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Ambient sesquiterpene concentration and its link to air ion measurements

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Abstract. Ambient air ion size distributions have been measured continuously at the Finnish boreal forest site in Hyytiälä since spring 2003. In general, these measurements show a maximum of air ions below 1.0 nm in diameter. But this physical characterization does not provide any information about the ion's chemical composition, which is one key question regarding the explanation of nucleation events observed. In this study we propose a link of the observed maximum of negative air ions between 0.56 and 0.75 nm to the so-called stabilised Criegee biradical, formed in the reaction of biogenic sesquiterpenes with ozone and predominantly destroyed by its reaction with ambient water vapour. Calculations of the electron and proton affinities of 120 kJ mol⁻¹ (1.24 eV) and of 960 kJ mol⁻¹ support this link. Other possible candidates such as sulphuric acid derived clusters are unable to explain the observations made. By using this approach, we are able to calculate the ambient concentration of sesquiterpenes at the air ion instrument inlet with a high time resolution on the daily and seasonal scale. The estimated concentration is found to reveal the same seasonal pattern as emission measurements conducted at shoot level. As expected for biogenic VOCs, the concentration is obtained highest during summer (maximum values of about 100 pptv) and smallest during winter (minimum less than 1 pptv). Because of the sesquiterpenes high reactivity and its low ambient concentrations, this approach can be a first step in understanding their emission and their impact on atmospheric chemistry in more detail. The findings presented are highly relevant for emission budgets too, since boreal forests are extended over large areas of the globe.

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