SUSTAINABLE ENERGY SYSTEMS
ENERGY AND CIRCULAR ECONOMY

# Emissions of particles and organic compounds from small and medium scaled biomass combustion

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# Abstract

The use of biomass for domestic heating is common in some parts of the world. Biomass is a Renewable Energy Source (RES) and it is considered as a climate friendly fuel since it is a $CO_2$ neutral energy source. However, combustion of wood biomass in the residential sector is a main contributor to pollution of the ambient air, mainly in terms of fine particles. This is a severe health problem and needs to be addressed to improve the air quality. There is also a gap between calculated air quality particle concentrations using the available emission data, measured at the stack, that needs to be addressed. The concentrations of organic particles in the atmosphere are higher than expected from reported emission factors, but there is also a gap between emission factors registered in different countries, emphasising the need of similar standards for individual countries, or at least more information around the emission data.

The residential combustion of wood biomasses is characterized by incomplete processes leading to high concentrations of gases and particles containing both organic and inorganic substances. The formation of organic particles is a temperature dependent process where Semi Volatile Organic Compounds (SVOC) condense on solid particles or form particles after nucleation. Thus, the temperature where particle emissions are measured is crucial to the result. There are many different standard methods for the measurement of combustion generated particle emission, some differ between different combustion sources. Most standards focus on the most straight forward measurement methods, collecting particles directly from the hot fumes, while in other standards the particles collection is after a dilution of the exhaust gases, taking into account the condensation of organic gases.

In particularly for residential biomass heating appliances there is no European harmonized standard for measuring particles and a few Member States provided with own method (to be performed during laboratory type testing). The methods are different each other: one method is adapted from the standard used to measure particles produced by big plant fed with fossil fuels (just collecting solid particles at high temperatures), the other method is based on the dilution of sampled gases to collect solid and condensed particles.

The aim of the present work is to provide background information both on different emission standards used in combustion appliances and on the development of the particles from the combustion zone to the ambient air at a larger distance from the source. The development of the particles change the amount of particles originating form biomass combustion by both number and mass. Input data for this study is taken from the literature and from a survey sent to laboratories engaged in emission measurement from small to medium scaled biomass combustion facilities.

Key words: Biomass combustion, emissions, particle, standards

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# Preface

This report presents the results from Activity A2.1.4 in Task 2.1 of *WP2: Biomass Combustion Emission Measurement* in the project *IMPRESS 2: Metrology for Air Pollutant Emissions*. The project is funded by EMPIR (European Metrology Programme for Innovation and Research) which is an initiative co-funded by the European Union's Horizon 2020 research and innovation programme and the EMPIR Participating States.

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# Abbreviations and nomenclature:

| | |
|---|---|
| SVOC | Semi Volatile Organic Compounds |
| VOC | Volatile Organic Compounds |
| NMVOC | Non-Methane Volatile Organic Compunds |
| OGC | Organic Gaseous Compounds |
| PAH | Poly Aromatic Hydrocarbons |
| CCN | Cloud Condensation Nuclei |
| BC | Black Carbon |
| PM | Particulate matter (no size segregation) |
| PM10 | Particles with a diameter less than 10 µm |
| PM2.5 | Particles with a diameter less than 2.5 µm |
| PM1 | Particles with a diameter less than 1 µm |
| FID | Flame Ionisation Detector |
| FTIR | Fourier-Transform Infra-Red |
| GC | Gas Chromatograph |
| TEOM | Tapered Element Oscillating Microbalance |
| ELPI | Electric Low-Pressure Impactor |
| SMPS | Scanning Mobility Particle Sizer |
| TDLAS | Tunable Diode Laser Absorption Spectroscopy |
| ESP | Electrostatic Precipitator |
| EEA/EMEP | European Energy Agency/Europeian Monitoring and Evaluation Programme |
| SPC | Solid Particle Counter |
| Solid particles | Often called primary particles in the literature. In this study particles present at high temperatures, i.e. above ca +200°C, are called solid particles. These particles does not include condensable matter |
| Condensable particles | Particles formed by gaseous compounds cooled from stack-temperature to ambient temperature. No chemical effects, atmospheric aging, is included in these particles. Sometimes called secondary particles (aerosols) in the literature. |
| Ambient particles | Often called secondary particles in the literature. In this study particles containing condensable matter which have been exposed to atmospheric aging are called ambient particles. |

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# 1 Background

Emissions from residential wood combustion has been in focus from an air quality perspective since many years, recently intensified by increased use of biomass combustion as a measure against climate effects. The reason to increase use of biomass is that biomass is reformed by nature at an immensely larger speed than fossil fuels. Still, combustion emissions have negative effects on the atmospheric air quality, while air pollution is the single largest health risk related to environmental issues. Air pollution alone is estimated to cause around 3 million annual premature deaths globally and half a million in Europe alone (EEA, 2017, WHO, 2016). Biomass combustion emitted particles also have a large effect on cloud formation, and other smoke related areas, including climate effects (Kreidenweis *et al.* 2008).

This study focuses on particles originating from wood biomass combustion in small to medium scale facilities and formed in the combustion process or by effluents from combustion after atmospheric processing. 44% of the total $PM_{2.5}$ emissions in the EU originate from residential wood combustion alone (EEA, 2013); and the biomass combustion contribution to PM concentration in the air quality is larger during winter than summer (Viana *et al.*, 2016). During winter residential combustion is common to solve the heating problems, while barbecue is more common during summer.

Condensable matter in the flue gas is added to the solid particles when the flue gas is cooled and diluted when entering the ambient air. Therefore, it is difficult to evaluate the impact of biomass heat generators, just taking solid particles emissions into account, with no information about organic gases content.

The emission factors depend on where the process the sampling point is located and what part of the smoke that is analyzed, which differ between in the measuring standards for each provider of data (Vicente and Alves, 2018). Thus, the available data on biomass emission vary between countries depending on standard used, on top of on real emission differences (Denier van der Gon *et al.*, 2015). This makes the understanding the full system of biomass combustion effects on air quality unnecessarily challenging and the need of updated standards for particle sampling of emissions from biomass combustion has therefore been pointed out in several studies (Vincent and Alves, 2018, Denier van den Gon *et al.*, 2015, Bruns *et al.*, 2016, Bäfver, 2008).

The aim of the present study is to provide background information needed for the development of a new standard method for particle emission measurement from domestic biomass combustion with a special emphasis on the condensable particulate matter. A literature study is performed where present standard methods, typical emission data and how ambient particle concentrations are related to emission data is reviewed. A survey is sent to notified bodies around Europe with the aim of collecting information regarding which standard methods that are being used and also to collect comments from the users of the standards. The data obtained in the literature study and survey is given in the result section and the report ends with a discussion and recommendations for emission measurements.

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# 2 Method

This study is a desktop study, including a literature review to view the development of the smoke and how the parameters are measured and regulated, and using a survey to gather information regarding which international standard methods that are being used by laboratories and notified bodies in Europe.

## 2.1 The literature study

The study is based on the existing standard methods of emission measurements for small and medium sized biomass combustion plants and complemented with a selection of related scientific literature. The existing standards are chosen *ad hoc* with a focus on what is used in Europe, with US standards as a complement.

The literature study is not a full review of the literature, but merely a way to describe the area of interest. The study is focusing on measurement methods of all species that in some way or another might end up in the particle phase in ambient air. Thus, both semivolatile species in the gas phase and gas phase species that after aging might form particle mass are discussed. A key issue of the study was the possibility to simplify the techniques to form a useable standard method for emission measurements useable to understand the formation of particles in ambient air and to characterize the plant in terms of its contribution to PM in ambient air.

The literature study resulted in a description of "The emission process" including measurement techniques, abatement strategies and regulations. The standards (both active and future) are described in the "Standards for PM and SVOC measurements".

## 2.2 The survey

A total of 15 standard measurement methods were included in the survey, with the possibility to add three additional standards based on your own choice. The standard methods included in the survey are listed below. The standards that were included in the survey was selected based on the experience from the project partners (RISE, DTI, INERIS, INNOVHUB and ENEA). All partners are mainly working with combustion related emissions, so the standards are intentionally biased towards this field. Other standard methods, *e.g.* for OGC (Organic Gaseous Compounds) emissions in other sectors, are available but not included in the survey. When similar standards have been found, not all were included in the survey. An example of this is ISO 12141 and ISO 9096 where the main difference is the weighing procedure of the filter blanks.

The survey was sent to 65 persons from 12 different countries in Europe with a higher representation from the project partners countries. The following standard methods were included in the survey:

1. ISO 12141
2. ISO 25597
3. ISO 13271
4. ISO/EN/SS 23210
5. ISO/EN/SS 11338-1

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6. EN 11338-2
7. CEN/TS 13649
8. EPA Method 5G
9. EPA Method 5H
10. EPA Method 17
11. EPA Method 28/28R
12. EPA Method 201A
13. EPA Method 202
14. ASTM E2515
15. ASTM E2780-10

An online tool (www.surveymonkey.com) was used for the survey where both multiple choice and free-text questions were asked. For each method a number of questions were asked starting with the awareness of the method, and to what extend the respondents have used it and in which application. They were also asked if they recommended the method and to add comments. All questions in the survey can be found in Appendix 1. The analysis of the responses to the survey was done both by looking at the individual answers and by calculating the statistics from the Yes/No answers. A few of the responses were not included in the evaluation due to the limited extent of answers, *i.e.* typically by only answering the questions on the first method. All comments are read and are, together with the recommendations, included in the evaluation of the methods.

# 3 The emission process

Particles that originate from biomass combustion are formed by several different processes. In the case of residential biomass combustion, the particle emissions are dominated by fine particles (< 1 µm). The solid particles emitted consists, depending on the combustion conditions, mainly of soot or alkali species (Bäfver, 2008). After the flue gas is emitted to the atmosphere through the stack, it is cooled to ambient temperature and diluted which leads to condensable matter adding to the particle mass by condensation of gaseous compounds. The condensed matter ends up both at surfaces within the stack, at the surface of other particles and as nucleated new particles if there is not enough surface area to deposit on. In the atmosphere particles can evaporate due to high dilution ratio but also increase in size and number by atmospheric processing of the flue gas. The amount of particles formed is affected by, *e.g.* the humidity of the air, the amount of light (*e.g.* long dark winters) and other constituents of the air (*e.g.* if passing clean or polluted areas, kind of pollution) (Hallquist *et al*, 2009, Janhäll *et al.*, 2012).

Measurements of particle emission factors for biomass combustion is complicated by the fact that the amount of particle mass change rapidly depending on where the particles are measured. When the flue gas has reached ambient temperatures outside of the chimney the amount of particle mass is often doubled or even ten-folded as compared to in the hot flue gas (Bäfver, 2008). After some time of aging in the plume, chemical and physical reactions change the amount of particle mass. The way to measure and what to include in the data depends on what you want to know about the emission source. Often the aim is to understand the source of the particles affecting the human health, to give

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the opportunity to abate the emissions giving the particles in air that humans breath. To highlight this discussion the process of emission, from the furnace to the ambient air is described in this section, including both measurement techniques, data treatment, abatement strategies and regulations of emissions.

## 3.1 The combustion

Solid fuel can be characterized by dividing its content into moisture, volatile matter, fixed carbon (char) and ash. During combustion of a solid fuel the fuel undergoes a series of processes initiated by drying of the fuel removing the water. The temperature then increases, and the volatile substances are released as gases. The remaining part of the fuel consists mainly of carbon and is finally combusted in the char combustion phase (Nussbaumer, 2003). Depending on the fuel type and composition of the ash, it may be released as gases or be left as solids together with the char. The ash content is in general very low, less than 2%, in conventional biomass fuels used in domestic appliances (Nussbaumer, 2003). The interest for alternative biomass fuels (*e.g.* energy crops) with a higher ash content are increasing and are often used in pelletized form. However, these fuels are mainly used in larger, non-residential, boilers where the pellets are milled and combusted in suspension fired systems. (Paulrud, 2004). Still, for conventional biomass fuels some alkali containing substances are often released into gas phase forming alkali salts, often in the form of potassium chloride (KCl), or potassium sulphate ($K_2SO_4$) (Nussbaumer, 2003, Boman *et al.*, 2004, Bäfver, 2008, Johansson *et al.*, 2003). High temperatures normally enhance the conversion of ash material to the gas phase and consequently the emission of inorganic particles (Nussbaumer *et al.*, 2008). It is mainly the hydrocarbons in the volatile matter of the fuel that are of interest in this work since it forms soot or tar-components. In oxygen lean zones of the combustion chamber hydrocarbons will form soot, through formation of aromatic rings (PAH) and later solid particles. PAHs with higher number of rings are normally present in the smallest size fraction, while larger particles consist of ash/soot with a condensed part of PAHs with fewer rings as those are more volatile in the first part of the cooling of the flue gas (Shen *et al.*, 2013). The size of these soot particles will also grow due to absorption by gaseous species on the surface and by agglomeration with other soot particles. PAHs have been shown to increase during smoldering, but in other studies also at high combustion temperature (Lim, 2015).

Domestic combustion of biomass typically consists of batches of wood logs being combusted in a non-continuous manner, but also of pelletized fuels which can be continuously fed to the combustion chamber. The lack of control systems in many appliances results in large variations during combustion of the wood logs. In many cases the start-up phase is responsible for more than 50% of the total PM emissions of a whole batch (Nussbaumer *et al.*, 2008). Particles from wood stoves operated under poor conditions consist mainly of soot and organic substances (Nussbaumer *et al.*, 2008) and the size of particles become smaller when the combustion conditions are improving (Janhäll *et al.,* 2010, Nussbaumer *et al.*, 2008).

## 3.2 Abatement of emissions

The emissions from large and medium scaled facilities depend mainly on particle removal equipment (Nussbaumer *et al.*, 2008), while mitigation technologies are not

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really available for domestic scale appliances (Lim *et al.,* 2015). Experiments testing additives to prevent particle formation and catalysts of different kinds have been performed without obvious success (Bäfver *et al.*, 2009, Andersson *et al.*, 2014, Kaivosoja *et al.*, 2012). The efficiency of the catalyst can be increased if adding an electrical heater close to the catalyst, and more studies in this direction are preformed (Lim *et al.*, 2015). Filter techniques is mainly not a solution as the filters will clog and domestic heater owners will not find it interesting to maintain and shift filters at a high enough rate (Lim *et al.*, 2015). The most promising solution, still in the need of further studies, is the small scale electrostatic precipitator (ESP) (Lim *et al.*, 2015), even though the high voltage needed can be a problem in domestic dwellings. There is also a possibility to use more of the abatement techniques used in vehicular exhaust emission abatement (Lim *et al.*, 2015). The availability of oxygen in the fuel bed has been identified as a promising primary measure for decreased emission of ash particles from biomass combustion (Nussbaumer, 2003). However, the decrease of primary air through the fuel bed may lead to increased emissions of carbon containing particles instead.

Still, at this stage the most efficient way to abate biomass combustion emissions from small and medium sized appliances is to focus on avoiding inappropriate operation of manual wood combustion appliances as this is the largest uncertainty and has the largest effect of all studied variables (Nussbaumer *et al.*, 2008, Fachinger *et al.*, 2017) and to improve the appliances *e.g.* by improved control of air inlets to the combustion area and by increased energy efficiency through using heat storage tanks (Nussbaumer *et al.*, 2008).

## 3.3 Development of the aerosol

Particles formed during combustion evolve and change, both in size and content from the furnace to deposition, Fig. 1. Thus, the techniques and locations used to measure the emissions will be important for the registered emissions. The registered emissions are also used in different ways and different aspects of the particles or relations needs to be registered together with the data. In this section the development of the particles is divided into three main processes, combustion, condensation and chemical transformation. Aspects of measurement techniques are discussed in all three parts.

**Figure 1**. Particle development from biomass combustion, *i.e.* solid particles in the hot flue gas, condensed material added (green) and after chemical development in the ambient are, secondary particle material formed (pink).

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### 3.3.1 Solid particles in chimney

The solid particles emitted through the chimney consist of soot or ash (alkali) particles. The vast majority of available emission data is collected as solid particles in the chimney at rather high temperatures, often a filter heated to 150-180°C (Lim et al, 2015). The flue gas temperature in the sample location may be both over and under this temperature depending on the application. The reason for heating the filter is to avoid condensing material both on the sample and on other surfaces complicating the sampling procedure. The standard methods included in this study often recommend a sampling temperature that is the same as the flue gas temperature in the sampling point or required to be above a certain temperature. Each chosen temperature includes or excludes parts of the organics that has their condensing temperature in range of sampling, and the amount of matter sampled depend heavily on temperature and on temperature stability at the sampling point.

The solid particles make up only a small fraction of the particle mass that is found in the atmosphere and related to biomass combustion (Denier van der Gon et al, 2017).

### 3.3.2 Condensed or evaporated material

During cooling of the flue gas some parts of the gaseous compounds will condensate. This cooling either takes place after emission from the chimney or within the flue gas system. Thus, the condensation of gases in the flue gas take place either with dilution or without dilution. The level and specific features of the dilution/cooling process affect the particle formation heavily, when the condensed matter form new particles through nucleation or deposit on existing particles or surfaces within the flue gas system.

The sampling taking also the condensed matter into account must, in this case, be performed at a lower temperature than for solid particles and the temperature needs to be appropriately chosen and kept stable. The standard methods studied either use a dilution tunnel, where the flue gas is mixed with clean air with lower temperature cooling the sample or using impingers to collect the condensate at distinct temperatures.

Dilution tunnels try to mimic the emission from the chimney to the ambient air. The amount of dilution air, the temperature of the dilution air and the speed of mixing all have effects on the resulting aerosol, as low dilution might overestimate the particle mass and high dilution might impose larger evaporation of the condensed material and underestimate the particle mass (Vincente and Alves, 2018). Thus, the dilution tunnel has to be thoroughly described according to dilution, including times and temperatures, in order to minimize uncertainty in the sampling (Vicente and Alves, 2018).

Most emission data are given as particle mass (Lim *et al.*, 2015), but also particle number emissions are reported. The particle number is to an even higher extent than the particle mass affected by dilution and by cooling rate, among other things nucleation can occur in the instruments that would increase the number of particles manifold. The main reason for achieving emission factors of particle number is the effect on clouds through cloud condensation nuclei (CCN), but also other processes are particle number dependent (Denier van der Gon *et al.*, 2015, Lim *et al.,* 2015). Lipsky and Robertson (2005) showed an example of large differences between two dilution tunnel samplers of different size and portability (portable dilution tunnel without residence tank and a

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conventional sampler based on Caltech design for $PM_{2.5}$ total mass). The effect was attributed to the difference in mixing rates between the systems as nucleation occurred in one of the samplers.

Another approach to measure or at least approximate the emissions of condensed particles from biomass combustion appliances is to analyze organics in the gaseous phase. This implies that the gaseous compounds must be differentiated between the ones condensing in ambient air from the ones still in the gas phase. If species are measured one by one this might be rather time consuming as many species are present in biomass flue gas. In other cases, the total amount of organics, measured in different way giving different description of the organics, is measured. This concept is used by the suggested method from the joint European Co-normative Research Project EN-PME-TEST (Fraboulet, 2015), where a FID is used to measure the total amount of hydrocarbons in the gas phase at chimney locations. This method will be tested in other parts of the IMPRESS2 project.

Denier van der Gon *et al.* (2015) calculated concentrations of particles at a European scale using emission factors from EEA/EMEP. The results were compared to measured concentrations of organic particles at different sites around Europe showing that the emission factors were too low to explain the obtained concentrations. The discrepancy between measured and calculated concentrations also differed between countries, which suggested that one source of error was that different standards are used. This was treated by adding assumed semivolatile components to the emissions based on Shrivastava *et al.* (2008), giving rather good results in the end. The EEA/EMEP emission inventory guidebook was updated in 2013 (EEA, 2013) with numbers of total particle emissions from residential wood combustion in line with this modelling study, but the recommendation was that primary aerosol inventories need to be revised to include the semivolatile organic aerosol that is formed almost instantaneously due to cooling and dilution of the flue gas or exhaust (Denier van der Gon *et al.*, 2015).

### 3.3.3 Chemical effects

After emission of the flue gas into the atmosphere, including condensation when cooled, the flue gas will be affected by many physical and chemical processes. These processes will lead to even larger particle mass measured in aged emissions from domestic biomass combustion. The main effect of chemistry in this case, is that different compounds have different boiling points and thus move mass between gas and particle phase.

Particles that form during atmospheric aging is naturally not possible to measure at the chimney, as we do not yet know all the paths from organic gases in the chimney to organic particles at a later stage. Some studies have tried to use this method to measure only a few different gaseous precursors to approximate the aged particles from biomass combustion.

Many studies measure levoglucosan in air quality PM as a tracer for biomass combustion. Levogucosan is rather stable in the atmosphere and the source is mainly pyrolysis of ligning and thus closely related to biomass combustion. Still, other sources e.g. combustion of brown coal emits levoglucosan of the same reason and in many studies other anhydrosaccarids, like mannosan and galactosan, can be used together with levoglucosan to discriminate between the sources (Achad *et al.*, 2018, Fabbri *et al.*,

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2009). According to Yttri *et al.* (2015) the methods for analyzing levoglucosan on filters has now improved to a level not very much worse than sulphate on filters, but still the mean percentage error is within ±10% for 62% of the laboratories and within ±20% for 85% of the laboratories. The ambient concentrations are 0.1-0.4 µg/m$^3$ measured with traditional equipment (Yttri *et al.*, 2015). Levoglucosan and other anhydrosaccarids can thus be useful to trace biomass emission in the ambient air.

Bruns *et al.* (2016) measured primary emissions of particles and some gases and compared to the measured organic aerosol after aging with OH in a laboratory study. This yield was then compared to the calculated yield from published yields related to each organic specie measured and the measured concentration in the primary emission. This calculated yield showed that 22 species in the primary emissions could explain 50-100% of the total yield of secondary organic aerosol. The calculation was based on published yields for 18 of the organic compounds emitted from 9 different publications, and four more was assumed from other data (Table 2, Bruns *et al.*, 2016). Phenol, naphthalene and benzene alone explained 20-60 % of the total SOA in the experiments, but around 80% of the total observed SOA globally (Bruns *et al.*, 2016). Of these, only benzene is normally included in the SOA precursors used in most models (Bruns *et al.,* 2016). There is also a need of updated yield of phenol, as only one of the publications referenced has measured yield including seeding particles (Bruns *et al.*, 2016).

Most of the aging occurs due to formation of radicals from solar radiation, but in wet aerosols and during times where solar radiation is not active the OH-reaction is limited and instead reaction products like (R-O-) and (-O-CH-O-) increase (dark chemistry) (Gilardoni *et al.*, 2016). This would happen during dark winters as in the Nordic countries. The dark chemistry affects both how fast aging occurs and the wavelengths that are absorbed in the aged aerosol (Gilardoni *et al.*, 2016).

Still, a large part of the aging of the smoke is due to reactions with OH radicals and this aging can be mimicked in a reaction chamber, by adding high concentrations of OH speeding up the process of aging and particle formation. This process takes only aging through OH reaction into account, and not reactions with other species present in the ambient air and different weather effects. The method is very useable as OH aging is most dominant in most plumes.

## 3.4 Measurement techniques

The following section presents measurement techniques of interest to this study. The particle measurement techniques are often used in the standard methods described. A short section with optical measurement techniques is also included as it is seen as an interesting future option for measurement of precursors of secondary organic aerosols.

Due to variations in the composition and temperature of the flue gas there is a need to normalize the emission factors to enable a fair comparison of emissions from different sources. This is typically done by normalizing the gas volume to standard temperature and pressure, 0 °C and 101,3 kPa. Also, to account for variations in dilution of the flue gas the emissions are often related to a specific $O_2$ concentration. The emission of particles can also be related to the amount of fuel that is fed to the combustion, in that case the emissions are given per kg or MJ of fuel that is combusted. There is a need to

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recalculate the emissions between emissions per kg wood and emissions per GJ and different conversion are used, *e.g.* Vicente and Alves (2018) who used 18 MJ $kg^{-1}$.

### 3.4.1 Particle measurement techniques

The most common way to measure emissions of particulate matter from combustion applications is to collect all particles on a filter and use gravimetrical analysis to determine the particle mass concentration. Typically, the gas flow through the system is measured using a gas meter and the concentration of particles are often presented in $mg/m^3$. There are several different types of filters used for this e.g. planar or tube filters. But the principle is the same for all setups, the filters are placed in a glass or metallic filter holder which can be heated if required (ISO 13284-1). If information on the particle size distribution is required (e.g. $PM_{10}$, $PM_{2.5}$ or $PM_1$) cyclones or impactors can be placed before the previously described filters. Cyclones are often used at high dust loads, whereas impactors are more suitable at low dust loads. In a cyclone the flue gas sample is led into a conical chamber where a swirl is induced. Particles larger than a pre-determined diameter (depending on velocity) will hit the wall due to the centrifugal force are separated from the smaller particles which will follow the gas. Several cyclones with different cut-off diameters can be placed in series to obtain a size distribution with higher resolution (ISO 25597:2013).

There are several different types of impactors *e.g.* round-nozzle (ISO 23210), virtual (ISO 13271) and low pressure (Hering *et al.*, 1978). All types of impactors utilize the inertia of the particles to separate them according to their aerodynamic diameter. The particles are gradually accelerated in several stages, and the particles with high enough inertia will deflect from the gas stream and hit a collection plate where they can be gravimetrically analysed. A size distribution based on the cut-off diameter of each stage in the impactor is thus obtained.

In the standard methods described in this study simple techniques such as filters, cyclones and impactors are used. More advanced methods for measurement of the particle size distribution such as TEOM (Tapered Element Oscillating Microbalance), ELPI (Electric Low Pressure Impactor) and SMPS (Scanning Mobility Particle Sizer) exists and is widely used in research applications relating to aerosols. However, these instruments are costly, requires experienced users and are hard to use in field measurements.

### 3.4.2 Optical measurement techniques

The majority of all studies made on characterization of VOC emissions from wood combustion is made by collecting a sample on a filter which is later washed, and the collected hydrocarbons can then be identified using GC (Gas Chromatograph), *e.g.* Refs. (Tissari, 2007, Barrefors and Persson, 1995, Avagyan *et al., 2016*, Shauer *et al.*, 2001, Shannigrahi *et al.*, 2014). It is, in general, a difficult task to use optical measurement techniques to measure hydrocarbons in flue gases due to overlapping bands of $H_2O$ and $CO_2$, but also overlaps with other hydrocarbons. For light hydrocarbons, such as methane, commercial FTIR systems can be used. But large molecules, including VOCs, are more difficult to analyse due to a broader spectral footprint with lower information content (Hodgingson and Tatam, 2013, and Ossler *et al.*, 2001). Additionally, the

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temperature of the gas cells in commercial FTIR systems are typically kept below 200 °C which causes problems of condensation of larger hydrocarbons such as PAH.

However, in research applications there are studies using optical techniques measuring VOCs. Fourier Transform Infra-Red (FTIR) spectroscopy and Tunable Tiode Laser Absorption Spectroscopy (TDLAS) can be used to measure benzene in vapor phase (Jeffers *et. al.,* 2004). Christian *et al.* (2004) used open path FTIR and proton transfer reaction mass spectroscopy (PTR-MS) to measure emissions from biomass combustion in a test facility. They obtained good results for benzene using the PTR-MS method, but the FTIR based method was mainly suitable for smaller hydrocarbons and benzene was not possible to measure with this setup. Also, no good results for measurement of Phenol was obtained with the PTR-MS or FTIR-technique (Christian *et al.,* 2004). Ossler *et al.*, (2001) studied fluorene, naphthalene, antracene and pyrene using picosecond laser-induced fluorescence showing that if advanced laser techniques are applied it is possible to measure individual PAH with optical techniques.

There is an ongoing work of improving the sensitivity and selectivity of optical measurement techniques. As stressed by Hodgingsson and Tatam (2013), the recent and ongoing development of tunable laser sources in the mid-IR region offers new possibilities of measuring species of interest in this study. More research and development are needed in this field before these techniques can be applied in any standard methods. But, in future TDLAS seems like an interesting option to measure precursors of secondary aerosols, where benzene appears to be the easiest of the three precursors (benzene, naphthalene and phenol) identified in this work.

Optical measurement techniques are still only used in research and more development work is needed before they can be used in the applications relevant for this study.

Thus, the complexity of adding measurements of organic compounds to a particle measurement method is delicate and what description of the organics that is interesting to the emission data needs to be understood in detail.

## 3.5 Emission factors

Nussbaumer *et al.* (2008) made a detailed survey and review of the various emission factors in use in Europe, also in relation to the type of measurement technique. They pointed towards a few differences between different measurement methods and *e.g.* showed how emission factors varied between measurement with and without start phase emissions and between measurements of solid particles (indicated as SP), solid particles plus condensables found in impinge bottles (SPC), and total particles collected in a dilution tunnel (Nussbaumer *et al.*, 2008). Denier van der Gon *et al.* (2015) showed that the emission data they needed for their modelling study partly originate from different methods that countries use and cannot be directly compared as no detailed description of measurement methodology is given. In a recent study, Kindbom *et al.* (2017) studied specifically emission factors for SLCP (Short Lived Climate Pollutants) from residential wood combustion in the Nordic countries. This included measurements of $PM_{2,5}$, EC, OC, $CH_4$, NMVOC and CO from some ten biomass boilers and some ten room-heaters, operated at varying conditions and with varying fuels. The emission factors derived from the measurement data were mostly different from the currently used national emissions factors, some considerably higher and some considerably lower. There were also

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significant differences between the emission factors used in the different countries. A main reason for this is that they are based on emission data derived using different measurement standards. In Table 1, emission factors for $PM_{2,5}$ from the above measurements are compared to literature values for different types of combustion equipment (from Refs. in Kindbom *et al.* (2017)).

**Table 1§.** Comparison of PM2,5 measurement results from Kindbom et al (2017) and literature values (from Refs. in Kindbom *et al.* (2017)) for different types of combustion equipment.

| **Technology** | **Kindbom *et al.* (2017)** **[g/GJ]** | **Literature values** **[g/GJ]** |
|---|---|---|
| Older/traditional wood boilers | 317 - 1975 | 95 - 1300 |
| Modern wood boilers | 24 - 89 | 11 - 450 |
| Conventional/traditional wood stove | 78 - 330 | 90 - 3000 |
| Modern wood stoves | 53 - 821 | 36 - 798 |

In summary some valuable data for varying operating conditions are available for residential wood combustion, but it is of prime importance to know the measurement method used. Data from larger plants mainly show the effect of the particle removal equipment and national or local emission limits (Nussbaumer *et al.*, 2008).

There is a need to differentiate between elemental carbon and organic carbon that has a large effect on air quality, e.g. Denier van der Gon *et al.* (2015) had to assume that EC/OC-fractions were independent of control technology as the reviewed literature did not allow further technology- dependent fractions of EC and OC. Nussbaumer *et al.* (2008) recommends that measurements of particle emissions should differentiate between organic condensables, that are related to combustion efficiency, and inorganic particle mass (mainly salts), that are more a result of the fuel.

Standard emission measurements for residential wood combustion includes carbon monoxide (CO), unburned organic compounds (OGC), nitrogen oxides ($NO_x$) and particles. As most of the particles in wood smoke fumes are below 2.5 µm in diameter, the size segregation is normally not used. Particles are as discussed in this text extremely variable in many other different aspects.

## 3.6 Legislation and governance

The legislation related to the emissions from biomass combustion for heating can be divided into emissions and concentrations in outdoor air. For concentrations legislation originates from the EU and is appointing the countries responsible for the air quality at sites where humans can reside. There are environmental quality standards interesting to this subject for particles in the form of $PM_{10}$ and of $PM_{2.5}$, and the standards can be hard to reach for areas with a large amount of residential combustion for heating (Herich *et al.*, 2014).

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For emissions the legislation focuses on the appliances, as they are manageable to control. Many countries have laws that force new installations to be approved for low emissions, and a few even have regulations for appliances already in use (Viana *et al.*, 2016). There is also a large market focused on ecolabelling and other types of labelling to improve energy efficiency and emission levels liberally (Viana *et al.*, 2016).

For residential appliances and medium combustion plants, there will in a few years be common requirements valid for all EU member states. For boilers up to 1000 kW and for residential room-heaters these are implemented under the Eco-design directive. For boilers, they include maximum emission levels on particles, OGC, CO and NOx and will enter into force on 1st of January 2020. The emission limit values for particulates are max 60 mg/m$^3$ at 10 % $O_2$ for wood log boilers and max 40 mg/m$^3$ at 10 % $O_2$ for pellet boilers. For room-heaters, requirements on the same parameters will enter into force on 1st of January 2022. The emission limit values for particulates varies between max 20 mg/m$^3$ at 10 % $O_2$ and max 50 mg/m$^3$ at 10 % $O_2$ depending on the type of equipment. For medium-sized combustion plants, *i.e.* plants between 1 and 50 MW, new emissions limit values for particulates, $NO_x$ and $SO_2$ will enter into force on 20th of December 2018 (new plants), 1st of January 2025 (existing plants >5 MW) or 1st of January 2030 (existing plants <5 MW). ). The emission limit values for particulates are max 20 mg/m$^3$ at 6 % $O_2$ for new plants (max 25 for plants <5 MW) and max 30 mg/m$^3$ at 6 % $O_2$ for existing plants (max45 for plants <5 MW).

The eco-design regulation and the few standards on particle emission sampling methods adopted by few member states are all concerning the type testing of the heating generator appliances. Just few Countries in Europe, as Austria and Germany, are provided with a standard addressed to measure gaseous emissions "in-field", that is in the place that the appliances has been installed. This is a critical point for air quality, in particular for small appliances, as the emission factors are strictly dependent on the quality of the installation (both of the appliances and of the exhaust system) and on managing the plant (characteristic of the fuel, maintenance procedure, air regulation, etc.).

There are also regulations for the total emission from each country in the EU, where biomass combustion emissions are included. The reporting of air quality emissions to the European Union follows the same system as reporting climate gas emissions and as air quality is a local or at least regional issue the data can be hard to use. This imposes a need to increase the usability of the emission data to both researchers and to local governmental institutions responsible for air quality (Gustafsson and Kindbom, 2017). Also, the effect of different measurement methods used in different countries can have a large effect on the approval of EU emission limits (Denier van der Gon, 2015).

Still, a large part of residential heating in the EU is due to combustion of fossil fuel, and a struggle to limit coal as an important fuel is presently in place. In Madrid a fund started 2007 and is still active, with the goal to reduce coal combustion turning many of the heaters into natural gas heaters. This reduces the climate emissions, but the fuel is still fossil and thus further steps need to be taken, probably into more biomass fuel used in the domestic heating sector. Natural gas (fossil) has a large part of the domestic heating market in the European Union (Viana *et al.*, 2016).

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# 4 Standards for PM and SVOC measurements

The following section provide a short principle description of the standards included in the survey, the description is mainly limited to the setup of the sampling system. Additional standards, not included in the survey, are also presented.

## 4.1 Included in the survey

### 4.1.1 ISO 12141:2002 – Determination of mass concentration of particulate matter (dust) at low concentrations – Manual gravimetric method

This international standard has been developed for measurements of dust up to 50 mg/m$^3$ with a special emphasis in the region around 5 mg/m$^3$. It has been developed for waste incinerators but can be used at any stationary sources. The measurements are performed by iso-kinetically extracting a flue gas sample from the stack. The extracted particles are then collected on a plane filter. The filter holder can either be placed in the probe tip, i.e. in the stack, or in a heated box outside the stack.

This standard resembles ISO 13284, but more emphasis is put on high volume sampling at low dust concentrations. It is also similar to ISO 9096:1992, but with extended range to also cover lower concentrations and requirements on how the filter blanks are treated.

### 4.1.2 ISO 25597:2013 – Test method for determining PM and PM10 Mass in Stack gases using Cyclone Samplers and Sample Dilution

This standard can be used to measure both primary and secondary aerosols in flue gases from stationary sources using cyclones. Two different sampling trains are described; one basic and one more advanced. In the basic setup primary aerosols are measured using $PM_{10}$ and $PM_{2.5}$ cyclones in series with a filter. All components are placed inside the stack allowing sampling at the same temperature as the flue gas. In the more advanced setup, where also secondary aerosols can be analyzed, the basic setup is completed with a dilution chamber, placed outside the stack and after the $PM_{2.5}$ cyclone. After the dilution chamber the solid (< 2.5 µm) and condensable particles are collected in a $PM_{2.5}$ cyclone and a filter.

Isokinetic sampling shall be applied. In the more advanced setup a minimum dilution ratio of 20:1 with a residence time of at least 10s in dilution chamber. The filter temperature shall be at least 42°C during the sampling.

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### 4.1.3 ISO 13271:2012 – Determination of PM10/PM2.5 mass concentration in flue gas – Measurement at higher concentration by use of virtual impactors

A method based on two-stage virtual impactors for measurement of $PM_{10}$ and $PM_{2.5}$ in flue gas of industrial sources is presented in this standard. The standard is especially suitable for high dust loads and for sampling of flue gas at harsh conditions (*e.g.* containing reactive compounds, at high temperature or high humidity). The typical dust concentration for this method is 1 – 200 mg/m$^3$ with a mean value of 40 mg/m$^3$.

The sampling system consists of a two-stage virtual impactor placed in the probe tip, *i.e.* in-stack, the components downstream the impactor shall comply with the specifications in ISO 12141. The size classification is done within the impactor where particles >10 µm are separated in a first stage, in the second stage particles between 2.5 and 10 µm are collected and particles smaller than 2.5 µm are collected in a backup filter. The separation curves of $PM_{10}$ and $PM_{2.5}$ shall in this method correspond to the separation curves presented in ISO 7708 for ambient air quality measurements.

### 4.1.4 ISO 23210 – Determination of PM10/PM2.5 mass concentration in flue gas – Measurement at low concentrations by use of impactors

This standard describes the use of two-stage round-nozzle impactors for measurement of $PM_{10}$ and $PM_{2.5}$ in flue gas with concentrations preferably lower than 40 mg/m$^3$. Isokinetic sampling shall be applied, and the impactor is placed in the probe-tip, i.e. in the stack. The impactor has two stages with cut-off of 2.5 and 10 µm and a back-up filter to collect all particles smaller than 2.5 µm. The separation curves of $PM_{10}$ and $PM_{2.5}$ shall in also in this method correspond to the separation curves presented in ISO 7708 for ambient air quality measurements.

### 4.1.5 ISO/EN/SS 11338-1 – Determination of gas and particle-phase polycyclic aromatic hydrocarbon – Part 1: Sampling

The ISO 11338 standard contains two parts which should to be combined to obtain a complete procedure for determining PAH emissions from stationary sources. This first part describes three different sampling methods. The three methods are; The dilution method (A), The heated filter/condenser/adsorber method (B) and the cooled probe/adsorber mehod (C). All methods are expected to give equivalent results but depending on the application the methods are more of less suitable. A table with various process characteristics are listed together with suggestions on which sampling method to use depending on the conditions.

In method A, the sample is extracted through a probe maintained at the same temperature as the flue gas. The extracted sample is then cooled via dilution with air to

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a temperature below 40 °C. A silicon-bonded glass fibre filter impregnated with paraffin oil is then used to collect PAH components with 4 to 7 rings. A solid absorbent can be used downstream to collect lighter PAH (2 or 3 rings).

In Method B, a heated filter maintained above dew point but below the flue gas temperature is used to collect solid particles. After that the sample gas is cooled to below 20 °C and led to impingers where gaseous PAH are condensed or adsorbed on solid absorbents. A cyclone or quartz wool filter can be used before the heated filter when sampling at high particle concentrations (*e.g.* > 100 mg/m$^3$).

Method C prescribes a water-cooled probe with an inner tube of glass in which the flue gas is condensed. The temperature of the probe is kept below 40 °C and the condensate is collected in a flask after the probe. Gaseous PAH are then collected in an adsorbtion stage with impingers, filled with *e.g.* diethylene glycol, and a solid adsorbent. A filter is normally used before the last adsorption stage.

Isokinetic sampling shall be applied in all methods and the sampling procedure of the flue gas should be performed in accordance with ISO 12141 or ISO 9096.

### 4.1.6 ISO/EN 11338-2 – Determination of gas and particle-phase polycyclic aromatic hydrocarbons – Part 2: Sample preparation, clean-up and determination

The second part of ISO 11338 describes how to prepare and analyze the PAH samples collected using Part 1 of the standard. The analysis of PAH is performed using either high performance liquid chromatography (HPLC) or gas chromatography-mass spectroscopy (GC-MS). The PAH sample can be either in particulate phase or in gas phase, collected on a filter or by an adsorbent respectively. The samples are then extracted using an organic solvent, all parts of the sampling equipment should also be cleaned using the solvent and added to the analysis.

### 4.1.7 CEN/TS 13649 – Determination of the mass concentration of individual gaseous organic compounds - Sorptive sapling method followed by solvent extraction or thermal desorption

The present specification describes a method for determining emissions of individual VOCs. The flue gas is extracted using a probe and a pump and the samples are normally collected by adsorption on sorbents which are extracted using solvents or thermodesorption and are finally analyzed by gas chromatography. Other methods may also be used to collect the VOCs, *e.g.* using a condensate trap. A decision tree is presented where the proper sampling procedure can be selected depending on the conditions in the emission source. A dust filter is used to prevent particles from contaminating the samples, but the particles collected on the filter is not analyzed.

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### 4.1.8 EPA Method 5G – Determination of particulate matter emissions from wood heaters (dilution tunnel sampling location)

This American emission standard describes a method for determination of particle emissions from wood heaters. All flue gas is collected in a hood where it is diluted with air, thereby allowing formation of secondary aerosols. A sample of the diluted flue gas is extracted using a stainless steel or glass probe. The particles are collected on two glass-fiber filters in series, the temperature shall be measured between the filters and the maximum allowed temperature is 32 °C.

### 4.1.9 EPA Method 5H – Determination of particulate matter emissions from wood heaters from a stack location

This method describes sampling of particulate matter and condensable species (SVOC) from wood heater stacks. A sample is extracted using a glass or stainless-steel probe, solid PM are then collected on a glass fiber filter maintained at a temperature of maximum 120 °C. After the filter, the gas is cooled to a temperature below 20 °C and led through an impinger system where condensable species are collected. A filter, also with a maximum temperature of 20 °C, are placed after the impinger system where particles formed during cooling/condensation of the gas are collected.

### 4.1.10 EPA Method 17 – Determination of particulate matter emissions from stationary sources

In contrast to Method 5 where the filter is maintained at a specified temperature, the filter holder is in this method placed in the stack thus collecting particles at the flue gas temperature. Since the filter holder is placed within the stack, the temperature of filter varies depending on the sampling conditions, it is stressed that Method 17 should only be used when the PM concentrations are known to be independent of temperature

### 4.1.11 EPA Method 28/28R – Certification and auditing of wood heaters

This method describes the procedure for testing of wood heaters. The particle sampling shall be performed using either Method 5G or Method 5H. Note that this method is not directly connected to the present study and should not have been included in the survey as no new test methods were described.

### 4.1.12 EPA Method 201A – Determination of PM10 and PM2.5 emissions from stationary sources (Constant sampling rate procedure)

Emissions of filterable particulate matter (i.e. materials in solid or liquid form) from stationary sources can be measured using this method. For condensable species, this

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method should be combined with Method 202. The particles are classified into PM10 and PM 2.5 using cyclones and a filter in series, all equipment is placed inside the stack.

### 4.1.13 EPA Method 202

If the filtration temperature of Method 201A, *i.e.* stack temperature, exceeds 30 °C this method should be combined with Method 202 to obtain information of filterable and condensable PM. This method can also be used in conjunction with Method 17. After the filterable PM has been removed, as described in other EPA methods, the sample gas is cooled in a condenser and then led to cooled dry impingers where condensable species are collected. A filter is also placed in the impinge-train where condensable aerosols are collected, the temperature of the filter shall be kept below 30 °C. After the sampling is completed, the impingers may be purged with pure nitrogen to remove dissolved $SO_2$ in the condensate.

### 4.1.14 ASTM E2515 – Determination of Particulate matter emissions collected by a dilution tunnel

This test method is used to measure particulate emissions from small scale appliances burning solid fuels. A dilution tunnel is placed above the exhaust of the test object, *i.e.* the total flow of flue gas is diluted with air. Sampling of the diluted flue gas shall be performed using dual sampling trains, each consisting of two glass fiber filters in series. The filter holders shall be maintained at a temperature of less than 32 °C. The average of the two samples are then used to present the particulate emissions from the test object.

### 4.1.15 ASTM E2780-10 – Determining Particulate matter emissions from wood heaters

This method describes how to operate the wood heater when performing particle emission testing. The fueling and operation protocol is described in detail. However, the particle sampling shall be made in conjuction with ASTM E2515 and no new test methods are described. So, as for EPA Method 28, this method should not have been part of the survey.

## 4.2 Standards not included in the survey

### 4.2.1 NS 3058-2 – Determination of particulate emission

This Norwegian standard is used for determination of particulate emissions from wood heaters. The method is based on measurements in a dilution tunnel, where the total flue gas flow is diluted with air. A sample is extracted from the diluted flue gas and the particles are collected on a two glass fibre filters placed in series.

### 4.2.2 EN-PME-TEST

The EN-PME-TEST is a method under development with the aim of establishing a common European standard for particle emission measurement from appliances and boilers (Fraboulet, 2015). The method is based on collection of primary particles on a heated filter and measurement of OGC using a FID. Both sampling lines are kept at a

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constant temperature of 180 °C. A long term goal with the method is to also include secondary aerosols in the method. But in the short-term suggestions, only measurement of OGC is included. However, this is a precursor of secondary aerosols and information on SOA is thus to some extent included in this method.

### 4.2.3 EN 12619

This is a European standard for measurement of total organic substances in gaseous or vaporized form from stationary sources. A gas sample is extracted using a probe and led to in a heated sampling line (minimum 180 °C) through a protective particle filter before being analysed using a FID. The results from the standard is expressed as mg/m$^3$ of total carbon (TVOC).

### 4.2.4 EN 13284

The EN 13284 standard method is primarily developed for measurements of total dust from waste incinerators at concentrations below 50 mg/m$^3$, but it can be applied to other sources and at higher concentrations. The principle of the measurements is that an iso-kinetic sample is extracted from the sampling point using a probe. The dust is then collected onto a pre-weighed filter which can either be placed in the probe tip or placed outside the stack. In the case of out-stack filtration the filter housing and suction tube shall be temperature controlled to avoid condensation before the filter.

### 4.2.5 VDI 2066

This German standard is identical to EN 13284 for measurements in the range up to 50 mg/m3.

## 4.3 Summary of standards

The typical sampling points of the different standards are schematically illustrated in Fig. 2. In this figure the division has been made between methods where the actual size classification instruments are placed inside the stack (denoted “In-stack”) and methods where flue gas is extracted from the stack. Also, methods using dilution chambers are distinguished. Here the division has been between methods where the total flue gas flow is diluted and methods where a flue gas sample is diluted in an external dilution chamber. The use of dilution enables secondary aerosol formation during cooling of the flue gas and thus sampling at conditions more resembling the conditions outside the stack.

There are six methods (ISO 12141, ISO 15597, ISO 13271, ISO 23210, EPA Method 17 and EPA Method 201A) based on placing the filtration device inside the stack. These are typically standards presenting size classification of the particle emissions in PM2.5 and PM10 and are measured using cyclones or impactors. These techniques are limited to larger combustion units due to the requirements on stack dimensions. However, as shown in the literature study, the characteristic size of particle emitted from combustion of wood in small units is $PM_1$, i.e. classifying the emissions into $PM_{10}$ and $PM_{2.5}$ is not necessary in these applications.

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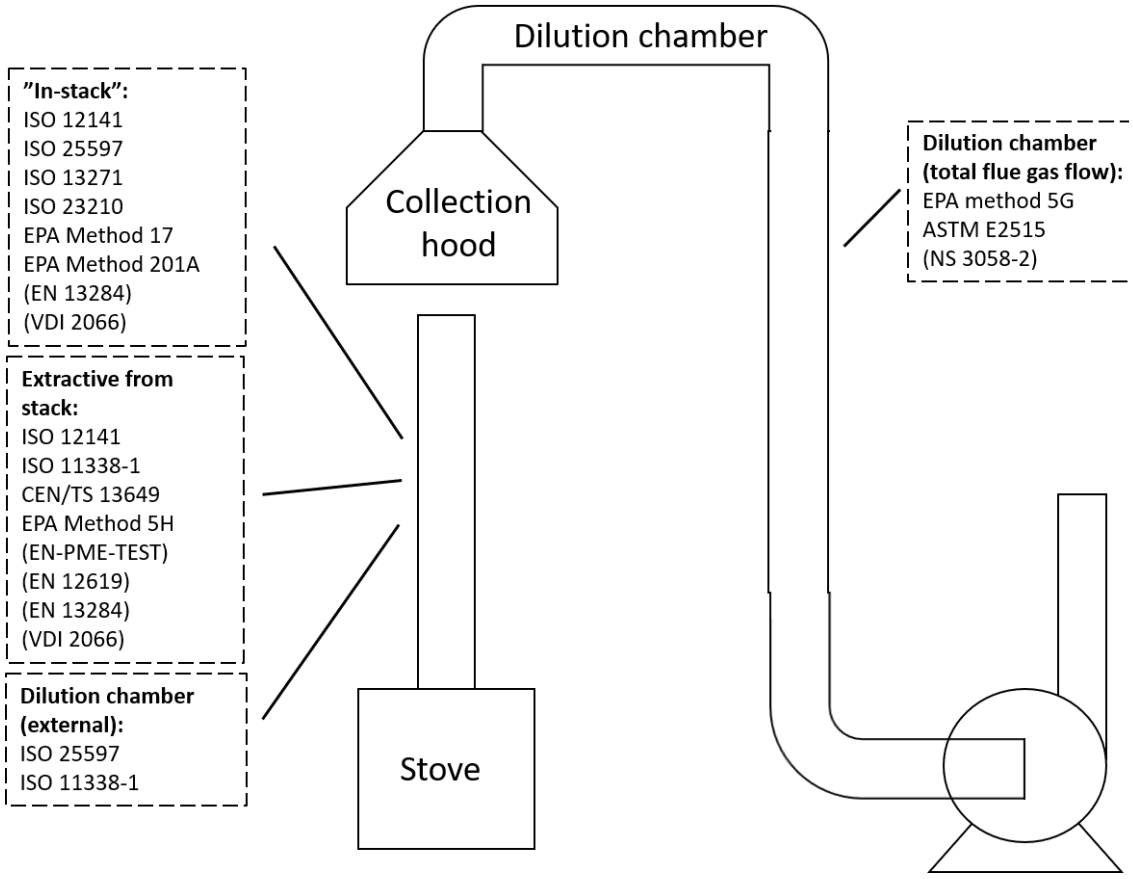


**Figure 2.** Schematic visualization of where the standard methods included in the survey are implemented in a typical test furnace.

The standard methods can also be grouped based on the quantities measured. Table 2 shows a representation of the methods based on this division. The methods with size classification into $PM_{10}$ and $PM_{2.5}$ are based on the use of cyclones or impactors whilst the $PM_{TOT}$ methods collect all filterable particle sizes on filters (glass or quartz) at the current temperature of the filter.

**Table 2.** Measured parameters for the standard methods described (the methods in italics were not present in the survey).

| PM10 and PM2.5 | $PM_{TOT}$ | Condensable particulates | OGC (PAH, SVOC) |
|---|---|---|---|
| ISO 25597<br>ISO 13271<br>ISO/EN 23210<br>EPA Method 201A | ISO 12141<br>EPA Method 5G<br>EPA Method 5H<br>EPA Method 17<br>*EN-PME-TEST*<br>*EN 13284*<br>*VDI 2066* | EPA Method 202<br>EPA Method 5H<br>ISO 25597<br>*NS 3058-2-* | ISO/EN 11338-1<br>ISO/EN 11338-2<br>CEN/TS 13649<br>*EN-PME-TEST*<br>*EN 12619* |

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## 4.4 Results from the survey

We received 20 responses to the online survey, representing 7 different countries in Europe (Denmark, France, Germany, Italy, Sweden and Switzerland) with 10 answers received from Italy. Note that the number of responses is too limited to show how these standards are used within Europe, and the survey is thus mainly used indicatively, excluding general conclusions. However, it gives an idea on which standards that are being used by the notified bodies which the survey was sent to. Also, the comments on the methods provides valuable information on observations made by the actual users of the methods.

To identify which of the methods were well known and to which extent they were used a number of questions were asked starting with if the person is aware of the method, then if it has been used and finally if the method has been used frequently.

Figure 3 shows the results from the first question ("Are you aware of this method?"). The results indicate that most methods are known, but no method is known by more than approximately 60% of the respondents. Most methods are known by approximately 50%, but the EPA Method 28/28R was only known by one person and the ISO 13271, EPA Method 202, ASTM E2515 and ASTM E2780-10 are known by less than 30 % of the respondents. In general, the International or European methods are more well-known than the American standard methods.

The order in which the methods appeared in the survey was the same for all, *i.e.* according to the numbering in Fig. 3, and it was not mandatory to answer all questions to proceed in the survey. This resulted in a decreasing number of responses to the questions later in the survey, due to persons skipping the last part of the survey. In this question we received 20 responses for the first method and 17 for the last method. The statistical certainty is therefore better for the first methods than for the last. The presented data is from the actual answers (Yes/No) and skipped answers were excluded from the results. The difference in result due to this is not regarded as significant but randomizing the order of the methods would have been better.

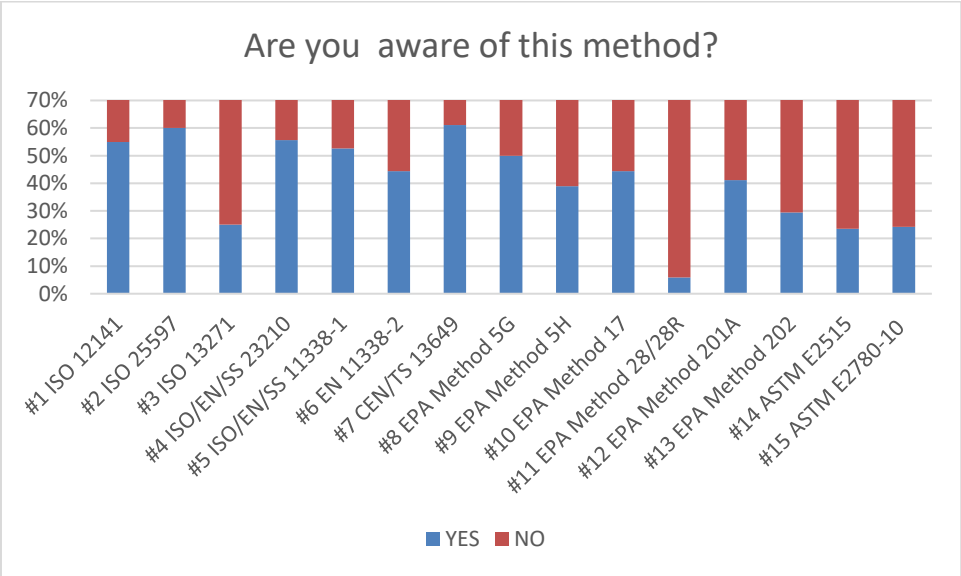


**Figure 3.** Survey answers on the question "Are you aware of this method?"

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The question "Have you used this method?" received between 20 and 15 responses. There are four methods (ISO/EN 23210, ISO/EN 11338-1 and -2 and CEN/TS 13649) which are clearly more used than the others, Fig 4. More than 35 % of the respondents have used them, whereas the other methods were used by less than 20. This shows that the respondents of the survey have some own experience from most of the methods included in the survey. The EPA Method 28/28R and ASTM E2780-10 has not been used by any of the respondents of the survey. These are methods describing how to operate the test objects and could have been excluded from this survey.

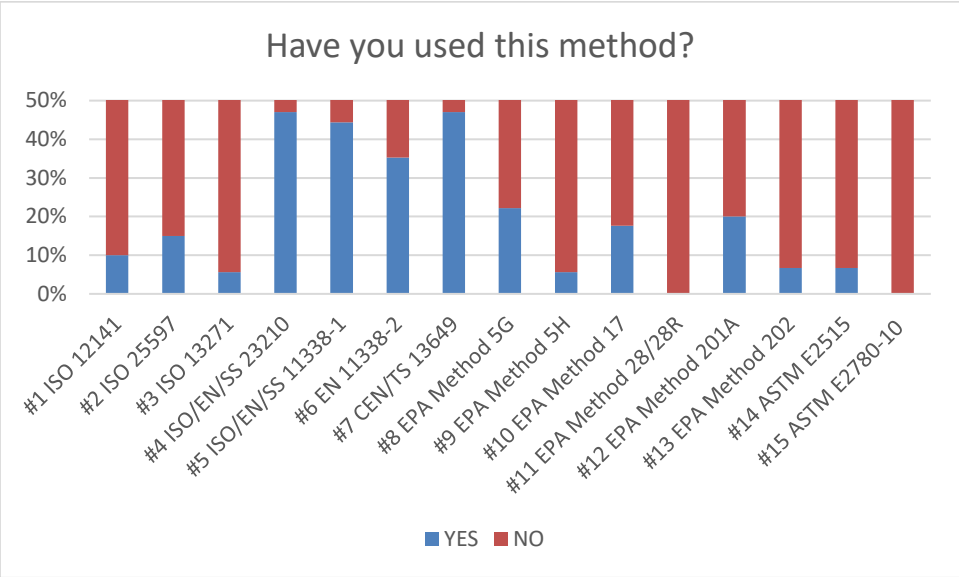


**Figure 4.** Survey answers to the question "Have you used this method?".

The methods that has been used frequently by more than 40 % is the ISO/EN 23210, ISO/EN 11338-1 and CEN/TS 13649, Fig. 5. The ISO/EN 23210 is used for determination of $PM_{10}/PM_{2.5}$ emissions by using in-stack round nozzle impactors and it is, in this study, the most popular method for size classified particle measurement. When comparing the comments on the strengths with this method with the other methods for $PM_{10}/PM_{2.5}$ measurement it seems to be a method which is relatively easy to learn and straightforward to operate. In common for all impactor and cyclone-based methods is that they may be more useable in larger facilities than residential combustion, due to the size and flow rates, as pointed out in some comments.

Both CEN/TS 13649 and ISO/EN 11338-1 are methods for measurement of gaseous organic compounds (only PAH for the latter). Comparing the comments in the survey on these two methods it is obvious that they are both cumbersome, but they are also recognized as interesting for this study due to the possibilities of measuring relevant gas species for condensable aerosol formation.

For the total PM-methods (ISO 12141, EPA Method 5G, 5H and 17). Comments were made on both the ISO 12141 and EPA Method 5H that they were complicated and required skilled users. Several positive comments were made on the EPA Method 5G, which is based on a dilution tunnel, including both that it accounts for condensable particles but also that the construction of the dilution tunnel is relatively easy and cheap.

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However, none of these methods has been used frequently by the respondents. One method that was not included in the survey but was added as an optional extra method by several persons was the EN 13284-1 which, if included in the statistics, is the most frequently used method for measurement of the mass concentration of dust. The new method EN-PME-TEST was also added as extra method where a fixed filter temperature and the possibility of measuring OGC using a FID were some of the positive features mentioned. The standard VDI 2066, used for type testing in Germany, were also frequently used by some but to less extent than the other standard methods. Worth noting when considering how frequently the methods have been used is that the survey was only sent to European labs and that the American standards are not used frequently is therefore natural.

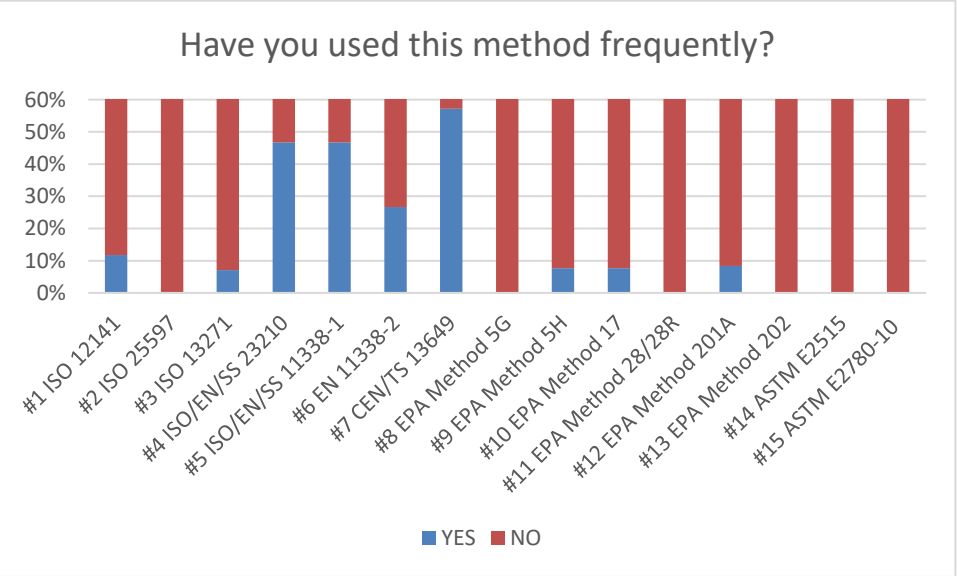


**Figure 5.** Survey answer to the question "Have you used this method Frequently?".

The question "Can you recommend this method?" was skipped by many, and the number of responses vary more than for the previous questions (between 2 and 9 responses), so instead of presenting the share of Yes and No answers the number is instead presented, Fig. 6. It does require some experience from the persons responding to the survey to make a recommendation on a method.

Looking at the individual answers, there is a tendency that it is the persons using the method frequently that has also answered this question. Despite the few answers the persons making the recommendations of the methods are considered well aware of the pros and cons of the method, since they are often frequent users, making the results valuable. It is the methods ISO/EN 11338-1 and -2 and CEN/TS 13649 that is the most recommended methods in this study. The frequently used method ISO/EN 23210 has a higher share of persons not recommending it compared to the others, but it is still the $PM_{10}/PM_{2.5}$-method with most recommendations. From the non-included methods, the EN13284 was recommended by three persons making it the most recommended standard for total dust measurement. EN-PME-TEST and VDI 2066 methods were both recommended by two persons.

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In retrospect, it would probably have been good to include more $PM_{TOT}$ methods in the survey, and in particular EN 13284, to increase the statistics also on these methods although this was not the main objective with this study. If the EN 13284 would have been included in the survey, this would likely have been recommended to a higher degree. The reason for this assumption is that only a few persons took the chance of adding extra methods in the end of the survey, and that the general trend was that the methods which were frequently used also received a higher share of responses to the question: "Can you recommend this method".

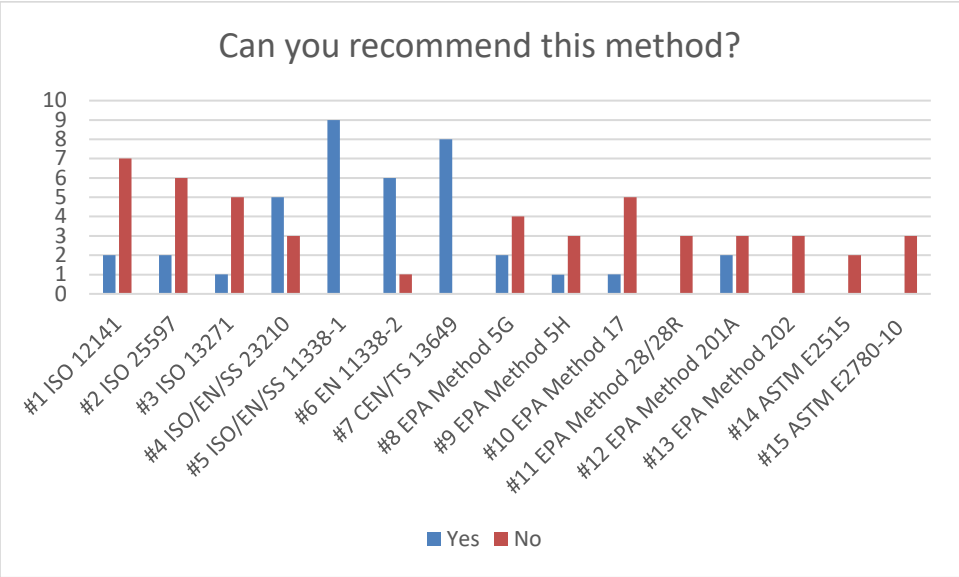


**Figure 6.** Survey answer to the question "Can you recommend this method?".

Three of the four most popular methods in the survey deals with analysis and sampling of gaseous organic compounds. This fact is positive considering the scope of the present project was that these compounds are often seen as precursors to secondary aerosol formation in the atmosphere. Even though these methods are a bit cumbersome in terms of sampling and analysis the methods are popular and frequently used which indicate that this analysis may be included in a future method without encountering severe resistance from the community using this type of standards, even though a more focused study/survey might be needed before this decision is made. Some comments were received on the difficulty of using these methods. But it was also mentioned that after some training, good results could be obtained from experienced users.

However, it may not be the test labs that will object against cumbersome measurement techniques used in the standards but instead the customers and manufacturers. The use of these techniques implies more advanced analysis and equipment and a more time-consuming sampling procedure. If required these methods will be applied by the test labs, but the cost of these measurement will be significantly higher compared to the simpler techniques. As the complexity of the methods used increases the need for control of the test labs increases, *e.g.* by round robin tests, to ensure that all methods are implemented correctly and that the analysis is made correct.

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There are large similarities between many of the standard methods described and considering the amount of work needed to be a notified body for a specific method it would be of great importance to make an effort in the standardization community to decrease the number of different standards.

# 5 Conclusions

This short review showed that:

- It is time to improve the measurement standards and the reporting of emission data in different countries, starting with the EU countries.
- Including emission data on e.g. phenol, naphthalene and benzene, would improve the estimates of formation of particle mass in biomass effluents (ambient particles).
- It would be preferable to measure inorganic and organic particles differently, as the inorganics is mainly related to the fuel and the organics more to the combustion facility, and thus the abatement strategies for the different parameters differ.
- Formation of secondary organic particles takes place both in the gas phase and the aqueous phase, and the resulting compounds differ. In the aqueous phase the reactions are included in dark chemistry and other compounds are important than in OH-driven chemistry (Gilardoni et al, 2018). This might imply benefits of measuring other species than phenol, naphthalene and benzene in some areas.
- The partitioning between the gas and the particle phase can be calculated assuming instantaneous thermodynamic equilibrium at sufficiently good results (max 7% error in Sinha et al, 2018).
- Particle number is important for cloud formation processes and thus important data, but still this data is highly related to dilution processes and thus these processes need much more research to be usable in a measurement standard at this level.
- If different standard methods on measuring particle emissions from biomass combustion is used, the reported emission data needs to include information on what type of emission factor that is presented.
- The laboratories answering the survey did not find it problematic to include some organic analysis in the standard, showing that it is time to improve the standard emission measurement standards at least to include the particles that are condensed close to the source.

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# Appendix 1 – Survey

EMPIR EURAMET

The EMPIR initiative is co-funded by the European Union's Horizon 2020 research and innovation programme and the EMPIR Participating States

Measurement methods for the characterization of PM and organic compounds emissions from domestic biomass combustion

## 1. Start

**The following methods are included in the survey:**

- ISO 12141
- ISO 25597
- ISO 13271
- ISO/EN/SS 23210
- ISO/EN/SS 11338-1
- EN 11338-2
- CEN/TS 13649

- EPA Method 5G
- EPA Method 5H
- EPA Method 17
- EPA Method 28/28R
- EPA Method 201A
- EPA Method 202
- ASTM E2515
- ASTM E2780-10

- Extra 1
- Extra 2
- Extra 3

RISE DANISH TECHNOLOGICAL INSTITUTE INERIS ENEA Italian National Agency for New Technologies, Energy and Sustainable Economic Development

## 1. Contact information

Name

Organization

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EMPIR EURAMET

The EMPIR initiative is co-funded by the European Union's Horizon 2020 research and innovation programme and the EMPIR Participating States

Measurement methods for the characterization of PM and organic compounds emissions from domestic biomass combustion

## 2. ISO 12141

**ISO 12141 - Stationary source emissions - Determination of mass concentration of particulate matter (dust) at low concentrations - Manual gravimetric method**

ISO 12141:2002 describes a reference method for the measurement of low dust content in ducted gaseous streams at concentrations below 50 mg/m3 under standard conditions.

### 1. Are you aware of this method?

- ◯ Yes
- ◯ No

### 2. Have you used this method?

- ◯ Yes
- ◯ No

### 3. Have you used this method frequently?

- ◯ Yes
- ◯ No

### 4. If you have used this method frequently, in which context? (Several answers can be selected)

- ☐ Genereral emissions R&D
- ☐ On site emission verification
- ☐ Product R&D
- ☐ Product verification
- ☐ Air quality
- ☐ Other

### 5. If other, please indicate which context?

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## 6. Which instrument type and manufacturer has been used?

## 7. In general, we recommend this method

- ◯ Yes
- ◯ No

## 8. We have the following comments on the method:

| | |
|---|---|
| User feasability | |
| Relevance | |
| Strenghts | |
| Weaknesses | |
| Other | |

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