

Estimating hydrocarbon gas fluxes through sedimentary basins with chemical proportions and stable isotopes fractionations

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Gases in sedimentary basins have two main origins. They can derive from deeper formations (mantle, deeper crust) or be produced *in situ* via thermal cracking or biological activity. Interactions between gases and sediments result in qualitative and quantitative changes that influence the mass balance of gases in sedimentary basins and have the potential to affect our understanding of global geochemical cycles. Estimating a gas budget in sedimentary basins has two basic interests: a better control of gas accumulation, with an energetic target, and an important clue in the global carbon budget in the Earth, with an environmental target.

In the majority of studied sedimentary basins, the mass of accumulated hydrocarbon gas in reservoirs is smaller by one or more order of magnitude than the estimate for hydrocarbon generated in source rocks. Causes for this poor yield may result from various processes taking place during genesis, expulsion, migration, but it is more likely the result of alteration and leakage from the sedimentary rocks. As gas caprocks are never absolute seals at geological time scales, leakage from reservoirs may be the main explanation for the poor yield of gas accumulation. Leakage may have considerable weight on the proportion of reduced carbon from sedimentary basins oxidized to CO₂ and recycled back to the atmosphere. A natural gas reservoir has then to be seen as an accumulation due a delay during gas transfer from its source to the atmosphere, rather than an absolute trap, not allowing the gas molecules to go further up.

Gas segregation and quantification of gas leakage

Geochemical gas characterization deals always with the problem of the significance of both chemical and isotopic signatures, in terms of genesis, transport and alteration. Several works have raised the problem of possible isotopic fractionation of methane during migration through porous media: Fuex (1980),

claiming that no significant fractionation could affect methane during transport, supposedly solved this debate. However, experiments (Hoering and Moore, 1958; May *et al.*, 1968; Galimov, 1975; Bondar, 1987; Pernaton *et al.*, 1996) and geological observations (Colombo *et al.*, 1970; Friedrich and Jüntgen, 1972; Neglia, 1979; Prinzhofer and Pernaton, 1997) have shown that some fractionation does occur in specific geological situations. The involved physical processes may be solubilization/diffusion of methane in water (Galimov, 1975; Pernaton *et al.*, 1996), or adsorption/desorption of methane from a solid network (Colombo *et al.*, 1970; Friedrich and Jüntgen, 1972). The migration of gases through porous rocks rises the question of the relative importance of gaseous Darcian flux, selective solubilization between fluid phases, diffusion, interactions between fluid and solid phases, etc...

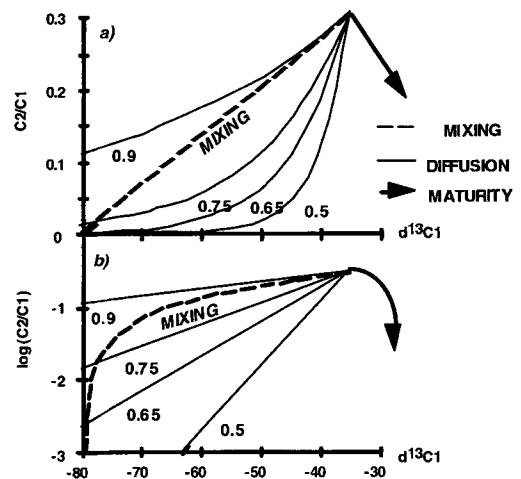


FIG. 1. Calculation of mixing and diffusion trends in a diagram C₂/C₁ versus δ¹³C₁, in linear and semilogarithmic scales (from Prinzhofer and Pernaton, 1997).

A C2/C1 diagram versus the $\delta^{13}\text{C}$ of methane enables to distinguish these three possible trends (Prinzhofer and Pernaton, 1997): a mixing trend between two end members (for example thermogenic and bacterial end-members) would give an almost straight line in linear scales. On the contrary, a diffusive fractionating migration would give a concave-shaped curve in the same linear scale (Fig. 1a). In a semi-logarithmic scale (Fig. 1b), the segregative trend will give apparent straight lines, when the mixing trend will be convex. For a pure maturity trend, the slope is always reversed to both bacterial contamination and segregation (Fig. 1a and b). Assuming a knowledge of the isotopic fractionation of methane according to the stratigraphic series, it is possible to calculate, from the Fick's law, the proportions of gases migrating through this kind of process.

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