

# Clean Biomass Combustion in Residential Heating: Particulate Measurements, Sampling and Physicochemical and Toxicological Characterization (BIOMASS-PM)

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## Project Summary

### *Background*

Residential biomass combustion has a substantial contribution to the total fine particulate (PM<sub>2.5</sub>; particle diameter < 2.5 µm) emissions in most European countries. This is mainly due to a widespread use of old, un-optimised small-scale biomass combustion systems for woody biomass in residential heating. Because of extensive adverse health effects associated with current ambient air PM<sub>2.5</sub> concentrations, there is pressure in the EU and many European countries to start or strengthen the regulation of combustion emissions in this sector. It has been recently estimated by the Clean Air for Europe (CAFE) Programme of the European Commission that in the year 2000, mainly due to long-term health effects of PM<sub>2.5</sub>, there were nearly 350,000 premature deaths annually due to cardiovascular and respiratory diseases and cancer in the 450-million populations of the 25 EU member states. Moreover, tens of millions of elderly subjects with chronic lung and heart disease, asthmatic subjects of all ages and young children

were estimated to frequently have an increased need for rescue medication and restricted daily activity due to health impairment provoked by PM<sub>2.5</sub>. The total economical losses due to the health damage in 2000 were estimated at 268-781 billion Euros (European Commission, 2005).

The present multidisciplinary project consortium consisted of ten internationally well-established research teams from four countries. The purpose of the BIOMASS-PM project was to strengthen the interdisciplinary scientific evidence on the advantages of new combustion technologies and emission after-treatment in small-scale biomass heating systems. Due to intensive research and development work carried out during the last decade, small-scale biomass combustion units have already reached a high technological standard in terms of thermal efficiency and low emissions. However, concerning the reduction of particulate emissions, still further work has to be done.

Summing up the results and conclusions from the country reports (Austria, Finland, Germany, Sweden), it can be outlined, that in all four countries small-scale biomass combustion technologies play a relevant role by providing 10% - 30% of the total energy demand for residential heating and hot water supply. Moreover, small-scale biomass combustion technologies are the dominating renewable energy source for residential heating. It is also similar for all four countries, that logwood is the most common biomass fuel used in stoves and boilers. However, wood pellets have gained rising interest as fuel for residential heating systems since their market introduction, and this trend is expected to continue.

In all four countries, old biomass combustion technologies (stoves and boilers) dominate the current stock of applications. Since these systems show significantly higher particulate emissions than modern small-scale combustion systems, residential biomass heating has often a relatively high contribution to the total national thoracic particle (PM<sub>10</sub>; particle diameter <10 µm) and PM<sub>2.5</sub> emissions and is, therefore, facing increasing criticism. Comprehensive R&D work on the optimisation of small-scale biomass combustion devices has been performed during the last decade and, as a result of this, advanced residential biomass combustion technologies, operating at significantly lower particulate emissions than the old technologies, are already available. The lower particulate emissions have been achieved by improving the combustion conditions in ways that reduce the formation of organic aerosols and soot. However, the changeover from old to new residential biomass combustion systems has, so far, not taken place. Also the development of secondary measures for particulate emission reduction is in all four countries just in the initial phase. Therefore, the same measures for the reduction of particulate emissions from small-scale biomass combustion can be proposed for Austria, Finland, Germany and Sweden:

- Financial support for the substitution of old combustion devices by modern systems.
- Provision of appropriate “user training” for non-automatically fed systems.
- Support of the R&D of low-particulate-emission combustion technologies.
- Support of the R&D and application of appropriate secondary particulate emission reduction technologies for residential biomass combustion systems.

There were significant differences between the four countries with regard to the existence of particulate emission limits for small-scale biomass combustion appliances. In Finland, no emission limits exist so far, and in Sweden particulate emission limits are fulfilled on a voluntary basis. In Austria, there are particulate emission limits, and it is expected that they will become more stringent within the coming years. The most ambitious emission limits in Europe with regard to particulate matter are soon expected to get into force in Germany (2015 target: 20 mg/Nm<sup>3</sup>, dry flue gas, 13 vol% O<sub>2</sub>). A major problem is that different units and different O<sub>2</sub> reference levels are applied in different countries. Therefore, a harmonisation on a European level is recommended.

One of the main objectives of the project was a determination of feasible methods for particulate emission measurements, sampling and physicochemical characterisation. An appropriate best practise procedure was worked out. One of the most relevant aspects of this best practice procedure should be that it covers the evaluation of the whole process chain, starting from the combustion process (combustion quality, performance of the furnace) followed by particulate sampling, the physicochemical characterisation of particulate emissions, and the evaluation of emission-related health risks. A future aim should be to link the information gained from the physicochemical characterisation of particulate emissions with the results of toxicological studies in order to estimate the health risk potential of the emissions in relation to technologically and economically feasible small-scale biomass combustion systems. Consequently, the approach taken in the BIOMASS-PM project deviates from the commonly applied particulate sampling and measurement techniques, since in these procedures the interface with the toxicological evaluation of particulate emissions has not been taken into account.

#### *Measurement of particulate emissions*

The present best practise recommendations must apply to bad burnout conditions as they represent the most important challenge for future R&D of technologies as well as for health protection. As the first step of the procedure, due to presence of the condensable organic species at stove/boiler outlet, a dilution of the flue gas with clean air is recommended. Only clean air (filtered air) should be used for dilution. The dilution ratio has to be monitored continuously by parallel CO<sub>2</sub> or NO<sub>x</sub> measurements in the diluted and undiluted flue gas.

The dilution ratio should be high enough to keep the temperature of the diluted flue gas <52°C in order to ensure a full yield of condensable species in particulate phase. The dilution ratio should be in a range of 20 for the full potential of particle formation by condensation of organic vapours. However, higher dilution ratios can be applied, e.g. in connection to toxicological health studies.

A second important step of the best practise procedure is the determination of the gas phase composition (O<sub>2</sub>, CO, CO<sub>2</sub>, NO<sub>x</sub>, OGC) with conventional flue gas analysers. These gas phase measurements should be performed in the undiluted flue gas at boiler/stove outlet, since conventional flue gas analysers are usually not designed for measurements in diluted flue gas flows.

The total emission of particulate matter should also be determined in undiluted gas due to two reasons. First, this procedure enables a comparison of the results from small-scale biomass combustion installations with those obtained from boiler tests that are usually performed without dilution. Second, losses of coarse fly ash particles occur during dilution, which means that a measurement in diluted flue gas would lead to an underestimation of the total particulate matter emission. The measurements should always be performed according to the respective test standards.

A stepwise chemical analysis procedure is recommended, so that the level of characterisation can be adjusted to different purposes. A basic fractionation into soot, organic and inorganic matter is currently highly relevant. There can be also fractionation of the carbonaceous matter into elemental (EC) and organic (OC) carbon. Further fractionation and speciation of the organic and inorganic matter can be performed, especially in scientific studies. The concentrations of known genotoxic PAHs in particulate phase emissions are of great interest.

Concerning all analysis methods mentioned above, it is important to adjust the pre-treatment, conditioning, handling and storage of the sampling substrates to the demands of the respective analysis method. As the dilution ratio applied during sampling may influence the chemical composition of particulate emissions, it should always be reported in connection to the results of chemical characterisation.

For automated furnaces in continuous operation, it is recommended to perform the measurements and particulate sampling at stable full-load and minimum load operations. For automated furnaces in on/off operation (especially relevant for Sweden), the tests should be done according to the Swedish P-marking regulations. Concerning batch combustion systems, also the first full batch or kindling wood ignition should be included in the test. Sampling during this first batch should start as soon as the CO<sub>2</sub> concentration exceeds 1 vol%. For all subsequent batches sampling should start at the beginning of the batch and should end as soon as the CO<sub>2</sub> concentrations decrease below 4 vol%.

When applying the present best practise procedure, the chemical and physical characterisation of particulate emissions from biomass combustion can be adjusted to the level of information needs for different purposes.

#### *Investigation of health effects of particulate emissions*

Aerosol exposure systems with on-line diluted flue gas from combustion installation are used in experimental human and animal studies. Also re-aerosolization of combustion emission particles collected in advance with, e.g. electrostatic precipitator, has been used in animal studies. Alternatively, one may collect size-segregated emission particles from diluted exhaust and instill them in aqueous medium directly to cell culture or under visual control to the lower airways of experimental animals e.g. mice or rats.

Common requirements for both the aerosol exposure and the particulate collection method are:

- Evaluate the characteristics of the exposure system in advance using standard aerosol monitoring and sampling methods as reference.
- Avoid carefully any external contamination of the combustion emission particles and use methodologically similar sham exposure to filtered air or blank samples that control all stages of particulate sample handling.
- Report always the combustion technology, fuel and condition as well as the dilution ratio, temperature and the results from gaseous and particulate monitoring and chemical speciation as background information in the toxicological study report.
- The most important size range to be investigated in toxicological studies is PM<sub>1</sub>. However, investigation of the coarse (> 1 µm) particles, mostly fly ash, or separation of the ultrafine particles (< 0.1/0.2 µm) from other submicron fine particles may be motivated in connection to some technologies or biofuels, or special health issues of interest.
- The non-toxicity of particulate sampling substrate needs to be confirmed in cell tests made in advance. The substrates should be pre-cleaned before sampling using the same solvent and cleaning protocol as in the actual particulate extraction. Blank substrates are processed similarly to the ones used in particulate sampling.
- Particulate collection needs to be done with dilution and cooling the flue gas <52 °C to include condensable material in particulate phase emission.
- After particulate sampling, collection substrates must be protected from sunlight and removed as quickly as possible for storage at -20 °C. For subsequent weighing and extraction, the frozen substrates and particulate material need be conditioned at room temperature for 4 hours in closed containers, followed by 16–18 hours in open containers before weighing.

Toxicological cell and animal studies need at least tens of milligrams of biomass combustion particles per size range and test condition, so that the same particulate material can be used for testing several different end-points as well as making chemical analyses of interest (e.g. ions, total and water-soluble elements, PAHs). Tens to hundreds of milligrams of size-segregated particulate mass have been recently collected from wood combustion experiments using a high-volume cascade impactor (HVCI). It collects particles at high volume (850 litres/min) in four size ranges (PM<sub>0.2</sub>, PM<sub>1-0.2</sub>, PM<sub>2.5-1</sub>, PM<sub>10-2.5</sub>), and the collected particulate mass can also be extracted from the sampling substrate, i.e. porous polyurethane foam, with a high 80-90% efficiency.

One of the main motivations for toxicological studies is to provide scientific evidence on the biological plausibility of adverse health effects reported in epidemiological studies. Experimental human and animal studies can be conducted on a limited number of research questions on biomass combustion aerosol due to ethical reasons and elaborate study setups, but toxicological cell studies can be utilized more flexibly to reveal the toxicity profiles of multiple combustion conditions. It can be stated that the traditional test of mutagenic activity in bacterial cell cultures (Ames' test) should be mostly replaced by modern test batteries in cultured mammalian cells like macrophages and respiratory epithelial cells. In all toxicological setups, the dose-dependency and

time-dependency of the different kind of response endpoints to biomass combustion particulate samples need to be known to avoid false negative findings.

The inflammatory activity of air particles has been linked for a long time to exacerbation of pulmonary diseases like asthma and chronic bronchitis, but more recently it has been suggested to contribute to atherosclerosis and acute vascular events such as cardiac infarction and stroke. Tissue damage induced in the lungs and other organs by combustion-derived particles can be also mediated, at least partly, by direct cytotoxicity, i.e. via increased apoptotic, i.e. programmed, cell death or un-programmed necrotic cell death, or via inhibition of DNA synthesis in the cells.

Genotoxicity is primarily regarded as the mechanism leading to increased carcinogenic risk. In connection to research on biomass combustion-derived particles, Comet assay can be used as a relatively easy and quick screening test of DNA damage. Its information can be complemented, e.g. by the more elaborate micronucleus test and PAH-DNA adduct test.

#### *Future directions*

It is obvious that the field of residential biomass combustion would much benefit from a European-wide harmonization of the emission test methods and procedures.

Interdisciplinary research involving both the combustion and aerosol scientists as well as toxicologists on several topics is needed:

- More detailed comparison of different sampling and dilution methods is needed to define a standard method for the measurement of fine particle emission from residential biomass combustion installations.
- Further technological R&D is needed to improve both the primary measures (combustion and control systems) and the secondary measures (emission after-treatment) to reduce particulate emissions from small-scale biomass combustion systems.
- More information on the impact of real-life user practices on particulate emissions is needed as well as on the overall impact of small-scale biomass combustion emissions on local and regional air quality.
- More information is needed about the association between different kinds of particulate matter emissions from biomass combustion installations and their adverse health effect potential as assessed by experimental human and animal studies. Cardiovascular endpoints should be investigated in addition to respiratory endpoints. This would increase information about the biological plausibility of adverse health effects reported in epidemiological studies.
- Cell studies should provide a generic concept on the association of inorganic and organic chemical constituents with the inflammatory, cytotoxic and genotoxic activities of particulate emissions from a series of combustion technologies and biofuels. The representativeness of the results should be confirmed in selected experimental animal and human studies. This information would help the regulator to give emission limits to some potentially highly toxic constituents (e.g. PAHs), the

industry to direct its product development towards health-wise cleaner and safer combustion installations, and the consumer to adapt for good operational practice.

It would be advantageous also to promote interdisciplinary research between the aerosol scientists and epidemiologists:

- New short-term panel studies with personal exposure monitoring and source-specific exposure modelling are needed. Contributions of biomass combustion source-specific outdoor  $PM_{2.5}$  to indoor  $PM_{2.5}$  should be assessed as well as lung dosimetry in relation to the physico-chemical properties of the particles (e.g. fresh emission nearby vs. aged regional emissions).
- GIS-based cohort studies on chronic respiratory and cardiovascular diseases and cancer are needed. Data on household heating appliances and relevant particulate emission factors need to be up-to-date, because they are in constant change due to altered appliance types and improvement of technologies.

The topics suggested for future research collaboration between the present research partners of the BIOMASS-PM project should be considered for funding in the 7th Framework Programme of the European Commission or ERANET-type collaboration of national funders.