Ultrafine and nanoparticle emissions from residential-scale heating appliances

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Extended summary

Interest in ultrafine (diameter < 0.1 µm) particles have grown of late because some health researchers proposed that these aerosol size fractions may have a greater biological effect than an equal mass of larger particles and can deposit in the lungs or penetrate into the interstitial sites and access blood circulatory system, moving from the lungs to other organs, thus producing pathologies in organs not directly exposed (Oberdorster et al., 2005).

Given these health effects, the determination of the ultrafine particle levels becomes very important especially in urban areas where much of the population is daily exposed to particle emissions from mobile sources and domestic heating. In urban areas, the spatial density and limited flue gas release height of these combustion sources may lead to important impacts on air quality, and unfavorable meteorological conditions may cause direct and prolonged exposure of the individuals to these emissions.

The main objective of this work is to provide field experiment data on ultrafine and nanoparticle (diameter < 0.05 µm) real-world emissions in the flue gas of small scale (heat output < 1MW) combustion installations. For this purpose measurements were carried out on a laboratory test bench for boilers at Stazione Sperimentale per i Combustibili.

The boilers investigated are representative of the of the typical residential scale heating appliance population in Italy, the first one being an advanced technology wood pellets boiler of 100 kW thermal output, equipped with an axial dust removal device. The boiler reaches high combustion efficiency and low emissions due to gasification of the fuel on a moving grate with minimal primary air and the combustion of the combustible gas-secondary air mixture in the rotary combustion chamber. The second boiler investigated is a two pass reverse flame boiler of 150 kW heat output, not provided with particulate matter control devices. This boiler is equipped with interchangeable burners for light oil and natural gas light. All measurements were conducted only after the boiler had reached the steady state.

The fuels used during testing are characterized by a low sulfur content, and, in particular, regarding the aerosol forming species the sulfur content for the light oil is 0.09$\%_w$, the water and sediments are less than 0.005$\%_v$; the wood pellets has a chlorine content of less than 0.01$\%_w$, a sulfur content around 0.03 $\%_w$, and moisture around 7$\%_w$.

The test cycles consisted of the sampling of the flue gas with and without dilution (hot sampling) and the measurement of size resolved number concentration of the particles with aerodynamic diameters in the range of 0.007-10 μm. Dilution sampling was applied for simulating, as close as possible, the behavior of the emissions under atmospheric dispersion, dilution and cooling conditions, and the results were compared with those of the hot sampling. While the undiluted gas samples contain the primary particles, the diluted samples may contain also the condensable particles. Such sampling strategy allows to evaluate the influence of the dilution and cooling of the flue gas on particle emissions. The influence of combustion efficiency is also investigated by operating the boiler under nominal (heat output and combustion air supply at design conditions) and non nominal (simulation of the worse combustion by decreasing the load or reducing the combustion air supply) conditions.

Figure 1 shows the sampling and measurement line used in the experimental investigation for hot sampling (Figure 1A) and for dilution sampling (Figure 1B). Hot sampling of flue gas at actual stack conditions was performed with a standard filterable particulate matter stack sampling configuration, modified by substituting the final hot filter for particulate collection with the particle number counting device: the sampled gas flows through the stainless steel heated probe, equipped with a PM2.5 cyclone, and reaches the particle measurement instrument approximately at stack gas temperature.

In dilution the flue gas is extracted isokinetically from the exhaust stream through a heated line, equipped with a PM10 and a PM2.5 cyclone in series positioned at the initial track of the sampling probe, and passed through a venturi into a conical mixing zone, where it is cooled and diluted with HEPA filtered and dehumidified air prior to the entrance into a Teflon coated tubular residence chamber (∅=102mm, L=559mm), with the particle measurement instrument connected at the rear end of the chamber. Cyclone separators and venturi are heated above stack gas temperature, in order to prevent condensation of the semivolatile species present in the gas stream. The dilution procedure, which followed the EPA CTM-039 (US EPA, 2004), is regulated automatically: the system operating design is adjusted to achieve dilution ratios ranging from 10 to 50, corresponding to residence times in the chamber of 2 to 0.5 s, and is provided with temperature and relative humidity sensors to control the required sampling conditions.

Number concentration and size resolved distributions are measured with an electrical low pressure impactor (ELPITM 10 lpm - Dekati Ltd., Finland), with 12 stages and a final filter stage (∅=47mm) for detecting particles with aerodynamic diameters in the range of 0.007-10 μm (Marjamaki et al., 1999). The operating principle is based on particle charging, inertial classification in a cascade impactor and electrical detection of the aerosol particles separated by impaction in every single stage (Keskinen et al., 1992). Greased aluminum foils were used as impactor substrates. The impactor was equipped with an external heating system to regulate the temperature of the impactor stages up to the stack gas temperature during hot sampling.

For all the sources investigated, results obtained for different dilution ratios were analyzed and compared with conventional hot sampling measurements, thus obtaining insights about the share of condensation effects on total particulates emitted, whereas monitoring of local background ambient concentration values were utilized for evaluating the corresponding contribution from combustion air. Particle number emission factors are determined based on fuel feed rate of the boiler.

The measurement results are expressed in terms of total particle number concentration (TN) and the relative contribution of UFP and NP to TN. Plots in Figure 2 report the statistics of interest derived from 1 sec measurement data (mean, median, inter-quartile range, minimum and maximum values) multiplied by the dilution ratio, and referred to dry gas at 0°C, 1 atm and reference flue gas oxygen content (6 $\%_v$ for solid fuels, 3 $\%_v$ for liquid and gas). Figure 3 shows lognormal distributions fitted to particle size distributions observed during dilution sampling.

For the wood pellet boiler operating average number concentrations measured result $6.8 \cdot 10^7$ cm^{-3} at low dilutions (DR=15-20), $6.6 \cdot 10^7$ cm^{-3} at medium dilutions (DR = 20-35) and $7.8 \cdot 10^7$ cm^{-3} at the highest dilutions applied (DR=35-50), with an overall average value of $7 \cdot 10^7$ cm^{-3}. The mode diameter of the size distributions is positioned around 0.072 μm, with the fraction of UFP accounting on average for nearly 95% of TN. Variation of applied DR is seen to have minor effects on number concentrations, however the contribution of NP to TN slightly increases with increasing dilutions. Hot sampling results in number concentrations exceeding the detection limits of the ELPI impactor (~6.7 cm^{-3} at sampling conditions). TN is observed to be higher than background combustion air levels for all the test conditions. Average number emission factors included between $2.6 \cdot 10^{14}$ to $3.6 \cdot 10^{14}$ particles/kg$_{fuel}$ are estimated.

TN measured from light fuel oil boiler shows a rather clear dependence with dilution. Increase in number concentrations from an average value of $1.1 \cdot 10^7$ cm^{-3} at the higher dilution ratios (DR=35-50) to $2.7 \cdot 10^7$ cm^{-3} for the medium ratio (DR=20-35) up to $7.7 \cdot 10^7$ cm^{-3} for the lowest

DR (DR=15-20) were observed, with the latter conditions resulting thus in values comparable with wood pellet measurements. Despite this, no measurable variations were detected in particle size distributions, with very similar results in the whole range of dilutions applied and without any apparent shift in the mode diameters, located around 0.02 µm for all dilutions. The particle number concentration measured during hot sampling of the light oil boiler under nominal operation, was on average $1.8 \cdot 10^6$ particles cm^{-3}, whereas during the non nominal boiler operation worsening combustion efficiency reflected on increased number concentrations ($7.7 \cdot 10^6$ cm^{-3}). The fraction of UFP accounts on average about 99% of TN for all test conditions. A clear increase is observed in the contribution of particles in the NP fraction which account for 37% during hot sampling and 92% on average for dilution sampling, thus pointing out the effect of flue gas dilution cooling on enhancing the new particle formation arising from nucleation/condensation processes. Average number emission factors included between $2.2 \cdot 10^{13}$ to $1.0 \cdot 10^{15}$ particles/kg_{fuel} are estimated.

TN levels measured for natural gas fired boiler result, as expected, in the lower range of values detected for all the experiments. An average TN of about $6 \cdot 10^3$ cm^{-3} is obtained when operating at a dilution ratio of 15, whereas non detectable values are registered at higher dilutions. Resulting concentration levels are almost an order of magnitude lower than background values measured in combustion air, included in the range $2\text{-}4 \cdot 10^4$ cm^{-3}. As for light fuel oil boiler the mode diameter is located around 0.02 µm. An average number emission factors of $9.3 \cdot 10^{10}$ particles/kg_{fuel} is estimated.

Conclusions

The emissions are in a strict relationship with the fuel type and boiler operating conditions, and range by about five orders of magnitude ($9 \cdot 10^{10}$ particles/kg_{fuel} to $1.0 \cdot 10^{15}$ particles/kg_{fuel}) from very low emission levels for natural gas boiler to higher levels observed for light oil and pellets boilers. The dilution sampling suggests enhanced particle formation upon dilution and cooling of the flue gas. In particular, an increase in the number concentration of particles with diameters less than 50 nm is observed after dilution. Average size distributions present mode diameters positioned mainly in the ultrafine particles which account for more than 90% of the total particle number concentration. The comparison of the emission results with local ambient air measurements indicate that small combustion installations, which usually are not equipped with flue gas cleaning devices, are not small emitters of ultrafine particles.

The findings highlight the dynamic nature of the particles, and the consequent potential underestimation of emission factors determined with conventional hot sampling methods. Dilution sampling approach such as used in this study helps to approximately estimate the fate of the particle emissions once released into the atmosphere.

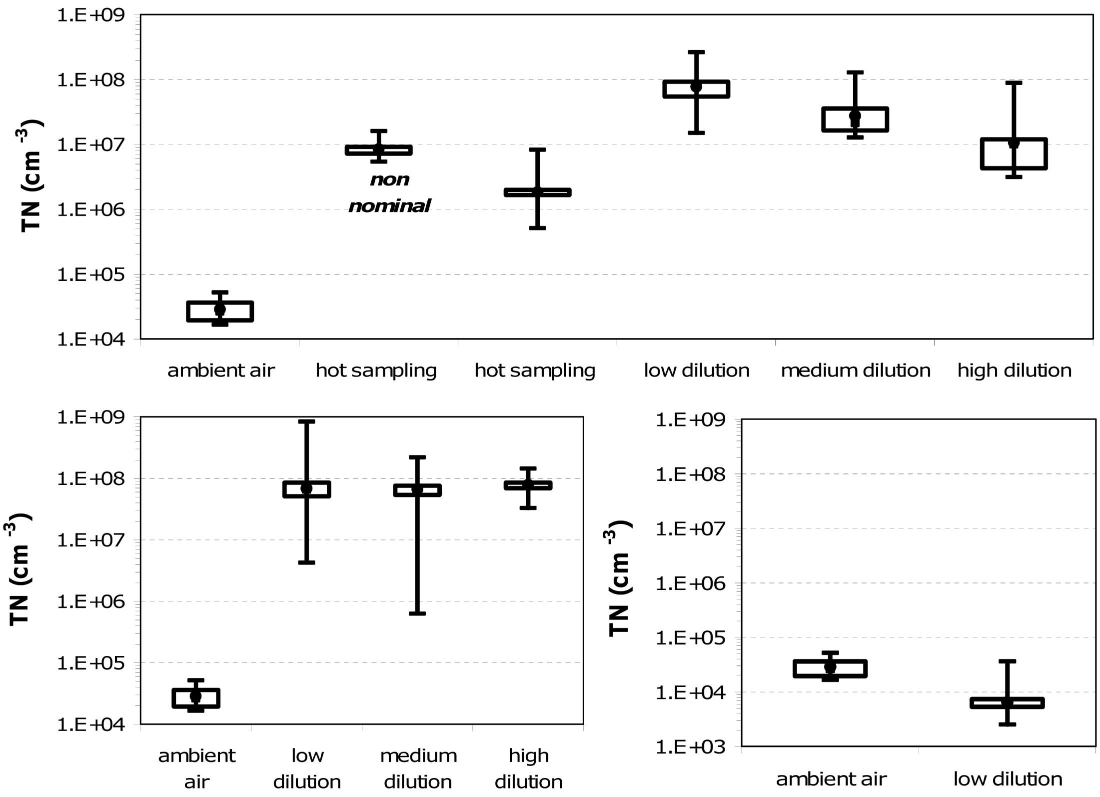

Figure 2. Total particle number concentrations (cm^{-3}) measured.

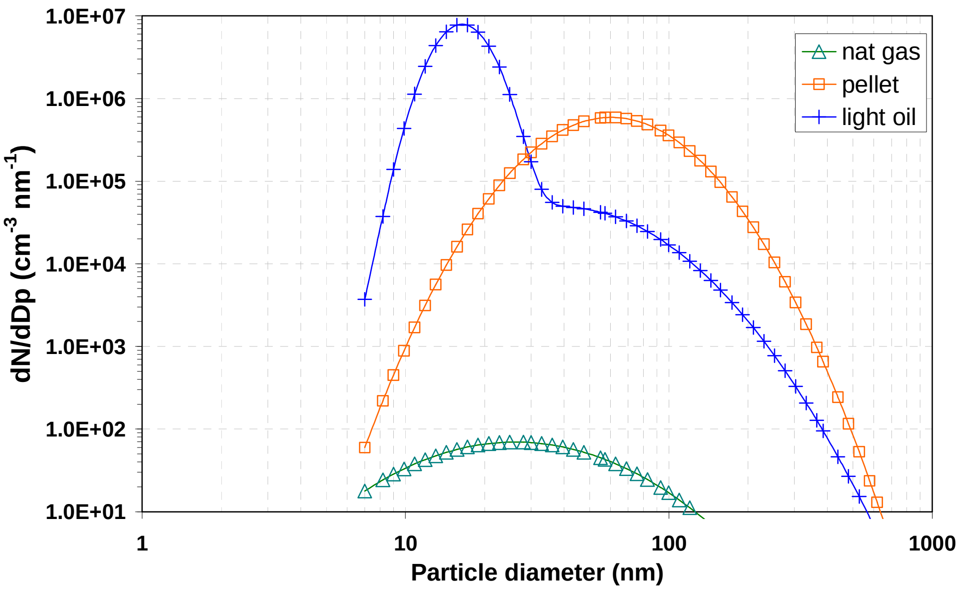

Figure 3: Lognormal distributions fitted to particle size distributions observed during dilution sampling.

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Ultrafine particle emissions from small scale combustion installations

Senem OZGEN

INTRODUCTION

Ultrafine particles (UFP)

- UFP < 100 nm and nanoparticle (NP) fraction < 50 nm
- health concerns (high toxicity, translocation effects)
- main environmental concerns
  - nanotechnologies
  - nanomaterials
  - indoor exposures
- recent attention to combustion emissions
  - most data available for vehicle exhaust
  - limited investigations for stationary sources
- measurement issues
  - significance in terms of number rather than mass
  - no standard protocol for stationary sources
  - condensable particulate matter

INTRODUCTION

Objectives of the study

- providing field experiment data of UFP emissions of small scale combustion installations
- sampling campaigns on full scale residential heating units working with conventional fuels (natural gas, light oil, pellets)

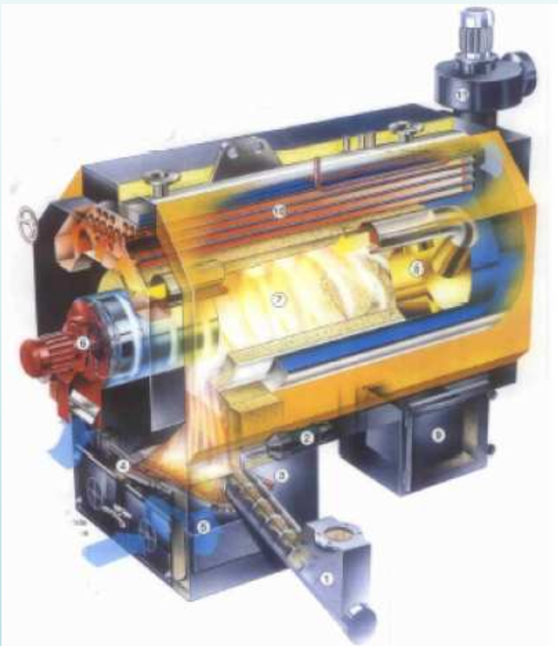
Small scale combustion installations

- residential heating - limited flue gas release height
- urban areas - high spatial density
- direct and prolonged exposure of the individuals
- no emission control device

MATERIALS AND METHODS

Sources investigated – Pellets boiler

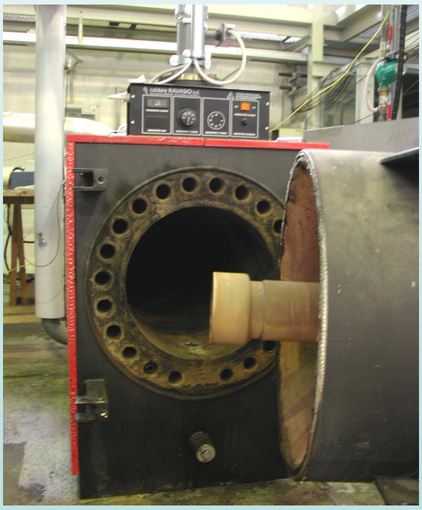
Advanced wood pellet boiler (output 100 kW) two stage combustion (moving grate secondary swirling vortex chamber) with flue gas recirculation, axial flow cyclone for particulate removal (pellet FR: 21.4 kg h^{-1})



FR: fuel feed rate

Sources investigated – Light oil / Natural gas boiler

Conventional design two-pass reverse flame boiler (150 kW) equipped with interchangeable burners for light oil and natural gas without any particulate control device (light oil FR: 13 kg h^{-1}; natural gas FR: 17.5 m_n^3 h^{-1})



FR: fuel feed rate

Sampling conditions

- hot sampling: determination of emissions at stack conditions (e.g, temperature, relative humidity)
- dilution sampling: simulation of the of the atmospheric dilution and cooling
  - low dilution DR=10-20
  - medium dilution DR=20-35
  - high dilution DR=35-50

Comparison gives condensable PM

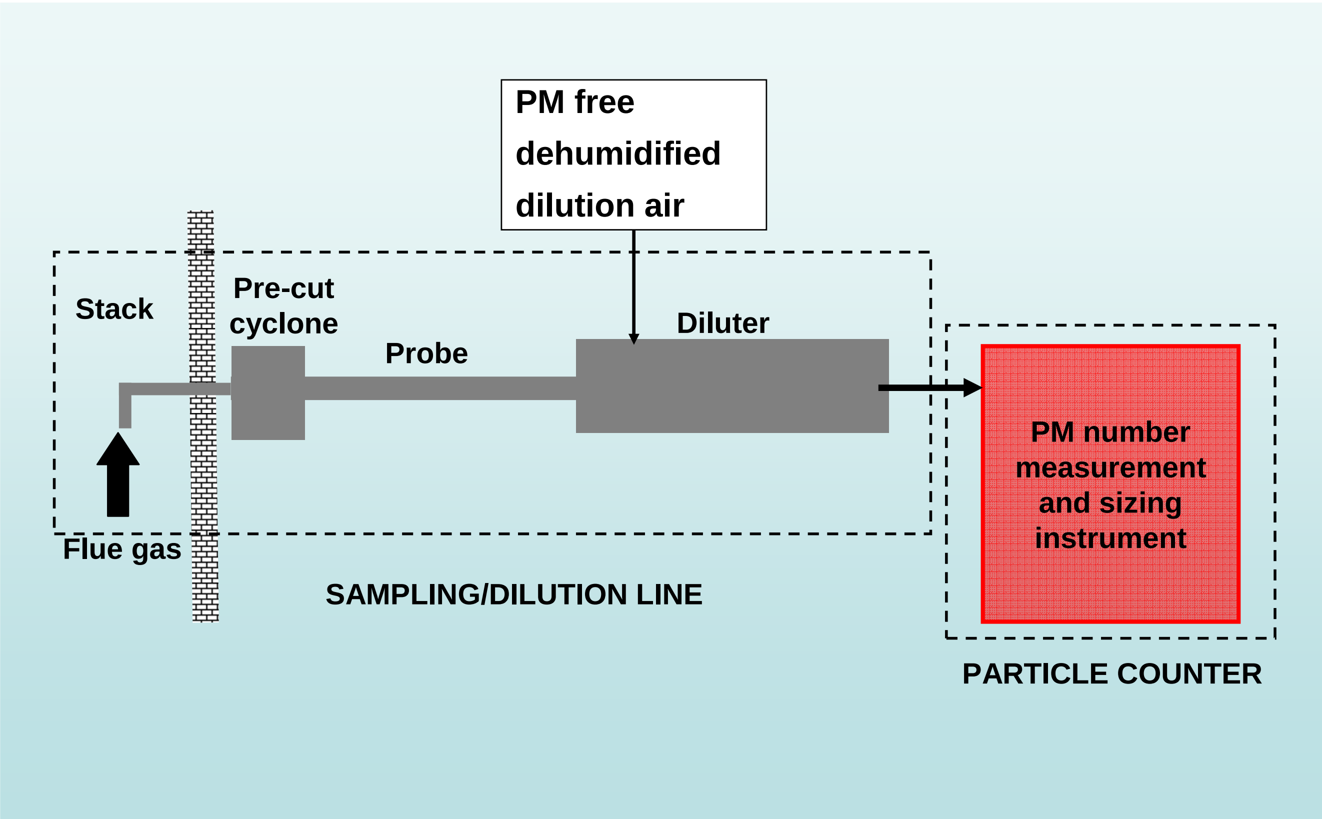
Boiler operation

- nominal operation at design conditions (full load, optimal air supply)
- non nominal operation (reduced load, reduced air supply)

Comparison gives the influence of combustion efficiency

MATERIALS AND METHODS

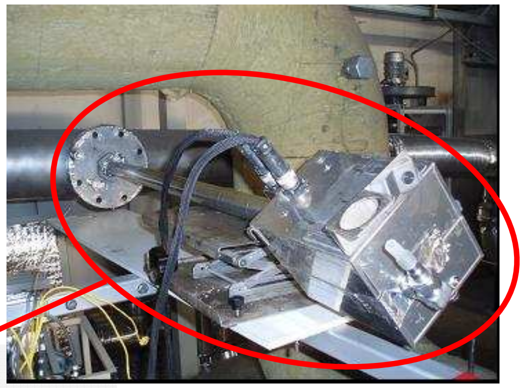
Sampling and measurement line - general scheme

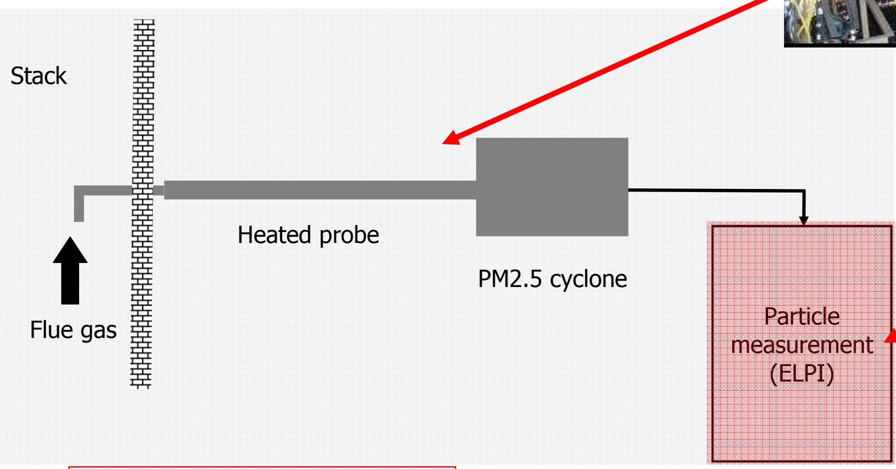


MATERIALS AND METHODS

Hot sampling at actual stack conditions

Hot sampling system with heated probe and precut cyclone



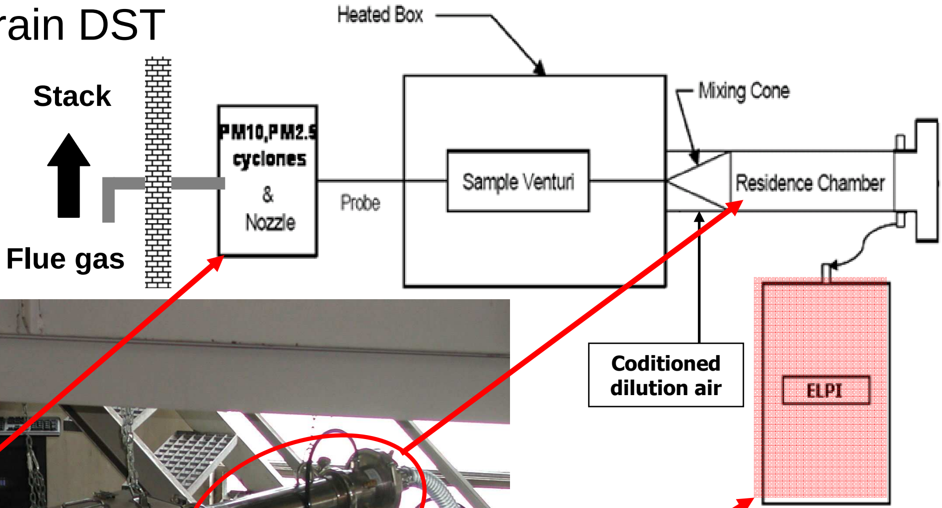


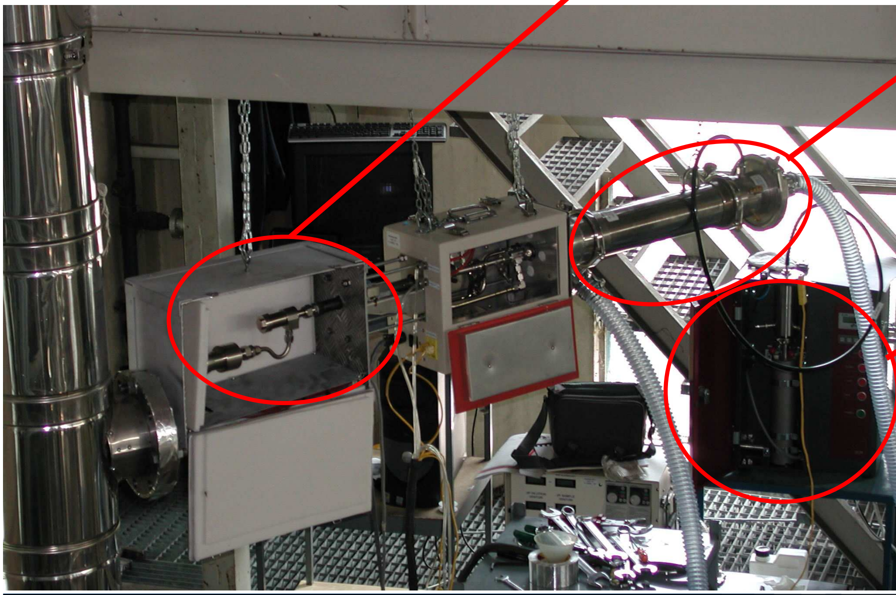
$T_{flue\ gas}$=150-180℃

MATERIALS AND METHODS

Dilution sampling train DST
(CTM039 – USEPA)

Dilution - cooling system with dry and clean air





Residence time
t~0.5-2 s

Diluted effluent temperature:
T~24-30℃

MATERIALS AND METHODS

Particle number concentration and size distribution measurements

Electrical low pressure impactor (ELPI, Dekati)

- Real time measurement
- Operation principles:
  1) Particle charging
  2) Inertial separation
  3) Electrical particle detection in the range 0.007-10 µm

Greased aluminum foils as impactor substrates.

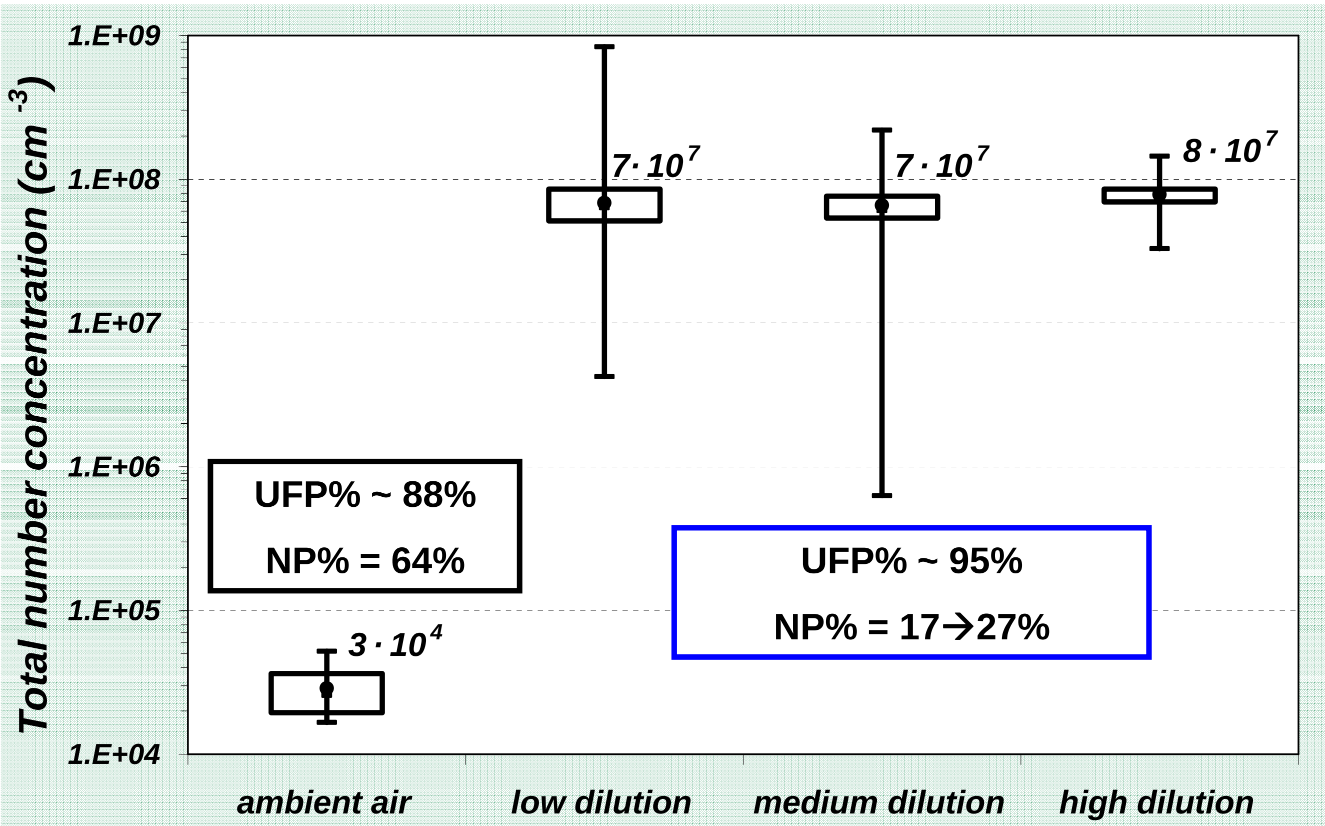
MATERIALS AND METHODS

Size resolved particle number concentration data

- statistics (mean, median, inter-quartile range, minimum and maximum values) of measured 1-sec of total number (TN) concentration data
- relative contribution (%) of UFP and NP to TN is provided
- concentrations considers dilution ratio, and refer to dry gas at 0℃, 1 atm and reference flue gas oxygen content (6 %v for solid fuels, 3 %v for liquid and gas)
- Observed particle size distributions

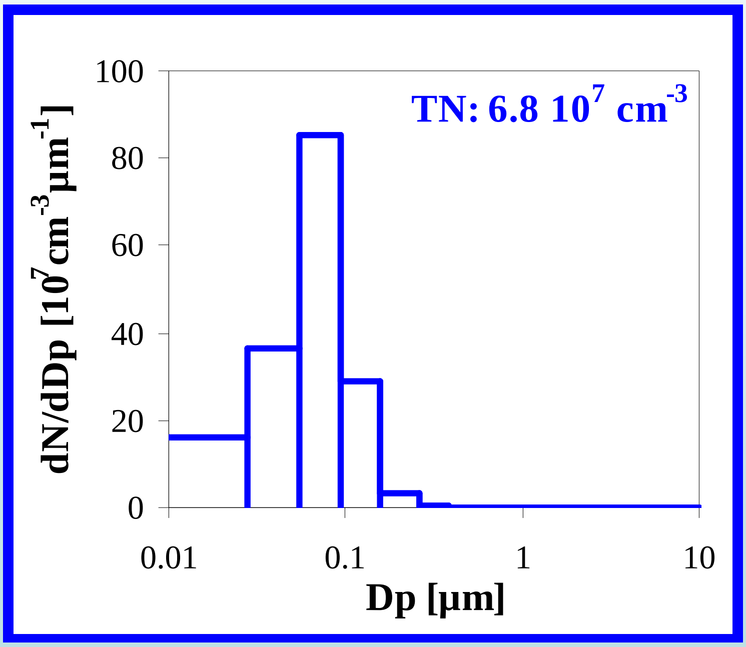
EXPERIMENTAL RESULTS

PELLETS BOILER – number concentration (cm^{-3})



EXPERIMENTAL RESULTS

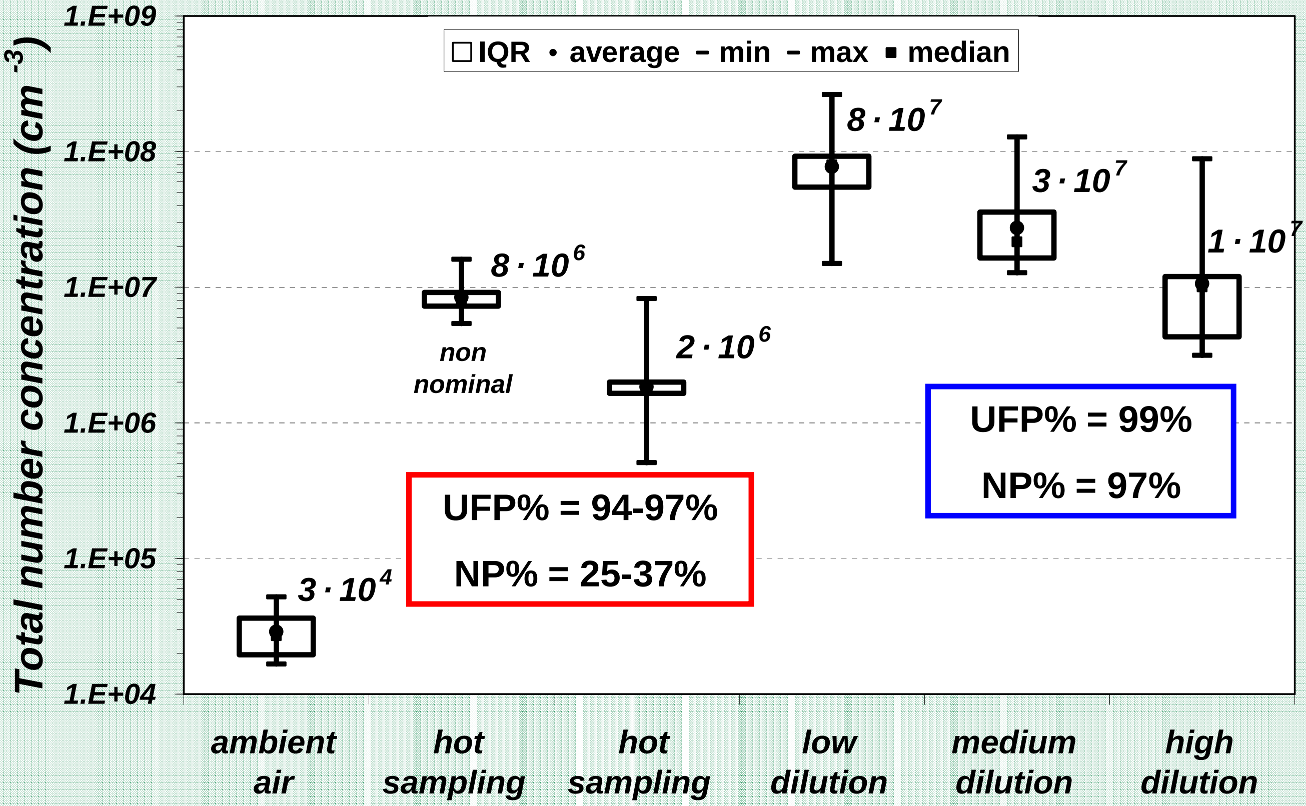
PELLETS BOILER – Size distribution



Low dilution

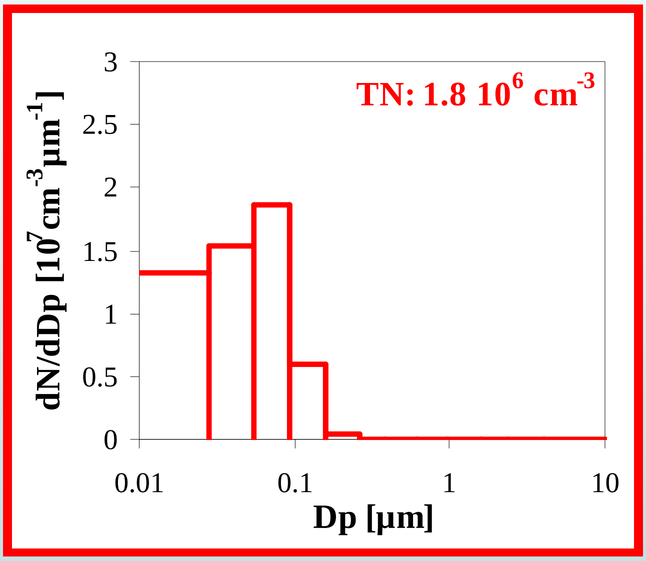
EXPERIMENTAL RESULTS

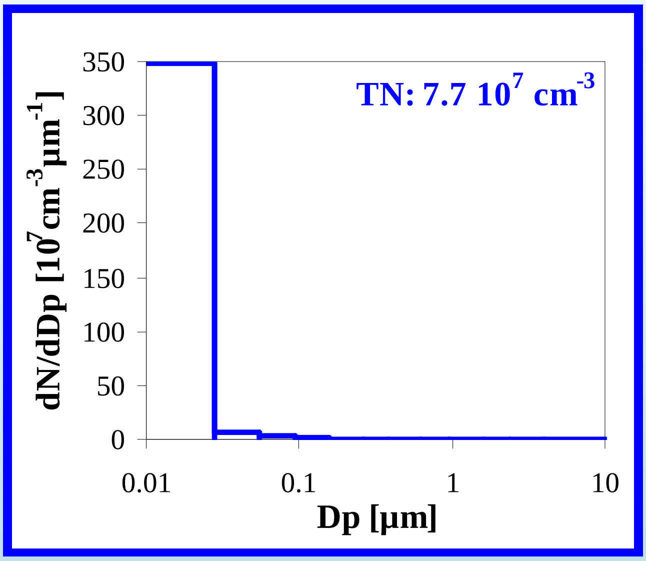
LIGHT OIL BOILER – number concentration (cm^{-3})



EXPERIMENTAL RESULTS

LIGHT OIL BOILER – Size distribution

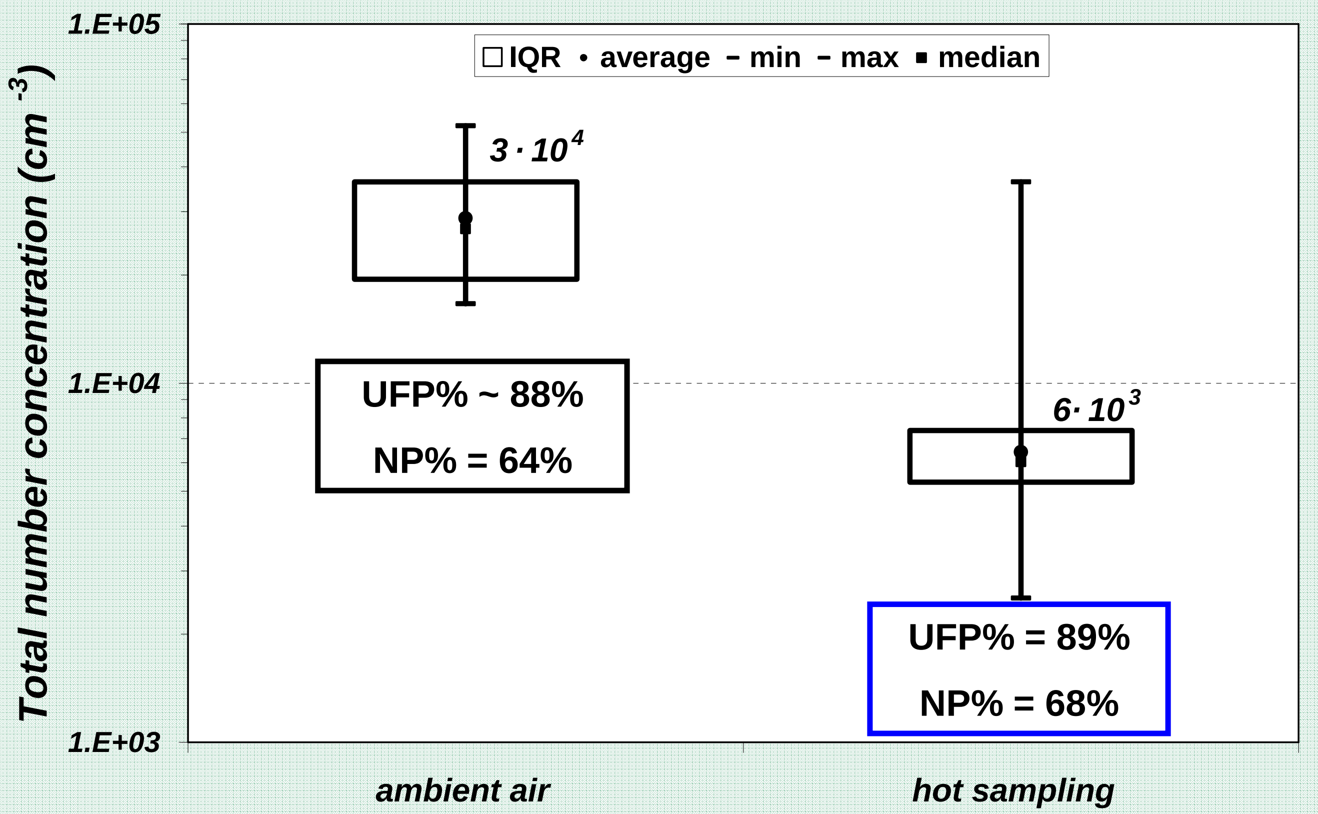




Hot sampling **Low dilution**

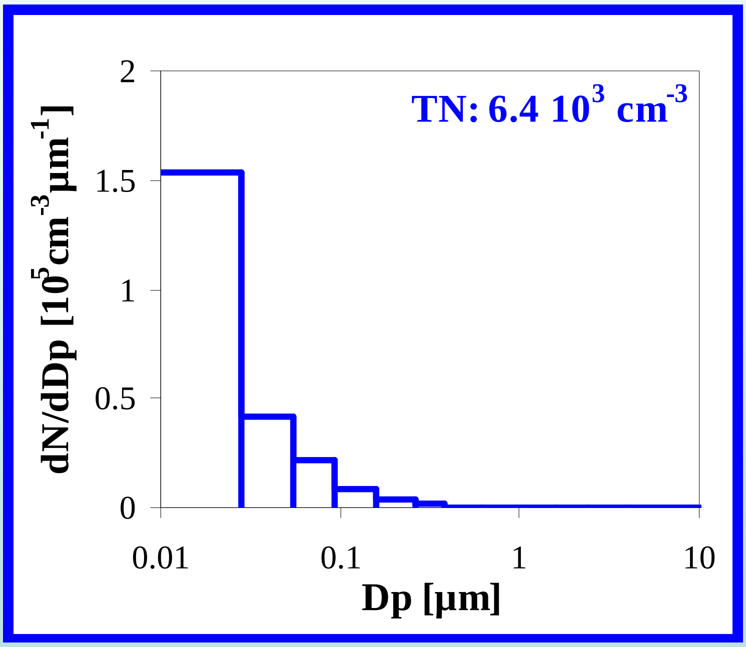
EXPERIMENTAL RESULTS

NATURAL GAS BOILER – number concentration (cm^{-3})



EXPERIMENTAL RESULTS

NATURAL GAS BOILER – Size distribution

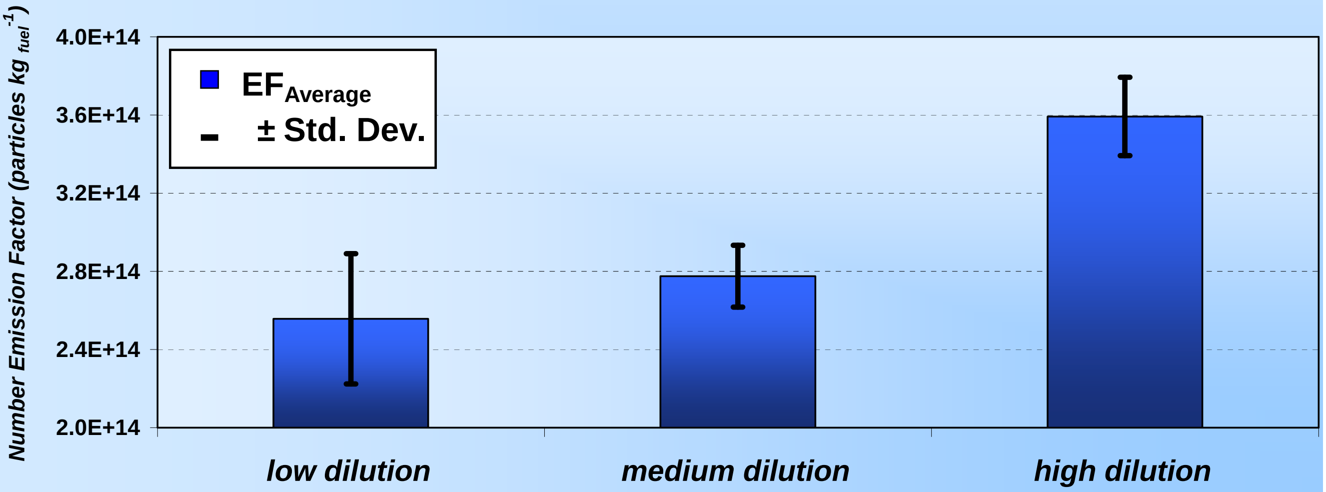


Low dilution

EMISSION FACTORS

PELLETS BOILER EMISSION FACTORS (@6%O_2)

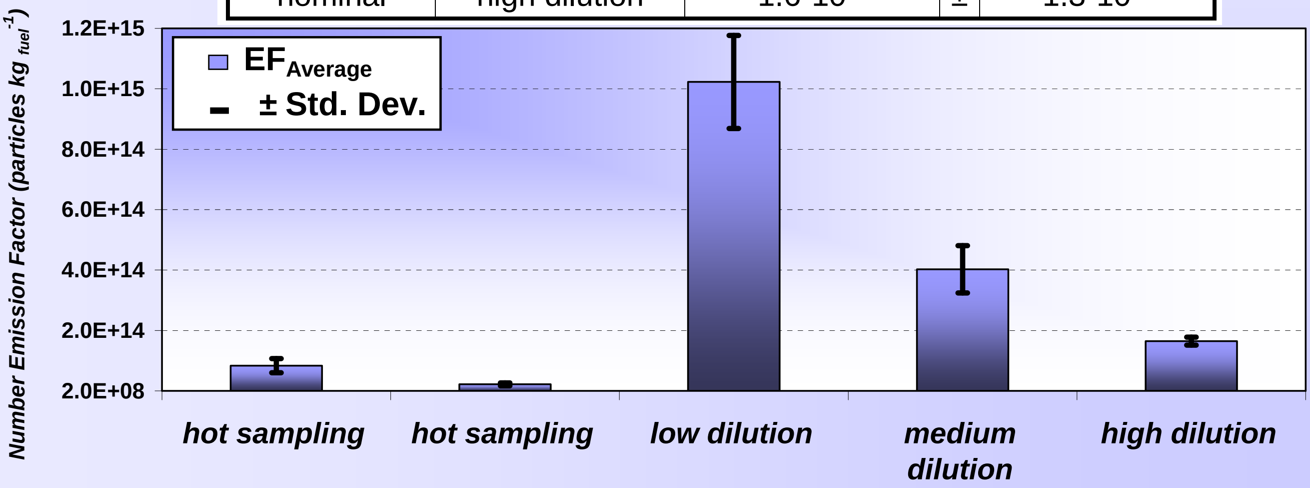
Boiler operation	Sampling conditions	Average emission factor (particles kg^{-1})		Standard deviation (particles kg^{-1})
nominal	low dilution	$2.6 \cdot 10^{14}$	±	$3.3 \cdot 10^{13}$
nominal	medium dilution	$2.8 \cdot 10^{14}$	±	$1.6 \cdot 10^{13}$
nominal	high dilution	$3.6 \cdot 10^{14}$	±	$2.0 \cdot 10^{13}$



EMISSION FACTORS

LIGHT OIL BOILER EMISSION FACTORS (@3%O_2)

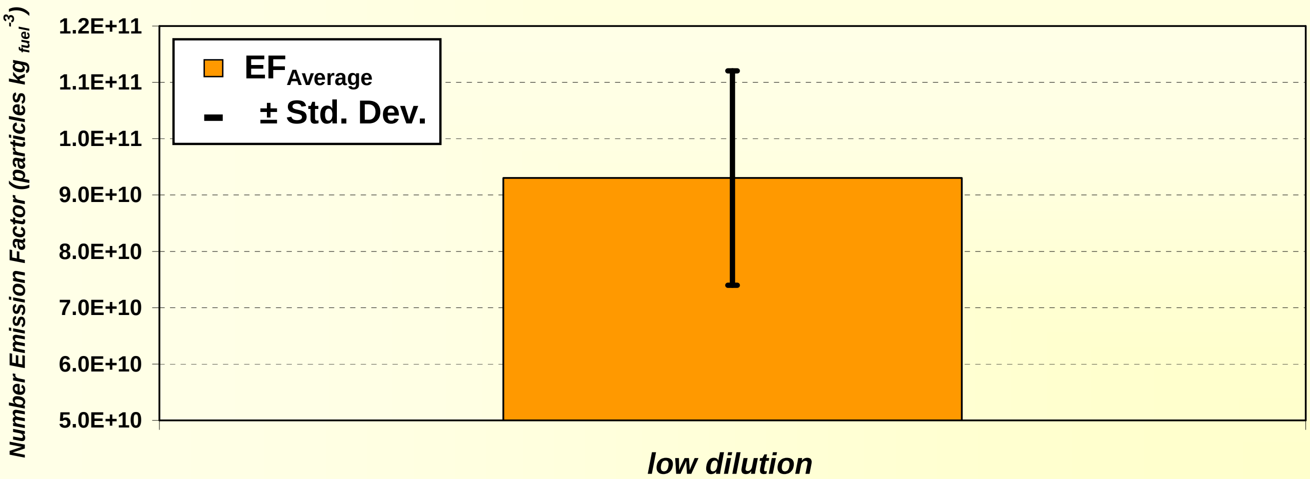
Boiler operation	Sampling conditions	Average emission factor (particles kg^{-1})		Standard deviation (particles kg^{-1})
non nominal	hot sampling	$8.3\cdot10^{13}$	±	$2.3\cdot10^{13}$
nominal	hot sampling	$2.2\cdot10^{13}$	±	$4.7\cdot10^{12}$
nominal	low dilution	$1.0\cdot10^{15}$	±	$1.5\cdot10^{14}$
nominal	medium dilution	$4.0\cdot10^{14}$	±	$7.8\cdot10^{13}$
nominal	high dilution	$1.6\cdot10^{14}$	±	$1.3\cdot10^{13}$



EMISSION FACTORS

NATURAL GAS BOILER EMISSION FACTORS (@3%O_2)

Boiler operation	Sampling conditions	Average emission factor (particles kg^{-3})		Standard deviation (particles m^{-3})
nominal	low dilution	$9.3\cdot10^{10}$	±	$1.9\cdot10^{10}$



CONCLUSIONS

- emissions in strict relationship with the fuel type and boiler operating conditions
- EFs range between $9 \cdot 10^{10}$ particles/kg_{fuel} for natural gas to $1.0 \cdot 10^{15}$ particles/kg_{fuel} for light oil boiler
- dilution and cooling enhances particle formation
- mode diameters positioned mainly < 50 nm
- UFP for more than 90% of TN
- small combustion installations, which usually are not equipped with flue gas cleaning devices, are not small emitters of ultrafine particles.

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