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Nucleation events: Annual and seasonal events and their link to atmospheric chemistry

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Dal Maso *et al.* (2005) have found the number of nucleation events at the boreal forest field site in Hyytiälä, Southern Finland (61.85° N, 24° E) to increase from 1997 to 2003, while it declined afterwards. Nucleation by the current sulphuric acid systems (binary, ternary or kinetic nucleation) fail to explain these number of nucleation events. But taking into account reactive volatile organic compounds (VOCs) such as sesquiterpenes, this seems to be possible. Nucleation in smog chamber studies on terpene oxidation are found to be caused predominantly by ozone, with the nucleation intensity decreasing by increasing amounts of water vapour. Thus, the ambient water vapour and ozone concentrations need to be taken into account. For Hyytiälä the annual behaviour of the nucleation events shows a similar behaviour as the concentration of ozone as well as the ratio of water vapour to ozone. This indicates the participation of terpenes in the process of new particle formation. Second, the seasonal behaviour (Hirsikko *et al.*, 2005) of nucleation events reveals two maxima one during spring (March, April and May) and a secondary one in autumn (August, September and October), interrupted by a minimum during summer. We used gas-phase simulations of the global chemical transport model MATCH-MPIC (Bonn *et al.*, 2005) to explain the seasonal behaviour of nucleation events as well as the growth at different aerosol sizes. Therefore, it was assumed that formaldehyde (HCHO) and pinonaldehyde (from a-pinene oxidation) cause nucleation, while carboxylic acids (formic acid, pinic and pinonic acid) suppress nucleation. Applying the nucleation mechanism of Bonn *et al.* (2002), the ratio of the carbonyl compounds (HCHO and pinonaldehyde) to the carboxylic acids multiplied with the corresponding reaction rate coefficients with the so-called stabilized Criegee intermediate should indicate a ‘nucleation probability’.

By using the data of the global chemistry transport model for the individual months, the same pattern is obtained as is observed for the ambient nucleation event number. While formaldehyde predominates in spring time, formic acid suppresses the nucleation during summer time and pinonaldehyde becomes of major importance during August. As a conclusion it can be stated that ambient nucleation is linked to the emission of terpenes, the presence of ozone and reduced pollution coming from Central or Eastern Europe, which transports the acids towards Fennoscandia.