High Temperature Condensation Particle Counter (HT-CPC)

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Outline

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Motivation (1)

• Diesel particles are getting a bad press “Lyon, France, June 12, 2012 -- After a week-long meeting of international experts, the International Agency for Research on Cancer (IARC), which is part of the World Health Organization (WHO), today classified diesel engine exhaust as carcinogenic to humans (Group 1), based on sufficient evidence that exposure is associated with an increased risk for lung cancer.”

• Particle number is here to satay

• Though recent studies suggest that for new vehicles, PM is heading towards the urban background, and NOx may be a bigger health concern.
Engine out, light-load, low soot conditions: Most of the number emissions are solid with $D_p < 23$ nm.
Spark ignition engines can also produce tiny solid nanoparticles, especially with metal additives.

Euro 3 passenger car, 10 ppm Mn in fuel, data courtesy Johnson-Matthey

Courtesy of Dr. Kittelson
Motivation (2)

- European legislated particle number method ("PMP") requires a complex system for the removal of volatile material, and cooling of the sample to ambient temperature, prior to measurement by a conventional CPC.

- This work is concerned with development of a HT-CPC that is insensitive to volatile particles (even though the PMP legislation would not allow the use of such an instrument as written), and permits “hot” inlet conditions. Whether or not such an instrument has relevance to PMP is not addressed here.
The HT-CPC – Requirements

Desirable characteristics

- Operating temperature sufficiently high to avoid nucleation of all volatile material, without the need for upstream removal.
- Mass diffusivity of fluid < thermal diffusivity of carrier gas
- Fluid stable at high temp, and in presence of oxygen etc. Non-toxic.
- Fluid vapour pressure appropriate at working temperature

Cooling Type Condenser
- High boiling point fluids are likely to have low mass diffusivity
- Optical Counter easier to keep at a safe temperature
How hot should a HT-CPC be?

- Difficult question! (Even without considering (hot) dilution in order to maintain single particle count mode.)

- Experience suggests that at $\geq 150 \, ^\circ C$, no nucleation particles from a real-life engine sample will survive.

- For example (borrowing the PMP tetracontane test) if $10^4 \, \#/cc \, C_{40}H_{82}$ particles with $d_p = 30 \, \text{nm}$ are fully vapourised, $p_{\text{sat}} \approx 4.5 \times 10^{-7} \, \text{Pa}$, corresponding to $t_{\text{sat}} \approx 90 \, ^\circ C \, (\approx 360 \, \text{K})$.

- Perhaps $150 \, ^\circ C$ would be “safe” ($p_{\text{sat}} \approx 4 \times 10^{-3} \, \text{Pa}$).
• The model we have developed solves heat and vapour transfer in the condenser for the profiles of temperature, vapour pressure and hence saturation ratio.

(Comparison – using Butanol as the working fluid, $T_{\text{saturator}} = 35 \, ^\circ\text{C}$, $T_{\text{condenser}} = 10 \, ^\circ\text{C}$)
Modelling – Counting efficiency

- Assuming that all activated particles grow to detectable sizes, the counting efficiency is equal to the activation efficiency.

- Using the contour of Kelvin-equivalent activation diameter and particle concentration profile due to diffusion, the activation efficiency is \#activated particles / \#total inlet particles.
Experiment

Experimental Set-up

Compressed Air → NaCl Atomiser → Diffusion Dryer → Aerosol Neutralizer → Differential Mobility Analyzer TSI DMA

Pump → TSI 3775 CPC

0.3 l/min → Atmospheric Pressure

Mono-disperse Aerosol

Spill → HT-CPC

Pump → Compressed Air

Sheath Flow → Hepa Filter

Nitrogen → Valve
Typical operating conditions

- Optical Counter
- Condenser
- Saturator

\[ T_{\text{saturator}} = 230 \, ^\circ\text{C} \]
\[ T_{\text{condenser}} = 180 \, ^\circ\text{C} \]

Current hardware limits temperature to 315 \(^\circ\text{C}\)

- \( Q_{\text{total}} = 0.3 \, \text{l/min} \)
- \( Q_{\text{aerosol}} = 6\% \) of \( Q_{\text{total}} \)

SiC – DPF brick

Nitrogen or air

Fluid

\( \sim 200 \, ^\circ\text{C} \)

\( < 315 \, ^\circ\text{C} \)
Fluid Candidates

- Di – Ethylhexyl Sebacate (DEHS)
- Perfluorocarbon
  - Fomblin Y 6/6
  - Fomblin Y 25/6
- 5-ring Polyphenyl-ether (Santavac 5)
- Tetraphenyl-tetramethyl-Trisilicane
  - DC704
  - K J Lesker 704
Counting Efficiency (data corrected for diffusion losses and counting efficiency)

- **Predicted Counting Efficiency for Solid Particles**
  - Tetracontane ( >10,000#/cc)
  - NaCl

**Measurement not possible with available DMA** which results in degraded counting efficiency of NaCl. However, it allowed significant test duration.

**DEHS decomposed over time**

**Temperatures:**
- $T_{\text{saturator}}$ 230 °C
- $T_{\text{condenser}}$ 180 °C
Perfluorocarbons and Polyphenylether

• Perfluorocarbons (Fomblin Y 6/6 and Fomblin Y 25/6) are stable up to about 290°C. However, they were found unable to grow NaCl particles under the homogeneous-nucleation-free conditions. Possibly because they do not wet the particles.

• Polyphenylether (Santavac 5) has an excellent decomposition temperature of 443°C. However, it was found unable to grow NaCl particles. Possibly due to low vapour pressure.
Tetraphenyl-tetramethyl-Trisilicane (DC704)

Counting Efficiency (data corrected for diffusion losses and counting efficiency)

- NaCl
- Tetracontane (>10000#/cc)
- DPG Soot
- Ambient particles

Measurement not possible with available DMA

T<sub>saturator</sub> 250 °C
T<sub>condenser</sub> 205 °C

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Tetraphenyl-tetramethyl-Trisilicane (Lesker704)

Counting Efficiency (data corrected for diffusion losses and counting efficiency)

Measurement not possible with available DMA

NaCl

Soot

Tetracontane

Ambient

Tetracontane (>10000 #/cc)

DPG Soot

Ambient Particles

Theory

T_{saturator} 250 °C

T_{condenser} 205 °C

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Tetr phenyl-tetramethyl-trisiloxane (Lesker705)

Counting Efficiency (data corrected for diffusion losses and counting efficiency)

- **NaCl**: Electrical mobility Diameter (nm)
  - NaCl particles
- **Soot**: Tetracontane (>10000 #/cc)
- **Ambient**
- Measurement not possible with available DMA

Electrical mobility Diameter (nm)

- Tetracontane
  - (Lesker705)
- DPG soot particles
- Ambient particles
- Theory

**T**
- **_{saturator}** 290 °C
- **_{condenser}** 215 °C

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Conclusions

- A HT-CPC using Di-Ethylhexyl-Sebacate (DEHS) as the working fluid running at 230/180 ºC has been built and tested.
- The measured counting efficiency for NaCl particles (solid particles) agrees well with model predictions.
- Tetracontane particles of electrical mobility diameter range 7nm – 310nm @ concentration higher than $10^4$#/cc were removed with about 99% efficiency, even without sample pre-heating or dilution.
- DEHS is not suitable due to thermal decomposition
- However, the DEHS CPC has successfully demonstrated the HT-CPC concept, though not as yet on combustion-generated particles.
Thank you

High Temperature Condensation Particle Counter

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