Sampling parameter effect on the particle size distribution during controlled dilution

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ABSTRACT

Fine Particle Sampler is a diluter optimised for fine particle sampling with adjustable and controlled dilution ratio and real-time data acquisition. Easy application to nucleation tendency studies with two stage dilution, adjustable residence time and proven repeatability in soot mode measurements make the Fine Particle Sampler a reliable and flexible sampling system for use in the automotive industry. In our earlier work, we have presented a comparison of Dekati sampling products in diesel exhaust sampling: Fine Particle Sampler, Dekati thermodenuder and Dekati diluter. In thermodenuder, the sample is first heated to volatilize any hydrocarbons. The volatile hydrocarbons are then adsorbed in a cooled active charcoal section. Dekati double diluter is a two-stage ejector diluter system. In which the first dilution stage is hot and the second stage is cold to bring the sample temperature to an ambient level. In this work, we add coagulation, condensation and loss calculations to further evaluate these results. In addition we will present calculations on the performance and on the effect on the resulting particle size distributions of different sampling systems in different vehicle exhaust conditions.

INTRODUCTION

Current vehicle emission standards limit the particulate mass emissions. However, some health studies propose that particle number has a stronger correlation with health than particle mass. Therefore, future standards may also focus on the number of particulates released from vehicles. This presents new challenges for sampling equipment. Soot mode number concentration measurement is repeatable if the sample is conditioned with a thermodenuder, but all volatile material is lost and the end result may represent real-life atmospheric dilution poorly. If the exhaust is diluted in a controlled manner with cooled dilution air and an adjustable dilution ratio the volatile matter can be forced to nucleate/condensate thus providing information about the behavior of volatile matter. The selection whether to measure soot or soot + volatile particles is important, because
nucleation can produce 10-100 fold increase in particle number in the alveolar deposition size range.

CONCLUSIONS

Particle distribution transformation was effectively governed with the FPS dilution system. Distinct nucleation and accumulation modes were produced in expected conditions.

Accumulation mode was repeatable even when produced with different sampling systems.

Aerosol coagulation and condensation models produced distributions which were in good agreement with measured results.

Total mass emissions are much less affected by sampling parameters, when compared to number emissions.
Sampling parameter effects on the particle size distribution (and concentration) during controlled dilution

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Motivation

• Future emission limits: currently mass concentration, number is of interest
  – How to maintain a particle in exhaust

• Health effects: what measure associates with observations?
  – how to measure it reliably

• Distinguished nucleation and soot modes

OR

• Only soot mode
Instrumentation

Diagram:

- FD: Flow Divider
- AC: Ageing Chamber
- P, T: Pressure, temperature recording
- PD: Primary Diluter (FPS)
- ED: Ejector Diluter (FPS)
- TD: Thermodenuuder
- DD: Double Diluter
- Valve box
- Cooling
- Sample in
- ELPI
- CPC
- PM-filter
Instrumentation: Operation principles

### Thermodenuder (TD)
- **Heater**
- **Adsorpter**

### Double Diluter (DD)
- **Ejector diluter HOT**
- **Ejector diluter COLD**

### Fine Particle Sampler (FPS)
- **Porous tube diluter HOT/COLD**
- **Aging**
- **Ejector diluter COLD**

**Legend:**
- **Red** - Heater
- **Purple** - Active char coal
- **Yellow** - VOC adsorption
- **Blue** - Dilution air
- **Black dots** - Soot particles with some condensed vapors
- **Small black dots** - Soot particles
- **Small brown dots** - Nucleation particles
Corrections: Thermodenuder losses

- Determined for monodisperse aerosol with two CPCs that were frequently cross checked
Instrumentation: Figures

Dekati Double Diluter (DD)

Dekati Thermodenuder (TD)

Dekati Fine Particle Sampler (FPS)
FPS probe

- Pressure transducers
- Dilution air
- Flow divider
- Sampling
  - Primary
  - Secondary
- Thermocouples
Instrumentation: Sampling parameters

- **Thermodenuder (TD)**
  - No DR
  - $T = 250^\circ C$
  - $t = 3\, s$

- **Double Diluter (DD)**
  - $DR \approx 100$
  - $T_1 = 250^\circ C$
  - $T_2 = \text{ambient}$
  - $t = 1\, s$

- **Fine Particle Sampler (FPS)**
  - $DR_1 = 3-15$
  - $DR_2 \approx 10$
  - $T_1 = 250^\circ C$
  - $T_1 = 32^\circ C$
  - $T_2 = \text{ambient}$
  - $t = 1\, s$ and $3\, s$

Processes:
- Dry soot
- Volatile adsorption
- Dry soot
- Volatile evaporation and/or condensation
- Nucleation
- Dry soot
- Volatile evaporation and/or condensation
**Instrumentation: Particle losses**

- **DD correction**
  - depends on particle size, but can be approximated with 5-10% for particles < 1 µm
- **TD particle penetration**
  - depends on particle size according to equation:
    \[
    1 - \eta = -9.7 \cdot \ln(D_p) - 0.5 \cdot Q + 68, \quad D_p < 70 \text{ nm}
    \]
    \[
    1 - \eta = -0.5 \cdot Q + 28, \quad 70 \text{ nm} \leq D_p \leq 500 \text{ nm}
    \]
  - for mass correction 21% at 15 lpm applied
- **FPS correction** under determination
Methods: Modelling

- Particle size distribution evolution was evaluated for
  - Particle growth by coagulation of particles
  - Vapour condensation/evaporation from/to particles
    according to Mason equation (e.g. Williams, M.M. and Loyalka, S.K. 1991. Aerosol Science: Theory and Practice. USA: Pergamon Press. 233 p.)
Modelling: Input

- Original particle number concentration $7 \cdot 10^7 / \text{cc}$ at 100 nm and GSD=2
  - a EURO 3 passenger vehicle

- Residence times: 0.1, 1 and 10 s

- Vapour concentration 5 v-%
  - in diluted sample

- Saturation ratio 1.05 and 1.1
  - depicts rapidness of cooling during dilution
Dilution ratio determination

- **Steady state**
  - dilution ratio stable and well defined

- **Dynamic conditions**
  - CVS – pressure variation minimal
    - dilution ratio well defined
  - high pressure variation occurs
    - serious trouble in determining exact dilution ratios with any sampling system
    - trace gas determination preferred

- FPS designed for dynamic sampling conditions with high pressure fluctuations
  - produces stable samples for pressure sensitive instruments, e.g. TEOM
Results: FPS dilution ratios

- HD engine test, no exhaust system (worst case)
Results: PM concentration results

- DD and TD results corrected for losses
- Effect of volatiles clearly seen (unstable source)
Results: Number concentrations

- ELPI and CPC concentrations agree
- Cooled FPS shows high nucleation at low load
- Not corrected for losses
Results: Typical number size distributions

- Low load: volatile concentration ca. 50% of PM
- Measured with ELPI for 5 min, 15 repetitions
- Soot mode repeatable
- Nucleation mode tendency can be studied

DD and FPS heated

TD

FPS cooled
### Results: Calculated size distributions

![Graph showing calculated size distributions with different cooling conditions](image)

- **Original**
- **0.1 s**
- **1 s**
- **10 s, rapid cooling**
- **10 s, moderate cooling**
Conclusions

- Exhaust sample transformations were successfully governed with a controlled dilution system
- Soot particles were measured repeatably
- Nucleation tendency could be studied, not formation mechanism
- Measurement results had good agreement with modelling results

- Soot mass measurement not strongly dependent on sampling parameters

- For other modes (nucleation mode, number concentration) sampling needs to be defined in detail for comparable and repeatable results