

Nucleation in a perforated tube diluter

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Abstract

This study investigates the effect of the dilution method on the formation of particles by nucleation during cooling. Nucleation in a perforated tube diluter was studied with experiments and models, using dibutylphthalate (DBP) as the nucleating agent. The experiments demonstrate that nucleation tendencies of various DBP concentrations can be observed in the diluter. Modelling studies were carried out using estimates of DBP nucleation rates from the literature. These indicate that nucleation mostly takes place inside the diluter either at the first orifice rings as some DBP vapour is trapped in low temperature regions or in a similar situation in turbulent eddies when high vapour concentration is still present. The complex turbulent dilution system generates broad particle size distributions.

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1. Introduction

The rates of many aerosol transformation processes are highly temperature dependent. This makes the cooling process an essential consideration. In many systems, such as exhaust emissions to the ambient air, cooling results predominantly from mixing with colder gases. In measurement systems, and in some industrial processes, cooling via dilution is used when temperature or concentration levels need to be lowered to the range of the measurement devices, or reactions or particle coagulation need to be quenched. Diluters are used as part of sampling systems from operating real-world devices or as part of experimental set-ups. Transformations during cooling by dilution are rate-dependent both on the cooling rate, and on the rate at which the concentrations decrease. Therefore, to have some understanding and control over the transformation processes during dilution, it is essential to consider the way dilution is achieved. Often, reactions and condensation of gas phase species onto external walls and/or particle deposition are a concern, and it is necessary to minimise these undesired effects of the walls. This study deals with a laboratory-scale diluter. The interest is in the appearance of a nucleation mode in the diluter when the sample contains condensable vapours. A nucleation mode can

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appear during dilution when the decrease in temperature pushes the saturation ratio of a condensable vapour to levels of appreciable nucleation at least in some location, and the decrease in the vapour concentration is not rapid enough to offset this effect.

There are a number of practical cases, when the appearance of a nucleation mode in a diluter is of interest. Nucleation generates new particles in exhaust plumes and diluters are used for assessing nucleation tendencies of various types of exhausts. Fine particles in exhaust gases have attracted interest due to their adverse health effects (e.g. Donaldson, Li, & MacNee, 1997; Peled et al., 2005; Pope et al., 2002). In terms of human exposure, vehicular emissions are considered to be significant. The particulate phase in these emissions consists of particles that form at high temperatures (the “soot” mode), and particles that form in exhaust plumes via nucleation during the release into the ambient air (the nucleation mode). Nucleation mode particles have been observed in on-road measurements (Giechaskiel et al., 2005; Vogt, Scheer, Casati, & Benter, 2003). Sulphuric acid is considered to be the usual nucleating agent for the nucleation mode, while the issue whether hydrocarbons can be significant as nucleating agents in some cases is still under investigation (Kittelson, 1998; Kulmala et al., 2004; Shi & Harrison, 1999; Vouitsis, Ntziachristos, & Samaras, 2005). After nucleation has taken place, hydrocarbons condense on the nucleated particles and account for most of the particle mass (Mathis, Kaegi, Mohr, & Zenobi, 2004; Sakurai et al., 2003; Tobias et al., 2001). Not much is known about the health effects of the nucleation mode particles. Besides vehicular emissions, sulphuric acid accounts for the initiation of nucleation mode in the exhausts of many other common combustion processes as well. In high-temperature measurements of combustion processes, the nucleation mode is formed during the sampling process from ash components that are in the vapour phase at the sampling location. Alkali chlorides in biomass combustion are an example of these (Mikkanen et al., 1998). Heavy organic compounds can nucleate when released as fugitive emissions, for instance during the peat drying process. Diluters can also be used in nanoparticle generation via aerosol methods to lower the temperature levels for after-treatment and collection (e.g. Ahonen, Richard, & Kauppinen, 2001).

The formation of particles via nucleation in exhaust plumes has been studied a fair amount. Even if the release as a jet into the ambient air involves complex mixing in turbulent flow, approximations can be used to provide a rough idea of the essential issues in this process. If the molecular diffusivities of heat and nucleating vapour are assumed to be equal, and vapour depletion and coagulation are negligible, the nucleation rate for simple one-component nucleation in any spot is only determined by the degree of mixing. In case of turbulent mixing, average values can be obtained if the local probability density function of turbulence is known. Lesniewski and Friedlander (1998) used this approach for the analysis of nucleation in the shear layer of a turbulent jet for which experimental probability density functions are available. Analyses beyond the $Le = 1$ assumption, such as the case of large low-diffusivity hydrocarbon molecules, are more complicated. When the diffusion of the vapour species is slow, areas where temperature has decreased but vapour concentration remains relatively high can form inside the turbulent eddies, thus increasing the overall average nucleation rate. This has been demonstrated in DNS simulations (Pyykönen, Murfield, & Garrick, 2004). In case of rapid dilution and relatively low number concentrations, nucleated particles do not possess enough scavenging power to significantly affect gas-phase concentrations, and nucleation is suppressed when the vapour concentration has been diluted to a sufficiently low level. With slower dilution and higher particle number concentrations, the vapour depletion by pre-existing and nucleated particles as well as coagulation can also be significant. Additional considerations include possible kinetics of gas phase reactions that precede nucleation and the kinetics of heterogeneous nucleation prior to the onset of condensation onto pre-existing particles. Detailed studies of sulphuric acid behaviour in vehicular exhaust trails have been made (Kim, Gautam, & Gera, 2002; Wu & Menon, 2001). The processes related to turbulent micromixing have been shown to increase particle number concentrations significantly.

Many diluter systems are available for cooling via dilution. The most common of these are probably various types of dilution tunnels and the ejector diluter that provides more control of the dilution process. A porous tube diluter also provides control over the dilution process (Newton, Carpenter, Yeh, & Peele, 1980; Ranade, Werle, & Wasan, 1976). It has the advantage over the ejector diluter that it does not have problems related to plugging of the flow channels and losses are minor even for supermicron particles. The control of the flow is not as straightforward as with the ejector diluter, though. A variant of the porous tube diluter has been developed by Dekati Ltd.: a small-diameter (3.5 mm i.d.) perforated tube diluter that can cope with time-dependent measurements from a source with varying pressure. The small dimensions eliminate the need for large flow rates to achieve rapid turbulent mixing. The walls of the diluter consist of a large number of small laser-drilled holes.

Often, the objective of sampling and dilution is to obtain the properties of the aerosol as it is at the measurement location. For instance, in vehicle measurements, the soot mode is usually of primary interest. A dilution method that

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