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Hydrothermal Systems of Kamchatka are Models of the Prebiotic Environment

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Abstract The composition of organic matter and fluctuations of thermodynamic parameters were investigated in the hydrothermal systems of the Kamchatka peninsula in the context of the origin of life. Organics were analyzed by gas-chromatography/mass spectrometry, and 111 organic compounds belonging to 14 homologous series (aromatic hydrocarbons, alkanes and isoalkanes, halogenated aromatic hydrocarbons, carboxylic acids, esters, etc.) were found in hot springs inhabited by Archaeal and Bacterial thermophiles. The organics detected in the sterile condensate of water-steam mixture taken from deep boreholes (temperature 108-175 °C) consisted of 69 compounds of 11 homologous series, with aromatic hydrocarbons and alkanes being prevalent. The organic material included important prebiotic components such as nitrogen-containing compounds and lipid precursors. A separate organic phase (oil) was discovered in the Uzon Caldera. A biogenic origin is supported by the presence of sterane and hopane biomarkers and the δ^{13} C value of the bulk oil; its age determined by 14 C measurements was 1030±40 years. Multilevel fluctuations of thermodynamic parameters proposed to be required for the origin of life were determined in the Mutnovsky and Pauzhetsky hydrothermal systems. The low-frequency component of the hydrothermal fluid pressure varied by up to 2 bars over periods of hours to days, while mid-frequency variations had regular micro-oscillations with periods of about 20 min; the high-frequency component displayed sharp changes of pressure and microfluctuations with periods less than 5 min. The correlation coefficient between pressure and temperature ranges from 0.89 to 0.99 (average 0.96). The natural regimes of pressure and temperature fluctuations in Kamchatka hydrothermal systems can guide future experiments on prebiotic chemistry under oscillating conditions.

Keywords Origin of life · Organic compound · Prebiotic chemistry · Pressure · Temperature · Fluctuation · Oil

Introduction

Geothermal environments on the early Earth are considered to be a plausible site for the origin of life (Corliss et al. 1981; Washington 2000; Holm and Andersson 2005; Russell et al. 2005;

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Mulkidjanian et al. 2012; Deamer 2013). In this context, the Kamchatka peninsula represents a useful analogue of the prebiotic environments because it contains variable types of hydrothermal discharges distributed over a vast territory. The Kamchatka peninsula is situated in the remote northeastern part of Russia and spans a region of approximately 400 by 1200 km (Fig. 1). This region contains 30 active volcanoes, together with 270 exposed hydrothermal sites. The most active hydrothermal systems are located within a narrow (30–50 km) zone along the axis of main deep faults that cross the volcanic belts of eastern and central Kamchatka.

We explored 14 hydrothermal fields in Kamchatka and the adjoining regions (intracontinental part of the Russian Far East and Kuril Islands): Mutnovsky, Pauzhetsky, Uzon, Karymsky, Paratunsky, Koshelevsky, Kuldur, and some others. The temperature and chemical composition in these thermal discharges vary: the pH ranges from 2.0 to 9.5, and the temperature from 55 to 98 °C in hot springs and up to 240 °C—in the water-steam mixture found in bore holes. Significant concentrations of Cl⁻, SO₄²⁻, HCO₃⁻, Na⁺, Ca²⁺, Mg²⁺, NH₄⁺ are found in the liquid phase.



Fig. 1 Map of the Kamchatka peninsula. Main hydrothermal fields: 1 – Uzon caldera and Valley of Geysers; 2 – Karymsky caldera lake; 3 – Mutnovsky; 4 – Pauzhetsky

- Exploration of the organic matter found in hydrothermal fluid. In the context of the origin
 of life problem, it was interesting to compare biogenic organics in hot springs, inhabited
 by (hyper) thermophiles, and in the lifeless condensate of water-steam mixture, where
 organic compounds could not appear as the result of contemporary microorganisms'
 destruction.
- 2. Analysis of pressure, temperature and some chemical components fluctuations in hydro-thermal systems. Substantial fluctuations of thermodynamic and physico-chemical parameters in the medium represent the fourth required condition for life origin (in addition to the three formerly stated conditions the presence of water, organic substances and a source of energy), as follows from the inversion concept (Kompanichenko 2012, 2014). This concept proposes that thermodynamic inversion can occur when available free energy prevails over entropic disorder. This is a key transformation that launches biological processes in a prebiotic microsystem by driving a specific nonequilibrium position within oscillations around the bifurcation point. Optimal fluctuations in the medium are a necessary condition for such oscillations to be maintained in the microsystem.

This paper expands and integrates the data published in our previous works (Kompanichenko et al. 2009, 2010; Kompanichenko and Shlyufman 2013; Poturay 2013), and includes important results from Kamchatka hydrothermal systems explored by other researchers in an origin of life context.

Organic Matter in Kamchatka Hydrothermal Systems

We investigated moderately volatile organic compounds in hot springs and water-steam mixtures from wells with a Shimadzu GCMS-QP2010S spectrometer. The C-18 cartridges that were used to extract organics from water samples (volume 200 ml) were then dried under an argon jet. The organics was extracted by adding methylene chloride (1 ml). The analyses were conducted in the regime of full ionic current, m/z values 50–350. With the full ionic current chromatograms moderately volatile organic compounds (mainly neutral and acid) were identified and their relative percentage abundances estimated. Phthalates were present in all samples, presumably as trace contaminants from plastic containers.

A total of 69 organic compounds that belong to 11 homologous series were analyzed in the condensate of water-steam mixtures taken from deep boreholes from the Mutnovsky and Pauzhetsky fields (Table 1). These organic compounds are characteristic of a deep sterile zone near active volcanoes of Mutnovsky and Koshelev. In them, the water-steam mixture is under a pressure of 1.7-8.2 bars at temperatures ranging from 108 to 175 °C. The pH varied between 4 and 8, and the borehole depths ranged from 600 to 2000 m. The most diverse homologous series in the condensate were aromatic hydrocarbons (naphthalene, 1,2-methylnaphtalene biphenyl, phenanthrene, fluorene, squalene, 1,3-diethylbenzene, trichlorobenzene, etc.) and alkanes (decane, dodecane, tridecane, tetradecane, pentadecane, hexadecane, heptadecane, etc.). The highest-temperature (175 °C) sample from the borehole contained only polycyclic aromatic hydrocarbons (naphthalene, biphenyl, phenanthrene, fluorene and 1-methylnaphtalene). Biphenyl and phenanthrene were absent in the samples from boreholes with lower temperature (108-124 °C). However, these samples contained other aromatic hydrocarbons (benzenes, xylenes), as well as compounds of other homologous series.

Temperature, °C	108–175	
Availability of (hyper)thermophiles	Absent	
Homologous series / number of compounds	Alkanes / 21	
	Isoalkanes / 7	
	Isoprenes / 2	
	Aromatic hydrocarbons / 26	
	Halogenated aromatic hydrocarbons / 2	
	Alcohols / 4	
	Aldehydes / 2	
	Ketones / 2	
	Carboxylic acid / 1	
	Terpenes / 1	
	Sulfur-containing hydrocarbons / 1	
Total	11 / 69	

 Table 1
 Moderately volatile organics in lifeless condensate of water-steam mixture (wells)

The list of organic compounds found in diverse hot springs and shallow boreholes is longer—111 compounds from 14 homologous series (Table 2). In addition to the aromatic hydrocarbons and alkanes detected in the lifeless condensate, these samples contain a lot of isoalkanes, halogenated aromatic hydrocarbons, carboxylic acids and esters. The temperature of the solutions in the springs of the Mutnovsky, Uzon and Karymsky systems ranged between 55 and 99 °C, and the pH between 2.5 and 7.0. The Kuldur hydrothermal system, located in the continental part of the Russian Far East (near the Kamchatka peninsula), is characterized by alkaline lower-temperature solutions (pH 9–9.5, temperature 55–72 °C). All these springs and subsurface areas are inhabited by thermophilic and hyperthermophilic *Archaea* and *Bacteria*.

 Table 2 Moderately volatile organics in the inhabited hot springs

Temperature, °C	55–99	
Availability of (hyper)thermophiles	Archaea and Bacteria	
Homologous series / number of compounds	Alkanes / 21	
	Isoalkanes / 17	
	Cycloalkanes (naphthenes) / 2	
	Alkenes / 7	
	Aromatic hydrocarbons / 13	
	Halogenated aromatic hydrocarbons / 9	
	Alcohols / 5	
	Aldehydes / 4	
	Ketones / 3	
	Carboxylic acid / 14	
	Esters / 11	
	Steroids / 3	
	Terpenes / 1	
	Lactams / 1	
Total	14 / 111	

The source of these compounds has not been established yet. They may represent preexisting organic material chemically degraded by pyrolysis. For instance, Simoneit et al. (2009) established that light oil associated with the Uzon caldera in Kamchatka was formed by pyrolysis of buried algal mats. It would be interesting to determine whether the aromatics and alkanes are products of Fischer-Tropsch type synthesis. The input of abiotic organics is possible due to the availability of Cl-alkanes in hot solutions, because these compounds are seldom produced by living organisms.

Volatile organics taken from gas-steam jets in several Kamchatka hydrothermal fields were explored by Isidorov et al. (1992). The authors determined 72 compounds from 12 homologous series, including alkanes (9 compounds), isoalkanes (16), aromatic hydrocarbons (9) and sulfur-containing organic compounds (13). In addition, they detected nitriles (2) that could be important for prebiotic chemistry. Mukhin and colleagues (1979) analyzed amino acids in several samples of water from hot springs inhabited by thermophiles, and from lifeless condensates of water-steam mixture. They found 12 biological amino acids in hot springs: Ala, Val, Gly, Ile, Ley, Pro, Thr, Ser, Phe, Asp, Glu and Lys. Their concentrations were between 0.07 and 0.19 mg/l. It is remarkable that some small concentration of possibly abiogenic glycine (0.001 mg/l) was detected in lifeless condensate of water-steam mixture taken from a deep borehole in the Pauzhetsky hydrothermal system.

The composition of organic matter can vary over time. We took three samples of water from the same boiling water-mud pool in the crater of the Mutnovsky volcano. The composition of organics in samples 1 and 2 (which were taken within an interval of 30 min) is very close, while the composition of sample 3 (taken 4 years later) it is quite different (Fig. 2). A highly variable composition of organic substances was noted in the alkaline hot solutions of the Kuldur hydrothermal field over the period of 2007–2011.

The consolidated Table 3 includes all the organic substance discovered in hot water and water-steam mixture in Kamchatka and its nearby areas, a total of 243 compounds that belong to 24 homologous series. This list includes important prebiotic components—nitrogen-containing compounds (amino acids, nitriles, amides, nitrogen cycles) and lipid precursors (carboxylic acids, esters, alcohols, aldehydes and other).

The segregation of organic substance into a separate oily phase was noted in the Uzon Caldera. The unique hydrothermal systems of the Uzon caldera and the Valley of Geysers are found in the vast Uzon-Geyser volcano-tectonic depression located in the south-east of Kamchatka. The depression is characterized by one of the most intense heat flows in Kamchatka. The zone of thermal water discharge extends across the caldera for~5 km and consists of several thermal lakes with boiling water and mud pools. The zone is located above a magma chamber, resulting in a high local temperature gradient. The estimated temperature at a depth of 500 m is 200–250 °C (Karpov 1988). Oil in the Uzon hydrothermal field was explored by several scientific teams, including our recent investigation (Simoneit et al. 2009). Gas chromatography-mass spectrometry (GC-MS) was used to determine the primary constituents, and the ¹³C and ¹⁴C compositions provided information about the potential source and age of the oils. The ${}^{14}C$ ages determined were 1030 ± 40 years (measured) or 940±40 years (conventional). The δ^{13} C value is 30.6‰ versus the PDB standard, a value consistent with a biological origin. The nearly contemporary age of the C content indicates a geologically recent origin from biogenic detritus and not by synthesis from mantle C. The biogenic origin is supported by the presence of sterane and hopane biomarkers and the δ^{13} C value of the bulk oil. The overall compositions of the oils indicate that they are derived from rapid hydrothermal alteration of algal/bacterial mat detritus buried by volcanic ashfall deposits of the Uzon Caldera. The oils represent the youngest hydrothermal petroleum reported to date.



Fig. 2 Change of organics in length of time (Mutnovsky spring)

Fluctuations of Thermodynamic and Physico-Chemical Parameters

We have characterized short-term and long-term fluctuations of pressure and temperature in Kamchatka hydrothermal systems mainly in the Mutnovsky hydrothermal field. It is situated 70 km southwest of Petropavlovsk, and is flanked by two active volcanoes, Mutnovsky on the south and Gorely on the west. The field itself is within an active fissure zone that connects to Mutnovsky's plumbing system. Consequently, the volcanic and seismic activities of the volcanoes may directly affect the fluctuations in the Mutnovsky geothermal system.

To estimate amplitudes, periods/frequencies of fluctuations and the correlation between pressure and temperature, data from the perennial monitoring in the boreholes was used. The mathematical methods of direct spectral analysis were employed for the database that was drawn from the monitoring of well N_{2} 30 in the Mutnovsky hydrothermal system (the time interval between measurements was 2 min). The well penetrates two zones of higher permeability, one at a depth of 825 m (with temperatures of 217–228 °C) and the other at 950 m (231–233 °C). The pressure in the well at 950 m has been between 26 and 28 bars in the most recent years (Kiryukhin et al. 2002). Pressure was measured at a depth of 950 m. The temperature and pressure gradients between this level and the surface are very high: the temperature of water-steam mixture at the mouth of well N_{2} 30 was 120 °C and the pressure

Class	Homologous series	Number of compounds
Hydrocarbons	Alkanes	29
	Isoalkanes	33
	Isoprenes	2
	Alkenes	12
	Isoalkenes	2
	Halogenated hydrocarbons	6
Lipid precursors	Alcohols	8
	Aldehydes	6
	Ketones	4
	Carboxylic acid	20
	Esters	28
	Terpenes	4
	Monoglycerides	1
	Steroids	3
Nitrogen-containing organic compounds	Nitriles	2
	Amides	2
	Amino acids	12
Organic sulfur compounds	Thiols	12
	Alkylsulfides	2
Carbocyclic compounds	Cycloalkanes (naphthenes)	4
	Aromatic hydrocarbons	35
	Halogenated aromatic hydrocarbons	9
Heterocyclic compounds	Nitrogen cycles	6
	Sulfur cycles	1
Total	24	243

Table 3 Organic compounds in hydrothermal systems of Kamchatka and nearby areas (consolidated table)

was 2 bars. Spectral analysis in combination with visual methods was applied to the study of pressure variations in steam–water mixture based on the 2004 monitoring observations (a total of 251,961 measurements) provided to us by A. Kiryukhin (Institute for Volcanology and Seismology, Petropavlovsk). The methods of spectral analysis yield an idea of the variation in the frequency domain in cases where this is difficult to do in the time domain. The method we selected from the spectral analysis tools was the Schuster periodogram. This is a direct spectral estimation method and can provide amplitude–frequency characteristics of the variation under study based on raw observational data. The results of this investigation are briefly given below (published in detail in: Kompanichenko and Shlyufinan 2013).

We identified large-scale and small-scale water-steam mixture pressure variability. The large-scale ones correspond to the dynamic low-frequency component, in the form of macro-fluctuations with considerable (up to 2 bars) drastic or smooth changes of pressure (Fig. 3). The small-scale changes represent both irregular and regular microfluctuations. In this variable framework it is possible to allocate mid-frequency and high-frequency components. The mid-frequency of 0.00083 Hz), the maximum change of pressure between the measurements being 0.7 bars (Fig. 3). The high-frequency component represents sharp changes of pressure (decrease, increase and emissions), as well as microfluctuations with periods less than



Fig. 3 Pressure dynamics of water-steam mixture in the Mutnovsky hydrothermal system (well $N \ge 30$, depth 950 m). High-amplitude irregular macrofluctuations (amplitudes up to 1–2 bars), and low-amplitude quite regular microoscillations of pressure (amplitudes 0.1-0.3 bars) with the period about 20 min are shown

5 min. The high-frequency and mid-frequency components in various combinations describe the fluctuations of pressure about this level. Spectral analysis of short pressure variation segments showed that the pressure fluctuations in many segments are gradual oscillations with frequencies in the medium range.

Another database was used to estimate the correlation coefficient between pressure and temperature. Once every 10 days pressure and temperature were measured at the mouths of 12 boreholes in the Mutnovsky field over 6 years. The temperature at the mouths ranged from about 100 to 240 °C, and the pressure from about 1 to 40 bars. The estimated correlation coefficient ranges from 0.89 to 0.99 (average 0.96).

The preliminary study of pressure dynamics of water-steam mixture in 11 borehole mouths of the Pauzhetsky hydrothermal system demonstrated the presence of similar fluctuations. The periods of regular pressure microfluctuations of the Pauzhetsky field ranged from 10 to 60 min. The amplitudes of macrofluctuations reached 1–2 bars; and the amplitudes of microfluctuations occurred within the interval of 0.1–0.3 bars.

Earthquakes and volcanic eruptions may strengthen thermodynamic and physico-chemical fluctuations. Thus, it was shown that three of four big earthquakes in Kamchatka initiated a rise in pressure oscillation amplitudes (Kiryukhin et al. 2002). Big earthquakes may also initiate a rise in light hydrocarbons outflow (CH₄, C₃H₆) and a significant change of inorganic component (Na⁺, Cl⁻) concentrations in Kamchatka groundwater systems (Lyubushkin et al. 1997).

We did not investigate the relationship between pressure fluctuations and the chemistry of the fluid, but there is a study of this relationship in a hot (58 °C) solution at well SOB-1 in the Mura thermal field in Slovenia (Kralj and Kralj 2000). This study revealed rather regular changes in temperature and pressure that occur with a period of 70 min. A strong negative correlation was found to exist between the two parameters. These authors also studied changes in brine chemistry during a single 70 min cycle. There was a shift of about 15 min in well-defined maxima of pH and concentrations of Na⁺, K⁺, Ca²⁺, Cl⁻, Br⁻ relative to the peak of maximum temperature (which coincided with the pressure minimum). Poorly defined peaks of the greatest concentrations of I⁻, F⁻, and CO₂ also coincided with these maxima, only in the case of Mg²⁺ were no definite trends detected.

Conclusion

The geothermal region of Kamchatka contains many geological sites that can be considered as possible analogs of prebiotic environments (both surface discharges and their subsurface

cracks). Terrestrial superficial thermal outcrops are diverse: geysers, boiling water and mud pools, hot or warm pools, thermal lakes and streams, outflow of thermal springs, and steamgas jets (fumaroles). Many submerged hot vents are at the bottom of lakes (Karymsky, Khodutka) and near a small Kuril volcanic island of Yankicha, situated not far from Kamchatka. Examples of the sites are shown in Fig. 4. Explorations of near-surface and subsurface zones (up to the depth of 2 km) of hydrothermal systems in Kamchatka demonstrate the availability of three accepted conditions for the origin of life: liquid environs, a source of energy (high temperature, substantial concentration gradients), and organic matter (243 compounds, including important prebiotic components—nitrogen-containing compounds and lipid precursors). In addition, the Kamchatka hydrothermal systems provide a fourth required condition, added by the present authors: significant fluctuations of thermodynamic and physico-chemical parameters. Multilevel irregular and regular fluctuations of pressure and temperature were detected in the Mutnovsky and Pauzhetsky hydrothermal systems. The fluctuations are periodically stimulated by earthquakes and volcanic eruptions.

Therefore, the Kamchatka geothermal region is a very suitable testing ground to investigate the process of life's origin on Earth. Experiments exploring self-assembly and polymerization reactions in Kamchatka natural environments was already carried out by Deamer et al. (2006). The mixture of representative organic solutes (amino acids, nucleobases, a fatty acid, glycerol) and phosphate were added to a high temperature, clay-lined pool in the Mutnovsky hydro-thermal field. The pool temperature was about 90 ° C and the pH was~3. Most of the added organics and phosphate were removed from solution with half-times measured in minutes to a few hours. The analysis of the clay showed that the organics were adsorbed to the mineral surfaces at the acidic pH of the pools, but could be released in basic solutions. Deamer and



Fig. 4 Types of possible analogs of prebiotic environments in Kamchatka: \mathbf{a} – boiling water pool (Uzon caldera); \mathbf{b} – boiling water-mud pool (Pauzhetsky field); \mathbf{c} – hot water pool (Mutnovsky field); \mathbf{d} – geyser Sakharny (Valley of Geysers); \mathbf{e} – hot stream (Pauzhetsky field); \mathbf{f} – Karymsky caldera lake with associated hot vents

collaborators conducted and developed laboratory experiments on prebiotic chemistry under conditions that simulated natural geothermal sites (Deamer 2013; Toppozini et al. 2013).

Another origin of life concept proposes that protocells must have evolved in habitats with a high K^+/Na^+ ratio, relatively high concentrations of Zn^{2+} , Mn^{2+} , and phosphorous compounds (Mulkidjanian et al. 2012). These authors proposed that the ionic composition conducive to the origin of cells could not have existed in marine settings but is compatible with emissions of vapor-dominated zones of inland geothermal systems. To support this view, they adduced geochemical analyses of vapor–gas jets (fumaroles) in Kamchatka, where K^+ concentrations are higher than Na^+ concentrations.

The necessity of thermodynamic and/or physico-chemical fluctuations in the origin-of-life medium that follows from the inversion concept suggests that laboratory simulations of prebiotic conditions should incorporate a fluctuating hydrothermal environment (Kompanichenko 2012, 2014). Variable oscillating conditions may lead to a significant prevalence of synthesis/oligomerization over degradation that depends on the scale and frequency of fluctuations. Thus, synthesis and oligomerization of glycine take place in simulated hydrothermal conditions, but synthesized (macro) molecules have a tendency to decompose (Cleaves et al. 2009). However, these molecules may reorganize under optimal fluctuations and changes of temperature into increasingly complex structures. Some laboratory experiments have demonstrated an acceleration of prebiotic chemical evolution under temperature oscillations. For instance, amplification of DNA, under temperature oscillations between 64 and 92° centigrade, accelerated the division of giant vesicles (Sugawara et al. 2012; Sugawara 2014). Changes of wet-dry conditions and oscillations of temperature around 85° (the model of a periodically evaporating and filling hydrothermal pond) promoted synthesis of RNA-like molecules from mononucleotides (Deamer 2013), and condensation of amino acids into peptides under a thermocycling regime around the boiling point of water was reported by Varfolomeev (2007). Computational models of thermal cycling also suggest the importance of oscillating conditions for prebiotic evolution (Osipovich et al. 2009; Barratt et al. 2010). Natural regimes of pressure and temperature fluctuations revealed in Kamchatka hydrothermal systems could be used to guide future experiments on prebiotic chemistry.

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