Nucleation in the Presence of Background Aerosol

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When condensable molecules are being produced, either by chemical reactions or cooling, it is important to know whether they will all condense on existing aerosol or form a new aerosol by nucleation. For example, ultrafine particles in the workplace can be highly injurious to health. The suppression of nucleation by existing aerosol is well known in the atmosphere, and is well illustrated by an analysis of a complete set of Finnish forest nucleation data observed in 1996 and 1997 (Clement *et al* 2001). Here we point out physical situations and processes where this suppression does not necessarily occur

In a homogeneous atmosphere where condensable molecules are being produced by chemical reactions, the level of supersaturation reached is always limited by condensation on, and coagulation of tiny nuclei with, existing aerosol. As the level is inversely proportional to the removal rate onto existing aerosol, suppression of nucleation is guaranteed for large amounts of existing aerosol. In situations where supersaturation is produced by cooling a vapour-gas mixture, particularly those involving mixing with another mixture, motion of vapour and aerosol takes place. Differences in their motion can lead to large local differences in amount of aerosol present and consequent supersaturation. It was previously pointed out (Clement and Mather 2005), that the observed presence of a small-sized sulphuric acid aerosol in a volcanic plume initially containing a large amount of large-sized aerosol could be attributed to this effect. The acid vapour diffuses into the surrounding air at a much greater rate than large aerosol particles so that nucleation is possible.

Very similar situations must have arisen in the many experiments performed by Buckle and collaborators (see Buckle and Marwella (1987) and references therein) who evaporated heated metals into gas flows to produce aerosols. The geometry is shown in Fig.1 where the metallic vapour diffuses into a laminar gas flow (Mg and Pb into argon in Buckle and Mawella (1987)). Observed size distributions of aerosol are often very wide and sometimes have two peaks, interpreted as arising from nucleation of both liquid droplets and solid particles. The continuing nucleation after the initial aerosol is formed will arise because the aerosol is carried upwards with the flow whereas the vapour continues to diffuse horizontally into virgin gas.

The experiments cannot be interpreted by the theory given in Buckle (1986) because of its essentially one-dimensional nature. All the mass current including both vapour and aerosol is moving in the same direction so that the aerosol will inhibit new nucleation. Instead, continuing nucleation uninhibited by aerosol already produced will give rise to a very wide size distribution which may, if the freezing point temperature is crossed be concentrated into two peaks for liquid and solid nuclei. Detailed analysis of the experimental results of Buckle requires two-dimensional calculations of heat and mass transfer.

The cooling of vapour-gas mixtures by turbulent mixing or in turbulent flows at cooled walls is another area where even considerable amounts of aerosol may not prevent nucleation. The basic reason is that rapid flows can produce large temperature differences across small distances, for example across a boundary layer at a wall. The consequent large temperature gradient can produce a large supersaturation in a small layer of width δ. A criterion (Clement 1987) for the amount of aerosol needed to prevent this is

$$[\, 4\pi N \langle R \rangle \,]^{1/2} \delta >> 1,$$

where N is the aerosol number concentration with mean radius $\langle R \rangle$.

The consequence of this criterion is that the onset of turbulence in flow through cooling tubes increases the likelihood of nucleation occurring. In general higher levels of turbulence will lead to smaller boundary layer distances and more nucleation.

Where many vapours are present, the lack of nucleation suppression can lead to a variety of aerosols being formed, and a nuclear example is discussed.

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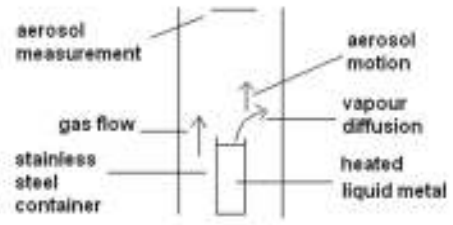

Fig. 1. Buckle apparatus for aerosol production.