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Nucleation in a Boreal Ecosystem: The Link to Terpenes and Sulphuric Acid

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Observed nucleation and particle formation rates in boreal forest ecosystems are known to be anticorrelated with water vapour concentration and correlated with ambient sulphuric acid concentration and. The correlation with sulphuric acid is found to be either linear or squared for the smallest clusters or particles around 1 nm in diameter. However, the meaning, variation and interpretation of the correlation coefficients is not understood so far. In this study we demonstrate that reaction products of sesquiterpenes and ozone are possible key parameters determining the value of the correlation coefficients and the occurrence of a nucleation event. Both ways, i.e. the linear and the squared one, are postulated to start with the formation of the stabilized Criegee intermediate in the reaction of sesquiterpenes with ozone. This intermediate can subsequently react with carbonyl compounds (linear dependency) and with sulphuric acid (squared dependency) or with water vapour (no nucleation). The initial products of both nucleation pathways (linear and squared dependency) are supposed to be reactive and to bind colliding sulphuric acid molecules for activation by chemical reaction. The derived expressions for both correlation coefficients are very similar to the observed ones. Nevertheless, there are serious uncertainties with a need for detailed investigation. The proposed nucleation mechanism has serious impacts on the valid atmospheric range of the postulated nucleation mechanism and on climate feedbacks in a future atmosphere.