Geophysical Research Abstracts Vol. 17, EGU2015-12455, 2015 EGU General Assembly 2015 © Author(s) 2015. CC Attribution 3.0 License.



## Removal of atmospheric methane in shallow subterranean environments

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Methane (CH4) is considered as the third most important greenhouse gas, after water and carbon dioxide, contributing substantially to radiative forcing. About 90% of the removal of CH4 from the atmosphere occurs through reaction with hydroxyl radicals. Moreover, secondary methane sink is related to soils by microbial oxidation in the aerobic zone of soils. Our monitoring results in subterranean environments have shown that there is an active remove of atmospheric methane without a significant intervention of methanotrophic bacteria. Several caves were monitored to identify the environmental factors controlling the gases exchange (CH4, CO<sub>2</sub> and 222Rn) between subterranean environments, soils and atmosphere. Real-time and spots measurements of these greenhouse gases were measured using a cavity ring-down spectroscopy (CRDS) technique. Our results determine that concentrations of 222Rn and CO<sub>2</sub> rise during the period of cave isolation (barely any exchange with the exterior atmosphere), contrary to the methane concentration decrease. The subterranean methane concentration was usually lower than the atmospheric and soil mean values. In addition, zero methane concentrations (ppm) were registered during several months in the most isolated caves. Our hypothesis is that an active process of methane oxidation is occurring in the underground atmosphere, akin to the photolysis effect that occurs in the troposphere-stratosphere region. Thus, negative and positive ions were measured inside the subterranean atmospheres to verify the correlation between the ionization by the 222Rn alpha particle decay and to the depletion of methane concentration. High negative correlations between negative ions and methane were obtained. Therefore, it is suggested that the oxidative gases  $(CO_2, O_2, H_2Ov...)$ , presented inside the subterranean environment, would be ionized by the energy released by 222Rn alpha particle decay, reacting and, consequently, oxidizing the atmospheric methane content.