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The Radiative Effect of Ion-Induced Inorganic Nucleation in the Free Troposphere

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Abstract. To determine the effect of cosmic rays on the Earth's climate via ion-induced nucleation, a parametrisation of inorganic nucleation was formulated based on experiments at the CERN CLOUD experiment. The parametrisation was implemented in the GLOMAP aerosol microphysics model and used to estimate the radiative effect of the change in ionisation experienced over an 11-year solar cycle.

Keywords: Nucleation, Modeling, Ion-Induced Nucleation, Inorganic Nucleation, Ammonia, Ions, Galactic Cosmic Rays
PACS: 64.60.Q-, 64.60.qe, 64.60.qj, 82.60.Nh, 82.70.Rr, 91.67.gp, 92.60.Mt, 96.50.S-

INTRODUCTION

Nucleation is known to be an important source of cloud condensation nuclei (CCN), which affect the Earth's climate directly and indirectly [2, 3]. It has been estimated that nucleation is responsible for up to 45% of global CCN [2]. However, the parametrisations of nucleation currently in use in global aerosol models are generally based on boundary-layer observations [4, 5], kinetic or thermodynamic models [6, 7], or theoretical calculations [8, 9]. These parametrisations produce very different nucleation rates from the same inputs [10], because of their different dependencies on factors including temperature, relative humidity, or concentration of H_2SO_4 or NH_3.

While classical nucleation theory suggests that nucleation rates observed in the boundary layer cannot be attributed to binary H_2SO_4-H_2O nucleation alone, until recent years there was no consensus as to whether the nucleation rate was being enhanced by the presence of a ternary vapour such as ammonia or organics [11], or ions [12]. Recent technological advancements have made it possible to measure contaminant concentrations at a parts-per-billion (ppb) or parts per trillion (ppt) level [13, 14], and to determine the composition of the smallest nucleating clusters [15]. As a result, the substances participating in new particle formation can be identified.

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THE CLOUD EXPERIMENT

The CLOUD experiment was conceived as a means of measuring neutral and ion-induced binary H_2SO_4-H_2O nucleation under free tropospheric conditions, at low temperatures and in the absence of contaminants. The pion beam from the CERN Proton Synchrotron (PS) can be used to generate ion concentrations at free tropospheric levels. During the earliest CLOUD experiments, the APi-TOF [15] uncovered trace amounts of NH_3 participating in the nucleation process [1]. This led to the CLOUD3 campaign, during which the ternary H_2SO_4-NH_3-H_2O system was examined. It was found that the presence of 100 ppt of NH_3 enhanced the nucleation rate by a factor of 100-1000 compared to binary nucleation [1].

A further set of experiments based on the inorganic nucleation system was then carried out at temperatures down to -65 °C during the CLOUD5 campaign. Measurements in the CLOUD3 and CLOUD5 campaigns covered temperatures from 208 to 298 K, H_2SO_4 concentrations from 2.5 $\times$ă10^5 cm^{-3}, and NH_3 mixing ratios from 0.1 to more than 10 ppt. Ultimately, the measurements made in the CLOUD experiments were used to formulate a parametrisation of inorganic nucleation for use in global aerosol and climate models. This work presents the form of the parametrisation, with particular emphasis on the atmospheric conditions under which neutral or ion-induced nucleation will dominate, as well as showing the simulated behaviour of the parametrisation in the atmosphere.

PARAMETRISATION OF INORGANIC NUCLEATION

This is the first parametrisation of inorganic nucleation based on laboratory experiments. It calculates the nucleation rate as a function of temperature, ionisation rate, and H_2SO_4 and NH_3 concentrations. The parametrisation calculates binary neutral, binary ion-induced, ternary neutral, and ternary ion-induced nucleation rates separately, making it possible to analyse the relative importance of ammonia and ions in different parts of the atmosphere.

[2] estimated that 78% of CCN at low-cloud level which had been produced via nucleation were formed in the free troposphere. The inorganic system does not produce high enough nucleation rates at boundary layer (BL) temperatures to reproduce observations, meaning that another ternary vapour is expected to participate in the BL nucleation process [1]. Although reproducing these observed BL nucleation rates is an important task for aerosol modellers due to their significant contribution to $PM_{2.5}$ concentrations which directly affect human health [16], accurately modelling free tropospheric nucleation is much more important in climate terms.

THE RADIATIVE EFFECT OVER A SOLAR CYCLE

Over a solar cycle, simulated ion concentrations in some regions of the atmosphere change by up to 20%. The sensitivity of the atmosphere to the resulting change in the nucleation rate depends on the proportion of nucleation which is ion-induced, and on the total nucleation rate. While secondary aerosol are a significant contributor to

CCN throughout the atmosphere, the production mechanism also has a self-dampening feedback, as freshly nucleated particles are lost to the sink of older nucleated particles which are in the process of growing to CCN sizes.

The Global Model of Aerosol Physics (GLOMAP) has been used to estimate the radiative effect of a solar cycle due to a change in the inorganic nucleation rate. GLOMAP-bin is a sectional two-moment global aerosol microphysics model which traces number and mass concentration of aerosol species. The sectional model is used instead of a computationally less expensive modal scheme because the sectional version makes no assumptions about the shape of the aerosol size distribution at the smallest sizes, which is particularly important when studying nucleation.

The look-up table of [17] was used to calculate the cosmic-ray-induced ionisation (CRII) rate during solar minimum and solar maximum. The change in the cloud droplet number from two otherwise-identical simulations which used $CRII_{min}$ and $CRII_{max}$ to estimate the nucleation rate was then used to calculate the radiative effect using the method described in [16].

CONCLUSIONS

The first parametrisation of inorganic nucleation based on laboratory measurements has been implemented in a global aerosol microphysics model. The model has been used to calculate the radiative effect due to the change in CRII over a solar cycle.

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