

Origin, migration and degradation of gases from mud volcanoes and surface seeps in the Niigata Prefecture

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Gases were sampled from 2 mud volcanoes (Kamo, Hachikoku) and 5 surface gas seeps (Kurokawa, Shibata, Niitsu, Myohoji, Osawa) in the Niigata Prefecture. Based on the gas compositions and carbon isotope compositions, origin, migration and degradation of gases are studied. The gas from the paddy field in Shibata is interpreted to be microbial, the gas from Myohoji is thermogenic, and the gas from Kurokawa is interpreted to be mixture of microbial and thermogenic gases. The gases from the mud volcanoes of Kamo and Hachikoku, and surface seeps from Niitsu and Osawa show heavy methane carbon isotopic compositions, heavier than -50 ‰, suggesting thermogenic origin but hydrocarbon ratios $C_1/(C_2 + C_3)$ are more than 100 higher than ordinary thermogenic gases. These gases are interpreted to have changed only the hydrocarbon ratios during the migration of thermogenic gases to the surface. Another explanation is possible that gases of microbial origin have changed their methane carbon isotope compositions to heavier values due to oxidative decomposition of methane by bacteria. The third possibility is that the gases are generated by methanogenic archaea reducing carbon dioxide generated by bacterial decomposition of crude oils. This process is called secondary methane generation. In Kamo, Hachikoku and Niitsu, the relatively large differences in carbon isotope composition of methane and carbon dioxide (about -50 to -65‰) indicate that the secondary methane production is possibly active. On the other hand, in Osawa and Shibata, the difference between methane and carbon dioxide isotopic composition is small, suggesting the methane could be oxidized. Based on the high $C_1/(C_2 + C_3)$, the primary origin of gas in Osawa is possibly microbial. The relationships between methane and ethane carbon isotope compositions suggest that the gas from Shibata consists of almost pure microbial methane. Based on the relationships between ethane and propane carbon isotope compositions, propane in gas samples from Hachikoku, Kamo and Niitsu is microbially degraded. The maturities of thermogenic components are estimated to be 0.9% in equivalent vitrinite reflectance (R_o) in Kurokawa and Hachikoku, 1% in Myohoji, 1.1 - 1.2% in Niitsu, and 1.6% in Kamo.

Water samples associated with the gas samples were taken from the mud volcano in Kamo and Hachikoku, and the surface seep in Osawa. Based on the ionic compositions of water and oxygen and hydrogen isotope compositions, the water in Kamo and Osawa is interpreted to be almost surface waters. Heavy oxygen isotope compositions in Hachikoku water suggest the seawater origin, but the chloride ion concentration is about one third of seawater. This water could be diluted with interlayer water of clay minerals.

Keywords: Mud volcano, Carbon isotope composition, Surface gas seep