

Atmospheric Methane Contributions from Fractured Bedrock Aquifers

D.L. Marrin, Principal Investigator

Along with carbon dioxide (CO₂), nitrous oxide (N₂O) and chlorofluorocarbons (CFCs), methane (CH₄) concentrations in the atmosphere have increased markedly since the industrial revolution. These so-called greenhouse gases, along with water vapor, are generally considered to be the major players in the current global warming trend. Just how much of this warming is due to anthropogenic contributions has not been resolved; however, recent increases in the atmospheric concentration of these gases are undeniable. The bulk of the atmospheric methane increase is attributed to [i] the burning of fossil fuels and biomass, [ii] the cultivation of rice and other flooded crops, and [iii] emissions from domesticated animals and landfills.

Groundwater is not generally considered to be an important contributor of atmospheric methane for two reasons. First, the organic carbon content of groundwater is normally lower than that of either surface waters or shallow soils; hence, there is often insufficient degradable carbon to sustain methanogenesis. In other words, the limited supply of organic carbon permits more oxidative biodegradation processes such as iron and sulfate reduction to prevail. Secondly, any methane that is produced in groundwater must partition into the overlying soil gas and diffuse through the unsaturated zone toward the atmosphere. If the soils overlying groundwater aquifers are relatively dry (i.e., moist but not saturated), they act as a fairly efficient methane sink. Methanotrophic microorganisms in soils are able to oxidize the methane to carbon dioxide, which is then released to the atmosphere as the major biogenic gas associated with subsurface biodegradation processes.

A current research project suggests that there are exceptions to the dogma regarding atmospheric contributions of methane by groundwater aquifers. These exceptions are related to both post-industrial human practices and to specific hydrogeologic settings. The normally limited supply of degradable organic substrates to groundwater has been augmented by the post-industrial practice of storing and transporting liquid petroleum products in the subsurface. This practice has resulted in the local introduction of organic carbon at concentrations that are orders-of-magnitude higher than the natural background. Not only are these petroleum products excellent substrates for biodegradation, but their ability to affect localized redox conditions favors methanogenesis over more oxidative biodegradation pathways. If the methane-producing groundwater is overlain by fractured bedrock, rather than by unconsolidated porous media, the opportunity for methanotrophic organisms to oxidize the methane is diminished. Upward movement of methane through discrete fractures limits the volume of soil into which CH₄ diffuses, thus reducing the surface area available for methanotrophic activity.

Preliminary results indicate that petroleum-contaminated groundwater in fractured bedrock settings may create another situation where post-industrial practices have led to an increase in the atmospheric loading of CH₄. The spatial distribution of methane in thin surface soils overlying bedrock suggests that methane flow through fractures or fracture networks does indeed occur. Moreover, a comparison CH₄ and CO₂ flux rates from groundwater with those from the ground surface indicates that while most of the aquifer-produced methane is oxidized before reaching the atmosphere, the oxidation efficiency is at least a factor of 50 less in these fractured rock settings than in most unconsolidated soils.

Atmospheric flux rates associated with petroleum-contaminated aquifers were on the order of several grams of carbon (as CH₄) per square meter, which is substantially less than that reported for major sources such as rice paddies. Nonetheless, these production rates represent a considerably higher flux than that associated with atmospheric methane consumption by soils. Atmospheric methane consumption refers to a process whereby the flux of CH₄ is actually into (rather than out of) soils, which constitute a sink as a result of resident microorganisms that perform biologically-mediated CH₄ oxidation. Although atmospheric methane contributions from contaminated aquifers overlain by fractured rock are probably minimal on a worldwide basis, they may constitute an important localized source and they most certainly have the potential of shifting soils from a sink to a source of atmospheric methane. Perhaps most interestingly, this research indicates a mechanism by which petroleum products increase the loading of methane (specifically) and greenhouse gases (generally) to the atmosphere without ever being combusted.