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Ice sheet related glacial/interglacial cyclicity of granitic tetrafluoromethane (CF_4) emissions before and after the Mid Brunhes

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CF_4 is a long-lived atmospheric trace gas that was thought to be emitted only by anthropogenic processes. However, small quantities of CF_4 are released from a natural source – chemical weathering of granitic rocks generate an atmospheric background concentration that is archived in polar ice. We measured CF_4 concentrations over the last 800 kyr and used an inversion to calculate CF_4 emission fluxes. We consistently found higher CF_4 fluxes for each interglacial, resulting in an increase of atmospheric CF_4 concentrations, while glacials show lower CF_4 fluxes and declining CF_4 concentrations. Different processes might be responsible for this pattern. First, higher CF_4 fluxes during warm conditions are expected as chemical weathering rates are known to increase with temperature and precipitation. Second, granitic rocks are not randomly distributed but preferentially located in high northern latitudes which are largely covered by continental ice sheets and permafrost during glacials inhibiting CF_4 release as weathering requires liquid water and a connection to the atmosphere. Thus, the waxing and waning of the northern hemispheric ice sheets has a larger leverage on CF_4 fluxes than expected from the area alone. Interestingly, the peaks of the CF_4 emission fluxes occurred at the starts of the interglacials. Our interpretation is that moraines left behind at the southern fringes of the retreating ice sheets provide easily weatherable material under already warm conditions. Conversely, from the late interglacials throughout the glacials we observe drops in CF_4 concentration. The minima of both CF_4 concentrations and CF_4 fluxes are located at the end of the glacials, i.e. before the deglaciations started. This observation helps to assess the activity of glaciers via their erosional grinding of bedrock which produces suspended fine materials, so-called "glacier flour". Because the mineral fluorite, which is typically enclosing CF_4 within the granite rock, is highly soluble in water, CF_4 would be quickly released after grinding since it should occur in wet conditions. Our data suggest that this process is small compared to the suppression of granite weathering via ice coverage, otherwise the maxima in CF_4 fluxes should have been found during glacial maxima.

On the long-term, our record reveals a marked rise in CF_4 fluxes after the Mid Brunhes event (MBE). Beginning with MIS 11, the first strong interglacial after a series of weak interglacials, the glacial/interglacial amplitudes in CF_4 emissions but also for CO_2 and ice volume increased. For the

430 kyr after the MBE the reconstructed CF_4 fluxes increased by ca. 8%, predominantly due to increasing interglacial emissions, especially for MIS 5, 9, 11. We discuss three possible scenarios for this post-MBE rise in granite weathering: First, higher temperatures in northern high latitudes. Second, the exposure of granitic rocks that was ice covered during previous weak interglacials. Third, a remaining fraction of the former regolith covering large parts of North America was eroded during MIS 12 initiating the climatic changes associated with of MBE.