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## Living microbial ecosystems within the active zone of catagenesis: Implications for feeding the deep biosphere

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## Abstract

Earth's largest reactive carbon pool, marine sedimentary organic matter, becomes increasingly recalcitrant during burial, making it almost inaccessible as a substrate for microorganisms, and thereby limiting metabolic activity in the deep biosphere. Because elevated temperature acting over geological time leads to the massive thermal breakdown of the organic matter into volatiles, including petroleum, the question arises whether microorganisms can directly utilize these maturation products as a substrate. While migrated thermogenic fluids are known to sustain microbial consortia in shallow sediments, an in situ coupling of abiotic generation and microbial utilization has not been demonstrated. Here we show, using a combination of basin modelling, kinetic modelling, geomicrobiology and biogeochemistry, that microorganisms inhabit the active generation zone in the Nankai Trough, offshore Japan. Three sites from ODP Leg 190 have been evaluated, namely 1173, 1174 and 1177, drilled in nearly undeformed Quaternary and Tertiary sedimentary sequences seaward of the Nankai Trough itself. Paleotemperatures were reconstructed based on subsidence profiles, compaction modelling, present-day heat flow, downhole temperature measurements and organic maturity parameters. Today's heat flow distribution can be considered mainly conductive, and is extremely high in places, reaching 180 mW/m<sup>2</sup>. The kinetic parameters describing total hydrocarbon generation, determined by laboratory pyrolysis experiments, were utilized by the model in order to predict the timing of generation in time and space. The model predicts that the onset of present day generation lies between 300 and 500 m below sea floor (5100-5300 m below mean sea level), depending on well location. In the case of Site 1174, 5-10% conversion has taken place by a present day temperature of ca. 85 °C. Predictions were largely validated by on-site hydrocarbon gas measurements. Viable organisms in the same depth range have been proven using <sup>14</sup>C-radiolabelled substrates for methanogenesis, bacterial cell counts and intact phospholipids. Altogether, these results point to an overlap of abiotic thermal degradation reactions going on in the same part of the sedimentary column as where a deep biosphere exists. The organic matter preserved in Nankai Trough sediments is of the type that generates putative feedstocks for microbial activity, namely oxygenated compounds and hydrocarbons. Furthermore, the rates of thermal degradation calculated from the kinetic model closely resemble rates

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of respiration and electron donor consumption independently measured in other deep biosphere environments. We deduce that abiotically driven degradation reactions have provided substrates for microbial activity in deep sediments at this convergent continental margin.

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## 1. Introduction

The biosphere is the part of planet Earth inhabited by life. It occurs in three overlapping zones: the *lithosphere*, comprising soils, sediments and sedimentary rocks; the hydrosphere, that part of the lithosphere either covered by water or containing water (within pores and fractures); and the atmosphere, the gaseous envelope surrounding the Earth. Microorganisms make up a major component of the Earth's biomass because they can grow under a wide range of conditions and have diverse metabolisms. Aerobic bacteria use free oxygen to degrade and metabolise labile organic substrates, but where oxygen has already been consumed and cannot be replenished, for instance in fine grained sediments, anaerobic microorganisms take over degradation using other terminal electron acceptors (oxidisers) such as sulphate, nitrate, manganese, iron and carbon dioxide. Anaerobes are the dominant inhabitants of the lithosphere. Their abundance in sedimentary basins shows a general decrease with increasing depth, eventually petering out as organic matter becomes too recalcitrant to be degraded or because water, nutrients and terminal electron acceptors cannot be supplied or because temperatures are too high.

In recent years it has come to light that surprisingly large indigenous microbial populations with considerable diversity are present in sedimentary rocks at hundreds or in some cases thousands of metres depth [1-4]. The organic matter supporting microbial metabolism in deep subsurface environments probably originates within the deep subsurface itself though the mechanisms involved remain poorly understood [5]. Fermentation products from indigenous organic matter in shales could diffuse to microbial communities residing in juxtaposed porous facies, for example [6]. Moreover, elevated temperatures (30-90 °C) acting over short periods (weeks and months) increase the rate of microbial degradation in laboratory experiments [7,8]. In the case of buried sedimentary organic matter in the natural system, residence times at these elevated temperatures (30-90 °C and beyond) extend way beyond human timescales, lasting millions of years. This has two ramifications.

Firstly, the aforementioned biological fermentations should be able to take place throughout long periods of basin history, with rates being temperature dependant. Secondly, kinetically controlled abiotic organic maturation reactions such as condensation and elimination (zone of diagenesis), cracking and aromatisation (zone of catagenesis) become significant, converting recalcitrant organic matter such as kerogen into volatile products which include petroleum [9]. The intriguing question is whether all or some of the abiotic reaction products can be utilised by microorganisms as substrates in the deep biosphere [8,10]. While migrated or migrating thermogenic fluids are already known to sustain microbial consortia at gas hydrates [11,12], petroleum reservoirs [13] and surface expressions of leakage such as carbonate mounds [14,15], an in situ coupling of abiotic generation and microbial utilization has not been demonstrated up to now. Indeed, classical geochemical concepts state that the two should be separated from one another in time and space but, as alluded to above, our concept of the biosphere is being driven to include greater depth intervals. It is therefore likely that some overlap between the biosphere and zones of thermal cracking will occur. Indeed, microorganisms have been detected in geological systems up to 100 °C [65] and cultured up to 121 °C [16], and the approximate present day temperature for the onset of petroleum generation for a "typical" petroliferous basin (heating rate 1-5 K/Ma) lies between 70 and 120 °C [17–19].

In this paper we show that deep microbial ecosystems and active hydrocarbon generation occur together in the Nankai Trough, and demonstrate that the rates of thermal degradation closely resemble rates of respiration and electron donor consumption in deep biosphere environments. We deduce that abiotically driven degradation reactions can provide substrates for microbial activity in deep sediments at this convergent continental margin.

## 2. Geological framework of the Nankai Trough

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