its total area.

Distinct from other oceans, with surface water Nd depletion, there is throughout the Arctic a pattern of high Nd concentrations, up to 58pM, at the surface that gradually diminish with depth to 15-18pM in the deep waters. A range of isotopic variations across the Arctic and within individual depth profiles reflects the different sources of waters. The dominant source of water and Nd is the Atlantic (ε_{Nd}= -10.7). Radiogenic isotope Nd signatures can be traced in Pacific water flowing into the Canada Basin and further into the Eurasian Basin (up to ε_{Nd}= -6.5). The variation of ε_{Nd} and concentration in the Arctic Ocean suggest that Nd input from rivers and shelf sediments is also of great importance.

Improving our understanding of the vast Siberian Shelves influence on Nd and trace element behaviour in the Arctic Ocean was one of the main objectives of the International Siberian Shelf Study 2008 (ISSS-08). The ISSS-08 cruise recovered filtered water (<0.2µm), particles and sediments from the Laptev and East Siberian Seas as well as estuarine and river water from Lena, Indigirka and Kolyma. Crucial processes, including loss of river water Nd in the estuarine region and shelf sediment-sea water exchange will be discussed in terms of controlling the Nd concentration and isotopic composition of sea water.

[1] Andersson *et al.* (2008) *GCA* **72**, 2854-2867. [2] Porcelli *et al.* (2009, in press) *GCA.*

A methodology to estimate Pitzer interaction parameters

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Most of the geochemical aqueous speciation models of electrolytic systems are based on the “ion pairing and complexing approach”, only suitable for aqueous solutions with ionic strengths up to about 0.8 eq/kgw. For highly saline solutions, an approach based on “specific-ion interactions” (named “Pitzer approach”) provides more accurate results.

Various thermodynamic databases supporting the “Pitzer approach” exist but they are often inconsistent and poorly documented. The number of chemical elements is limited because of the difficulties to measure relevant parameters for large ranges of temperature, ionic strength and pH. These limitations prevent from satisfying applications of the Pitzer formalism to simulate the geochemical behaviour of solutions and gas-brine-rock interactions in deep reservoir conditions. Consequently, databases have to be continuously updated and improved to better represent these geochemical processes.

This study presents the development of a new methodology for determining Pitzer interaction parameters. This methodology is based on the use of PEST [1], a nonlinear parameter estimation and optimization package, coupled to the PHREEQC code [2]. PEST takes control of PHREEQC, running it as many times as necessary while adjusting the Pitzer interaction parameters. The fitting procedure stops when the discrepancies between selected model outputs and a complementary set of field or laboratory measurements are reduced to a minimum.

This methodology was applied with success to different saline solutions (NaCl, KCl, HCl…). The computations were performed using measurements of water activities and mineral solubilities. All these calculations demonstrate that, if the number of measured data is sufficiently important, Pitzer interaction parameters can be determined for temperatures ranging between 25 and 300°C. Over the good fitting of calculation results with measured data, the smooth and monotonous variations of the interaction parameters values with temperature confirm the consistent adjustment of the determined parameters, allowing extrapolation potential to higher temperatures. This methodology to determine Pitzer interaction parameters is also applied to divalent salts and ternary interaction coefficients.

[1] Doherty (2004) *User Manual*: 5th Edition. [2] Parkhurst & Appelo (1999) *Wat. Res. Investigations Report* 99-4259.