



## Investigation of gas seepages in Thessaloniki mud volcano in the Mediterranean Sea

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### ABSTRACT

Gas seepages are commonly observed in marine environment. Especially, gas seepages due to anthropogenic gas hydrate dissociation are big concerns recently. In the Eastern Mediterranean Sea, Thessaloniki mud volcano was detected. Gas hydrate stability conditions in this mud volcano is very fragile. For this reason, in this study, gas seepages were predicted by using HydrateResSim at different seafloor temperature increments varying from 1 to 5 °C and different sediment permeability values varying from 0.1 mD to 5mD in Thessaloniki mud volcano. Both the effect of temperature increment (above 1 °C increment) and the increase in permeability cause faster gas hydrate dissociation. The gas seepages on the seafloor of Thessaloniki mud volcano was investigated in this study by using the bubble rise theory. It was found that the effect of gas bubble diameter is high on the height of gas flare in the study area. The effect of permeability and lithology near seafloor on gas release after gas hydrate dissociation is huge. Generally, in Thessaloniki MV, clays are dominant so it is advantageous for environment because even if gas hydrate dissociates, free gas reaches to the seafloor slowly. Moreover, since the next 100 years, it is expected that temperature will increase by + 2 °C on the seafloor of Thessaloniki MV. It was indicated that gas release will be obvious if temperature increment is above 1 °C in this area according to the numerical simulations with HydrateResSim.

### 1. Introduction

There are two confident things acquainted about the quantity of methane (CH<sub>4</sub>) hydrates worldwide; there are many CH<sub>4</sub> hydrate reservoirs and there is a lot of uncertainty about exactly how much (Gabbitto and Tsouris, 2010). At present, the opinions about the estimates continue to differ over several classes of magnitude; generating uncertainties about not only the volume of gas hydrates but also for its possible role in ongoing climate change (Boswell and Collett, 2011). Prior to 1998, the resources of hydrates were often declared much greater than all fossil fuels, while in 2000, the USGS reduced drastically its estimates to a level that hydrate accumulations may only antagonize the known reserves of conventional gas (Laherrere, 2000). Lee and Holder (2001) supported that the amounts of gas hydrates under the ocean and beneath Arctic permafrost represent an estimated of 53% of all fossil fuel (coal, oil and natural gas) reserves on Earth. From the standpoint of locations, the maximum amount of gas hydrates is located in continental margins of Alaska, Peru, Chile, Japan, Argentina, Indonesia, Taiwan and Gulf of Oman up to 157 kg/m<sup>2</sup> stored gas hydrates while Foskolos et al. (2011) supposed that the region of Mediterranean

and especially below the island of Crete in Hellas, in an area covered close to 200 kg/m<sup>2</sup> is found the maximum amount of gas hydrates. It is obvious that the gas hydrate inventory varies remarkably according to different investigations in the literature; nevertheless, the amount is still very large (Hester and Brewer, 2009).

Gas hydrates are crystalline compounds formed from water and suitable sized gas molecules. Depending on which gas molecules are present, hydrates form different crystal structures. Cubic structure I (sI), structure II (sII) and hexagonal structure H (sH) are the three common gas hydrate structures (Sloan and Koh, 2008). Structure I hydrate has two types of cavity: a small pentagonal dodecahedral cavity consisting of 12 pentagonal rings of water and a large tetrakaidhedral cavity consisting of 12 pentagonal and two hexagonal rings of water. The unit cell consists of water molecules ( $2 \times 5^{12} + 6 \times 5^{12} 6^2 = 46$ ). Structure II hydrate also has two cavity sizes, the pentagonal dodecahedral cavity and larger hexakaidecahedral cavity consisting of 12 pentagonal and four hexagonal rings of water. The unit cell consists of water molecules ( $16 \times 5^{12} + 8 \times 5^{12} 6^4 = 136$ ) (Nixdorf, 1996). Structure H hydrate, it has hexagonal crystals having three pentagonal dodecahedron cavities, two small irregular dodecahedron cavities, one large icosahedron

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cavity, and 34 water molecules per unit cell. The unit cell consists of water molecules ( $3 \times 5^{12} + 2 \times 4^3 5^6 6^3 + 1 \times 5^{12} 6^8 = 34$ ) (Carroll, 2009).

The type of gas hydrate structure depends on the molecular size of guest molecules. Approximately 130 compounds form hydrates. For example, methane (CH<sub>4</sub>), ethane (C<sub>2</sub>H<sub>6</sub>), carbon dioxide (CO<sub>2</sub>), and hydrogen sulfide (H<sub>2</sub>S) form sI hydrate. Propane (C<sub>3</sub>H<sub>8</sub>), nitrogen (N<sub>2</sub>) and i-butane (i-C<sub>4</sub>H<sub>10</sub>) form sII hydrate. I-pentane and other larger hydrocarbons form sH hydrate with help gas such as CH<sub>4</sub>, (Giavarini and Hester, 2011). Hydrates can be considered as a huge source of natural gas, because one cubic foot of solid gas hydrates contains an amount of gas which is 150–170 times higher (Gudmundsson, 1995). Gas hydrates are widespread in sea sediments hundreds of meters below the seafloor along the continental margins, as well as in Arctic permafrost (Collett et al., 2009). United States Geological Survey (USGS) predicted that about 27% of land regions and 90% of marine areas are favorable for the formation of gas hydrates. On the other side, an estimation of 99% of gas hydrates occurs in the sediments of continental margins at saturations as high as 20%–80% in some lithology; the remaining 1% mostly is associated with sediments in the beneath areas of high altitude, continuous permafrost (Kvenvolden and Lorenson, 2001). Most of GH reservoirs are deposited in marine sediments (at least 95%) and permafrost regions (less than 5%) according to Max and Johnson (2016). The presence of gas hydrates in marine sediments is stunted to the depth interval where the temperature and pressure of the sediment is into the thermodynamic stability zone, called the gas hydrate stability zone (GHSZ). Below GHSZ, gas hydrate equilibrium conditions are not satisfied so in porous space, there are free gas and free water (Chatterjee et al., 2016; Kumar et al., 2016).

Although the most importance is given on gas hydrates about only production, it is quite significant to comprehend gas hydrates for environmental purposes as well (Lu, 2015; Sain, 2017; Kondori et al., 2017). The large volumes of CH<sub>4</sub> contained in gas hydrate reservoirs and the dependence of their stability on temperature and pressure indicate that the fluctuations in the quantity of gas hydrate inventory could give alterations in atmospheric greenhouse gas levels, and influence the Earth's climate (Römer, 2011). Early studies focused on the role of melting methane hydrates in quaternary 'glacial to interglacial' transitions which methane can be rapidly released from decomposing (high-latitude) hydrate reservoirs after a small triggering with subsequent temperature increase (Nisbet, 1990; Loehle, 1993). Besides temperature variations in the deep ocean, the ocean circulation also promotes the methane release from gas hydrate (Thomsen et al., 2012). In any, natural or anthropogenic release of methane gas from hydrate during production tests or during drilling, completion and stimulation activities in marine environment is also a crucial disquiet as far as it concerns the sea floor instability due to changes of hydrate volumes and morphologies of the sediment (Zhao et al., 2017).

Climate studies have become augmentingly significant the last years and concentrate on the impact of Late Pleistocene to synchronous climate concern on the stability of methane hydrate deposits. The target must be firstly to comprehend how much and if any, methane hydrate dissociates on Earth and how this dissociation; influences the process of climate change and secondly there should be an evaluation of the quantity of methane that would reach to atmosphere from such degassing (USGS, 2018). A significant premise in precipitation of gas hydrates is the supersaturation of hydrate forming gases in the sediment pore water (Kashchiev and Firoozabadi, 2002) which is a function of pressure, temperature, and dissolved ions (Masoudi and Tohidi, 2005; Sun and Duan, 2007). The formation of gas hydrate will take place when the concentration of the former (CH<sub>4</sub>) surpass solubility. The lack of hydrates in shallow marine sediments within the GHSZ can accrue from elopement of CH<sub>4</sub> into the ocean, which is much under-saturated with CH<sub>4</sub> (Klapp, 2009).

Gas hydrates are stable at low temperature (from -5 °C to 20 °C) and high pressure conditions (from 1 MPa to 30 MPa). At least 95% of gas

hydrates are deposited in marine environment and the rest is deposited in permafrost environment. The sources of gas hydrates in marine sediments might be biogenic gas, thermogenic gas or mixed of them. Generally, methane (CH<sub>4</sub>) concentration is dominant in gas hydrates and it ranges from 90% to 99% (other impurities such as ethane (C<sub>2</sub>H<sub>6</sub>), propane (C<sub>3</sub>H<sub>8</sub>), n-butane (C<sub>4</sub>H<sub>10</sub>), carbon dioxide (CO<sub>2</sub>), etc.) (Arora et al., 2015).

Understanding the reasons of the fluid seepages in marine environment is essential for environmental studies. Moreover, the pathway of these fluid seepages is important. It is crucial to determine how much of these seepages are dissolved and consumed in seawater before releasing to the atmosphere. Recently, there is an argument that anthropogenic effects (i.e. global warming) cause the dissociation of gas hydrates near seafloor (Anka et al., 2012; Johnson, 2016). It was proposed by some scientists that huge amount of CH<sub>4</sub> might be released to the atmosphere due to the dissociation of gas hydrates widely distributed in marine and permafrost environments caused by global warming (IPCC, 2001; Bohannon, 2008, Krey et al., 2009, Mascarelli, 2009). Although only small portion of released CH<sub>4</sub> is expected to reach to the atmosphere, it is considered that still it may cause additional greenhouse effect and endanger sea life (Archer et al., 2009). However, the uncertainty in gas in-place calculations in gas hydrates is a big problem because this causes huge variations on the expected CH<sub>4</sub> release to the atmosphere in future (Ruppel et al., 2011). According to IPCC (2007), even if 0.1% (approximately 3 standards trillion cubic (tcm) of CH<sub>4</sub>) of gas hydrates in the world is released to the atmosphere, CH<sub>4</sub> concentration in the atmosphere is expected to increase from 1774 parts per billion (ppb) in 2005–2900 ppb. This will fasten climate change because CH<sub>4</sub> is ~20 times more potent than CO<sub>2</sub> as a greenhouse gas (Lelieveld et al., 1998; Ruppel et al., 2011). Kretschmer et al. (2016) proposed that with the current global warming projectile, gas hydrates in marine slopes are likely to dissociate next 100 years. Compared to pressure alteration, temperature alteration is much more important for gas hydrate stability so the effect of sea level changes is small compared to temperature increase (Tishchenko et al., 2005). If gas hydrate dissociates and releases huge amount of CH<sub>4</sub> to the atmosphere within next 100 years, this will augment atmospheric greenhouse gas and this will defy the dissociation of further gas hydrates due to global warming. This is defined as the climate hydrate feedback loop as occurred in the past. (Kvenvolden, 1998; Mestdagha et al., 2017).

Fig. 1 gives the timescale of gas hydrate dissociation and the history of gas seepages. As seen in Fig. 1, gas hydrate might dissociate between next 100 and 10<sup>4</sup> years. The oxidation of CH<sub>4</sub> in the atmosphere might take approximately 100 years. If huge amount of CH<sub>4</sub> is released to the atmosphere, it will be very dangerous for the environment. Therefore, it is essential to make prediction about the gas hydrate dissociation and gas seepage characteristics. Moreover, it is important to predict how much of CH<sub>4</sub> reaches to the atmosphere. As shown in Fig. 1, the released CH<sub>4</sub> in seafloor will not reach to sea level immediately. Firstly, high portion of CH<sub>4</sub> is oxidized and dissolved in seawater (Mestdagha et al., 2017).

In the past, vast CH<sub>4</sub> hydrate dissociation occurred during Quaternary climate change (Kennett et al., 2003; Krey et al., 2009). According to Praeg et al. (2011), the seafloor temperature of the Mediterranean Sea was 4 °C cooler (better for GHSZ- 25% thicker) during glacial stages. Similar impacts were also observed in the Black Sea, where temperature was 2–5.5 °C cooler but with the increase of temperature, 15–60% of gas hydrates in the Black Sea dissociated (Poort et al., 2005). Hence, climate changes due to human activities may cause the dissociation of huge amount of gas hydrates in future (Krey et al., 2009). Krey et al. (2009) calculated the global distribution of the ensemble mean trend in seafloor temperatures for next 100 years. According to Krey et al. (2009), in the Mediterranean Sea, the seafloor temperature increment might be 2 °C within next 100 years.

When gas hydrate is out of gas hydrate equilibrium conditions, all CH<sub>4</sub> will not suddenly reach to the atmosphere. This is a very slow

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