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Aqueous-phase ozonolysis of methacrolein and methyl vinyl ketone: a potentially important source of atmospheric aqueous oxidants

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Abstract. Recent studies indicate that isoprene and its gas-phase oxidation products could contribute a considerable amount of aerosol through aqueous-phase acid-catalyzed oxidation with hydrogen peroxide (H_2O_2), although the source of H_2O_2 is unclear. The present study revealed a potentially important route to the formation of aqueous oxidants, including H_2O_2 , from the aqueous-phase ozonolysis of methacrolein (MAC) and methyl vinyl ketone (MVK). Laboratory simulation was used to perform the atmospheric aqueous-phase ozonolysis at different pHs and temperatures. Unexpectedly high molar yields of the products, including hydroxymethyl hydroperoxide (HMHP), formaldehyde (HCHO) and methylglyoxal (MG), of both of these reaction systems have been seen. Moreover, these yields are almost independent of pH and temperature and are as follows: (i) for $\text{MAC}-\text{O}_3$, $70.3\pm 6.3\%$ HMHP, $32.3\pm 5.8\%$ HCHO and $98.6\pm 5.4\%$ MG; and (ii) for $\text{MVK}-\text{O}_3$, $68.9\pm 9.7\%$ HMHP, $13.3\pm 5.8\%$ HCHO and $75.4\pm 7.9\%$ MG. A yield of $24.2\pm 3.6\%$ pyruvic acid has been detected for $\text{MVK}-\text{O}_3$. HMHP is unstable in the aqueous phase and can transform into H_2O_2 and HCHO with a yield of 100%. We suggest that the aqueous-phase ozonolysis of MAC and MVK can contribute a considerable amount of oxidants in a direct and indirect mode to the aqueous phase and that these compounds might be the main source of aqueous-phase oxidants. The formation of oxidants in the aqueous-phase ozonolysis of MAC and MVK can lead to substantial aerosol formation from the aqueous-phase acid-catalyzed reaction of H_2O_2 with MAC, even if there are no other sources of oxidants.

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