KINETICS OF BINARY NUCLEATION IN AIRCRAFT EXHAUST PLUME

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Civil aviation releases various components that can affect natural atmospheric processes. In particularly, the sulfur and water vapours emitted by engines may be converted to liquid and ice aerosol particles that may act as cloud condensation nucleus and also impact on atmospheric chemistry. The key crucial point in this process is an initial phase of nucleation of numerous new particles in the exhaust plume during its cooling and expansion in the ambient atmosphere. In this article, the kinetics of non–steady nucleation and time lags for binary homogeneous nucleation of sulfuric acid-water aerosols is considered in contrary to the usually used classical steady-state nucleation theory. Classical nucleation analysis assumes: (1) that the timescale for establishing a steady-state sub-critical clusters population is very short compared the rate of change of the nucleation rate which in turn out depends on the temperature and gas species concentrations change (steady-state clusters population approach); (2) that the concentration of monomers is so much high than the concentration of sub-critical clusters, that the rate of cluster-cluster collisions is negligible compared to the rate of monomer-cluster collisions (monomer-cluster collision approach); (3) that the nucleating system is near the equilibrium and the concentration of monomers required to establish the steady-state clusters population is much high than the total concentration of monomers incorporated in clusters (monomers reservoir approach). The purpose of this paper is to investigate these assumptions for the practically important case of the binary nucleation during the aircraft exhaust plume cooling. For this, a model which directly consider the dynamics of clusters population (birth-death equations including monomers) with accounting for the cluster-cluster collisions and cluster dissociation into two smaller clusters together with the cooling of gaseous molecule-cluster system is proposed. It is shown that for many typical conditions the assumptions leading to the classical nucleation rate become increasingly questionable. For example, may be important collisions of two sub-critical clusters, which results in the cluster formation of critical or even larger size. Also, there is a relatively slow clusters build-up compared to plume expansion and cooling rate. Finally, the comparison between different models to describe the energetics of clusters formation, have shown that considered nucleating process of neutral clusters may be limited by the initial nucleation steps (i.e., dimer, trimer etc. formation).

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