POSSIBLE OBSERVATION OF THE $^3\mathrm{A'}$ - $^1\mathrm{A'}$ ELECTRONIC TRANSITION OF THE METHYLENE PEROXY CRIEGEE INTERMEDIATE

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It is possible that we have observed for the first time the transition from the ground ${}^{1}A'$ state to the low lying ${}^{3}A'$ state of the methylene peroxy Criegee intermediate, CH_2O_2 , which is otherwise known as carbonyl or formaldehyde oxide. Criegee intermediates are reactive intermediates that are formed in the ozonolysis of olefins in both liquid and gaseous phases with the methylene peroxy intermediate being formed specifically from the ozonolysis of ethene. In the atmosphere Criegee intermediates are formed in reactions that lead to secondary organic aerosols and participate in reactions with SO₂ and NO₂. A peroxy-like spectrum was observed in the near-IR using cavity ringdown spectroscopy after photolysis of a diiodomethane precursor at 248 nm followed by a reaction with O₂. Possible assignment of the spectrum to CH_2O_2 is based on a strong analogy between the electronic structure of methylene peroxy and ozone. However experiments and electronic structure calculations are continuing to positively attribute the spectrum to either CH_2O_2 or CH_2IO_2 which could also be formed by this chemistry.