Atmospheric Behaviour of Ultrafine Particles Arising from Road Traffic

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Abstract Airborne particles are a well recognised cause of increased morbidity and mortality in the general population. While the precise influences of particle size and chemical composition upon toxicity have yet to be fully elucidated, there has been a great deal of interest in the ultrafine fraction of less than 100 nanometres diameter which show high toxicity per unit mass. By far the greatest abundance of ultrafine particles in the urban atmosphere arises from emissions from road traffic and particularly from diesel engines, and recent work in our group has provided clear evidence that such particles are semi-volatile and that they progressively evaporate as they travel away from the source of emission. However, current knowledge of their chemical composition and rate of evaporation is very weak and this project aims to provide the necessary fundamental information to allow better prediction of their behaviour. This will involve sampling particles from test engines, analysis of chemical composition, alongside field experiments in which the evolution of the ultrafine particle fraction will be observed using advanced aerosol instrumentation. Numerical models will also be designed to simulate the oxidation of the vapours produced, and the consequent formation of secondary organic aerosol.

Introduction. Airborne particles are a topic of international interest because of their adverse effects on human health. In this context there has been particular attention paid to the ultrafine or nanoparticle fraction of less than 100 nanometres diameter because of its high toxicity per unit mass. There has been considerable concern over manufactured nanoparticles but currently the main source of nanoparticles in the urban atmosphere are those present in the emissions from road vehicles. Our recent work has shown that these are made up of two different types of particle, one of which referred to as the nucleation mode is semi-volatile, i.e., it slowly evaporates as it moves into cleaner air. Recent work from the United States has suggested that the semi-volatile compounds associated with such particles not only desorb into the vapour phase but are subsequently oxidised to form less volatile compounds which recondense onto the solid particles. These processes cause not only an increase in mass (because of the oxidation process) but also lead to pollution by secondary organic compounds at great distances from the source regions. However, much of this work is theoretical in nature and requires an experimental underpinning which is the primary aim of this project.

Aims. The purpose of the project is to gain an improved and more quantitative understanding of the composition of semi-volatile nucleation particles from diesel exhaust, their behaviour in the atmosphere and consequences for regional air quality. Associated with this are specific objectives to sample and characterise particles from engine exhaust in the laboratory to measure their concentrations and their evolution as they age in the atmosphere and to determine the vapour phase products arising from them and the consequences for air quality downwind.

Methods. The initial work will require sampling of diluted engine exhaust using the University's advanced engine test bed facilities. Size-fractionated samples will be collected using Micro-Orifice Uniform Deposit Impactors, and after thermal desorption will be analysed by gas chromatography/ mass spectrometry using state-of-the-art instrumentation. Electron microscopy will also be used to investigate possible solid cores to the primarily semi-volatile liquid particles. Methods will also be developed to collect the vapour phase components both in the engine laboratory and in the field on absorption tubes which will be analysed by thermal desorption and GCMS. Field experiments will also be conducted using the measurement of size distributions by Scanning Mobility Particle Sizers in order to determine the evolution of particle size distributions downwind from an automotive source and the extent to which vapour phase compounds are released. The final part of the programme will involve designing a simple parameterisation of the formation of secondary organic aerosol from the particle-derived vapour and using a photochemical trajectory model to estimate its impact on downwind air quality.

Training. The student will gain experience of a range of advanced instrumental techniques. This will include air and exhaust sampling using a Micro-Orifice Uniform Deposit Impactor and measurements of particle size distributions by Scanning Mobility Particle Sizer. In addition, training will be given in the use of thermal desorption GC/MS for the analysis of semi-volatile hydrocarbons both in vehicle exhaust and the atmosphere. The student will also take part in collaborative field campaigns in which much can be learnt from the activities of other participants. In the final part of the project, the student will gain experience of atmospheric chemical modelling with a state-of-the-art photochemical trajectory model.

Selected References.

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