New Approach on Salt and Methane Generation*

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Search and Discovery Article #51391 (2017)**
Posted June 26, 2017

*Adapted from extended abstract based on oral presentation given at 2016 AAPG International Conference & Exhibition, Barcelona Spain, April 3-6, 2016. Please see closely related article, <u>"Parallel Salt and Methane Generation, Possible Paradigm Shifts for Salt Generation in Deep Sea Processes"</u>, Search and Discovery article #51392. **Datapages © 2017 Serial rights given by author. For all other rights contact author directly.

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Abstract

Salt generation has been considered solely attributable to and rendered by the theory of evaporation. Since salt basins overlap some of the most productive gas provinces, this presentation aims at a new approach on salt and methane generation. It highlights the need to reconsider partially the classical approach to salt and methane generation on new observations. The existence of actual deep marine hypersaline anoxic basins (DHAB) is a well-documented fact. The analogy between the recent and ancient DHABs allows us to presume that methane has been generated by euryhaline bacteria. Hence we presume that the non-crystalized, over-pressured, buried salty brine is the material appropriate to trap and host methane. This viscous, gas-saturated brine can be an engine for diapir formation prior to the crystalline phase. This new approach may launch debate and will require further consideration as a probable alternative for salt and parallel methane generation, coupled with salt diapir formation in particular salt basins.

Introduction

The first firedamp explosion occurred on 9 September 1664; it was recorded in Hallstatt (Austria), in a salt mine, not in a coal mine (Harris 1908). Prior to the installation of a proper ventilation system, the so-called "pentinents" were responsible for burning off the methane that would accumulate in the ceilings of the mine chambers from Wieliczka Salt Mine. In the Szolotvina Salt Mine (Aknaszlatina, today in Ukraine), the methane from salt was used for illumination (Wanek, 2008) in the early 19th century. Nowadays salt mines are equipped with intensive ventilation to prevent similar firedamp explosions.

In the classical view, even the name "evaporites" shows that salt is considered as a product of evaporation: dry-out of sedimentary basins, as stated by Van't Hoff (Arrhenius and Lachman, 2003). Further evaporitic minerals, such as gypsum, potash salt, etc. result from this phenomenon (Clarke, 1920). It is also well known that numerous hydrocarbon accumulations are linked to salt basins. For successful

exploration, it is crucial to understand the deposition and generation of the evaporites, and how the subsequent deformation history can influence salt basin petroleum systems (Thomas, 2008).

The structural dynamics of salt systems is tackled in rich theoretical and applied science literature (Jackson, 1994), yet the theory describing how salt bodies may have been generated has not been unambiguously challenged to date; Arrhenius and Lachman (2003) point out that no large-scale salt deposits are being formed under current geological conditions.

Evidence

Deep Hypersaline Anoxic Basins/Lakes (DHAB)

The aim of the MEDRIFF (Mediterranean Ridge Fluid Flow) Project granted by the EU within MASTIII Programme was bathymetric mapping of the Mediterranean Sea (MedRiff Consortium, 1995). In the eighties the research was developed by several cruises organized by Italian researchers with the co-operation of European and non-European colleagues. The research cruises of MEDRIFF produced the discovery of new hypersaline anoxic basins, namely Tyro Basin (1983), Bannock Basin (1984), Urania, L'Atalante Basin, and Discovery Basin (1993-1994).

Further, several similar brines were identified worldwide: Gulf of Mexico (Orca Basin, Brine Pool NR-1/GC233), Red Sea (Conrad-; Oceanographer-; Shaban-; Kebrit-; Nereus-; Wando-; Albatross-; Atlantis II-; Chain-; Discovery-; Shagara-; Valdivia-; Erba-; Port Sudan-; Suakin Deep) (Antunes et al., 2011).

The first outstanding feature to spot these brines was a sonic response in the course of bathymetric sonic surveying of the Mediterranean domain. On several deep zones (~3000-4000 m), the bottom presented a "flat" reflection. A second important feature was high viscosity of the brine inasmuch as the "flat" reflection surface can and does support the deposited fine-grained fractions (Corselli et al., 1995).

The MEDRIFF project covered various physical parameters measuring conductivity along the total seawater profile. With Discovery Basin they found abruptly increasing conductivity on brine surface (Corselli et al., 1995) from 40 to 125 mS/cm. In the same basin, the salinity abruptly increases at the same depth from 40 ppt to 125 ppt. In consequence, the salinity and the conductivity properties present strong regression all over the profile, resulting in a superficial tension between normal sea water and salt brine.

Along physical properties, biological characteristics were also identified. The methanogenesis in the brines was described in several articles; Borin et al. (2008) mention that methanogenesis greatly exceeds sulphate reduction in most of the saline layers in Urania DHAB, where extremely high bacterial abundance varies from layer to layer, labelled as "hot-spot of microbial activity".

For the DHABs from the Mediterranean range, Karisiddaiah (2000) published high methane concentrations varying between 128-2692 x 10³ nM (2.048-43.072 mg/L). In Lake Madee (Eastern Mediterranean), an intensive and stratified bacterial life was identified (Yakimov et al., 2013). Furthermore, besides the bacterial phylogeny, a detailed chemical composition is documented, where a daily biogenic methane production rate and acetate content is displayed.

Discussion

Since the generation of DHAB is evidence, the following immediate questions occur:

- 1. Why does diffusion not work over that "flat" reflection mentioned by Corselli et al. (1995)?
- 2. How is that surface formed? What is the role of it?
- 3. Can this phenomenon occur in geological time scale?

Membrane polarisation and superficial tension

In our view, it is this that happens: at a certain depth, the depositing argillaceous particles (of negative electric charge) will generate membrane polarisation. This creates a potential field in which the positive ions (Ca²+ and Mg²+) will be forced to migrate, hence causing the increased conductivity detected. This occurs due to the coagulation and flocculation of the argillaceous particles in the presence of the Ca²+ and Mg²+ ions, a process also met in the article "Relative factors influencing membrane filtration effects in geologic environments" by Frederick A.F. Berry (1969).

The membrane generated by this polarisation will function as an osmotic, semipermeable membrane, allowing only certain ions to pass through. Considering the above mentioned depth (~3000 m) and pressure (>300 atm [~4,500 psi]), the phenomenon will be one of a reverse osmotic process. This facilitates the solvent, freshwater to pass through this membrane. Meanwhile, ions will remain behind, thus increasing the concentration. This is how brine will be generated, reaching high salinity and increased viscosity. The diffusion between normal sea water and brine is blocked by the superficial tension created by membrane polarisation.

Buried DHAB

This viscous brine will be able to support the deposited sediments above, as mentioned by MedRiff Consortium (1995). Thinking in geological time, huge amounts of DHAB might be buried and isolated in a sedimentary basin. These hidden, over-pressured brines of high salinity and viscosity could preserve their properties for geological ages, meanwhile supporting huge sediment sequences above them.

The Methane Content of the DHAB

If we acknowledge methane content of current DHABs to be a fact (Karisiddaiah, 2000; MYakimov et.al. 2013), then substantial biogenic methane content is probable to be trapped in the buried brines (\sim 40 mg/L). Neither can the outstanding acetate concentration be neglected, since acetate decomposes into methane and water: (CH₃–COOH⁻ = CH₄ + H₂O), presenting a different source for yielding methane in brine (\sim 500 μ mol/L = \sim 30 mg/L).

Thus, the primary bacterial \sim 40 mg/L and the secondary acetate decomposition \sim 30 mg/L will increase the aggregated methane concentration to \sim 70 mg/L in buried brine. Upon the volume of the buried brine, this concentration facilitates estimating the biogenic methane quantity

generated in brine. As a consequence, these brines holding methane can serve as sources for hydrocarbon accumulation (whether primary or secondary, it is to be confirmed by further research).

New Hypotheses Relating to Diapir Formation

Once the over-pressured, methane-containing salt brine is formed in a deep hypersaline anoxic basin (DHAB) and it is covered by a considerable sediment package, it will bear the weight of the deposits. If this initial equilibrium is lost due to an event, i.e. earthquake, this (geological scale) load over the viscous, non-crystalized brine will generate diapirs. The shock of energy forces the brine to penetrate the sedimentary package. The over-pressured, ascending brine injects the gas and water into the reservoir packages. It is this de-gasification and dehydration that starts salt crystallization meanwhile preserving the folds generated during the penetration and ascending movement. During this process xenoliths form in salt bodies by incorporating foreign bodies, rock fragments, from the covering deposits.

Conclusions

- The current scientific knowledge about recent deep marine processes and phenomena described thereof, especially the evidence on Deep Hypersaline Anoxic Basins/Lakes, steered us to formulate a new hypothesis on salt and biogenic methane generation.
- It is well-known that normal salinity and diffused sea water properties change at a particular depth, creating abrupt increase in conductivity and salinity concentration. It is the phenomenon of membrane polarisation that is responsible for the increase of conductivity taking place between the argillaceous ions of negative charge and the positive Ca²⁺ and Mg²⁺ ions.
- Membrane polarisation creates a semipermeable membrane, and generates, at a specific pressure, the reverse osmotic process. This is a phenomenon responsible for the increase of the salinity underneath, generating the DHABs. The semipermeable membrane is responsible for the superficial tension between normal sea water and brine, serving as a physical barrier against diffusion.
- The oversaturated, viscous brine is an ideal environment both for the halophile bacteria, i.e. methane generating bacteria, and for acetate decomposition (CH4 \sim 40 mg/l bacterial + \sim 30 mg/l Acetate).
- The viscous brine surface can support the deposited sediments. This is why the brines can be isolated and buried in geological times, generating an overpressure-buried, non-crystalized body with high methane and acetate concentration (\sim 70 mg/l).
- The over-pressured brine with methane content, buried in geological time, is exposed to the impact of various tectonic events, causing the trapped methane and water to release into the porous reservoirs; as a result, the crystalline salt diapirs are formed.

Acknowledgements

This work was supported by Sandhill Petroleum BV., and Oil & Gas Development (OGD) Ltd. Hungary. We express special thanks to Pete Nolan (senior consultant) and Abhi Manerikar (Exploration Director at OGD), Steve Pearson (vice-president New Venture and Exploration Operations) for permanent conversation and encouraging support, debates and essential expert advice. We express our gratitude to the colleagues for the critical opinion on listening to our new hypotheses.

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NEW APPROACH on SALT and METHANE GENERATION

by Zoltán UNGER & David LeCLAIR

University of West Hungary & Oil&Gas Development Ltd. Hungary

waste water recycling

- reverse osmosis
- coagulation and flocculation
- membrane filtration, superficial tension
- microbial activity

GEOLOGICAL TIME SCALE

Sedimentation processes + overburdening + over pressuring

+ dehidratation and degasification

the result can be **SALT & CH₄**

"To this day, we have better maps of Venus, Mars, and the far-side of the moon than we do of much of underwater [America]".

Robert D. Ballard

Classical view on salt formation:

The name "evaporite" implies that salt is presumed a product of evaporation i.e. drying out of sedimentary basins

(Ochsenius&Van't Hoff theory in Arrhenius & Lachman, 2003).

Experiment: Saint Francisco Bay, California table salt production;

First precipitation: CALCITE [CaCO₃] 75% water evaporated

Second precipitation: GYPSUM [Ca SO₄ * 2H₂O] 90% water evaporated

Third precipitation: HALITE [NaCl] 96-97% water evaporated

Fourth precipitation: **SYLVITE** [KCI]

http://omp.gso.uri.edu/ompweb/

We are presenting a new approach to salt and methane generation Common knowledge: salt basins overlap productive gas provinces;

For successful exploration, it is crucial to understand:

- the deposition and generation of "evaporites",
- how the subsequent deformation history
 can influence salt basin petroleum systems

(Thomas, 2008).

The structural dynamics of salt systems are tackled in both theoretical and

applied science literature yet the theory describing

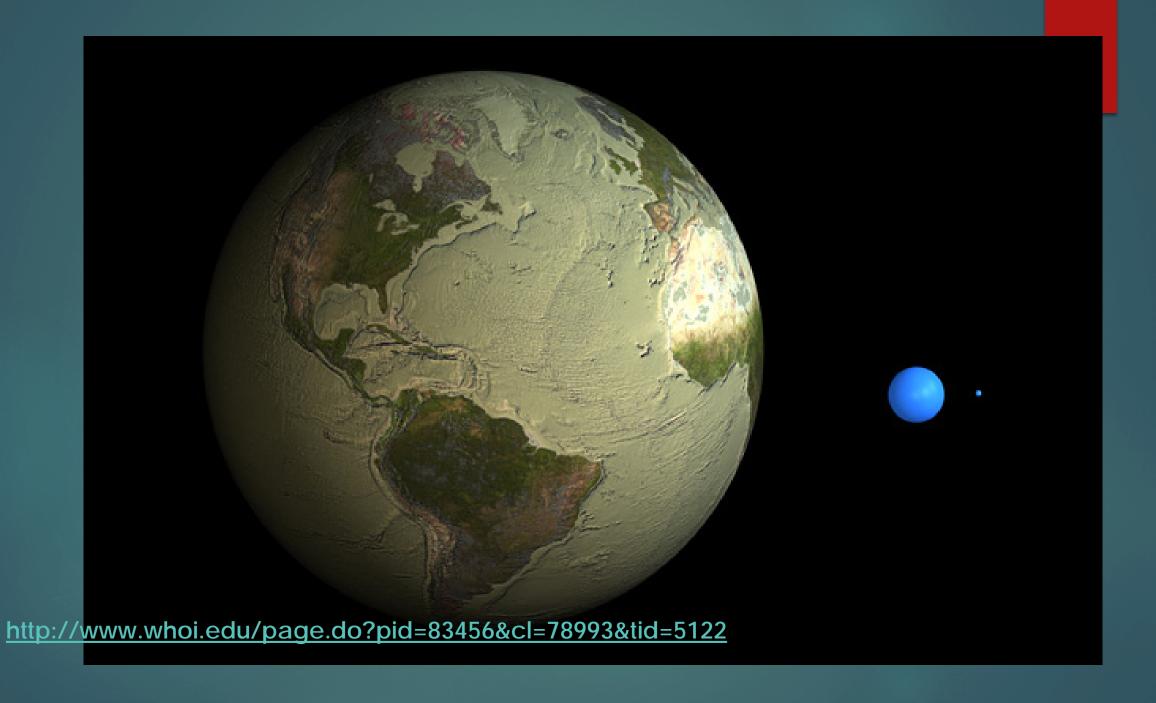
how salt bodies may have been generated

has not been unambiguously challenged to date.

(Trusheim 1961; Jackson, 1994),

Arrhenius & Lachman (2003): <u>no large-scale salt deposits</u> are being formed under current geological conditions.

NB: We may have to look for recent processes, environments to explain this enormous salt generation.



Contradictions in salt evaporation processes:

- spatial identity: spread of the salt deposits and spread of deep marine sediments;
- lack of erosion-products, no fossils in the desiccated sea/lake;
- 100 feet (30.48m) sea water produces 2 feet (0.61m) salt;
- folding of the salt independent from deformation and diagenesis of the covering rocks;

The well-known theory of Ochsenius

(i.e. the model of drying out of Kara Bugas bay)

for salt generation in great masses

should be doubted

What kind of climate can produce such evaporation and when in geological record?

- Messinian crisis?

Evaporitic age: 13.6-13.4 Ma = > was no longer than 0.4 Ma.

(Parathetys range)

(Gautier et. al. 1994)

Then, where is the salt coming from?

Evidences:

• 1980s: research of Mediterranean Sea bottom:

DHAB-s = Deep Hypersaline Anoxic Basin

- 1983: Tyro Basin;
- 1984: Bannock Basin;
- 1993: Urania- and L'Atalante Basin,
- 1993-94: Discovery Basin

Earth Science

Salty lakes hide deep in Mediterranean

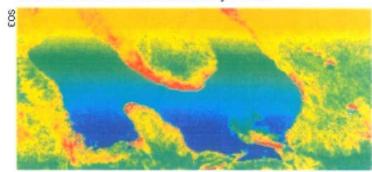
Geoscientists have discovered three lakes of extremely salty brine lying in pockets on the floor of the Mediterranean Sea, southwest of the island of Crete. Such lakes could help explain why the Mediterranean is saltier than typical ocean water.

The three brine lakes, situated more than 3,300 meters below the sea surface, were discovered in 1993 and 1994 by a team of European scientists conducting the Mediterranean Ridge Fluid Flow (MEDRIFF) project. Oceanographers had previously identified two other brine lakes in the Mediterranean during expeditions a decade ago.

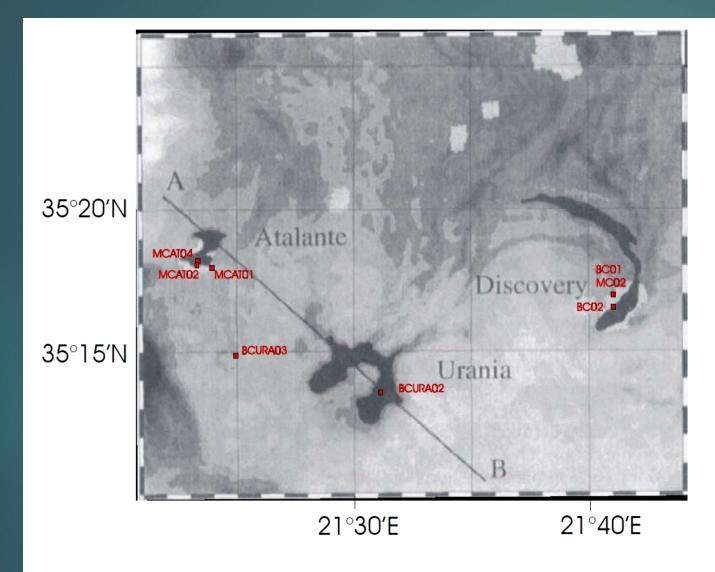
MEDRIFF researchers found the lakes using echo sonar, which sends down pulses of sound to probe the seafloor. The boundary between the dense brine fluids and the normal seawater above creates a flat lake surface that stands out amid the rougher surrounding seafloor. The density contrast prevents the brine from mixing easily with the seawater.

The scientists focused attention on a horseshoe-shaped lake, called Urania, which had an average depth of 80 meters. At one end, the lake reached 200 meters deep. The chloride content in the Urania brine measured about 5 times that of Mediterranean seawater. The lakes formed when seawater dissolved deposits of salt-rich rocks along the seafloor.

If many such lakes exist at the bottom of the Mediterranean, they could add salt to the seawater, perhaps explaining the Mediterranean's unusually high salt content, say the scientists. Current theories hold that high rates of evaporation produce the Mediterranean's extra salty waters.



Sonograph shows smooth surface of brine lake (in blue).





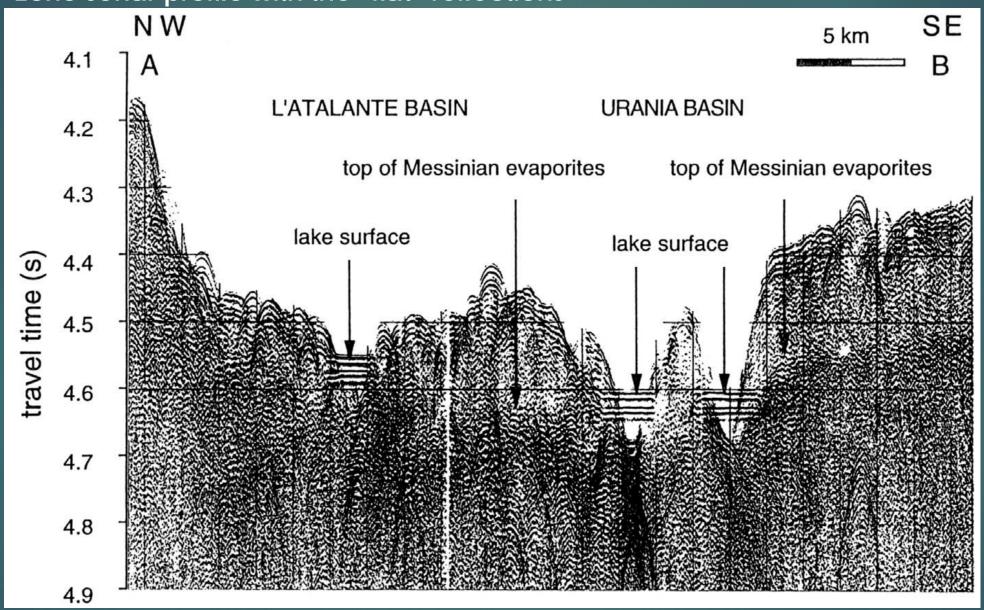
23°E

25°E

22°E

Map from MEDRIFF Consortium, in: Eos, Trans. AGU, 76, 313, 1995

Echo sonar profile with the "flat" reflections



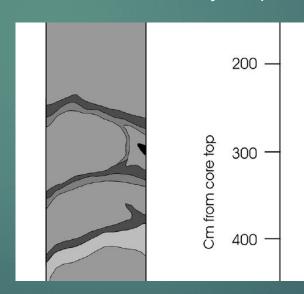
Brine characteristics:

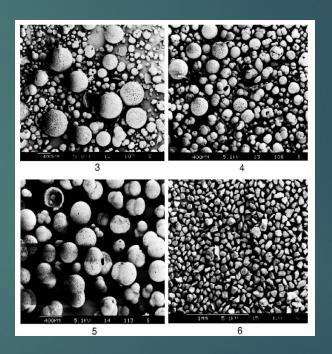
- high viscosity (>300 atm seems "solid" = non-Newtonian fluid
 i.e. supports deposited fine-grained sediments);
- sand fraction (> 64 µm in 4g between 4-11%) from planktonic nannofossils +

foraminifers = 61-78% carbonate content;

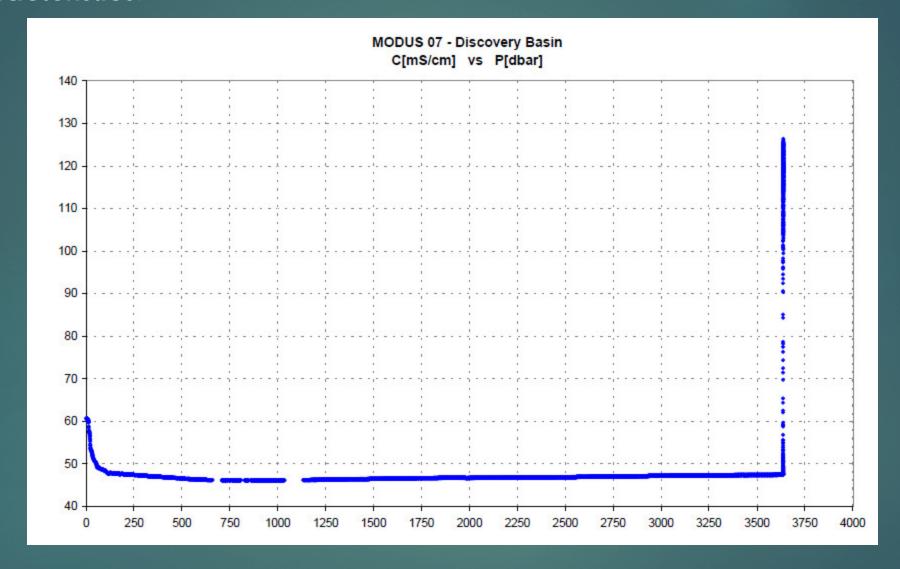
- superficial tension (between the sea water and salt brine layers);
- sediment injections;
- "hot-spots" of microbial activity
 (i.e. intensive and stratified
 methanogenic bacterial ecosystem);
- methane concentration:

128-2692x103nM (2.048-43.072 mg/L)

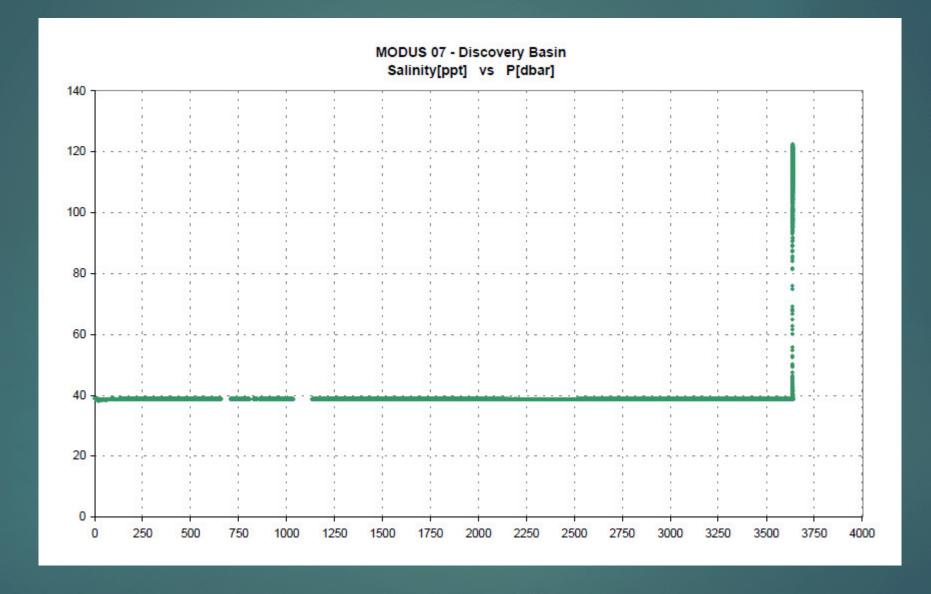




Brine characteristics:



Brine characteristics:



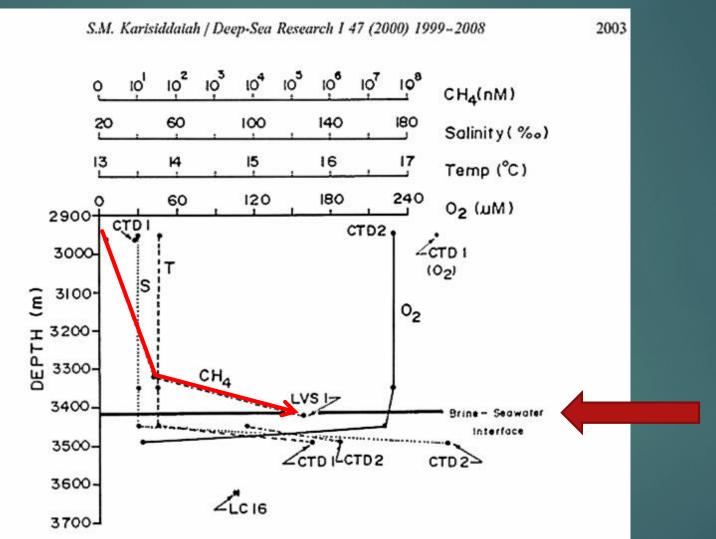


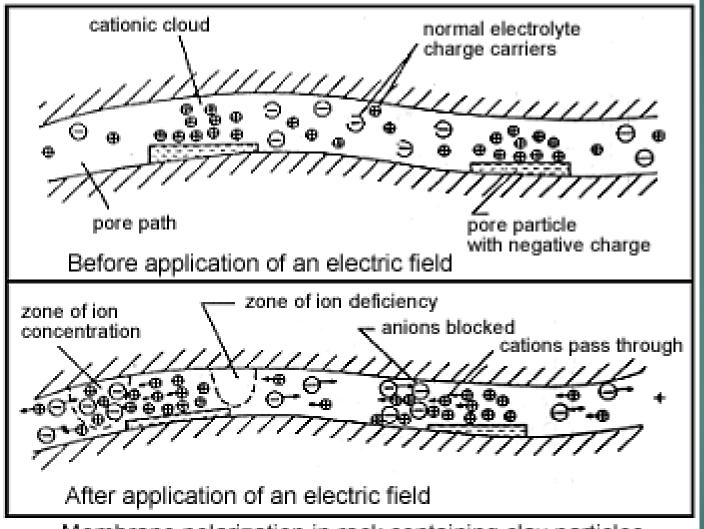
Fig. 2. Vertical profiles of dissolved oxygen (O₂ in μM); potential temperature (°C); salinity and dissolved methane (CH₄ in nM) in brine-sea water interface of Bannock and Urania Basins and of core LC 16 below the anoxic brine.

DISCUSSION:

There is sufficient evidence that DHABs exist. Questions to follow:

- 1. Why does diffusion not work across the "flat" reflection surface to mix sea water and hypersaline brines?
- 2. How is that surface formed and maintained, and what sediment load can it support?
- 3. Can this phenomenon occur over geological time scales?

Family of electro-kinetic phenomena



Membrane polarization in rock containing clay particles from S. Ward, 1990

Membrane polarisation and superficial tension

At a certain depth in the deep sea, the falling argillaceous particles as polar particles will aggregate and generate coagulation and flocculation.

This creates a potential field in which the positive ions

(Ca²+ and Mg²+)

will be forced to migrate, hence causing the increased conductivity detected – a phenomenon called:

membrane polarization.

Frederick A.F. Berry (1969) Relative factors influencing membrane filtration effects in geologic environments

in: Chemical Geology, Volume 4, Issue 1, Pages 295-301

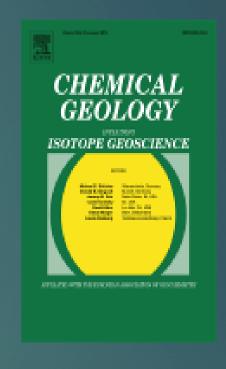
Abstract

"Shales serve as semi-permeable membranes.

These membrane properties result from the <u>electrostatic properties of</u> <u>clays and kerogen</u>. [...]

The probable effect of temperature, pressure, and chemical concentrations of the aqueous solutions on the total efficiency of the shale membrane and the relative passage rates of the dissolved species also are discussed.

Important parameters for a better understanding of membrane behavior that require experimental and field investigation are presented."



NB: It is flocculation and the mentioned electrostatic properties that generate membrane polarization.

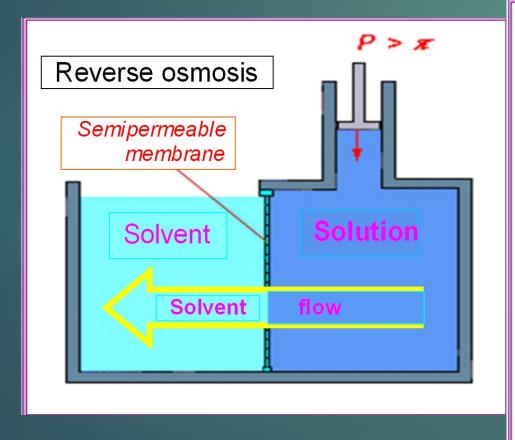
The result is a semipermeable membrane.

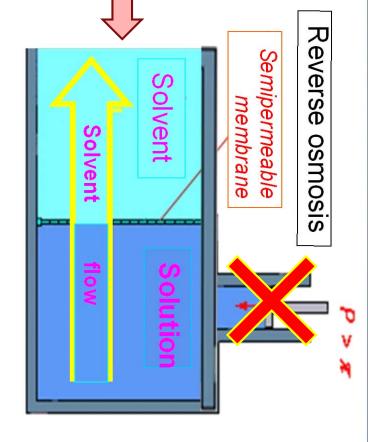
REVERSE OSMOTIC PRESSURE:

HIDROSTATIC PRESSURE (~300atm=4500psi > 25atm=363 psi)

The surface between the hypersaline and normal saline water presumably works as a semipermeable membrane due to the deposited clay minerals

osmotic pressure of the sea water





REVERSE OSMOTIC PRESSURE

The solvent = <u>water</u> is released; underneath, the NaCl concentration Increases, generating brine, the DHAB

Buried DHAB:

This viscous brine is able to <u>support deposited sediments</u>, as mentioned by the MedRiff Consortium (1995).

Over geological time, a large number of DHABs might be buried and isolated within a sedimentary basin.

These hidden, over-pressured brine pools of high viscosity could preserve their internal properties,

while behaving as <u>fluid-supported</u> formations and underlying large basin fill sedimentary sequences

Table 1 | Average chemical composition of the *Medee* Lake brines. All concentrations in mmol kg^{-1} unless otherwise stated. Reported geochemical values are mean \pm 5% (n = 6) obtained during 2010–2012 years unless otherwise stated. Abbreviations used: GB, glycine betaine; MPR, methane production rates; n.d., not determined

Parameters	Brine L1 2,940 m	Brine L2 2,975 m	Brine L3 3,010 m	Brine L4 3,102 m
Density, kg dm ⁻³	1.19	1.21	1.22	1.22
Temperature, °C	14.45	14.73	15.32	15.44
Salinity	304	314	325	345
Na ⁺	4,022	4,110	4,165	4,178
C -	4,684	4,833	4,830	5,259
Mg ²⁺	603	630	773	788
Mg ²⁺ K ⁺	331	363	462	471
Ca ²⁺	2.4	2.6	3.0	2.8
Cα ²⁺ SO ₄ ²⁻ HS ⁻	140.4	146	166.9	201
HS ⁻	0.67	0.93	0.97	1.64
Br ⁻	49.0	53.3	62.6	65.3
H ₃ BO ₃	1.9	2.0	2.2	2.3
NH ₄ ⁺	2.31	2.27	2.45	2.35
Li ⁺ μmol L ⁻¹	149	160	166	163
CH ₄ μmol L ⁻¹	18.0 ± 3.1	70.3 ± 2.3	24.1 ± 3.3	13.9 ± 1.4
Acetate μmol L ⁻¹	132 ± 21	539 ± 42	508 ± 37	n.d.
GB nmol L ⁻¹	170 ± 9	n.d.	44 ± 7	0*
MPR, μmol L ⁻¹ day ⁻¹	2.1 ± 0.2	3.1 ± 0.4	1.5 ± 0.6	0.5 ± 0.4

^{*}The values correspond to the glycine betaine concentration found in the sediments collected at the depth of 3,105 m.

Methane content in the DHAB:

If methane content of current DHABs continues to be confirmed (Karisiddaiah [2000] and M. Yakimov et.al. [2013]),

then substantial volumes of biogenic methane are likely trapped in buried brines (~40mg/L).

Since acetate decomposes into methane and water,

 $CH_3-COOH^- = CH_4 + CO_2$, an additional source for yielding methane in the brine (~500 μ mol/L = ~30mg/L) is available.

Thus, the primary bacterial generation ~40 mg/L and

the <u>secondary acetate decomposition</u> ~30 mg/L will increase

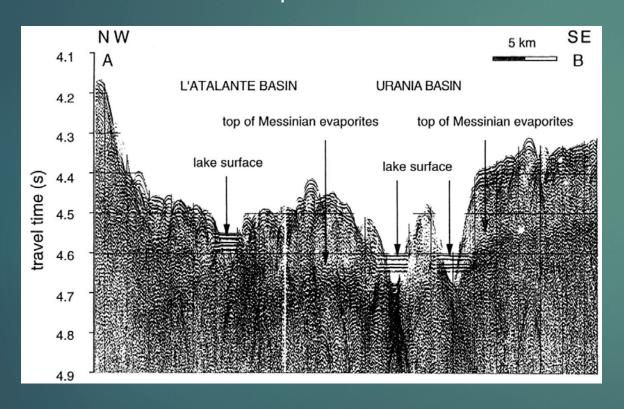
the <u>aggregated methane concentration</u> to ~70mg/L in buried brine.

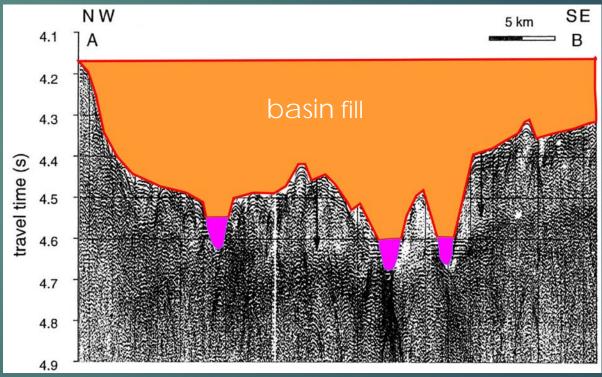
As a consequence:

these brines can serve as sources for hydrocarbon resources !!!

New hypotheses relating diapir formation:

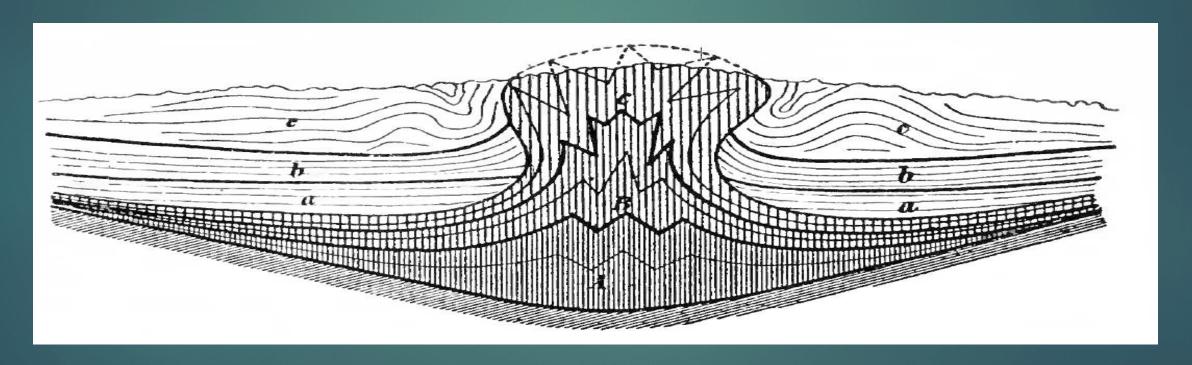
As the **methane-containing brine** is generated (DHAB) and covered by a significant sediment volume, the considerable weight of the deposits will create overpressure in the brine layers.





This initial equilibrium might be lost due to an event: an earthquake.

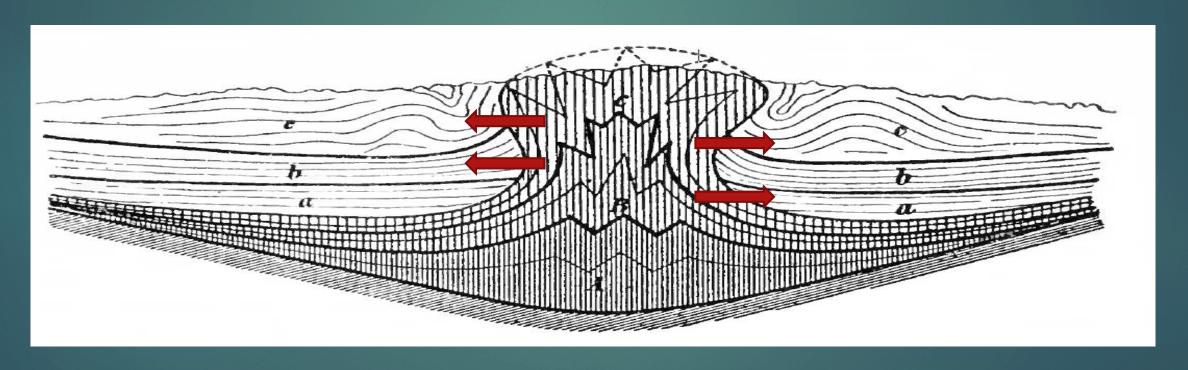
The breach of the trap, allowing for the escape of the over-pressured, viscous, methane-saturated brine into the overburden layers, will initiate salt diapir formation.



Franz Posepny (1871, p.178.) in his study Studien aus dem Salinargebiete Siebenbürgen.

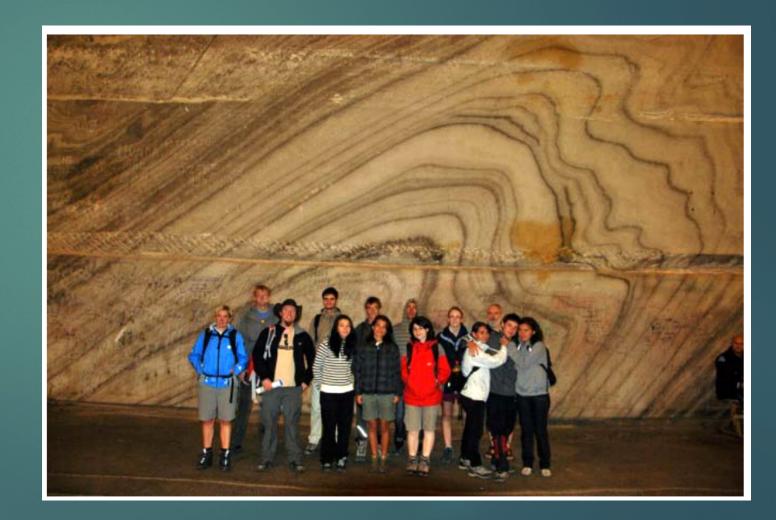
The over-pressured, ascending brine <u>releases gas and water</u> into <u>sandy</u> reservoir packages.

Meanwhile, the salt body suffers volume decrease, such as the collapse of volcanic structures.



Franz Posepny (1871, p.178.) in: Studien aus dem Salinargebiete Siebenbürgen.

The de-gasification and dehydration of the hypersaline brine initiates salt crystallization, meanwhile preserving the folds generated during the penetration and ascending movement.



This ascending process facilitates incorporating rock fragments from the covering deposits.



CONCLUSION I

- The recent scientific knowledge about deep marine processes and phenomena described thereof, especially the evidence of <u>Deep Hypersaline Anoxic Basins/Lakes</u>, drove us to formulate a <u>new hypothesis on salt and biogenic methane generation</u>.
- It has been shown that normal salinity and diffused sea water properties change at particular depths, creating abrupt increase in conductivity and salinity concentration.
 - It is believed that the phenomenon of <u>membrane polarisation</u> is responsible for the increase of conductivity due to argillaceous ions of negative charge and the positive Ca²⁺ and Mg²⁺ ions.

CONCLUSION II

- Membrane polarisation creates a semipermeable membrane, and generates, above a specific pressure, a <u>reverse osmotic process</u>.
 - This is the phenomenon responsible for the <u>increase of the salinity</u> beneath the membrane layer, generating the DHABs.
 - The semipermeable membrane is responsible for the superficial tension between normal sea water and brine, serving as a physical barrier against diffusion.
- The oversaturated, viscous brine is an ideal environment both for <u>methane generating bacteria</u> (~ 40 mg/L), and for <u>acetate decomposition</u> (~30 mg/L),

resulting totally ~70 mg/L methane concentration.

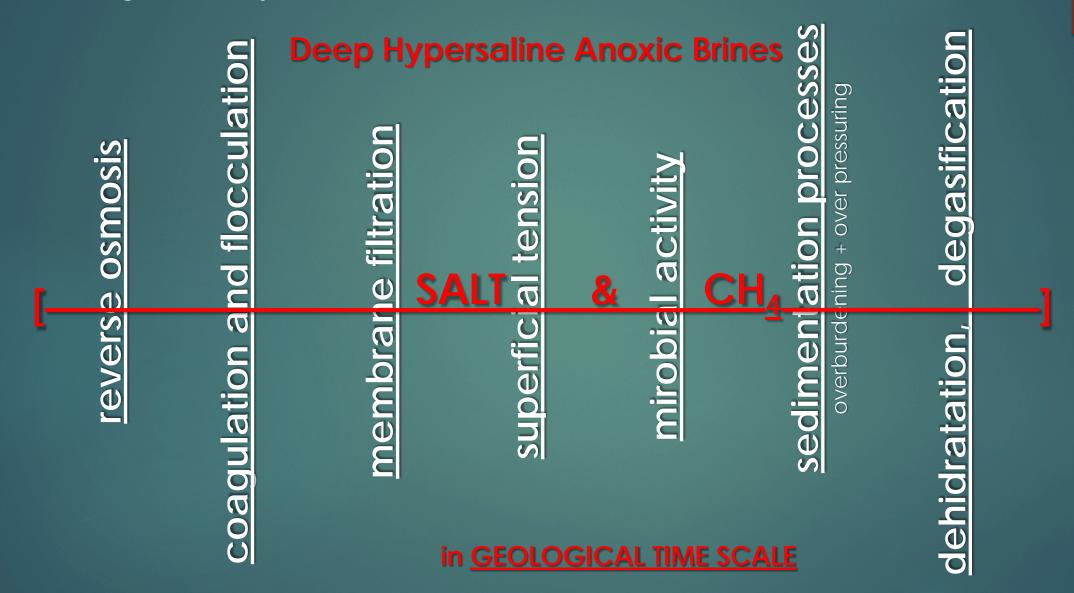
CONCLUSION III

 As found by MedRIFF, the viscous brine surface can support deposited sediments.

This allows the brines to be trapped and buried over geological times, generating an over-pressured, non-crystalized body of high methane concentration (~70mg/l).

Once the buried, over-pressured brine with methane content
is exposed to tectonic events, the trapped methane and water
breach the seal and are expelled into the sedimentary
layers above.

As a result, <u>crystalline salt diapirs are formed</u> and, if <u>porous reservoir rock</u> is encountered, it can become <u>charged with methane gas</u>.



So, as you see, instead of an astronaut

I became a geologist

exploring not the highest

but the deepest, like divers do.

Thank you for your kind attention!

ACKNOWLEDGMENTS:

This work was supported by Sandhill Petroleum BV.,
and Oil&Gas Development (OGD) Ltd. Hungary.

We express special thanks to Mr. Pete NOLAN (senior consultant) and Mr. Abhi MANERIKAR (Exploration Director at OGD),

Mr. Steve PEARSON (vice-president New Venture and Exploration Operations)

for permanent conversation and encouraging support,
debates and essential expert advice.

We express our gratitude to the colleagues for the critical opinion on listening to our new hypotheses.

I also thank to my family who had to listen all my detailed ideas and special thank to my wife who brushed up my English with more or less success.

https://www.youtube.com/watch?v=7t2KoQu5nUA

https://www.youtube.com/watch?v=3zoTKXXNQIU

Background

The first recorded firedamp explosion occurred on 9 September 1664 in Hallstatt (Austria) salt mine, not in a coal mine (Harris 1908).

Prior to the installation of a proper ventilation system, special workers were responsible for burning off the methane that would accumulate in the ceilings of the mine chambers in the Wieliczka Salt Mine (Poland).

In Szolotvina Salt Mine (Aknaszlatina, Ukraine), the methane from salt was used for illumination (Wanek 2008) in the early 19th century.

Nowadays, salt mines are equipped with extensive ventilation systems to prevent similar firedamp explosions.