Separation of solid and volatile fraction by thermodesorption and hot dilution

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Abstract

The performance of several devices to either prevent the volatile fraction in diesel exhaust from forming nucleation particles, or to remove volatile particles in order to measure the solid fraction only has been tested in laboratory experiments. The devices include three thermodesorbers (TSI, Dekati, Fraunhofer Institute of Toxicology) and a thermodiluter (Matter Engineering). Particle losses and the removal efficiency of volatile particles were measured for different operating conditions. The results show that it is possible to prevent nucleation but also that there is a certain optimization potential for the envisaged application in the type approval test of diesel engines.
Chassis dynamometer measurements were performed with two thermodesorbers and the thermodiluter. Results from measurements at a stationary operating point show that both system types efficiently remove the volatile particles. The total number of solid particles emitted during a new European drive cycle (NEDC) as measured by a CPC behind thermodesorber/thermodiluter is reproducible.

Introduction

The European particulate measurement programme (PMP) was established to investigate alternatives to the current mass-based emission standards for particulate matter from vehicle exhaust. A candidate method is the measurement of particulate number. However, there is a difficulty in the measurement of particulate number: When the exhaust gas cools down, volatile substances in the gas can form particles by condensation. This formation mechanism is sensitive to the particular sampling conditions in a given system, and therefore makes a pure number-based measurement unreliable. This problem can be overcome by removing the volatile fraction from the exhaust. We tested the performance of 4 systems to remove the volatile fraction in the laboratory: Two commercial thermodesorbers (TSI, Dekati), one experimental thermodesorber operating with a ceramic catalyst substrate instead of activated charcoal (Fraunhofer Institute of Toxicology) and a thermodiluter (Matter Engineering). The Dekati, Fraunhofer and Matter systems were then also used at a chassis dynamometer. The TSI thermodenuder was not included in the chassis dynamometer tests, because we encountered several problems with the device which was supplied to us. An improved version of the TSI thermodenuder has been sent to us after this study finished; the problems mentioned above seem to have been corrected.

Our laboratory tests show that the losses in these devices are generally in the region of 10-25% in the size range from 20-100 nm, for properly working devices when operating within the instrument specification, which we believe to be tolerable. When operating outside the specifications, losses can be as high as 50%. If the particle size distribution is approximately known (which is usually the case for engine emissions), one can compensate for the losses.

The removal efficiency was tested with two heavy hydrocarbons, Triacontane ($C_{30}H_{62}$) and Tetracontane ($C_{40}H_{82}$). Particles made up of these substances were passed through the devices. Triacontane was completely removed even at very high concentrations (10^7 pt/ccm with a mean diameter of ~80nm). Tetracontane is harder to remove from the gas stream due to it's higher boiling point, lower vapor pressure and lower diffusion constant. It was removed at similar concentrations as the Triacontane by the thermodilution system, while the thermodesorbers failed at high inlet concentrations.

We performed chassis dynamometer tests with a candidate system for solid number concentration measurement. At an operation point with high speed and high load, the volatile fraction could be completely removed both with the thermodiluter and with a thermodesorber. We believe that the failure of the thermodesorbers in the laboratory test is due to unrealistically high inlet concentrations of a very heavy hydrocarbon, which does not reflect real-world conditions. A second test showed that the total number concentration over the NEDC as measured by the candidate system is repeatable.

Conclusions

The volatile fraction can be removed reliably both by thermodesorption and by thermodilution. Under extreme conditions, thermodesorbers may fail. However, in our chassis dynamometer experiments, this never happened, although we purposely measured at an extreme operation point where a very high number of volatiles appear. Losses occur both in thermodesorbers and in the thermodiluter (and of course also in the entire sampling system). The losses are not very high, and can be partially compensated for with a calibration if the particle size distribution is approximately known. Total number concentrations measured over the NEDC were repeatable, showing less deviation than the gravimetric measurements done in parallel.

The final report can be downloaded at: http://wanda.fh-aargau.ch/iss/veroeffentlichungen/solid-voaltile_separation_report-final.html

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About PMP

- PMP (Particulate Measurement Programme) is a government-sponsored programme, which was established to develop a new system for measuring ultrafine particles emitted from vehicles to complement or replace existing particulate mass measurement systems
- The number concentration of solid particles in the exhaust is a good candidate because.... Learn more tomorrow evening!

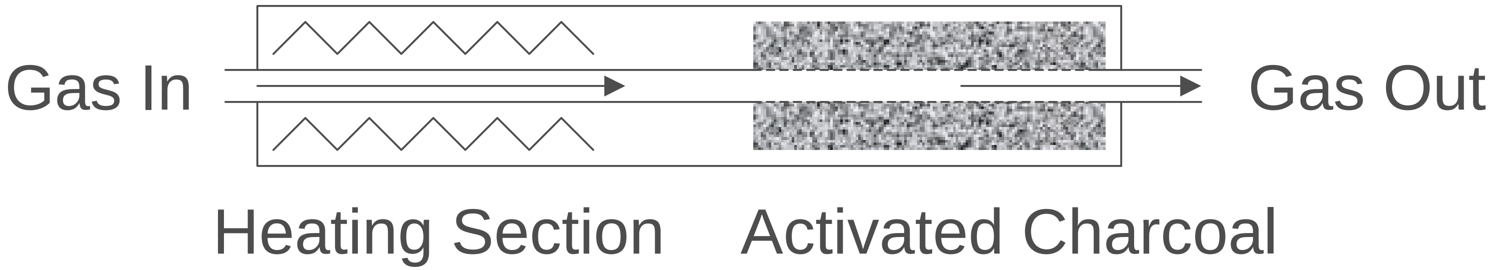
The Problem

- Volatile substances can form particles by nucleation when the exhaust gas is cooled
- The number of volatile particles can be orders of magnitude larger than number of solid particles under „bad“ conditions
- For a reliable number concentration measurement the volatile fraction must be removed

Thermodesorber Principle

e.g. Dekati, TSI, (Fraunhofer Prototype)

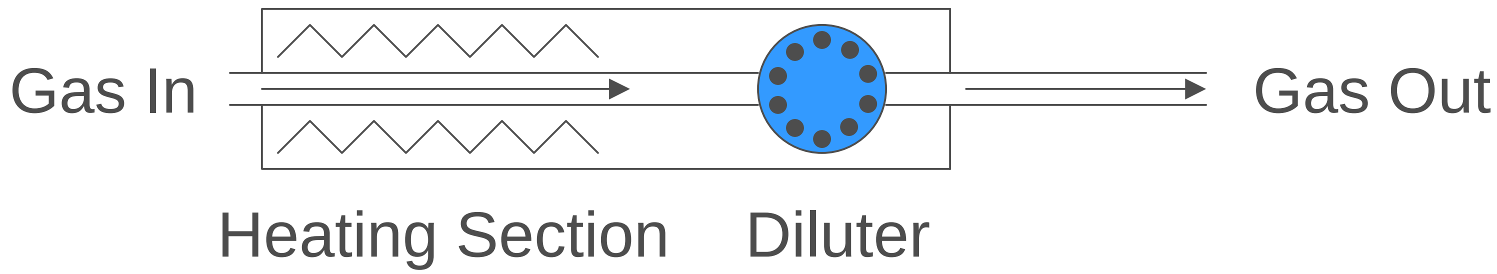
Volatiles are evaporated in a heating section. The hot gas flows past activated charcoal; the volatile vapor diffuses into the charcoal section and is trapped.



Thermodiluter Principle

Matter Engineering Prototype:

Volatiles are evaporated in a heating section, the vapor is diluted, and due to the dilution no recondensation takes place



Device Characterization

Laboratory Experiments (all 4 devices):

- Particle losses
- Removal Efficiency
- Temperature Profiles

Chassis Dynamometer Experiments (only Dekati thermodesorber and ME thermodiluter prototype)

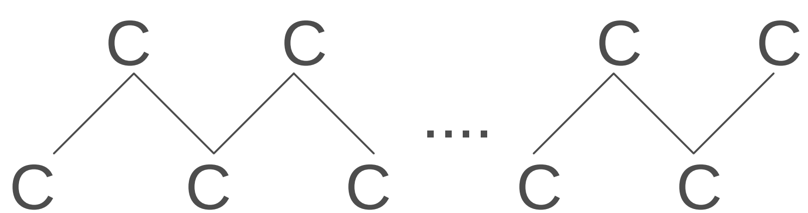
- Fixed operating point with high volatile fraction
- European drive cycle with/without volatile trap

Particle Losses

- Losses are a function of particle size, flow rate and temperature
- Losses are typically between 10 and 50%
- Losses are not very critical: If the particle size distribution is known (as is usually the case for vehicle exhaust), the losses can be compensated for with a suitable calibration

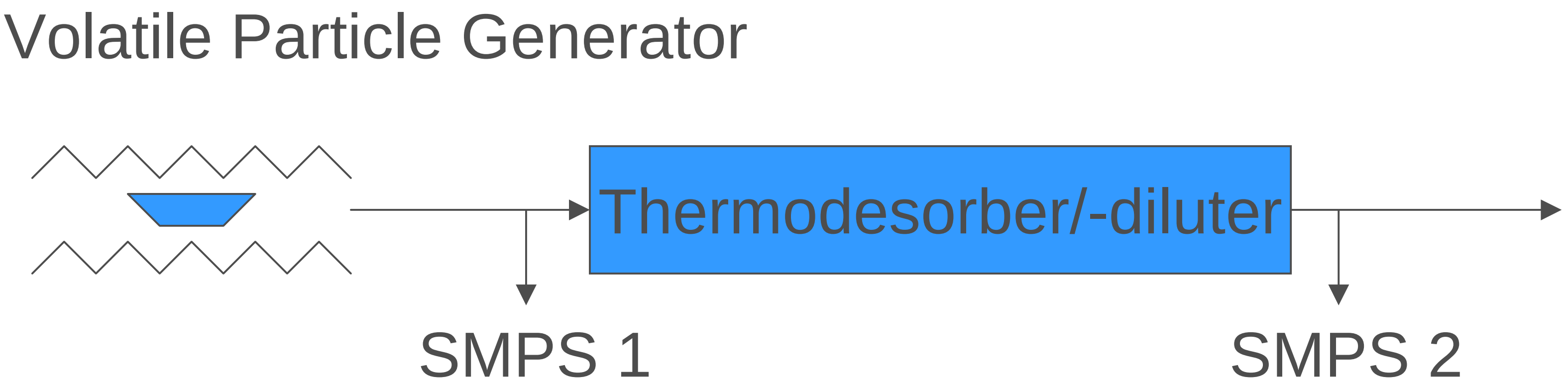
Removal Efficiency (1)

- What concentration of volatile particles can be completely removed?
- What happens when the volatile concentration is too high – how do devices fail?
- We used Triacontane ($C_{30}H_{62}$) and Tetracontane ($C_{40}H_{82}$) Particles:



Removal Efficiency (2)

- **Experimental Setup to determine removal efficiency:**

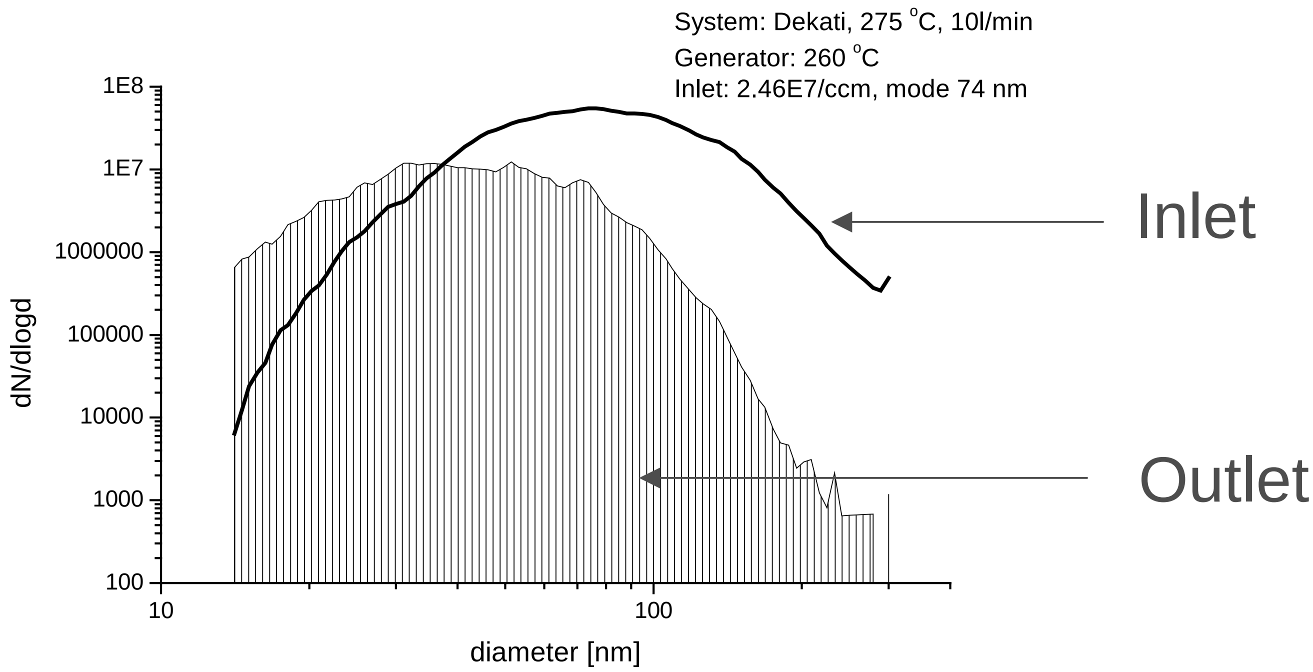


Removal Efficiency (3)

- Triacontane was completely removed by all devices at concentrations of 10^7/ccm and particle diameters of ~80nm
- Tetracontane was completely removed by the thermodiluter at all concentrations. The thermodesorbers were unable to remove (unrealistically?) high concentrations of tetracontane

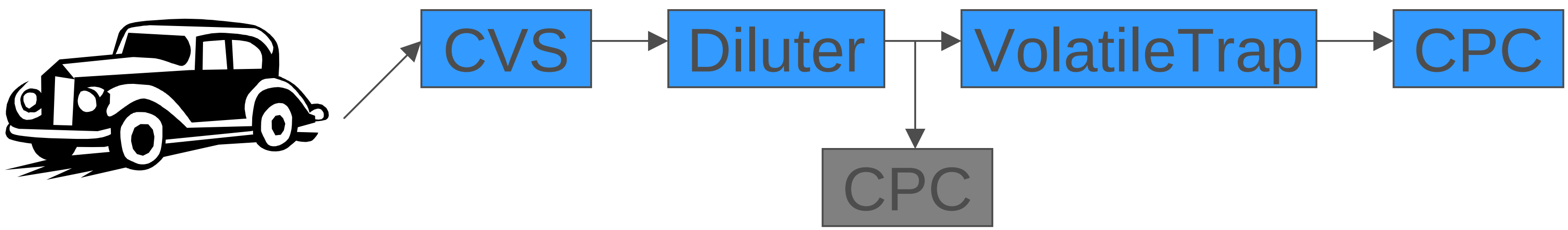
Removal Efficiency (4)

- **Example: Dekati Thermodesorber at 10l/min**



Chassis Dynamometer Tests

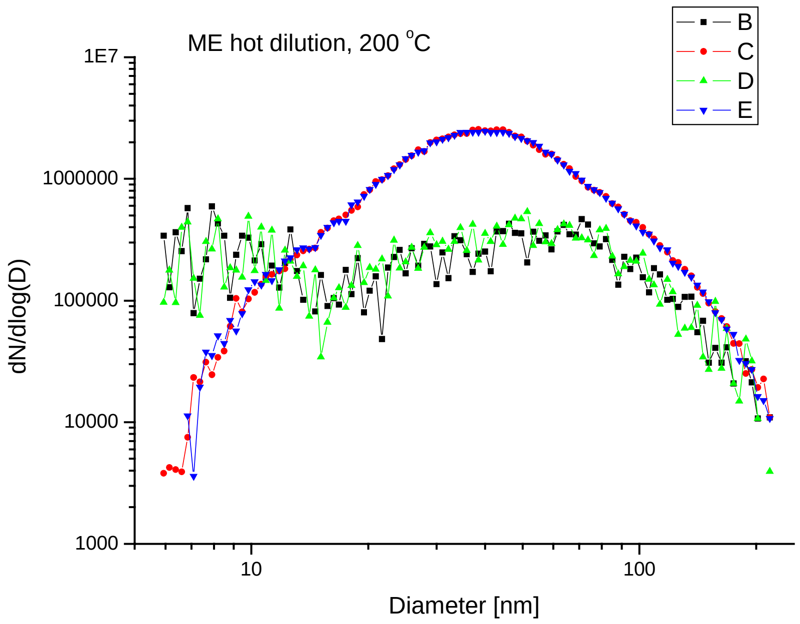
- Test a PMP candidate system to see...



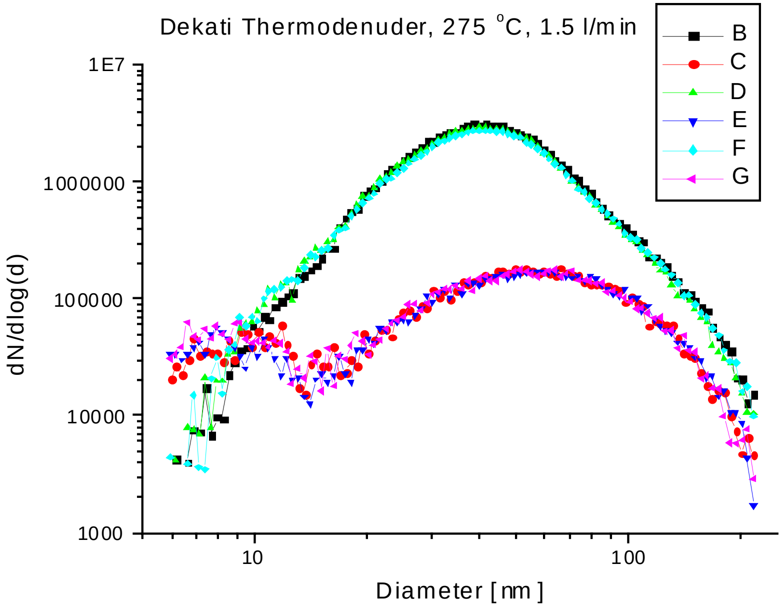
- Can all volatiles be removed?
- Are repeatable number concentration measurements possible in drive cycles?

High Speed & High Load

Thermodiluter

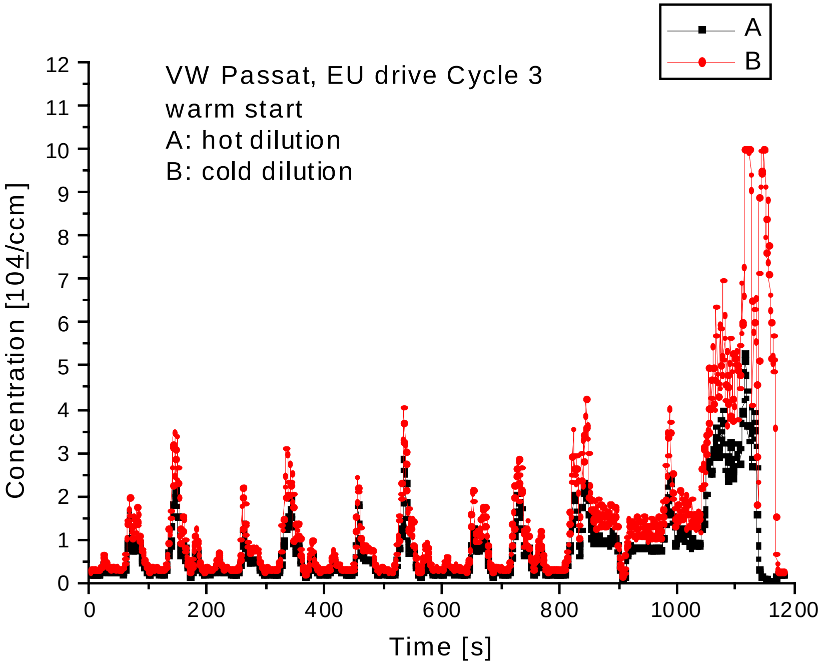


Dekati Thermodesorber

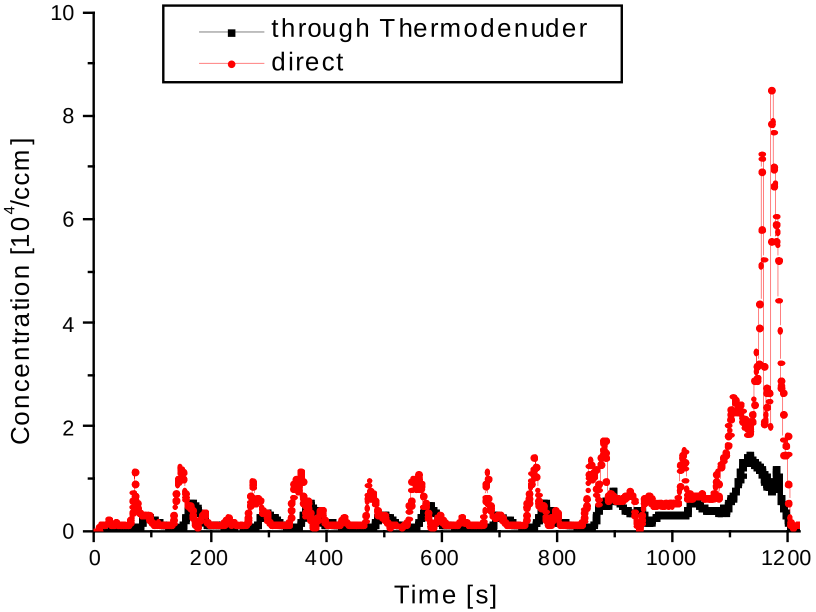


European Drive Cycle (1)

Thermodiluter



Dekati Thermodesorber



European Drive Cycle (2)

- Comparison of solid number concentration measured through thermodiluter and gravimetric analysis: it‘s repeatable!

Cycle Number	Number Concentration [a.u.]	Gravimetry [g/km]
1	7.4	0.025
2	7.6	0.028
3	7.6	0.030

Conclusions

- The volatile fraction can be reliably removed with a thermodiluter. Thermodesorbers can fail at very high concentrations of material with low volatility; this did not happen during our chassis dynamometer tests despite extreme conditions
- Particle losses occur in both thermodiluters and thermodesorbers; they can be compensated for with a suitable calibration
- Repeatable number concentration measurement in the European drive cycle is possible with both thermodesorbers and thermodiluters