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Cosmic ray induced formation of aerosol particles and cloud condensation nuclei: Evidence from the first detection of large negative and positive cluster ions in the upper troposphere

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Cosmic rays are potentially important in mediating the formation of aerosol particles and cloud condensation nuclei (CCN) and therefore may eventually influence climate. We report on aircraft-based atmospheric measurements of large cluster ions which indicate that aerosol formation via ion-induced nucleation (INU) is operative in the upper troposphere. For the first time we have detected in the upper troposphere large negative and positive ions using an advanced air craft-based large ion-mass spectrometer. The observed large ions represent key intermediates involved in INU. From the measured ions we have inferred the total concentration of highly supersaturated nucleating trace gas species X which ranges mostly from 1-4x10(6) per cm3 and which is comparable to gaseous H2SO4 concentrations previously measured by our MPIK group. This strongly suggests an identification of X as gaseous H2SO4. Building on (X)=(H2SO4) we have calculated the rates of H2SO4 induced INU and HONU (homogeneous nucleation). We also have made model simulations of H2SO4 induced condensational growth of new particles leading to CCN. We conclude that in relatively clean upper troposphere air masses INU is more efficient than homogeneous nucleation and that some fraction of the aerosol particles formed via upper troposphere INU can grow to sufficiently large sizes to act as CCN.