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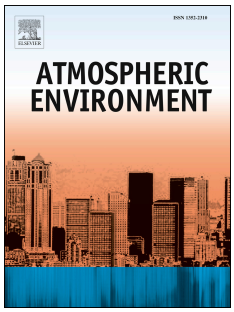

Current state of aerosol nucleation parameterizations for air-quality and climate modeling

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Current State of Aerosol Nucleation Parameterizations for Air-Quality and Climate Modeling

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Abstract

Aerosol nucleation parameterization models commonly used in 3-D air quality and climate models have serious limitations. This includes classical nucleation theory based variants, empirical models and other formulations. Recent work based on detailed and extensive laboratory measurements and improved quantum chemistry computation has substantially advanced the state of nucleation parameterizations. In terms of inorganic nucleation involving BHN and THN including ion effects these new models should be considered as worthwhile replacements for the old models. However, the contribution of organic species to nucleation remains poorly quantified. New particle formation consists of a distinct post-nucleation growth regime which is characterized by a strong Kelvin curvature effect and is thus dependent on availability of very low volatility organic species or sulfuric acid. There have been advances in the understanding of the multiphase chemistry of biogenic and anthropogenic organic compounds which facilitate to overcome the initial aerosol growth barrier. Implementation of processes influencing new particle formation is challenging in 3-D models and there is a lack of comprehensive parameterizations. This review considers the existing models and recent innovations.

Keywords: Aerosol nucleation, SOA, New particle formation

1. Introduction

New particle formation (NPF) and nanoparticle growth in the atmosphere have recently received increasing experimental and theoretical interest due to the relevance of the number density of submicron size aerosols for direct and indirect effects on climate (Stocker et al., 2013). Nanoparticles also have a significant impact on human health (Ibald-Mulli et al., 2002; Stölzel et al., 2006; Terzano et al., 2010). Both primary and secondary aerosols exist in the atmosphere. The former result from direct emissions from lithogenic (both land and ocean), biogenic and anthropogenic sources. Secondary aerosols are formed in the atmosphere by nucleation, condensation and chemistry of vapours including sulfuric acid and low volatility organic compounds which originate primarily from biogenic and anthropogenic sources. The formation of new aerosol particles which populate the accumulation mode with diameters typically smaller than 1 micron takes place frequently and at a broad variety of atmospheric conditions and geographic locations. NPF has been observed at numerous places with very different climatologic conditions, such as boreal forests, marine coasts, urban and polar regions and the free troposphere (Kulmala et al., 2004b; Kulmala and Kerminen, 2008; Nie et al., 2014; Bianchi et al., 2016; Sipilä et al., 2016).

There is a significant growth barrier for freshly formed particles or molecular clusters due to the Kelvin curvature effect which increases rapidly with the inverse of the particle diameter. Due to this barrier only the lowest volatility gas phase constituents can partition into the aerosol phase at the smallest sizes. Freshly nucleated particles do not all proceed to grow out of the sub 3 nm diameter region of the nucleation mode (which we define as the aerosol population with diameters be-

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