

Evidence for the microbial in situ conversion of oil to methane in the Dagang oilfield

NÚRIA JIMÉNEZ¹, MINMIN CAI^{1,2}, NONTJE STRAATEN³, BRANDON E L MORRIS¹, JUN YAO², HANS H RICHNOW¹ AND MARTIN KRÜGER³.

¹Helmholtz Centre for Environmental Research (UFZ), 04318 Leipzig, Germany

²School of Civil & Environment Engineering, University of Science and Technology Beijing, P. R. China

³Federal Institute for Geosciences and Natural Resources (BGR), 30655 Hannover, Germany

The microbial degradation of hydrocarbons in oil reservoirs affects the quality and economic value of recovered petroleum products. Recent studies suggest that anaerobic biodegradation may play a significant role in situ and the biodegradation of residual oil constituents under methanogenic conditions has been reported. Methane, like other gases, may aid in oil viscosity reduction and enhance flow characteristics through the reservoir matrix. In addition, methane may be used as a energy source.

The aim of this study was to assess the ability of indigenous microbial communities from a thermophilic oil reservoir (Dagang oilfield, China) to produce methane from crude oil under environmental conditions. The isotopic composition of reservoir fluids (H₂O, CO₂, CH₄) was analyzed, and GC-MS fingerprinting was applied to assess the oil composition in the reservoir. Microbial cell numbers were assessed by qPCR. Microcosms with ¹³C-labelled hydrocarbons were inoculated with production and injection waters to characterize these processes in vitro.

Analysis of oil samples confirmed that the majority of the oils from Dagang are highly weathered. Geochemical data from reservoir oil, water and gas are consistent with in situ biogenic methane production linked to aliphatic and aromatic hydrocarbon degradation. The bulk isotopic discrimination between methane and CO₂ was in accordance with previously reported results for methane formation during hydrocarbon degradation. Degradation experiments revealed that autochthonous microbiota is capable of producing heavy methane from ¹³C-labelled n-hexadecane or 2-methylnaphthalene. Microbial numbers in oil/water samples from production wells were abundant and the communities diverse, including methanogenic Archaea and hydrocarbon-degrading bacteria. In summary, the investigated areas of the Dagang reservoir may have significant potential for testing the viability of in situ conversion of oil to methane as an enhanced recovery method.

Occurrence of aerobic and methanogenic oil biodegradation in a water-flooded oil field

N. JIMÉNEZ^{1*}, M. CAI², Y. JUN², M. KRÜGER³ AND H.H. RICHNOW¹

¹UFZ, Permoserstraße 15, 04318 Leipzig, Germany

(*Correspondence: nuria.jimenez-garcia@ufz.de)

²University of Science and Technology Beijing - No. 30 Xueyuan Road, Haidian District, Beijing, P. R. China

³BGR, Stilleweg 2, 30655 Hannover, Germany

Water flooding is a secondary oil recovery method intending to enhance oil recovery by increasing the pressure in the oil-bearing strata. However, it can accelerate microbial oil degradation processes leading to a reduction of its quality (by changing its viscosity, sulfur content, etc.). In undisturbed subsurface oil reservoirs biodegradation is thought to be primarily anaerobic, as oxygen is practically unavailable, and methanogenesis may play a predominant role. Nevertheless, injection waters could be an oxygen source, causing alterations in the indigenous microbial communities. The conversion of oil to methane might be of interest because it does not require the supply of additional electron acceptors and methane could be recovered using the existing production infrastructure. On the other hand, aerobic oil biodegradation can involve the production of biosurfactants and other oil-releasing agents, which might favor oil production.

Oil biodegradation was investigated in a highly degraded water-flooded oil reservoir [1]. Isotopic composition of its gases was consistent with a biological origin ($\Delta\delta^{13}\text{C}$ between CH₄ and CO₂ ranging from 32 to 65‰). The reservoir contained an active microbial community, rich in syntrophic bacteria and methanogenic archaea, able to produce ¹³CH₄ and ¹³CO₂ ($\delta^{13}\text{C}$ up to 550‰) from ¹³C-labelled hydrocarbons *in vitro*. Additional laboratory experiments, using production waters, showed oil biodegradation potential not only under methanogenic but also under aerobic conditions. In both cases biodegradation patterns similar to those observed *in situ* were found: *n*-alkanes, *n*-alkyltoluenes and *n*-alkylbenzenes were removed before PAHs and biomarkers. Degradation decreased with increasing alkylation and ring number. A selective removal of some compounds like steranes or alkylated naphthalenes was observed. Further experiments will focus on studying biosurfactant production by indigenous oil degrading communities.

[1] Jiménez, Morris, Cai, Gründger, Yao, Richnow, Krüger (2012) *Organic Geochemistry* **52**, 44-54.