Characterization of fine and ultrafine particles in emissions from CHP Plants in Denmark

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Keywords: Combustion, aerosols, emissions, characterization

Introduction

CHP (Combined Heat and Power) plants are being increasingly used in Denmark due to their ability to utilize the heat generated from energy production. The type and magnitude of air emissions from CHP plants vary widely according to the energy source used and technology implemented for combustion and emission reduction. Ultrafine and fine particles emitted from nine Danish CHP Plants have been measured and characterized. The plants selected were fired by waste (WTE), biomass (BM), gas (GF) or gasoil (GO). A detailed description of the project is given in Fuglsang et al. (2009).

Methods

The methodology used for characterization of particles emitted from CHP plants were based on the following techniques:

1. Sampling of particles from the flue gas in a 12-stage low pressure cascade impactor (ELPI). Emissions of PM$_{0.1}$, PM$_1$ and PM$_{2.5}$ were calculated from the real-time number concentrations measured by means of ELPI.
2. Analysis of the collected particles by:
   - EDX (Energy dispersive X-ray spectroscopy): Analysis of elements in selected clusters of particles.
   - FIB-SEM (Focused Ion Beam-SEM): 3D cross section analysis of 2-3 selected particles.

Results and conclusions

The most abundant elements found by the EDX analysis in the different size fractions from the waste incineration plants and from the gas fired plants are shown in figure 1 and 2. The figures show representative results for these two plant types. For a description of the results from all plant types involved in the study, reference is given to Fuglsang et al. (2009).

In the figures below, the mass concentration in each size interval is indicated by marking the elements as follows: Bold: [10;100] % w/w; normal font: [2;10] % w/w; brackets: [0.2;2] % w/w.

Figure 1. Most abundant elements found by EDX analysis of the particles emitted from waste incineration plant WTE2. Particles sampled after the plant’s ESP.
Figure 2. Most abundant elements found by EDX analysis of the particles emitted from the landfill gas fired CHP plant GF2.

<table>
<thead>
<tr>
<th>Number</th>
<th>Mass</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.0E+00</td>
<td>1.0E+03</td>
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<tr>
<td>0.01</td>
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<tr>
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<td>10</td>
<td>10</td>
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</table>

FIB-SEM analysis of selected particles showed relatively large, hollow soot particles in the samples from gas fired engines. An example of a hollow soot particle is shown in figure 3. These hollow particles could be formed from rapid combustion of small droplets of lubrication oil entering the combustion chamber from the piston cylinder walls. Solid soot particles found by FIB-SEM from plant GF2 seemed to consist of a solid nucleus with a surface layer, probably formed by heterogeneous condensation of organic compounds during the combustion process.

Figure 3. Hollow, carbonaceous particle from gas fired plant GF1, cut through by means of FIB.

Key findings were:

- A significant enrichment of metals (e.g. Fe, Mn, Cu) in the PM0.1 and the PM1 fractions were found for particles from both WTE and BM plants.
- The particles from the WTE plants were generally found to be crystalline and porous in structure. A relatively high fraction of K and Na were found in the fraction between 0.1 µm and 1 µm. For PM > 1 µm, a high content of Ca, S, O and C were found. This is explained by the formation of CaSO4 during the desulphurization process at the WTE plants.
- Carbon and to some extend also oxygen dominates in the PM0.1 and PM1 fractions from the biomass, gas- and gasoil fired plants.
- Spherical soot particles dominate in the particles from the gas- and gasoil fired plants, and to a lesser extend also from the BM plants.
- The high number of particles emitted from the GF plants is most probably caused by the use of lubrication oil in the plants’ gas engines.
- The FIB-SEM analysis of particles from the GF plants showed that spherical soot particles of a diameter of 150 nm – 1500 nm from these plants may be hollow as well as solid.

This work was supported by Energinet.dk under the project framework ForskEL.

Characterization of fine and ultrafine particles from Combined Heat and Power (CHP) plants in Denmark

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ETH Conference on Combustion Generated Nanoparticles, Zürich, 4 Aug. 2010
<table>
<thead>
<tr>
<th>CHP Plant type</th>
<th>Plant id.</th>
<th>Emission control system</th>
</tr>
</thead>
<tbody>
<tr>
<td>Waste to Energy (Municipal solid waste incineration)</td>
<td>WTE1</td>
<td>DeNOx (SNCR) → desulphurization (2-stage scrubber incl. CaO addition) → addition of activated charcoal → agglomeration filter (venturi jet system)</td>
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<tr>
<td></td>
<td>WTE2</td>
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<td>WTE3</td>
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<tr>
<td>Straw-fired</td>
<td>BM1</td>
<td>Bag filter</td>
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<tr>
<td></td>
<td>BM2</td>
<td></td>
</tr>
<tr>
<td>Wood chips/ sawdust combustion</td>
<td>BM3</td>
<td></td>
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<tr>
<td>Natural gas</td>
<td>GF1</td>
<td>2 gas fired</td>
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<tr>
<td>Biogas</td>
<td>GF2</td>
<td></td>
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<tr>
<td>Gasoil fired</td>
<td>GO</td>
<td>1 gasoil fired</td>
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</tbody>
</table>
Sampling setup

[Diagram of sampling setup with labels for Flue gas, Pressurised Air Heater, DILUTION STEP, Clean, pressurized air, Corona charger, ELPI, Computer and control electronics, Electrometers, Impactor with insulators and contact needles, Vacuum pump, ASCII datafile, RS-Z32 serial]
Sampling setup

• Akkrediteret måling af PM10, NOx, SO2, CO, O3
Methods used for characterization

- SEM (Scanning Electron Microscopy)
- EDX (Energy Dispersive X-ray analysis)
- FIB-SEM (Focused Ion Beam-SEM)
FIB-SEM analysis
RESULTS for waste- and gas fired plants
Morphology - size distributions - analysis of elements
Waste fired CHP plants:
SEM analysis of representative particles
Waste fired CHP plants WTE2 (bag filter)
Average size distribution

- **dN/dlogDp** [1/cm³(n)]
- **dM/dlogDp** [mg/m³(n)]

**Number** — **Mass**

N/A
Waste fired CHP plants:
Most abundant elements found by EDX - plant WTE2

- Number
- Mass

- K, Cl, O, C, Na
- Fe, Mn, O, Cu
- Ca, O, C, S, (Na), (K), (Fe), (Si)

N/A
Gas fired CHP plants: SEM analysis of representative particles
FIB-SEM 3D analysis: Plant GF2
FIB-SEM 3D analysis: Plant GF1
Gas fired CHP plants:
Most abundant elements in particles from landfill gas fired plant GF2
CONCLUSION (1)

WTE plants:

– The particles were generally crystalline and porous in structure.
– A significant enrichment of metals (e.g. Fe, Mn, Cu) were found in the PM0.1 and the PM1 fractions.
– A relatively high fraction of K and Na were found in the fraction between 0.1 µm and 1 µm.
– A high content of Ca, S, O and C in PM > 1 µm may be explained by the formation of CaSO₄ and CaCO₃ during the desulphurization process.
Gas fired plants:

- The high number of particles emitted is most probably caused by the addition of lubrication oil to the gas fired engines.
- Soot particles emitted may be hollow as well as solid.
- Soot particles consisting of a solid nucleus and a surface layer were probably formed by heterogeneous condensation during the combustion process.
- The formation mechanism for hollow particles could be related to rapid combustion of lubrication droplets in the engine.
Acknowledgements

Energinet.dk

Co-authors

– Jacob Marcussen
– Karina Brotoft
– Thue Grønhøj Frederiksen
Report:

Characterization of ultrafine and fine particles from CHP Plants (2009). ForskEL project No. 2008-1-0071

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