Geophysical Research Abstracts Vol. 20, EGU2018-6115, 2018 EGU General Assembly 2018 © Author(s) 2018. CC Attribution 4.0 license.



## Occurrence and flux of crustal methane in the Fennoscandian Shield

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Methane ( $CH_4$ ) seems to be pervasive in deep groundwaters within the Fennoscandian Shield, where its concentrations often reach tens of mM (Kietäväinen et al. 2017). In Finland alone,  $CH_4$  has been detected at least in 20 separate localities, six of which contain more than 70 vol-%  $CH_4$  in the gas phase. The occurrence of  $CH_4$  is closely linked with lithology, such that the highest  $CH_4$  concentrations occur in association with ophiolites and metasedimentary rocks, graphite rich black schist in particular. Distinct isotope composition of  $CH_4$  and residence times of fluids on the order of tens of millions of years, compared to the age of the rocks (ca 2 Ga) and thermal history of the shield, seem to preclude thermogenic origin. Instead microbial methanogenesis and abiotic formation, probably from  $H_2$  and graphite, are suggested to be the main mechanisms in this environment (Kietäväinen et al. 2017).

In order to further constrain  $CH_4$  occurrence in the Fennoscandian Shield, we performed new isotopic and molecular composition analyses of gas and measured  $CH_4$  flux from two deep boreholes in Eastern Finland, Outokumpu Deep Drill Hole (2516 m, completed in 2005) and Juuka 116 (1118 m, drilled in 1982). Both sites have mica schist, black schist and serpentinized ultramafics as the main rock types, and visible bubbling of gases at the wellhead. In addition, flux measurements were performed in springs located at or close to major fracture zones and serpentinites in the Outokumpu-Jormua Ophiolite Belt, which could potentially provide natural source and paths for crustal  $CH_4$  to the surface.

Based on closed chamber measurements at the wellhead, about 40,000 liters of  $CH_4$  are released from the Outokumpu and Juuka wells to the atmosphere every day ( $\sim 10$  tons/year). These fluxes are considerably high, taking into account the age and organic carbon deprived nature of the highly metamorphosed crystalline bedrock. The studied springs do not carry crustal  $CH_4$  (only  $^{12}C$ -enriched microbial  $CH_4$ , likely related to wetland ecosystem, was found), which suggest that their hydrologic circuits are shallower than gas reservoirs. This is also inferred by the distinct  $^2H$ - and  $^{18}O$ -enriched isotope composition of crustal  $CH_4$ -related groundwaters compared to spring waters.

Isotope analysis of CH<sub>4</sub> released at the wellhead of the Outokumpu ( $\delta^{13}$ C -42.34‰,  $\delta^{2}$ H -287.0‰) and Juuka ( $\delta^{13}$ C -28.59‰,  $\delta^{2}$ H -281.5‰) boreholes are in line with previous measurements (Kietäväinen et al. 2017), even those acquired more than 20 years ago (Sherwood Lollar et al. 1993). Methane flux and stability of its isotope composition suggest that the gas source did not change over time and a pressurized gas reservoir must exist to support the strong emissions along the boreholes to the surface.

## References:

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