Contribution of ion-assisted nucleation to new particle formation in a tropical boundary layer

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Aerosols are ubiquitous in Earth's atmosphere and their interactions with clouds via activation of cloud condensation nuclei (CCN) constitute the largest uncertainty in climate forcing. Nucleation (as a result of gas-to-particle conversion) is a predominant source of new particles in the atmosphere and thus is an important process that controls aerosol and cloud abundance.

New particle formation (NPF) occurs nearly everywhere in the Earth's atmosphere [Kulmala et al., 2004]. A variety of species may contribute to nucleation and growth processes, such as sulfuric acid (H_2SO_4) [Kuang et al., 2008], ammonia (NH_3) [Ball et al., 1999], amines [Yu et al., 2011], organic acids [Metzger et al., 2010], and ion clusters [Yu, 2010]. Previous field study by Boy et al. [2008] reported ~15% contribution of ion-assisted nucleation to NPF in the boundary layer in Hyytiälä, Finland. On the contrary, modeling approach including the association of ion–ion recombination to neutral clusters showed 100% contribution of ion–mediated nucleation (IMN) to NPF in Hyytiälä [Yu and Turco, 2008]. A recent cloud chamber study revealed that ion-assisted nucleation contribution to the boundary layer NPF is negligible but is nevertheless taking place [Kirkby et al., 2011].

We conducted measurements of particle size distributions in an Indian tropical boundary layer during March 20–May 23, 2012. Eighteen regional NPF events were observed, all of these likely occurred under the influence of transported anthropogenic plumes. We used particle growth and nucleation (PARGAN) model to calculate total particle nucleation rate (J_1) from the measured particle size distributions along with aerosol microphysics box model [Kanawade and Tripathi, 2006] incorporated with binary homogeneous nucleation (BHN) [Vehkamaki et al., 2002], ion-induced nucleation (IIN) [Modgil et al., 2005] and IMN [Yu, 2010] schemes to simulate observed NPF events. The aerosol microphysical model simulations revealed that none of these nucleation mechanisms could explain NPF solely, suggesting that two or more processes occurring simultaneously. The other nucleation mechanisms, involving species such as NH_3 or organics, may be crucial in the boundary layer NPF but these were not possible to examine. For example, NH_3 concentrations in such heavily polluted environment vary from ~3 to 42 ppbv (Singh and Kulshrestha, 2012), which exceeds the limit currently treated in the ternary homogeneous nucleation (THN) parameterization.

Our simulations further indicated that ion-induced and –mediated nucleation could be an important contributor to NPF at this location whereas BHN failed to explain NPF at all. The median contributions of IIN and IMN were about 28% and 87%, respectively . These results demonstrate the importance of ion-assisted nucleation to the tropical boundary layer NPF.

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