

spermathecal sac (*B* in the figure).

It was impossible to estimate the number of sperm cells for comparison between the control and group 1, but it is unlikely that sperm had been removed in the course of the short copulations because the inseminatory canal is very narrow, about the diameter of a sperm cell⁴, and relatively long.

We doubt that there was a transfer of some substance that could selectively incapacitate the sperm of other males⁵ or cause its removal by the female⁶ during the first phase of copulation. Such substance, if deposited by the first four males in group 2, should have remained active and caused considerable decrease in female fecundity. But this was not the case (see table).

We conclude that during copulation in *C. berlessei* (and, presumably, in other acarid mites) males first transfer some substance which can push the previously deposited sperm away from the basal part of the spermatheca and the efferent

ducts leading to the ovaries, and only then sperm is transferred. The chance that sperm repositioned and dispersed in the spermatheca will reach the efferent ducts again must be very low. It seems possible that this sperm is decomposed and finally resorbed by microvilli rich walls of the spermatheca⁴.

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Terpenes and forest decline

SIR - Becker *et al.* imply¹ that reactions of terpenes with ozone could have a more important role in the formation of hydrogen peroxide than the disproportionation reaction of peroxy radicals, thereby contributing to forest decline. The importance of the oxidation of terpenes to the atmospheric H₂O₂ budget can be estimated by using the authors' reported molar yields and terpene fluxes from coniferous forests. Lamb *et al.*² reported α -pinene fluxes of 1.15×10^{-5} mol m⁻² h⁻¹ for a Loblolly pine forest (*T* = 30 °C). If one assumes a mixing height of 1 km and that β -pinene, Δ^3 -carene and *d*-limonene fluxes are similar, the resulting H₂O₂ production rate would be less than 6 p.p.t.v. (parts per 10¹² by volume) h⁻¹. The disproportionation of peroxy radical HO₂· + HO₂· → H₂O₂ would produce 25-700 p.p.t.v. H₂O₂ h⁻¹ (*k_r* = 5.5×10^{12} cm³ per molecule s⁻¹; *T* = 25 °C; relative humidity of 50%)³ at HO₂ levels of 10-15 p.p.t.v. (which are thought to be typical midday levels)⁴. Hence these calculations indicate that H₂O₂ production from the oxidation of terpenes may not be as important a source of atmospheric H₂O₂ as Becker *et al.* suggest.

The concentrations of reactants used by Becker *et al.* [*O*₃]₀ = 2-60 p.p.m.v., [alkene]₀ = 5-35 p.p.m.v.) are not relevant to the atmosphere. Concentrations of O₃ in rural areas are generally of the order of 30-70 p.p.b.v.⁵; concentrations of terpenes, both in absolute terms and relative to O₃ levels, in the ambient atmosphere are also much lower. Measurements indicate that total terpene

concentrations in forest air during the summer are of the order of 0.1-10 p.p.b.v.^{3,6}. Highest concentrations are generally found at night since convection and vertical transport are lower; total terpene concentrations at midday are seldom larger than 1 p.p.b.v. Fluxes of terpenes from forests are dependent on various factors, including the plant species, time of year and leaf temperature⁷.

Becker *et al.* also noted that H₂O₂ production increased with addition of water to the reaction chamber. They attributed this to the acid/base catalysed reaction of the Criegee radical with water to form H₂O₂. However, with their experimental design and the blank tests reported, one cannot exclude reactions of water and O₃ on glass surfaces to account for a portion of the increases in H₂O₂ concentrations, even though "rapid" appearance of H₂O₂ was observed. Such reactions are known to cause serious errors in H₂O₂ measurements^{8,9}.

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BECKER *ET AL.* REPLY - Ross *et al.* imply that the reactions of terpenes with ozone do not play an important role in the formation of hydroperoxides in the atmosphere and consequently in forest decline. Their estimation of the atmospheric hydrogen-peroxide budget using total terpene concentrations of about 1 p.p.b.v. is incorrect. The total terpene concentrations at midday are seldom higher than 1 p.p.b.v. because of the high oxidation rates. The estimation cannot be simply deduced from our re-

ported values from the H₂O₂-yield table; these values were measured under high concentrations of O₃ and terpenes, which result in high losses for the Criegee biradical by reactions with other species (such as HCHO and higher aldehydes). The estimation of the H₂O₂ formation rate, resulting from the reaction of the biradicals with water vapour, is only possible by a full kinetic simulation of forest-air chemistry. These simulation calculations must include the emission rates of the terpenes^{10,11}, the rate constants for the reactions R₂COO + H₂O → H₂O₂ and the yield for H₂O₂ formation for the different terpenes.

Ross *et al.* mention that the high concentrations used in our experiments were not relevant to the atmosphere. The relevance to atmospheric chemistry is given not by the concentrations used but by the occurrence of the reaction R₂COO + H₂O → H₂O₂ in atmospheric photo-oxidant systems. The reactions were carefully analysed by evaluating "concentration-independent" kinetic parameters in the case of C₂H₄ and tetramethylethene and O₃. The clearly established new reaction of water with biradicals seems to be an important H₂O₂ source in forest air and can possibly contribute to forest decline.

Heterogeneous formation of H₂O₂ on glass surfaces was ruled out by extensive blank tests. Details of these tests and their consequences for the kinetics of the reaction of the Criegee biradical with water were not reported in our paper¹. Blank tests with even higher ozone concentrations (up to 400 p.p.m.v.) in the presence of water vapour (1-10 torr) have shown no H₂O₂ formation. References 8 and 9 are irrelevant to our work. These describe experiments in which O₃ was bubbled through water. H₂O₂ formation in such systems can lead to interference in wet-chemical methods for H₂O₂ measurement, but certainly plays no role in our experiments. The high H₂O₂-yields recently observed with the wet-fluorescence method in the reactions of biogenic hydrocarbons with ozone in the presence of water vapour¹² are possibly due to such interference. An

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