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UV-Raman spectroscopic study of new gas hydrates collected from the Marmara sea.

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Sediments on continental margins hold enormous quantities of low molecular weight hydrocarbons as free gas, dissolved gas or gas hydrates. Fluid expulsions may have implications for structural geology, natural hazards, climate change or petroleum geology. The MARNAUT cruise (2007) in the Marmara sea was a multidisciplinary project with objectives centered on the relationships between active faults, fluid emissions and landslides. Numerous fluids escapes were observed in relation with active faults and the deep origin of these fluids is attested by a relatively high concentration of hydrocarbons (ethane, propane, iso-butane...) mixed with methane. Moreover, new gas hydrates specimens associated with these complex fluids were collected during the MARNAUT cruise at low water depth and relatively high temperature. These new samples are analyzed by UV-Raman spectroscopy and exhibit a clathrate structure II (sII), composed of 16 pentagonal dodecahedron cavities (a 12-face polyhedron constituted by regular pentagons, noted 5^{12}) and 8 hexakaidecahedron cavities (constituted by 12 pentagonal faces and 4 hexagonal faces, noted $5^{12}6^4$). The Raman analysis reveals the enclathration of guest molecules into various cages. These results will be discussed in comparison with other gas hydrates collected from African and Norwegian margins which exhibit a preponderance of clathrate structure I (sI), composed of two pentagonal dodecahedron cavities and six tetrakaidecahedron cavities (a 14-face

polyhedron having 12 regular pentagons and two regular hexagons, noted $5^{12}6^{2}$). The presence of type I structure and high methane concentration (98 %) rather indicates a microbial origin for methane in hydrates from African and Norwegian margins. In contrast, additional components (ethane, propane, iso-butane or carbon dioxide) identified spectroscopically in the samples from the Marmara sea suggest a thermogenic origin. Evidences exist that the origin of fluids from the Marmara sea and the presence of other hydrocarbons mixed with methane have a compositional effect on the relative cage occupancy of the methane.