



## **Nucleation mechanism proposed for boreal forest sites in Finland**

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Ambient nucleation events continuously measured since one decade in Hyytiälä, Southern Finland, are believed to be a surrogate for the large area covered by boreal forests around the globe. Thus, they have a significant impact on climate. Several studies have tried to explain the occurrence by either sulphuric acid nucleation mechanisms (binary, ternary, kinetic) or by ions. Further studies showed a linear or squared correlation of nucleation with sulphuric acid, indicating two competing mechanisms leading to new particle formation. In this study we present an alternative mechanism focusing on the oxidation products of biogenic hydrocarbons, i.e. sesquiterpenes, with ozone and its interaction with sulphuric acid in two stages of the nucleation process. In this mechanism the so-called stabilized Criegee intermediate molecules, which are not scavenged by water vapour, react (i) directly with sulphuric acid forming a water soluble organic sulfate that serves as a nucleation nuclei (NN) for sulphuric acid (quadratic correlation to sulphuric acid). Alternatively, these stabilized Criegee intermediate molecules can react (ii) with carbonyl compounds forming secondary ozonides, compounds, which are supposed to decompose by a further collision e.g. with sulphuric acid (linear correlation with sulphuric acid). Assuming stabilized Criegee intermediate concentrations of about  $10^5$  molecules  $\text{cm}^{-3}$  or more (sesquiterpene ozonolysis) and sulphuric acid concentrations of about  $1 \times 10^7$  molecules  $\text{cm}^{-3}$ , this gives nucleation rates of minimum  $1 \text{ cm}^{-3} \text{ s}^{-1}$ , which depends on the ambient terpene, ozone and water vapour concentrations. The reaction (ii) with formaldehyde, which is the major carbonyl compound (here aldehyde) in spring, (ca. 0.1 ppbv) will yield about  $150 \text{ cm}^{-3} \text{ s}^{-1}$ . Consequently, pathway (i) will be predominant at high sulphuric acid concentrations and only gentle pollution. Pathway (ii) will dominate under opposite conditions, where formaldehyde is mainly transported by anthropogenic pollution towards Finland and has to compete with the higher condensational sink in these

air masses.