



Evidence for a biological origin of isotopically heavy hydrocarbon gases in alkaline intrusions

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The occurrence of isotopically heavy methane, $\delta^{13}\text{C} > -10\text{‰}$ in alkaline intrusions notably the Khibina and Lovozero massives on the Kola Peninsula, Russia and the Ilimaussaq complex, SW Greenland, has played a major role in the theory of abiogenic hydrocarbons of magmatic origin [1]. Higher hydrocarbon gases plus disseminated bitumen also present in the rocks were hypothesized to have formed by condensation reactions during a late stage magmatic process [1, 2], though stable carbon isotopic ratio $\delta^{13}\text{C}$: $-32 - -24\text{‰}$ indicated a biological origin of the bitumen [3, 4]. Furthermore, biomarkers present in the extracts of bitumen in the Ilimaussaq rocks supports the assumption of a biological origin [5]. Specific biomarker analysis using GC/MS/MS indicate that the bitumen may be a residue from oils that entered the rocks of Ilimaussaq during or after the Late Cretaceous, when the area was most likely covered by a thick pile of sediments according the thermal history of nearby rocks [6]. Given the unusually high hydrocarbon gas content in certain minerals corresponding to 500 ppm of carbon and the relatively high proportion of higher hydrocarbon gases up to 20 percent plus the strong evidence for a biological origin of the bitumen it seems fair to assume that the hydrocarbon gases too are organic in origin, though the heavy isotopic data of methane remains to be explained. It has previously been suggested that isotopic fractionation caused by diffusion of gases out of the rocks may account for the heavy isotopic signature of the remaining methane [5]. Fractionation by diffusion could also explain the higher isotopic values of methane compared to ethane and propane. The fractionation-by-diffusion hypothesis has been supported by recent studies of gas release from rock samples of the two intrusions on the Kola Peninsula [7]. Furthermore, extending the area of investigation of the Khibina intrusion a wider range of methane isotopic values $\delta^{13}\text{C}$: $-22 - -5\text{‰}$ were observed, which was interpreted as being mixtures of biogenic and abiogenic gases [8]. Biogenic methane ($\delta^{13}\text{C}$: $-43 - -35\text{‰}$) in the Ilimaussaq intrusion has only been observed in fluid inclusions hosted in quartz [9], which may be considered more robust with respect to loss of gas compared to the type of minerals hosting fluid inclusions with isotopically heavy methane. Thus, it is concluded that the hydrocarbon gases are biological in origin likely to have entered the alkaline rocks during burial beneath sedimentary rocks. Later, partial loss of gas by diffusion probably during uplift and erosion was responsible for the occurrence of isotopically heavy methane, previously taken as evidence of a magmatic origin. Finally, the higher abundance of hydrocarbons in the alkaline intrusions compared to other rocks in the above areas is more likely due their higher potential for entrapment of circulating fluids due to the particular mineralogy of alkaline rocks, rather than specific redox conditions during their magmatic evolution.

[1] Petersilie & Sørensen (1970) *Lithos* **3**, 59-76. [2] Chukanov et al. (2006) *Geochemistry International* **44**, 715-728. [3] Konnerup-Madsen et al. (1988) *Bull. Minéral.* **111**, 567-576. [4] Galimov (2006) *Org Geochem* **37**, 1-63. [5] Laier & Nytoft (1995) *Proceedings of the 17th International meeting of Organic Geochemistry (San Sebastian)*, 1109-1111. [6] Laier & Nytoft (2009) *Goldschmidt Conference Abstracts (Davos)*, A715. [7] Nivin (2009) *Geochemistry International* **47**, 714-733. [8] Beeskov et al. (2006) *Lithos* **91**, 1-18. [9] Graser et al. (2008) *Lithos* **106**, 207-221.