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# Noble gas anomalies related to high-intensity methane gas seeps in the Black Sea

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#### Abstract

Dissolved noble gases and tritium were analyzed at a series of high-intensity methane gas seeps in the Black Sea to study the transport and gas exchange induced by bubble-streams in the water column. These processes affect marine methane emissions to the atmosphere and are therefore relevant to climate warming. The seep areas investigated are located in the Dnepr paleo-delta, west of Crimea, and in the Sorokin Trough mud volcano area, south-east of Crimea. Noble gas concentration profiles at active seep sites revealed prominent anomalies compared to reference profiles that are unaffected by outgassing. Supersaturations of the light noble gases helium and neon observed relatively close to the sea floor are interpreted as effects of gas exchange between the water and the rising bubbles. Depletions of the heavy noble gases argon, krypton and xenon that were detected above an active, bubble-releasing mud volcano appear to be related to the injection of fluids depleted in noble gases that undergo vertical transport in the water column due to small density differences. In both cases, the noble gas anomalies clearly document seep-specific processes which are difficult to detect by other methods. Helium is generally enriched in the deep water of the Black Sea due to terrigenic input. Although exceptionally high helium concentrations observed in one seep area indicate a locally elevated helium flux, most of the seeps studied seem to be negligible sources of terrigenic helium. Noble gas analyses of sediment pore waters from the vicinity of a mud volcano showed large vertical gradients in helium concentrations. The helium isotope signature of the pore waters points to a crustal origin for helium, whereas the deep water of the Black Sea also contains a small mantle-type component.

Keywords: noble gases; tritium; terrigenic helium; methane gas seeps; mud volcanoes; plumes; bubble-streams; depletion; gas-exchange; Black Sea

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#### 1. Introduction

Methane (CH<sub>4</sub>) gas seeps in marine and lacustrine environments are currently of scientific interest due to their potential influence on global carbon cycles and climate warming (e.g. Judd, 2004; Walter et al., 2006).

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Gas release from seeps and rising bubbles in the water column can be detected by hydroacoustic systems (e.g. Hornafius et al., 1999; Artemov, 2006; Naudts et al., 2006; Greinert et al., 2006). Using this technology, marine CH<sub>4</sub> gas seeps have been documented worldwide, but detailed studies of gas exchange between rising CH<sub>4</sub> bubbles and the water column are rare (e.g. Leifer et al., 2000; Leifer and Patro, 2002; Clark et al., 2003; McGinnis et al., 2006a) and are commonly restricted to shallow seeps. Hence, more information on the processes controlling gas/water partitioning during the rising of CH<sub>4</sub> bubbles in the open water column is essential to better quantify the contribution of marine seeps to CH<sub>4</sub> in the atmosphere. We present noble gas analyses from different active seep sites that give new insights into CH<sub>4</sub> transport in the Black Sea.

In the Black Sea, intense gas seepage has been observed on the northern shelf and slope, as well as from mud volcanoes on the abyssal plain (Fig. 1). Within the EC-funded project CRIMEA ("Contribution of high-intensity gas seeps to the methane emission to the atmosphere", http://www.crimea-info.org/), numerous seeps releasing gas bubbles into the Black Sea were characterized using various oceanographic and geochemical techniques to evaluate the potential effects of

these 'high-intensity gas seeps' on the atmosphere (Schmale et al., 2005; Kourtidis et al., 2006). The Black Sea is a unique environment in which to study marine CH<sub>4</sub> emissions because the water column is permanently stratified, with anoxic conditions and strong CH<sub>4</sub> accumulation below the chemocline at 100-150 m depth (e.g. Reeburgh et al., 1991). CH<sub>4</sub> concentrations in the isolated deep water body of the Black Sea reach values of up to  $\sim 12 \,\mu\text{M}$ . The majority of the CH<sub>4</sub> seeps observed during the CRIMEA cruises are situated at water depths shallower than 725 m. This depth limit corresponds to the upper boundary of the stability zone for pure methane hydrates at the ambient temperature and salinity conditions prevailing in the Black Sea (Naudts et al., 2006). Hence it appears that the seepage of CH<sub>4</sub> bubbles from the sediments is inhibited by the formation of gas hydrate layers in the sediment. Additionally, several gas-emitting mud volcanoes were studied south-east of the Crimea peninsula at about 2000 m water depth. These deep seeps occur within the gas hydrate stability zone. The mud volcano structures seem to provide migration pathways where gaseous or dissolved CH<sub>4</sub> may be released from the sediments without being trapped in the gas-hydrates. Indications of CH<sub>4</sub> release and the presence of gas-hydrates have

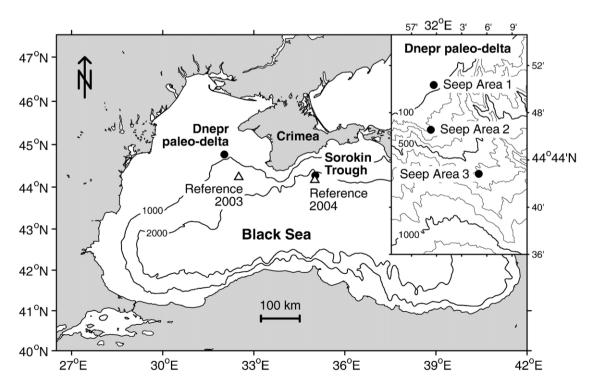


Fig. 1. Map of the Black Sea showing the study sites. Our work focused on two areas of intense gas seepage (●): the Dnepr paleo-delta region and the Sorokin Trough. The inset shows a detailed map of the Dnepr paleo-delta with the locations of Seep Areas 1–3. The deep seep site in the Sorokin Trough (Vodyanitskiy mud volcano) and two reference sites (△, no influence of gas seepage) are indicated in the main panel.

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