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Microseepage in drylands: Flux and implications in the global atmospheric source/sink budget of methane

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ABSTRACT

Drylands are considered a net sink for atmospheric methane and a main item of the global inventories of the greenhouse gas budget. It is outlined here, however, that a significant portion of drylands occur over sedimentary basins hosting natural gas and oil reservoirs, where gas migration to the surface takes place, producing positive fluxes of methane into the atmosphere. New field surveys, in different hydrocarbon-prone basins, confirm that microseepage, enhanced by faults and fractures in the rocks, overcomes the methanotrophic consumption occurring in dry soil throughout large areas, especially in the winter season. Fluxes of a few units to some tens of mg m⁻² day⁻¹ are frequent over oil-gas fields, whose global extent is estimated at 3.5–4.2 million km²; higher fluxes (>50 mg m⁻² day⁻¹) are primarily, but not exclusively, found in basins characterized by macro-seeps. Microseepage may however potentially exist over a wider area (~8 million km², i.e. 15% of global drylands), including the Total Petroleum Systems, coal measures and portions of sedimentary basins that have experienced thermogenesis. Based on a relatively large and geographically dispersed data-set (563 measurements) from different hydrocarbon-prone basins in USA and Europe, upscaling suggests that global microseepage emission exceeding 10 Tg year⁻¹ is very likely. Microseepage is then only one component of a wider class of geological sources, including mud volcanoes, seeps, geothermal and marine seepage, which cannot be ignored in the atmospheric methane budget.

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1. Introduction

Dryland soil is considered a net biotic sink of atmospheric methane, with a global uptake on the order of 30 ± 15 Tg year⁻¹ (IPCC, 2001) or 20 ± 3 Tg year⁻¹ (Potter et al., 1996). There are however projections falling within the range of 5–58 Tg year⁻¹ (Dorr et al., 1993), indicating that there is still substantial uncertainty over the magnitude of this global sink. The negative gas flux, generally on the order of -5 to -1 mg m⁻² day⁻¹ (Dong et al., 1998), is due to methanotrophic oxidation by CH₄-consuming bacteria in the soil. Methanotrophic oxidation occurs in grassland, temperate and boreal forest soil, desert soils, fertilized soil, humisol, moss-derived peat soils, tundra soils and unflooded paddy soils (Minami and Takata, 1997). The soil is considered a source of methane only in wet conditions, in the presence of methanogenic bacteria (in all wetlands, including rice paddies, bogs and flooded soils; Batjes and Bridges, 1994).

In the 1980s and 1990s, some anomalies with respect to the expected dryland behaviour (i.e., positive fluxes instead of negative fluxes) were found in South America. Unexpected emissions of methane

into the atmosphere $(>1 \text{ mg m}^{-2} \text{ day}^{-1})$ were found in two dry grasslands or savanna soils within the Orinoco Valley and in the Guyana Shield of northeastern Venezuela (Hao et al., 1988: Scharffe et al., 1990). These measurements were criticised and considered erroneous by other researchers, as the authors had no explanation for the positive methane flux (Crutzen, personal communication). Hao et al. (1988), however, suggested the possibility of gas release through upward diffusion from underground natural gas reservoirs near Chaguarama, which is in the region investigated. Hao et al. (1988) were quite right as their "biological" survey was actually conducted over what some years later would be recognized as one the largest petroleum systems in the world (the Orinoco Petroleum Belt; Erlich and Barrett, 1992). The area investigated by Scharffe et al. (1990), near the Guri dam, south of the Orinoco Belt, is located in association with important SW-NE deep fault systems containing highly fractured and permeable mylonites, characterising the regional brittle tectonics of the Guyana Shield (Bellizzia et al., 1976). This location apparently has deep-sourced gas migration processes operating.

Later, positive fluxes of methane in dry lands were reported in several sites in the USA in the framework of studies on hydrocarbon seepage from sedimentary basins (Klusman et al., 1998, 2000a). Indeed, the occurrence of methane and light alkane anomalies in dry soil has been extensively used by geologists and geochemists as a

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Fig. 1. Sketch of the microseepage process, from a hydrocarbon reservoir, through the unsaturated zone, to the atmosphere.

tool for oil and gas exploration since 1930s (Laubmeyer, 1933; then, more recently: Jones and Drozd, 1983; Davidson, 1986; Schumacher and Abrams, 1996; Klusman, 1993; Tedesco, 1995; Hunt, 1996; Matthews, 1996; Schumacher and LeSchack, 2002; Abrams, 2005). Indirect methods, such as microbial prospecting (e.g., Tucker and Hitzman, 1996; Wagner et al., 2002), remote sensing (e.g., Van der Meer et al., 2002) and magnetic measurements (e.g., Liu et al., 2004) have also shown the existence of microseepage throughout large areas over oil-gas fields on various continents. Nevertheless, these studies focused exclusively on the detection in the soil of anomalous concentration of methane and light alkanes (and associated geophysical or geochemical indicators); the soil-atmosphere flux measurement, being not necessary for oil/gas exploration, was never carried out. Understanding the impact on the atmosphere was not an objective. Consequently the available data-set on microseepage flux is rather poor. Only recently, since 2002, a large number of flux data have been acquired throughout dry soil areas in hydrocarbon-prone sedimentary basins of Europe and Asia, specifically in Italy, Romania, Greece, Azerbaijan and China (Etiope et al., 2002; Etiope et al., 2004a,b; Etiope et al., 2006; Tang et al., 2007; Tang et al., 2008). These surveys and the US surveys performed by the Colorado School of Mines (e.g., Klusman, 2006; and references therein) form the only systematic programme of measurements of microseepage flux to the atmosphere.

Today, it is known that the positive flux of methane, or microseepage, can reach levels of tens, hundreds and thousands of mg m⁻² day⁻¹ throughout large areas, especially around macro-seeps such as occur associated with mud volcanoes (Etiope et al., 2004a,b; Etiope and Milkov, 2004). At lower rates, microseepage is quite common and pervasive within petroliferous and sedimentary basins.

Finally, positive fluxes of methane from the soil can also occur in geothermal areas (Hernandez et al., 1998; Etiope, 1999; Klusman et al., 2000b; Etiope et al., 2007a), where methane is produced by high temperature inorganic reactions (Etiope and Klusman, 2002).

All these facts pose some key questions:

- 1) Has the occurrence of microseepage ever been considered in the estimates on global soil sink?
- 2) How large is the dryland area potentially affected by microseepage?
- 3) How large is the global microseepage emission into the atmosphere?
- 4) What are the implications on the global greenhouse gas budget?



Fig. 2. Photos of the closed-chamber systems used in microseepage surveys in USA (a) and Europe (b, c). Methane concentration increase is measured by GC analysis of samples taken manually by syringes (a, b) or by direct detection by laser sensor (c).

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