

## Natural occurrences of liquid CO<sub>2</sub> emissions in the Okinawa Trough: Geological setting and variations of gas composition

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Three out of four sites known worldwide where liquid CO<sub>2</sub> is accumulated at – and/or emanating from – the seafloor, are located in the Okinawa Trough, South China Sea, west of Japan. All liquid CO<sub>2</sub> seep sites known so far are situated in Back-Arc hydrothermal systems in water depths between ~ 1200 and 1700 m. Recently, these occurrences have reached interest for applied marine environmental sciences as “natural laboratories” to evaluate the potential reactions of the marine system to deliberate storage of manmade liquefied CO<sub>2</sub> in the deep sea and deep-sea sediments, i.e., carbon capture and storage (CCS).

The expedition SO 196 of the German RV SONNE in March, 2008, visited two of these sites, the Hatoma Knoll and Yonaguni Knoll IV area, the latter being the working area of several of the presentations of this session.

We describe the geological setting of liquid CO<sub>2</sub> emissions in the Okinawa Trough. The condensed CO<sub>2</sub> is generated by complex phase separation processes of the hydrothermal fluids, with various additional components present, in particular CH<sub>4</sub> and H<sub>2</sub>S. Data from Hatoma Knoll show distinct variations in gas composition at different vent sites on small spatial scale. We evaluate the phase transition and transport processes of the liquid CO<sub>2</sub>-rich phase which may cause these variations, and emphasize the great potential as well as some caveats of the use of natural CO<sub>2</sub>-emitting sites as analogues for future deliberate marine carbon storage scenarios.

## Tl isotope constraints on the origin of the Earth's Pb paradox

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The Earth's Pb isotope systematics have been the subject of debate since Patterson's pioneering work provided the first accurate estimate of the age of the solar system [1]. A particularly contentious issue is the origin of the radiogenic <sup>206</sup>Pb/<sup>204</sup>Pb and <sup>207</sup>Pb/<sup>204</sup>Pb isotope ratios of the Earth's mantle. It is unlikely that this signature is solely due to the accretion of the Earth from volatile depleted matter. The radiogenic Pb isotope composition of the bulk silicate Earth (BSE) is thus thought to reflect U/Pb fractionations that occurred during the accretion, differentiation or chemical evolution of the Earth. Recent partitioning experiments that have investigated the behavior of Pb during metal/sulfide-silicate segregation at high pressures and temperatures have yielded conflicting results, however.

Lagos *et al.* [2] contend that Pb is not sufficiently siderophile and chalcophile to account for the high U/Pb ratio of the BSE. In this case, an alternative mechanism of Pb depletion is required and it has been suggested that this may involve degassing of Pb to a hot silicate atmosphere, which was subsequently lost during the giant impact. Such a scenario can be evaluated with the aid of the extinct <sup>205</sup>Pb-<sup>205</sup>Tl decay system [3] in the context of current models of terrestrial accretion [4]. Studies of volcanic systems furthermore demonstrate that Tl is significantly more volatile during volcanic processes than Pb. Taken together, the data indicate that loss of Pb by degassing cannot have played a major role in establishing the U/Pb ratio of the BSE.

Other recent partitioning studies have shown that both Pb and Tl are moderately siderophile at high pressures and temperatures [5, 6]. Accretion models that apply such behavior can readily account for the Pb and Tl isotope systematics of the BSE, without requiring late volatile loss of Pb. At present, such a scenario thus provides the most reasonable solution to the terrestrial Pb paradox.

[1] Patterson (1956) *GCA* **10**, 230. [2] Lagos *et al.* (2008) *Nature* **456**, 89. [3] Nielsen *et al.* (2006) *GCA* **70**, 2643. [4] Halliday (2008) *Phil Trans R Soc A* **366**, 4163. [5] Wood *et al.* (2008) *EPSL* **269**, 326. [6] Baker *et al.* (2008) *GCA* **72**, A46.