Natural CH 4 gas seeps in the French Alps: characteristics, typology and contribution to CH 4 natural emissions to the atmosphere

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Abstract

Quantification of greenhouse gases that naturally leak into the atmosphere has to be considered in lien with mitigation of CO_2 emissions, underground storage of CO_2 and conventional and unconventional gas exploitation. Among the long lived greenhouse gases, methane is one of the most critical specie regarding its global warming potential, far greater than the one of CO_2. Methane can be released in the atmosphere through human activities (agricultural practices, landfills) but it can also be released from natural processes occurring *e.g.* in wetlands. Another source is constituted by natural gas seeps existing all over the world. Their contribution has been quantified in Europe to account for about a third of the yearly methane emissions (Etiope, 2009, Atmospheric Environment 43, 1430-1443).

The global methane budget still needs completion and refining, taking into account undocumented macro-seeps and, in particular, micro-seeps, difficult to measure and to account for. Such unreported seeps are located in the French Alps, near Grenoble. Although some of these emanations are known since Roman times, they have not been investigated since, except in the early XXth century for potential industrial exploitation of CH_4. Here we report first investigations at the *Fontaine Ardente* (Le Gua) and the *Rochasson* (Meylan) natural gas seeps. Investigations were designed to understand the patterns of gas leak and obtain samples for isotope characterisations. The present study is focused on site investigations.

These two gas seeps are set within Middle Jurassic clays formations along the eastern border of the large synclinal structures of the Vercors and the Chartreuse massifs. The *Fontaine Ardente* seep occurs in a small thalweg and has been intensively modified in the past in the aim to protect the main vent from the small stream flowing nearby. The *Rochasson* gas seep has been discovered only recently (40 years ago) and is probably the consequence of a small landslide that occurred in the clayey formations.

Investigations on site include soil gas quantification in the field using portable infrared gas analysers for CO_2, CH_4 and O_2 concentrations. Subsequent characterisation of the gas phase was done once a day using gas chromatography (N_2, Ar, helium and alkanes up to C_3H_8). Some tentative

evaluations of the CH_4 and CO_2 fluxes were done using a plastic funnel and the record of gas accumulation into this pseudo-chamber.

The gas of *Fontaine Ardente* seep is CH_4-rich (84%) with significant contribution of CO_2 (11.5%) and minor contribution of air or deep-seated nitrogen (4% N_2 and <1% O_2). Traces of C_2H_6 are present (max. 0.06%) together with helium (max. 0.025%). The gas of *Rochasson* seep has nearly the same composition in CH_4, O_2 and N_2, but is depleted in CO_2 (<0.5 %), enriched in C_2H_6 (2%) and in C_3H_8 (0.33%). Rough evaluation of the gas flux at the mouth of the seeps suggests that fluxes may reach 130 kg/day/m² of CH_4 and 38 kg/day/m² of CO_2 at *Fontaine Ardente* and 4.3 kg/day/m² of CH_4 at *Rochasson*. The value for *Fontaine Ardente* seems to be in reasonable agreement with previous evaluation of the leakage rate established around 130 l/min of CH_4 during borehole drilling in the 90's. As such, this gas seep can be considered as an important one regarding the database established by Etiope (2009).

Even if high fluxes are measured at the mouth of the gas seeps, leakage of CH_4 – and CO_2 – is not spatially restricted to the vent itself. Soil gas investigations allowed defining at very detailed scale (1 m grid) the geometry of gas emanation.

For *Fontaine Ardente,* it is possible to define an area of several square meters (6x4 m) where CH_4 concentrations at 1 m depth exceed 70%. In the centre, CH_4 concentrations are greater than 80%, with high CO_2 (> 10%), the presence of C_2H_6 and a strong depletion in O_2 and N_2. The replacement of one gas phase by another is not homogeneous even at such small scale. Samples taken near the gas seep show CH_4/O_2, CH_4/N_2 and CO_2/O_2 relationships suggesting that O_2 and N_2 replace CH_4 and CO_2. For each mole of CH_4 introduced in the system, 1/5 mole of O_2 and 4/5 mole of N_2 disappear, balanced by CO_2 (1 mole of CO_2 introduced and 2 moles of O_2 disappeared). With some distance (a few meters) from the seep, the relation is different as CH_4 concentrations remain under 8% while O_2 concentrations decrease from 21% to 8%. Once this pivot point achieved, then the CH_4 concentration rises exponentially. This second mechanism is not yet fully understood – nonetheless, points affected by this process are distributed in a halo shape suggesting that the seep, with high CH_4 flux, sweeps away atmospheric gases, whereas with some distance, there may be some mixing with soil gas. Halo distributions are known around some CO_2 gas vents (Gal *et al.*, 2011, IJGGC 5, 1099-1118).

Situation is different for the *Rochasson* seep. The CH_4 + C_2H_6 vent is narrower (2x1 m) and associated with strong O_2 and N_2 depletion. High CO_2 concentrations are not measured above the vent but few meters away. This may be the consequence of a CO_2-O_2 molar replacement without CH_4 influence, a process different from the mechanism found for *Fontaine Ardente*.

These seeps will be investigated in more details, to better quantify fluxes to the atmosphere and to better describe underlying processes. Another challenge will be to evaluate if other unknown macro- or micro-seeps can be discovered in this region, to evaluate if natural degassing of Callovian-Oxfordian shale is restricted to the known point sources or active at wider scale.

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