
Automotive particulate matter

The ongoing search for basic knowledge

Particulate matter (PM) in the air continues to be the focus of increased attention due to the concern of potential health effects. Accordingly, under the EU Air Quality Framework Directive an air quality standard has been defined with respect to PM₁₀ (particulate with an aerodynamic diameter less than or equal to 10 nm) with a review planned in 2003.

There is concern that automotive tailpipe emissions, as one of the many emitting sources, may make a substantial contribution to ambient particulate concentrations especially within urban areas. Legislation is in place to measure and control the mass of automotive particulate emitted at the tailpipe and tighter limits have been progressively introduced over the years. While there is evidence that adverse health effects are associated with current ambient PM concentrations, it is, as yet, uncertain which feature of the particulate matter, be it chemical or physical, has the most relevance for health studies. Thus, attention has concentrated on the number-based size distribution of the ambient particles.

GUIDANCE STILL REQUIRED ESPECIALLY FROM HEALTH SCIENCE

At present there is no proven mechanism whereby low-level ambient PM could cause either early death or morbidity and, in terms of plausibility for increased mortality, there are no toxicology data to allow any conclusion.

Guidance from the health and atmospheric scientific community is still awaited on the particulate characteristics relevant for further abatement. Nevertheless it is important that different sectors of industry making significant contributions to ambient particle concentrations have a good understanding of the characterization of the particulate emitted. Automotive particulate emissions appear to have been studied most extensively and a wide range of information is now available.

SCOPING EXERCISE ON AUTOMOTIVE PARTICLES COMPLETED

CONCAWE embarked very early on the study of the automotive particulate emissions by mass, number and size. CONCAWE report 96/56 presented a thorough literature study to identify suitable analytical methodology for the measurement techniques of both mass and number distributions of the particles. This work was followed by a scoping exercise to develop an understanding in the area of automotive particulate emissions focusing on a range of light duty diesel and gasoline vehicle technologies and using wide range of marketed fuel specifications (CONCAWE report 98/51, SAE 982600). With a test programme on heavy-duty (HD) engines of two technology levels (Euro 2 and 3) and a fuel matrix matching that used for the light-duty (LD) diesel vehicle study as closely as possible, CONCAWE has now completed its scoping exercise in this field. Results from the heavy-duty study were published recently (SAE 2000-01-2000) and the full CONCAWE report is in its editorial stage. The major findings are reviewed below and put into the context of CONCAWE's current understanding on particle aspects.

THE HEAVY-DUTY ENGINE STUDY SHOWED THAT SAMPLING CONDITIONS STRONGLY INFLUENCE THE MEASURED DISTRIBUTION OF NUCLEATION MODE PARTICLES

The investigations of HD engine particulate sizing were based on experience and understanding gained from the earlier work as well as information obtained by contact with the scientific community researching in this area. Thus particle measurements were extended down to 3 nm and sampling effects were addressed as well. The wider range of particles (3–1000 nm) was covered using a Dual Differential Mobility Particle Spectrometer consisting of two analysers applied in parallel to sample simultaneously during the short scanning times available during the steady-state modes of the European HD engine test cycles.

As done in the LD work (and in the absence of any standardized sampling methodology for particle measurements) legislated test and sampling procedures for particulate mass were used for this HD engine study.

The study showed that sampling conditions, such as dilution ratio in the tunnel, residence time of the aerosol from the engine manifold to the inlet of the measurement device and stabilization time for sampling, strongly affect the particle size distribution and integrated total number of particles. The number distribution is bimodal with an accumulation mode (>30 nm) and a nucleation mode (<30 nm). While the accumulation mode particles gave consistent data, nucleation mode particles showed considerable variation with the change of sampling conditions (example given in Figure 1).

In this study only small differences between the tested fuels were observed within the accumulation mode, while larger differences between the fuels were observed in the nucleation mode (nanoparticles) (Figure 2). But such differences were not confirmed by the mass distribution data using a low-pressure cascade impactor.

Figure 1
Sampling conditions, such as dilution ratio and temperature, influence the particle distribution strongly in the nucleation mode (<30 nm), while accumulation mode particles (>30 nm) are only slightly affected. (Results from a Euro 2 engine at one mode condition.)

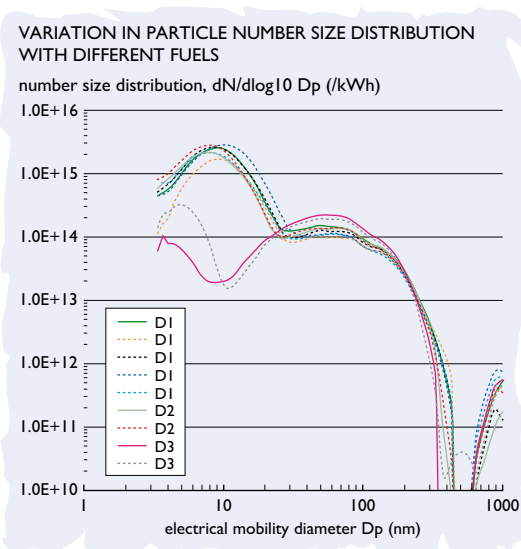
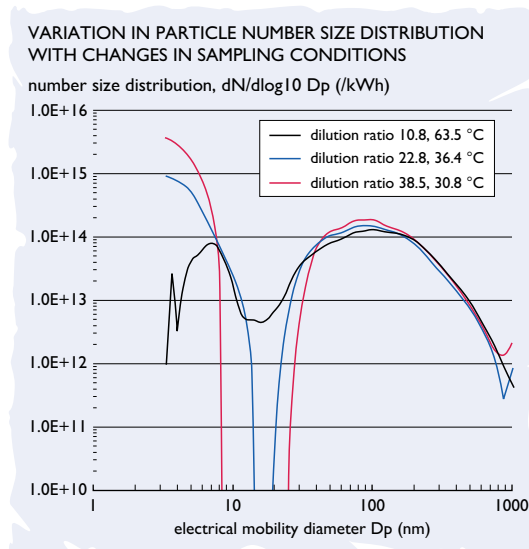


Figure 2
Differences in fuels were observed for nucleation mode particles which are strongly influenced by the sampling conditions (Figure 1). (ESC weighted averages with Euro 3 engine and individual test runs.)

From the foregoing it is concluded that there is no clear indication how to accurately measure nucleation mode particles. Particle measurements conducted in an atmospheric environment in other current research programmes will help to establish relationships between ‘real world’ particles and those measured under laboratory conditions. However, early ‘real world’ measurements also show that accumulation mode particles are quite insensitive to sampling and ambient conditions, while nucleation mode particle measurements are very sensitive, e.g. to ambient temperature.

NUCLEATION MODE PARTICLES NEED MORE RESEARCH

For accumulation mode particles the focus of further work should be on precision of size and number and/or mass measurement.

More research is required to better understand the basic generation mechanisms of nucleation mode particles. They are formed by condensation and therefore are observed to be very sensitive to sampling and ambient conditions. Reliable measurements do not seem to be achievable as yet. Further research will have to include engine design and operating conditions including specific fuel properties. Clarification is most urgently required on the contribution of nucleation mode particles to the ambient particulate matter and their relevance to human health.

NUCLEATION PROBLEM EXISTS WITH TRAPS AS WELL

Particulate traps can substantially reduce carbonaceous particulate (i.e. accumulation particles). However, drastically reducing the carbonaceous material will also remove the possibility of condensation of volatile particles. This would further increase the difficulties of controlled sampling and measurement since both particle modes are at a different level of understanding. As observed in the CONCAWE study, the reduction of fuel sulphur would not necessarily solve the problem of nucleation of particles. Even with very low levels of sulphur (< 10 mg/kg) large numbers of particles can be produced if the engine is equipped with a highly effective oxidation catalyst in the exhaust. Such effects were observed when a Euro 3 HD engine was equipped with a CRT (Continuous Regenerating Trap) to meet Euro 4 emission limits (Figure 3).

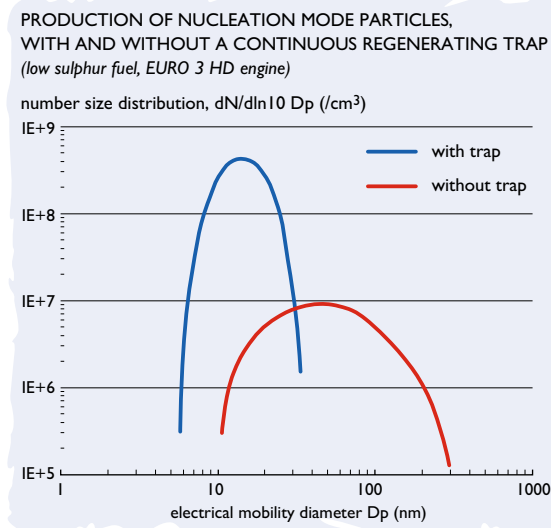


Figure 3 Particulate filter trap equipped Euro 3 engine adjusted to meet Euro 4 shows strong increase in nucleation mode particles even with a very low sulphur (<10ppm) containing fuel. Accumulation mode particles are completely trapped versus the Euro 3 set-up without the trap. Data given for medium operating condition (ESC mode 4). (Sampling effect, see Figure 1.)

A PRAGMATIC APPROACH

The CONCAWE study provides further evidence of the complexity of the sampling and measurement of the full range of particles. In view of these results reported also by other workers and also observed under 'real world' conditions, it might be appropriate to consider the work on these issues in segments. There is currently a greater understanding of the practical handling of accumulation mode particles, while more research is needed to understand the complex nucleation processes and the resulting nucleation mode particles. Of course it has to be kept in mind that both areas of the automotive particle phenomenon have to be put into perspective with regard to other sources and their relevance to human health.

CONCAWE continues to be involved in the work on particle aspects as e.g. shown in its participation in the DETR/SMMT/CONCAWE PM Research Programme and in the PM Consortium managed by the European Commission's Directorate for Transport and Energy.