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Why does high relative humidity suppress continental nucleation events?

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It has been observed in different continental locations that nucleation events take preferentially place at low relative humidities (RH) (e.g. Hyvönen at al., 2005; Hamed et al., 2007; Laaksonen et al., 2008). Several possibilities have been suggested for the RH dependence of nucleation, but no firm conclusions have been drawn so far. These possibilities include enhanced coagulation scavenging of sub 3-nm clusters at high RH, interference of water vapour with gas-phase chemistry producing condensable organics, diminished solar radiation at high RH leading to diminished gas-phase oxidation chemistry, and increased condensation sink (CS) of condensable gases due to hygroscopic growth of pre-existing particles. In this presentation we study the 2003 QUEST spring campaign data from Hyytiälä measurement station in Finland to examine some of these possibilities in detail.

Figure 1a shows the observed decrease of solar radiation at high RH during the 2003 campaign. Both RH and solar irradiation show a clear diurnal cycle: in the morning, as sunlight warms the ground layer, RH often starts to decrease, and will increase again when sunset approaches and air begins to cool. Hence, there is an anti-correlation between RH and solar irradiation. At an annual level this anti-correlation may be smeared, as the daily peak intensity of the irradiation varies strongly from midwinter to midsummer, but at a monthly level it should be detectable. In addition, solar irradiation can be attenuated at high relative humidity, both due to increased cloudiness, and due to enhanced scattering of sunlight by hygroscopicity grown particles (haze). This causes the production of OH radicals to decrease, and thus the production term of H_2SO_4 decreases (the same is valid of course for the production terms of condensable organics species).

It can be readily seen from Fig. 1a that the highest radiation values are limited to about 60% RH and below, and that at higher humidities the maximum values decrease as a function of RH quite steeply, especially above 90% RH. It is thus evident that high RH's limit the production of H_2SO_4 and other condensable gases, as is also seen in Figure 1b.

Figure 1b shows that the maximum H_2SO_4 concentrations decrease by more than an order of magnitude between 60-90% RH. This strongly suggests that the main reason for the suppression of nucleation at humid conditions is due to the effects by which high RH tends to decrease the source term and increase the sink term of H_2SO_4. Very likely high RH poses similar influences on condensable organics. However, it depends on the role of the organics in new particle formation whether RH's suppressing action on new particle formation is mediated also via them. If the organics participate only in particle growth, then there is only relatively minor influence on the observed nucleation rate "J_3" (at higher growth rate "GR", the probability of a cluster to survive to 3 nm increases). However, if they participate in the actual nucleation, increasing J_1, then there should be a similar influence as via H_2SO_4. Moreover, we have studied RH effect on CS and coagulation sink and they cannot explain event suppression

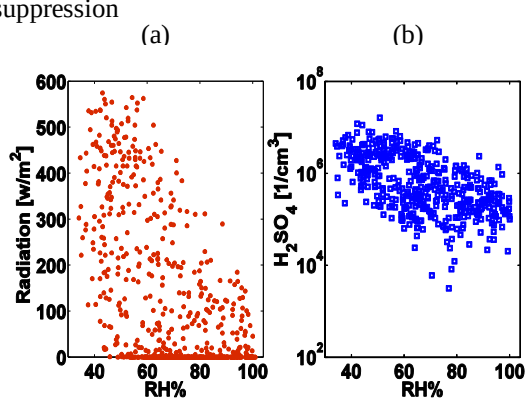

Figure1.(a) RH% versus solar irradiation (Wm^{-2}) (red colour) and (b) RH% versus H_2SO_4 concentrations ($1/cm^3$) (blue colour) observed during spring 2003 QUEST field campaign in Hyytiälä, Finland.

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