

Observation-based constraints on the present-day atmospheric methane budget

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The present-day methane source/sink budget is still poorly quantified given the unexpected and still largely unexplained decadal decline and, recently, possible recovery of the global annual growth rate of atmospheric methane. At the same time different emission estimates for present-day and past times circulate in the scientific literature. Scientific understanding can be enhanced by a better integration of knowledge that is being acquired in different disciplines, including geology, geochemistry, biology, ecology, isotope studies, and atmospheric chemistry, and by optimally combining past and present-day ice core data of methane and its stable isotopes with present-day spatio-temporal surface-based observations and satellite remote sensing. Concerning the present-day methane budget we need to better constrain the relative contributions of anthropogenic emissions (A) and present-day net natural fluxes (N) into the atmosphere, including their geographical and seasonal variations, and secondly, determine how the recently observed variations in the methane annual growth rate relate to trends and inter-annual changes in atmospheric lifetime (T), N, and A, respectively.

In this presentation we present budget constraints from a two-step approach using (i) optimal estimation of published present-day bottom-up emission estimates, and (ii), detailed spatio-temporal evaluation of forward and inverse global chemistry-transport model simulations. Using an optimal estimation technique based on the Ensemble Kalman Filter South-Pole concentrations and isotope fractionations for present-day, the pre-industrial, the pre-agricultural, and the Last-Glacial Maximum are combined with (uncertainties in) emission and sink isotope fractionations. We estimate the most likely present-day natural and anthropogenic global contributions from biogenic, fossil, and pyrogenic processes, respectively. The optimized scenarios, still fundamentally different, all satisfy past and present-day observations. In a next step we use surface-based and SCIAMACHY satellite observations of CH₄ for the year 2004 to evaluate global chemistry-transport simulations fed by natural sources and sink distributions depending on actual meteorology. Atmospheric lifetime is derived and constrained by multi-species model evaluations. Inverse modelling finally provides optimized categorized spatio-temporal methane emission distributions for the year 2004.

Raman spectroscopic characteristics of carbonaceous material in Archean rocks; Implications for early life studies

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Despite a range of *in situ* analytical techniques that enable detailed chemical and isotopic characterization of microscopic organic structures, the interpretation of ancient microfossils and other remnants of early life in Archean rocks has remained notoriously controversial. This is in large part due to the combined effects metamorphism, which caused thermal alteration and migration of biologic compounds as kerogen or graphite. Particularly difficult is the interpretation of carbonaceous structures in highly metamorphosed terrains (T>500°C), since COH-fluids could have directly produced graphite-filled fluid-inclusions, graphene-films on mineral catalyst surfaces, or highly disordered carbon at crystal boundaries within the rock matrix. Confocal Raman spectroscopy is a particularly useful technique for the recognition of all these features, as it enables the characterization of carbon structure within the direct mineral framework of the rock. Example cases will be discussed of migrated forms of kerogen in 3.3 Ga black cherts from the Barberton Greenstone Belt (South Africa), distinct pools of kerogen (syndimentary and hydrothermal) in a drill core from the 3.5 Ga Dresser Formation (Pilbara, Western Australia), and epigenetic graphite in fluid inclusions and disordered carbon on grain boundaries in a 3.8 Ga quartz-pyroxene rock from Akilia Island (Greenland).