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Atmospheric methane sources linked to anaerobic hydrocarbon degradation in sedimentary basins during Pleistocene glaciation.

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Methane is an important greenhouse gas and amplifier of climate change. Nevertheless, direct link between subsurface biogeochemical processes and atmospheric methane variations over glacial-interglacial cycles remain unresolved. Several terrestrial sources of methane have been proposed, including increases in emissions due to expansion of wetlands and decomposition of methane clathrates, however, here we consider a previously unrecognized source of atmospheric methane derived from the anaerobic biodegradation of sedimentary organic matter. This atmospheric source of methane may have been previously overlooked in current isotope mass balance models of atmospheric methane variability due to similar isotopic compositions measured for emissions from tropical wetlands. We propose that microbial methanogenesis occurring along the margins of sedimentary basins provides a source of atmospheric methane temporally coupled to the advance and retreat of continental ice sheets. Extensive anaerobic hydrocarbon biodegradation in the Late Devonian Antrim Shale, Michigan, U.S.A., is the result of an active subsurface consortium of fermentative and methanogenic microorganisms directly related to Pleistocene glaciations. The generation and release of this biogenic methane was initiated by the advancement of Pleistocene ice sheets driving glacial meltwater into the Michigan Basin. This meltwater incursion diluted the saline basinal brines stimulating the production of biogenic gas and biodegradation of the host sedimentary organic matter. Hydrostatic pressure created by the overriding ice sheet allowed for the accumulation and trapping of the

produced gas. Calculated estimates suggest that gas accumulated at a rate of approximately 1 Tg methane per 1000 years. Ultimately, ice sheet melt and retreat permitted the release of the accumulated biogenic gas to the atmosphere. Our estimates suggest that at present the Antrim Shale only contains 12 – 25% of the cumulative mass of methane generated over the Pleistocene, indicating that the methane that had accumulated during glaciation has been released. While this volume of gas is only a small portion of the global atmospheric budget, and represents a small portion of the geologic sources of atmospheric methane, the Antrim Shale is one of a number of sedimentary basins hosting methane reserves that were overridden by Pleistocene ice sheets. The cumulative effect of these basins may be a major previously unrecognized source of methane emissions to the atmosphere on glacial-interglacial timescales.