High concentration of adsorbed hydrocarbon gas in authigenic carbonate

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1.Introduction

Hydrocarbon gases in the marine sediments are important energy source. While, methane that dominates in the hydrocarbon gases is also an effective greenhouse gas when emitted in the atmosphere. Thus, to clarify the processes of both migration and concentration of the gases in sediments is important.

There are several storage forms of the hydrocarbon gases in the marine sediments. These are termed as dissolved, free gases in the interstices of sediments, and gases bound or adsorbed in the either organics or minerals in sediments. In these forms, the adsorbed gases defined as liberated gases by adding acid to sediments have not been received much attention, though the methane concentrations of the gases are often much higher than those of the other storage forms. Furthermore, how and where the adsorbed gases are preserved within the sediment matrix is still uncertain. Brekke et al. (1997) assumed that the adsorbed gases are strongly associated with the authigenic carbonates within sediments. On the other hand, Knies et al. (2004) assumed that the adsorbed gases are trapped within meso-pores (less than 10 nm diameter) of clay minerals within sediments.

In this study, we found that significant amount of hydrocarbon gases were liberated by adding acid to authigenic carbonate concretions as in the case of the adsorbed gases in sediments. If the gases were the same with those adsorbed in sediments in general, our observation would evidence the hypothesis that the adsorbed gases are strongly associated with the authigenic carbonates in sediments. Thus, we compared the concentrations, molecular compositions, and carbon isotopic compositions of the hydrocarbon gases liberated from the authigenic carbonate concretions with those of the ordinary adsorbed hydrocarbon gases in the surrounding sediments, and argued their origin and storage mechanism in the authigenic carbonate concretions.

2.Sample

In 1998, Ocean Drilling Program (ODP) Leg 186 targeted the Japan Trench forearc offshore Honshu Island, and two holes, Sites 1150 and 1151, were drilled. At both sites, several authigenic carbonate concretions were recovered. The X-ray diffraction analysis data indicated that these are predominantly dolomitic. The extractions of hydrocarbon gases from authignic carbonates were attempted by using these authigenic carbonate samples collected on board, four samples at Site 1150 and eight samples at Site 1151. Besides, adsorbed hydrocarbon gases were extracted from nine sediment samples at Site 1150 and eleven sediment samples at Site 1151, that were retrieved on board for monitoring of light hydrocarbon gases by using the standard headspace sampling method described by Kvenvolden and McDonald (1986).

3.Result and discussion

The carbon isotopic compositions of methane in the gases liberated from the carbonate samples and the adsorbed gases in the sediments showed analogous trend with depth; the isotopic compositions increase from -80 to -60 (permil VPDB) with increasing depth. Also, C1/C2 ratios of the gases liberated from the carbonate samples were analogous to those of the adsorbed gases in the sediments (ranged from 75 to 3300). The chemical characteristics of the gases liberated from the carbonate samples coincide well with those of the adsorbed gases in the surrounding sediments. We can, therefore, conclude that the liberated gases from the carbonate samples were the adsorbed gases.

The concentrations of methane adsorbed in the carbonate samples (291-4,527 nmol/g) were 10-100 times higher than those of the adsorbed methane in the sediment samples (5-1,304 nmol/g), while those in the headspace gases were orders of magnitudes lower than those of the adsorbed gases in the sediment samples. The enrichment of the adsorbed hydrocarbon gases in the carbonate samples supports the hypothesis that the adsorbed hydrocarbon gases are strongly associated with authigenic carbonates.