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Methoxyl groups of plant pectin as a precursor compound for atmospheric methane: evidence from deuterium labelling studies.

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It is well known that plants emit a broad range of volatile organic compounds (VOCs) such as isoprenoids and oxygenated compounds (e.g. methanol and acetone) to the atmosphere (Kesselmeier & Staudt, 1999). However, before it was not known that plants produce methane in an oxygen rich environment and release it to the atmosphere. We recently have demonstrated that intact living plants, as well as plant litter, produce methane under aerobic conditions (Keppler et al., 2006). We also observed that methane emissions were sensitive to both temperature and natural sunlight irradiation. The observation that plants produce methane (CH_4) under aerobic conditions has caused considerable controversy in the scientific community and the general public. It not only led to much discussion and debate on its contribution to the global CH_4 budget but also on the authenticity of the observation itself (e.g. Schiermeier, 2006, Evans, 2007, Dueck et al., 2007, Kirschbaum et al., 2006). However, on the other hand, a very recent study by Wang et al. (2007) reported emissions of CH_4 from several shrubs of the Mongolian Steppe confirming the finding of aerobic methane formation in plants. Apart from the observation that vegetation releases methane the mechanism of its formation remains unknown. Elucidation of the underlying biochemical pathway for aerobic CH_4 is crucial for gaining full understanding of its role and possible global significance.

As a first step it is important to gain information about precursor compounds in plants that could give rise to CH_4. Based on previous results we suggested the possibility of the involvement of the methyl moiety of the esterified carboxyl group (methoxyl group) of pectin (Keppler et al., 2004). Indeed, in experiments with apple pectin we not only observed emission of CH_4 but also noted that the emission rate was broadly similar to that measured with detached leaves (Keppler *et al.*, 2006). However, even though those results indicated a role for pectin they provide no proof for the involvement of the pectin methoxyl group in CH_4 formation.

We now have investigated pectin and polygalacturonic acid with varying degrees of deuterium labelled esterified methyl groups to prove our hypothesis that methoxyl groups are indeed a source of methane in an oxic environment. The stable hydrogen isotope values of the methane released in experiments with deuterium labelled compounds were very different from those which were non-labelled. Our results provide unambiguous isotope evidence that methoxyl groups of pectin can act as a source of atmospheric CH_4 under aerobic conditions. As previously shown (Keppler et al., 2006) emissions of CH_4 from pectin are strongly dependent on temperature and exposure to light, in particular in the UV range. Although the mechanism is still unknown our study is an important first step to gain more information about potential plant precursor components and thus to start investigating the reaction mechanism which is needed to understand the environmental importance of aerobic methane formation.

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