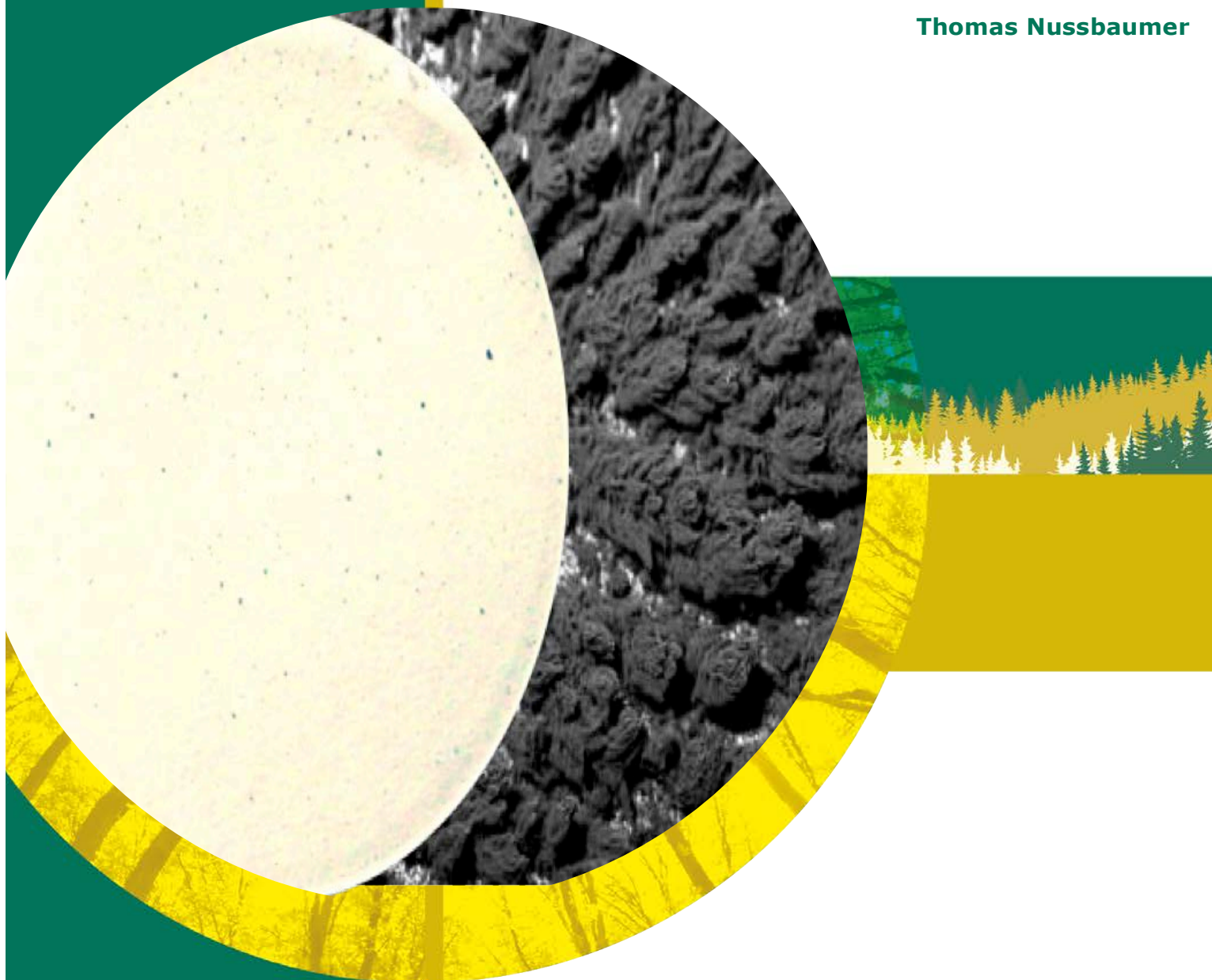# Aerosols from Biomass Combustion

Technical report on behalf of the IEA Bioenergy Task 32

Thomas Nussbaumer

Particles from near-complete biomass combustion collected on a filter (left) and soot agglomerates from incomplete combustion trapped in an electrostatic precipitator (right). Photos: Bioenergy Research Group, Lucerne University of Applied Sciences and Arts, Switzerland.

IEA Bioenergy

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Technical report on behalf of the IEA Bioenergy Task 32

Thomas Nussbaumer

Swiss Delegate in the IEA Bioenergy Task 32 on behalf of the Swiss Federal Office of Energy
Verenum Research, Zurich, and Lucerne University of Applied Sciences and Arts, Horw, Switzerland

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Content

Abstract

The use of biomass for heat and power production is supported in many IEA countries since biomass as a renewable fuel can effectively substitute fossil fuels and consequently reduce fossil CO_2. During biomass combustion, inhalable particulate matter smaller than 10 micrometres (PM_{10}) can be generated which can cause adverse health impacts. CO_2 mitigation strategy involving biomass therefore needs to consider potential health impacts and ensure low PM emissions. This report summarises the current knowledge on the health relevance of combustion generated PM, describes the mechanisms which can cause PM in biomass combustion, describes different particle types, and provides information on measures to reduce PM from biomass combustion. It is important to distinguish carbonaceous and inorganic pollutants as well as primary and secondary aerosols. Organic pollutants are particularly relevant to residential biomass combustion and can be avoided at near-complete combustion conditions. Particles resulting from incomplete combustion in manual devices exhibit a high cytotoxicity, while particles from properly operated automated biomass boilers and furnaces are mainly inorganic (derived from ash constituents in the biomass) and exhibit significantly lower or even undetectable cytotoxicity. In addition, inorganic particles can be removed effectively by air pollution control equipment such as electrostatic precipitators or fabric filters. In summary, biomass can play an important role in future energy supply in an environmentally friendly manner through implementation of state-of-the-art combustion devices and their appropriate operation.

Keywords: Particulate matter (PM), organic aerosol, inorganic aerosol, particle size, health effects.

Aerosol statement (executive summary)

Background

Biomass fuel is used as a renewable energy carrier to substitute fossil fuels for heat and power production. Modern biomass boilers, which comply with today's emission limits and that are operated appropriately, exhibit low environmental impact. There are, however, situations leading to lowering air quality caused by incomplete combustion. The most relevant pollutants affecting the air quality are volatile organic compounds (VOC) and inhalable particulate matter in the size range smaller than 10 micrometres (PM_{10}) and smaller than 2.5 micrometres ($PM_{2.5}$). Incomplete combustion also results in carbon monoxide (CO), which is a toxic gas leading to fatigue, serious health problems even fatality when exposed to high concentrations. During biomass combustion other gaseous pollutants such as nitric oxides (NO_X) and sulphur oxides (SO_X) are produced, and in the case of contaminated biomass, heavy metals and organic chlorine compounds can be formed. These pollutants need to be considered in industrial applications, while they are of less importance for clean virgin woody biomass fuels.

Epidemiologic investigations reveal strong evidence that PM_{10} and $PM_{2.5}$ can cause cardiopulmonary and cardiovascular diseases, and that PM exposure can lead to increased mortality. Among aerosol compounds in the ambient air, combustion generated particles are generally regarded as harmful as they can partly be consisted of toxic matter. Furthermore, they act as carrier of noxious substances, such as polycyclic aromatic hydrocarbons (PAH) that can be adsorbed on the surface of particles. A significant fraction of combustion particles is found in the size range of ultrafine particles (UFP) smaller than 0.1 micrometres, which exhibit a large specific surface area and a particularly high mobility in the human body. Although all types of combustion particles are generally undesirable, different particle types need to be distinguished in assessing the impact of biomass combustion on ambient air:

1. Carbonaceous products from incomplete combustion (PIC) – comprised of black carbon (often denoted as soot) and primary organic aerosol (POA) including condensable organic compounds (COC, also denoted as tar). PM_{10} in the atmosphere can additionally contain secondary organic aerosol (SOA) formed from VOC. POA and SOA are potentially harmful and can be avoided by a complete combustion. Conditions that are necessary for complete combustion include homogeneous mixing between combustible gases and combustion air in a reasonable air-fuel ratio (enabling a low oxygen content in the flue gas), followed by a sufficient residence time (more than 0.3 s to 0.5 s in the combustion chamber), and sufficient flame temperature (exceeding 800 °C). In case primary measures are not sufficient, secondary measures can be applied, such as a consecutive oxidation of PIC. Secondary control measures such as catalytic oxidation and particle precipitation are not suitable or exhibit a limited applicability for biomass flue gas laden with PIC and they increase the fire risk leading to reduced reliability.

2. Primary inorganic aerosol is comprised of ash constituents in the biomass fuel, such as potassium, calcium, chlorine and other salt forming elements, and result from near-complete combustion. Although investigations reveal that in general direct toxic effects of ash constituents in cell tests are low, their emission to the atmosphere is not desirable. Except for residential applications, non-carbonaceous particles can be captured by precipitation.

Position of IEA Task 32 on aerosols from biomass combustion

Due to significance of particulate matter on the ambient air quality, the International Energy Agency (IEA) Bioenergy Task 32 has long acknowledged the need to evaluate the role of biomass combustion on PM and to identify priorities for improvements to reduce PM emissions. As a result, IEA task 32 has organised technical seminars on the subject first held in in 2002 [1], and repeated in 2005 [2], 2007 [3], and 2016 [4]. Based on the collected findings, the IEA Bioenergy Task 32 concludes that the following topics need to be supported to avoid an adverse impact of biomass combustion to ambient air quality and to promote the implementation of bioenergy in an environmentally friendly way:

1. Particularly high exposure levels to biomass combustion particles in indoor air are caused by open fires and poor combustion conditions in appliances used for cooking and heating in developing countries. Consequently, it is of high priority to implement measures to substitute such combustion principles with improved concepts such as gasifier stoves or to replace such applications with cleaner technologies and fuels in order to reduce the global mortality caused by biomass smoke.

2. Operation of combustion devices has a direct and strong influence on emissions of air pollutants. This is true for all devices, however, most relevant for manually operated stoves, furnaces and boilers. One important issue is the use of appropriate fuel with respect to its moisture, size, and ash content for meeting the design and operational requirements of the combustion device. Another important aspect is establishing appropriate start-up regime. Various investigations show that an "ignition from the top" is usually advantageous for conventional wood stoves and fire-places. In addition, the amount of wood for each loading batch needs to match the size of the combustion chamber. Finally, sufficient combustion air, although not exceeding a reasonable amount, needs to be supplied, while air throttling during the combustion phase needs to be strictly avoided. Secondary measures such as catalytic converters and electrostatic precipitators are available for wood stoves and boilers and may assist reducing pollutants in certain cases. Never-theless, these measures should only be considered as an add-on to near-complete combustion as they can be limited in their effectiveness.

3. Standardisation of biomass fuels, combustion devices, test types, and measurement methods and devices can assist target-oriented development furthering the quality of biomass combustion and ensure low impact on air quality. New standards should not only consider ideal situations of steady state conditions but also mimic real-life operations.

4. Automated biomass combustion plants for heat and/or power production incorporate advanced design and sophisticated controls conditions to achieve near-complete combustion. Hence very low emissions of carbonaceous pollutants can be achieved with the appropriate operation of the systems. Flue gas cleaning such as precipitation can be effectively applied to reduce inorganic particle emissions to an acceptable level. For automated plants, quality management of plant planning including design guidelines, appropriate sizing of combustion system, heat storage to avoid frequent start-ups and load changes, and fuel quality management plan are pre-requisites for ideal operation. Furthermore, plant monitoring can assist the plant operation.

5. The IEA Bioenergy Task 32 supports the implementation of modern automated boilers which are equipped with combustion control and efficient particle precipitation, used for heat and combined heat and power production in medium and large scale. Residential applications can also be justi-fied if specific quality criteria for fuel and equipment are met. These include operating modern pellet boilers with high quality wood pellets and without too frequent start-ups or appropriately designing and operating log wood boilers, using seasoned dry wood logs, equipped with an

electronic control and a heat storage tank. Applications causing high emissions such as open fireplaces and old log wood appliances without combustion control should, however, be discouraged especially in urban areas.

6. Different types of primary aerosols, with significantly different properties and health impacts are formed during biomass combustion. In particular, there is evidence that organic pollutants from incomplete combustion as emitted from inappropriately operated manual devices exhibit strong cytotoxicity, while inorganic particles as emitted from properly operated automated biomass combustion reveal a far lower or even undetectable effect on the cell viability. In addition to primary particles, secondary aerosols can be formed in the atmosphere from volatile organic precursors, which are also present in flue gases from incomplete combustion. Consequently, future regulations should consider the health relevance of different aerosol components and take into account the potential impact of secondary aerosols.

7. An international exchange among all the stakeholders from research, industry, energy economics, and regulatory authorities can assist in furthering the advancements on a continuous manner. Establishing regulations on energy standards and air quality plays an important role and needs to be enforced on a national basis.

1   Introduction

Biomass contributes with approximately 51 EJ/a or 11 % to the global primary energy supply (reported in 2009 by [5]) and is currently the most important renewable energy carrier. A more recent study reports 57.7 EJ/a or 10 % of the global demand, which reflects that the bioenergy use is increasing, however not its share to the total energy consumption [6]. With an estimated technical potential of 104 EJ/a by [5], its contribution can be roughly doubled in the next decades [7]. Estimations on the theoretical potential, however, vary in a broad range from 100 EJ/a using residues only and an ultimate technical potential of 1500 EJ/a [7]. If a cost-efficient supply is assumed, the biomass supply is estimated to reach to 50–250 EJ/a in 2050, whereas the global primary energy use is predicted to be 600–1400 EJ/a [7]. In general, annual food crops are assumed to be less suitable as prime feedstock for bioenergy than perennial lignocellulosic crops, not only in terms of their potentials but also with respect to sustainability criteria [7].

Woody biomass and other biogenic fuels with moderate moisture content are mainly used in combustion processes to generate heat and to a less extent electricity. Combustion technologies are available in a broad range from a few kW for residential heating and cooking up to the size range of more than 100 MW applied in thermal power plants usually operated for combined heat and power (CHP) production [8]. Life cycle analyses show that state-of-the art biomass combustion systems achieve energy yield factors[1] of 8 to 14 for providing heat and combined heat and power [9]. When compared to the energy yield factor of around 0.70 for modern oil heating systems, biomass combustion enables a reduction up to 90 % of fossil CO_2 [10]. Another comparison of different woody energy applications shows that the largest sustainable CO_2 reduction could be achieved by using woody biomass for coal substitution, while refining wood to transportation fuels achieves only half of the reduction at a higher cost [11]. Hence, biomass combustion is a highly effective option to replace fossil fuels in buildings, district heating, process heat supply, and power production.

On the other hand, biomass combustion causes air pollutant emissions such as nitric oxides (NO_X), volatile organic compounds (VOC), and aerosols. Consequently, biomass combustion contributes to particulate matter (PM), which causes adverse health effects. These include respiratory and cardiovascular diseases and leading to mortality as shown in the six cities studies [12] and subsequent epidemiological investigations [13]. It is shown that long-term exposure to combustion-related particles increases the risk of cardiopulmonary and lung cancer mortality [14]. Consequently, the World Health Organisation introduced quality guidelines for ambient air considering PM and other components [15]. According to WHO air pollution causes 7 million premature deaths annually [16]. These data indicate direct link between air pollution and health impacts.

Commonly used indicators for the air quality are inhalable particles with an aerodynamic diameter smaller than 10 micrometres (μm) denoted as PM_{10}, the respirable fraction referred to as 'fine particles' smaller than 2.5 μm denoted as $PM_{2.5}$, and 'coarse PM' defined as difference between PM_{10} and $PM_{2.5}$ [17]. Further, PM_1 is denoted as 'ultrafine particles (UFP)', while particles smaller than 0.1 μm (100 nanometres) are referred to as 'nanoparticles'.

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[1]   The energy yield factor is defined as the ratio of the produced useful energy to the energy input of non-renewable sources during the lifetime of the respective energy conversion system.

Emission limit values for particulate matter from stationary sources most commonly refer to mass concentrations and do not distinguish between toxic and less harmful substances. However, combustion particles are generally regarded as potentially harmful since they can be carriers of toxic and carcinogenic substances such as polycyclic aromatic hydrocarbons (PAH). Moreover, the main mass fraction of combustion particles is often found in the size range smaller than 1 μm and often even smaller than 0.1 μm. In many studies, such nanoparticles are considered as particularly health relevant due to their large specific surfaces and the increased mobility in the human body [18], [19].

Due to the contribution to PM_{10} in the ambient air in particular with organic compounds [20], a trade-off between climate protection and air pollution mitigation may arise. Therefore, particle emission limits from biomass combustion were decreased in many countries in the recent years and additional measures as far as prohibition of residential wood combustion in urban areas were announced by some governments. In Switzerland, the emission limits for biomass combustion devices were reduced in 2007 [21]. Since then, automated biomass combustion systems greater than 70 kW were usually equipped with particle removal mostly by electrostatic precipitators or for larger plants also by fabric filters. For residential wood combustion smaller than 70 kW, regulations are also becoming more stringent. Their effect on the ambient air is, however, uncertain, since the operation of manual wood combustion devices has a strong impact on the pollutant formation and real-life emissions can significantly exceed the emissions under type-test conditions [22]–[25].

2 Fundamentals of biomass combustion

2.1 Types of biomass combustion particles

Depending on the fuel type and the combustion regime, different particles from biomass combustion can be distinguished as follows:

1. **Products from incomplete combustion (PIC):**

    1.1 a) Primary solid particles as soot consisting of elemental carbon (EC), black carbon (BC), and primary organic aerosol (POA) in the stack.

    b) (Primary) condensable organic compounds (COC) as liquid droplets or condensed on solid particles contributing to brown carbon (C_{brown}) in the atmosphere. COC result from non-methane volatile organic compounds (NMVOC) in the flue gas and contribute to POA, since COC are either found in the solid or liquid phase in the flue gas or formed by condensation when diluted with ambient air.

    1.2 Volatile organic compounds (VOC) in the flue gas act as precursors for secondary organic aerosol (SOA) in the atmosphere [29].

2. **Particles resulting from non-carbonaceous fuel constituents:**

    2.1 Ash constituents (K, Na, Ca, Zn, Cl, S, Mn, Mg, P and others) lead to inorganic particles (salts like KCl and oxygen containing compounds such as K_2SO_4, $CaCO_3$, CaO). In addition, and with increased concentrations in waste wood, heavy metals in the fuel result in emissions of Cu, Pb, and Cr and in low concentrations of Cd.

    2.2 Fuel nitrogen forms nitric oxide emissions (NO and NO_2 summarised as NO_X), which can contribute to secondary inorganic particles due to the formation of ammonium (NH_4^+) and nitrates (NO_3^-). For wood combustion, the potential impact of these products is, however, of lesser importance compared to both, PIC and also the before mentioned products from ash constituents.

As shown in Figure 1, different particle types can be assigned to different combustion regimes. The particle composition from different combustion conditions can vary widely as illustrated for various examples in Figure 2. These can occur in biomass combustion at different oxygen availability in the flame and are influenced by the air-fuel equivalence ratio or excess air ratio λ (lambda)[2]. Beside the influence of λ, different combustion phases occur in a batch-wise operated combustion, which can also strongly influence the combustion conditions. This is of specific importance for manually operated combustion devices, where a start-up phase from cold conditions (cold-start), a stationary phase at high temperature (also denoted as flaming phase), and a (char) burn-out phase can be distinguished. Further, conditions of warm-start can occur in case of fuel reload at operation temperature. For automated combustion systems, the different combustion phases are also of importance, since increased emissions often occur during start-up and during burn-out (or shut-down in automated devices). With respect to emissions, however, start-up and burn-out regimes need to be distinguished. While start-up

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[2] The air-fuel equivalence ratio (also denoted as air-fuel ratio or excess air ratio) λ is the ratio of the actual air-fuel ratio to the stoichiometric air-fuel ratio. $\lambda = 1$ describes stoichiometric conditions, $\lambda < 1$ air deficiency (or fuel-rich conditions), and $\lambda > 1.0$ air excess (or fuel-lean conditions).

at low temperatures most commonly result in increased emissions of carbon monoxide (CO) and organic pollutants from biomass pyrolysis, burn-out of charcoal causes high CO concentrations, however, may result in low organic components if only charcoal is available during burn-out [30].

While PIC can be avoided by a near-complete combustion, particles from ash constituents can be lowered to a certain degree by reducing the evaporation and the entrainment of ash constituents (by low temperatures and low gas velocities in the fuel bed). Beside such primary measures particle removal from the flue gas can be an effective measure for automated combustion [31]. Particle removal in residential stoves and boilers is under development [32]. Concerning NO_X, its contribution to near-ground ozone is commonly more critical than its contribution to secondary inorganic aerosol. NO_X reduction, however, can be achieved by staged combustion as primary measure, or by secondary measures with the use of a reducing agent either at high temperature in a SNCR process or by a catalytic conversion in SCR technologies.
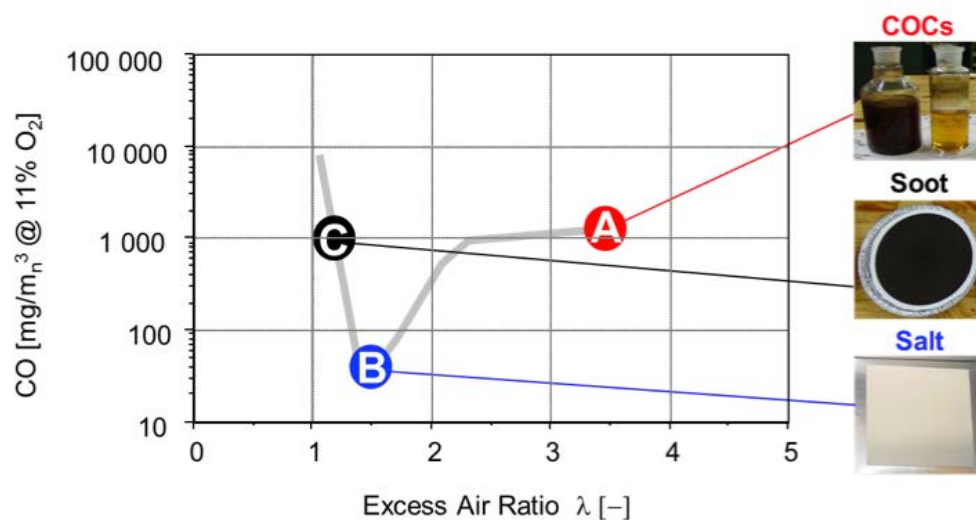

Figure 1   Carbon monoxide in the flue gas as function of the excess air ratio (λ) during biomass combustion for a downdraught log wood boiler [33] For such conditions, three different combustion regimes can often be distinguished. Regime A: Low temperature, e.g. occurring during cold start-up resulting in high COC concentrations. Regime B: Appropriate operation with optimum conditions resulting in a near-complete combustion with low PIC but relevant concentrations of mainly salts found in the particle phase. Regime C: Lack of oxygen at elevated temperature in the flame e.g. due to inappropriate operation by throttled air inlet causing high concentrations of soot.
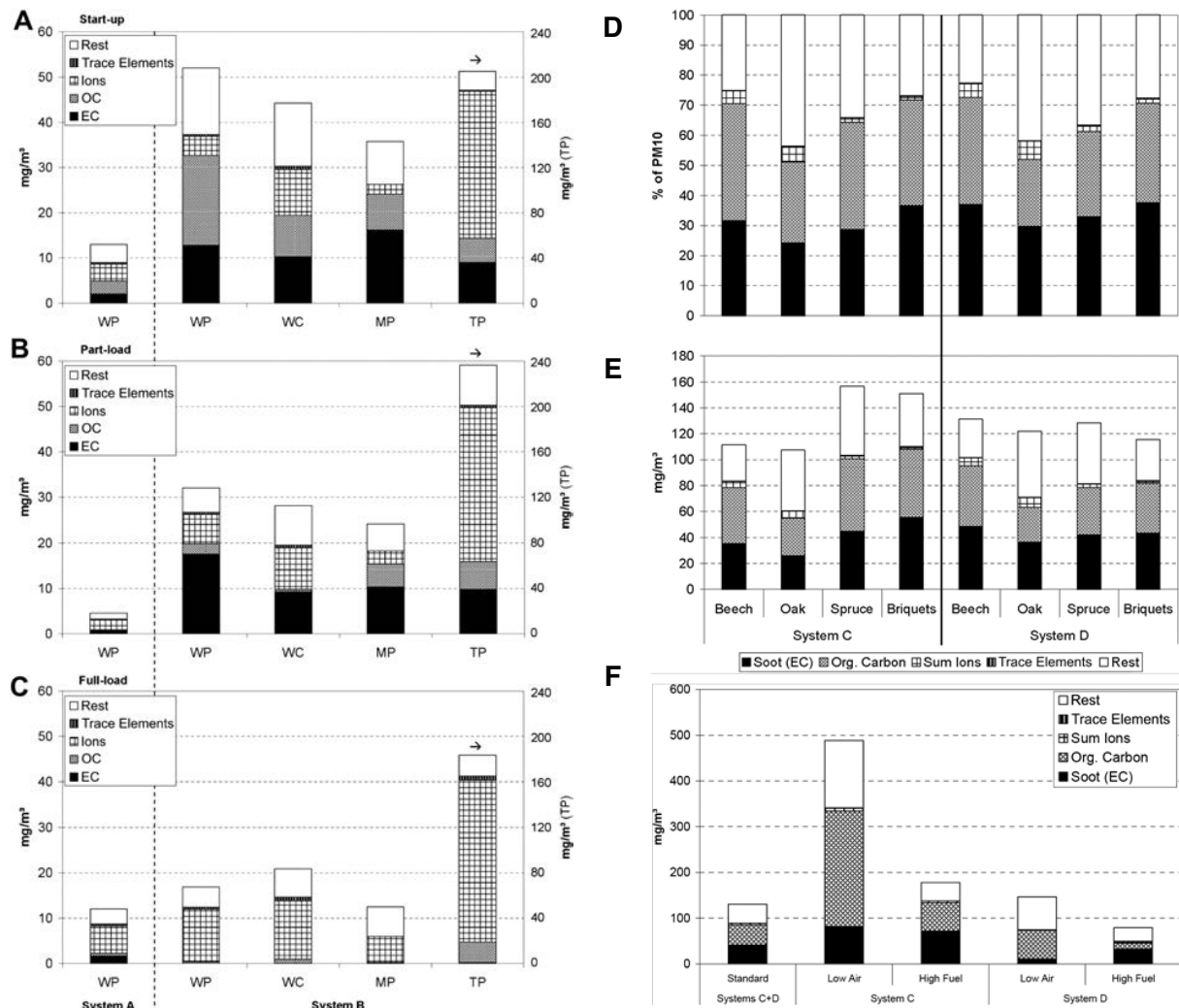
Figure 2   Particulate emissions and their composition from different biomass combustion conditions. Panels A-D: Automated systems A (pellet stove) and B (biomass boiler) and manual systems C (log wood stove) and D (advanced log wood stove). Panel F shows the comparison of standard test and mal-operation PM_{10} composition of manually operated systems C and D. [34] (adopted).
WP wood pellets, WC wood chips, TP triticale pellets, MP miscanthus pellets.

The main formation pathways of PM from biomass combustion are summarised in Figure 3. For the inorganic fraction of particles from ash constituents, evaporation at high temperatures can cause fine salt particles via the solid-vapour-particle-path. Entrainment at high gas velocities in the fuel bed can additionally contribute to coarse particles by the solid-particle-path. Considering carbonaceous particles, local temperature, oxygen and residence time in different combustion zones strongly influence the carbon conversion to CO_2, CO, VOC, COC, soot (including PAH which may be adsorbed on the particle surfaces), and char. For modern combustion devices, primary air is usually injected for the solid fuel conversion in a first combustion zone, while secondary air is added upstream for the oxidation of combustible components in a consecutive combustion chamber as shown in Figure 3. When released to the atmosphere, consecutive secondary reactions can occur, which may form additional aerosol, in particular SOA from VOC and inorganic salts from NO_X. For combustion conditions with high VOC emissions SOA can even exceed primary emissions, as shown in Figure 4.

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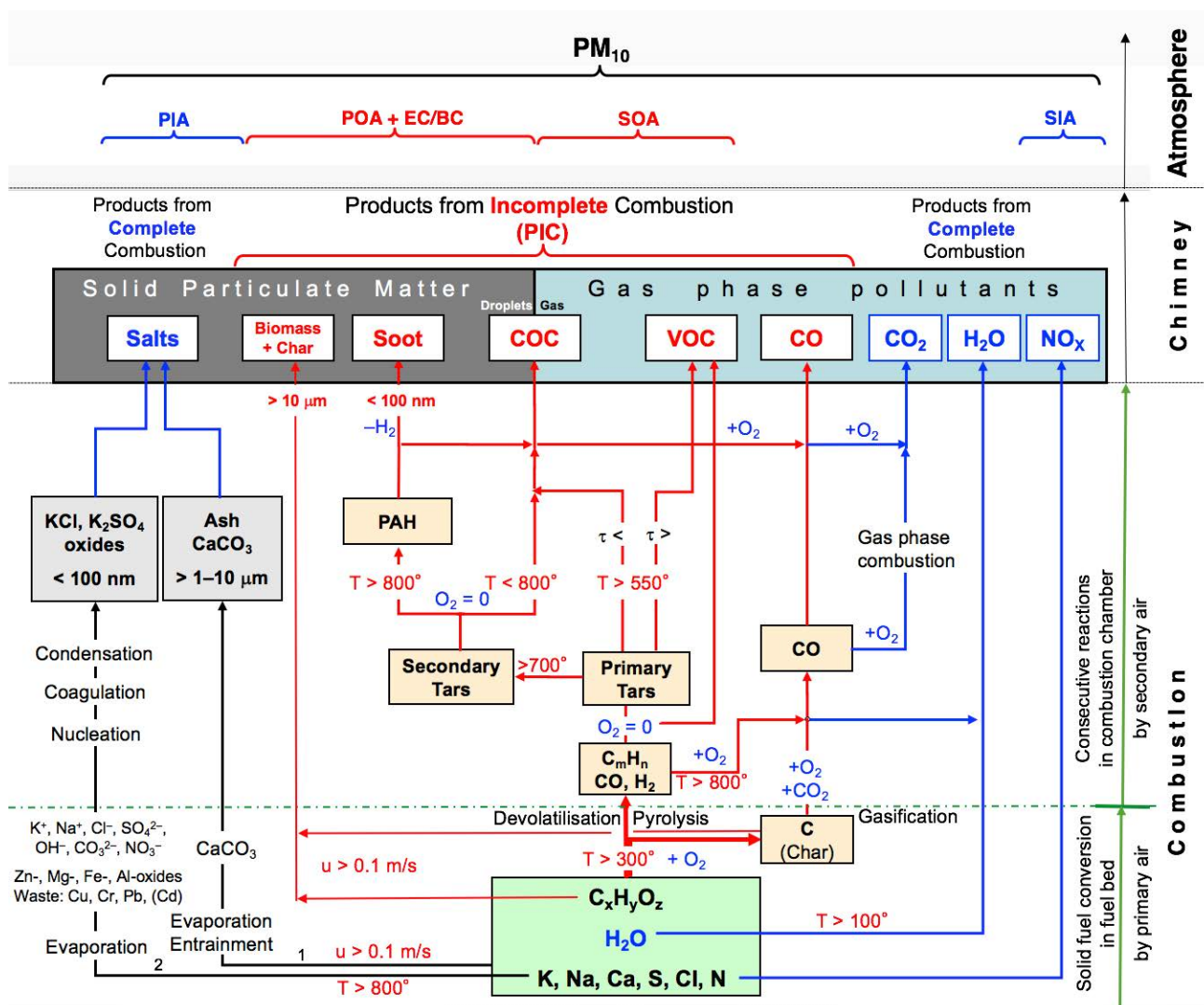Figure 3   Particle formation mechanisms in biomass combustion and consecutive secondary aerosol formation in the atmosphere. u Gas velocity, τ Residence time (< denoting short residence time, > denoting long residence time). 1 Solid-particle-path, 2 Solid-vapour-particle-path.
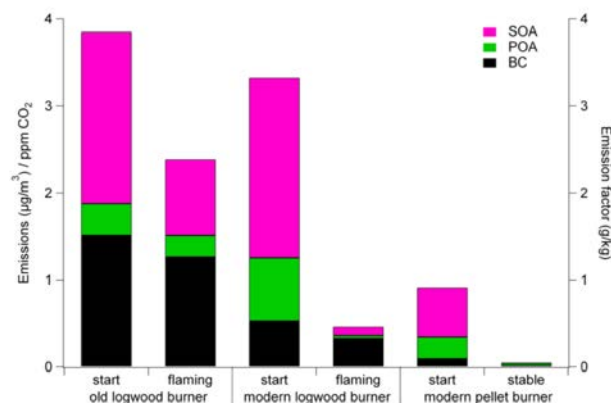

Figure 4   Average concentrations of BC, POA and SOA after 5 h of aging in a smog chamber for three combustion devices [35].

2.2   Particle size of primary particles

For most combustion conditions, the particles in the size range between 50 nm and 200 nm are dominant (e.g. [23], [36], [37], [40], and Figure 5, Figure 6 to Figure 8). At near-complete combustion, wood with low ash content exhibits a unimodal size distribution with nanoparticles smaller than 100 nm contributing up to 90 % of the particle mass. The particle size distribution and the particle mass concentration are, however, strongly influenced by the fuel, the combustion regime, and the combustion type as illustrated in Figure 5 to Figure 8:

- Particles from K, S, Cl, Zn, and Na are mainly found in the size range around 100 nm and commonly lead to one peak for low ash wood fuels, which in case of low particle concentrations can also dominate the total particle mass. Ca, however, as less volatile compound tends to form coarse particles with a potential second peak with particle diameters greater than 5 micrometres. The same is true for Mn, Mg, and P, however at lower concentrations. Due to high ash content including Ca, bark can cause a second peak in the micrometre range thus leading to a bimodal size distribution as e.g. shown in Figure 7. The contribution of the main compounds in the case of a pellet stove is shown in Figure 5: K is the dominant source of primary particles followed by S, Cl, Zn, and Na in the micrometre range, while Ca is dominant in 10 micrometre particles.

- Start-up often leads to increased emissions thus resulting in an increase of the particle mass concentration and the mean particle diameter as shown in Figure 6.

- An incomplete combustion, as commonly experienced in simple devices or caused by mal-operation leads to an increase in the size and the total mass of particles due to unburnt products as shown in Figure 8. Due to the particle growth at increased concentrations, the particle number concentration may reach to a maximum or is at least significantly less pronounced than the increase of the particle mass.

- Incomplete combustion can also cause COC exceeding soot and other primary solid particles by up to one order of magnitude. This is most relevant for residential combustion and it has been demonstrated by flue gas quenching in liquid sampling [40] or dilution with cold air ([41], Figure 9).

Due to the different formation mechanisms, not only the particle mass, but also the fraction of soot and COC can vary in a wide range. Therefore, to estimate the contribution of biomass to ambient PM, solid particles and COC need to be considered and potential SOA formation from VOC needs to be additionally accounted for.
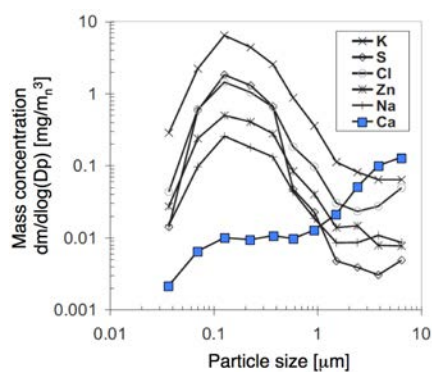
Figure 5   Particle mass concentration as function of the particle size from a pellet stove measured by a low-pressure impactor [23].
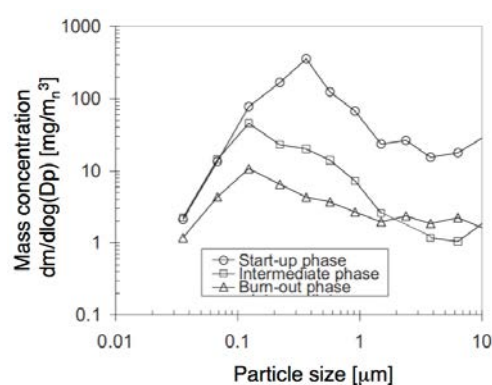
Figure 6   Particle mass concentration as function of the particle size from a wood stove during start, intermediate, and burn-out phase [23].
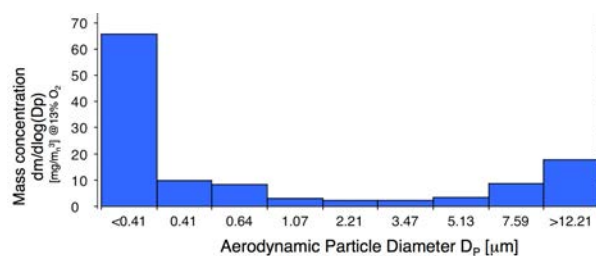

Figure 7   Particle mass concentration as function of the particle size from wood stoves measured by a cascade impactor [36].
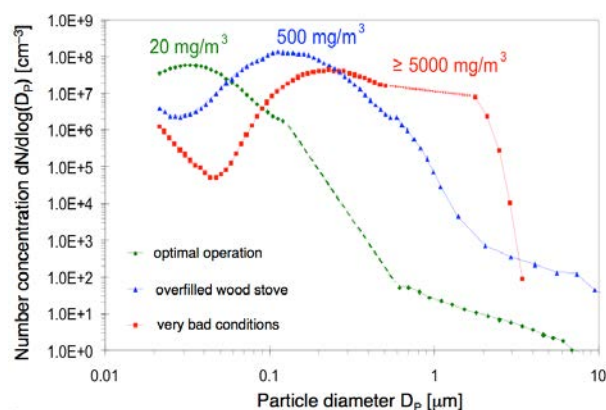

Figure 8   Particle number concentration as function of the particle size from wood stoves measured by a Scanning Mobility Particle Sizer (SMPS) and Optical Particle Counter (OPC) [40].
Optimal operation: well-designed wood stove properly operated with dry logs. Overfilled wood stove: wood stove with combustion chamber filled with more than 50 %, which often occurs in real life operation. Very bad operation: air throttling after start-up phase causing very high emissions.
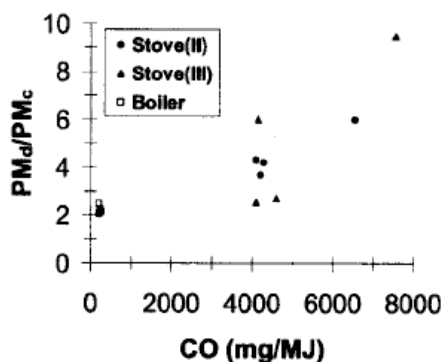

Figure 9   Ratio of particle mass concentration in diluted flue gas and in chimney (PM_d/PM_c) as function of the CO content in the flue gas [41].

2.3   Carbon monoxide as indicator for the combustion quality

Due to the complex formation mechanisms of different pollutants described in Figure 3, specific patterns of pollutants can occur at different combustion conditions. Nevertheless, carbon monoxide is often used as a basic indicator for the burn-out quality of the flue gases. For many combustion conditions, there exists a correlation between volatile organic pollutants and CO. Figure 10 displays a relationship between CO and NMVOC for wood pellets, bark pellets, wood logs and wood briquettes, while Figure 11 shows the correlation between COC and CO in a log wood boiler at various combustion conditions. These ratios may vary for different combustion systems. In addition, the ratio can decline for a batch operated combustion in the burn-out phase, when charcoal is remaining while organic matter from the initial biomass is not available any more as it has been converted during the flaming phase. As shown in Figure 12, under certain conditions, CO can also be used as an indicator for PM in the flue gas. This, however, does not necessarily apply to automated boilers, where mainly inorganic PM is found in the flue gas, which can even increase at low CO concentration. Although the described correlations are useful under common conditions, measuring CO only cannot replace detailed information on other pollutants.
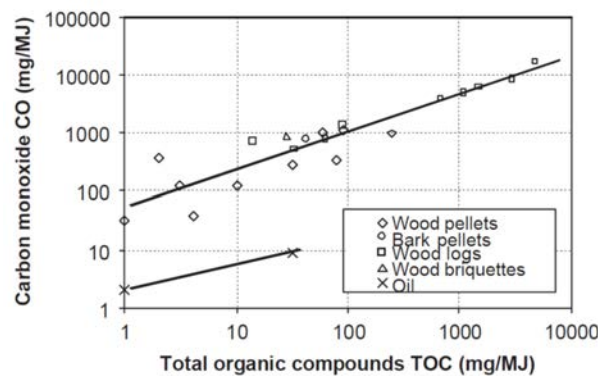

Figure 10   Carbon monoxide versus total organic compounds (methane-equivalents, measured with flame ionization detection) emissions for two old and three modern wood boilers, three pellet burners, one pellet boiler and two oil burners [42].
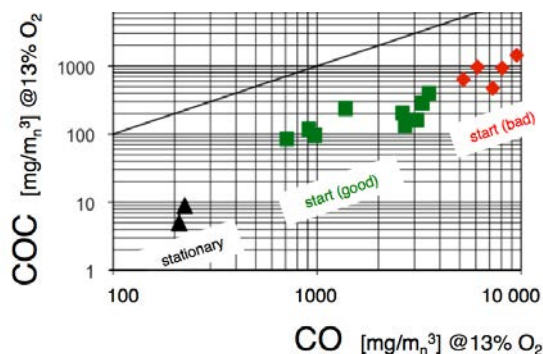

Figure 11   Condensable Organic Compounds (COC) as function of carbon monoxide during stationary combustion and during good and bad start-up in a downdraught log wood boiler with two-stage combustion [24].
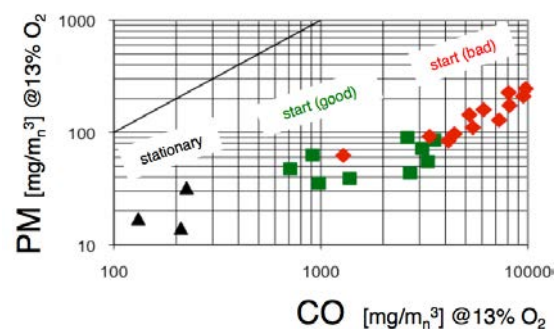
Figure 12   Solid Particulate Matter PM collected by hot filter sampling in the flue gas as function of carbon monoxide during stationary combustion and during good and bad start-up in a downdraught log wood boiler with two-stage combustion [24].

2.4 Technologies for biomass combustion

When accounting for local air pollution, non-technical applications such as natural fires, open field burning, bonfires, barbecue, or simple cooking applications with biomass can be the dominant source of carbonaceous PM in the ambient air [43]. Beside such sources, however, manual combustion devices can be an important source of PM emissions. Traditional manual combustion devices such as old-type wood stoves based on an up-draught principle often exhibit one single air inlet to the combustion zone and are operated with natural draught ([8], [30]). Numerous investigations reveal that this type of combustion, referred to as one-stage combustion, can cause significant emissions of unburnt pollutants, hence the content of carbonaceous compounds in primary PM can exceed 50 wt.%. Such combustion devices contribute significantly to BC, POA and SOA in the atmosphere. In addition, the total pollutant emissions strongly depend on the operation mode and the fuel type. While relatively moderate emissions of 10–50 mg/MJ of solid primary PM are attainable at appropriate combustion conditions and fuel (i.e., dry log wood), inappropriate operation can lead to more than 10 times higher solid PM in the flue gas thus significantly exceeding 100 mg/MJ [38]. This is true for traditional start-up with ignition of the fuel from the bottom or it can be a result of insufficient combustion air or use of wet wood and other types of mal-operation. In traditional stoves, also an undesirable heat extraction from the combustion zone can result in low combustion temperatures and high POA and VOC. Beside solid primary PM, VOC are emitted with a typical share of 40 % methane and 60 % non-methane VOC (NMVOC) [38]. NMVOC can exceed primary PM ([39], [40]). PM measured in diluted, cold flue gas can exceed PM sampled on hot filters by a factor of two to ten ([41] and Figure 13, [25]). In manually operated up-draught combustion devices, the start-up phase often causes increased PIC (e.g. Figure 14, [26]). To reduce PIC from wood stoves, ignition from the top can be applied [22]. Nevertheless, the use of appropriate fuel, i.e. dry wood logs, appropriate log size (Figure 15, [27]) is essential for such combustion devices [26]. Furthermore, overfilling and air throttling need to be avoided.

To further reduce pollutants from incomplete combustion in manual devices, advanced two-stage combustion has been developed for log wood boilers. In these devices, primary air is injected to the solid fuel zone to act as gasification agent. In a consecutive step, the combustible gases released from the fuel bed are mixed with secondary air to enable a near-complete burnout in a post-combustion chamber. To enable a clear separation of solid fuel conversion and consecutive gas phase combustion, a down-draught combustion is applied. Since an immediate heat extraction is avoided, combustion temperatures up to more than 850 °C are achieved, enabling low VOC, soot and POA concentrations. To ensure a near-complete combustion, mixing of combustible gases with combustion air is improved by aerodynamic measures and by means of forced draught enabling increased pressure drops. Further, electronic control of the combustion air can be applied.

Two-stage combustion is also applied in automated boilers for pellets and bulk solid fuels such as forestry wood chips. Thanks to high temperature and near-complete combustion, BC, POA and VOC are further reduced to emission levels of typically well below 5 mg/MJ. For such combustion conditions, high temperatures in the solid fuel conversion zone enhance the formation of particulate matter from inorganic fuel constituents. Consequently, automated wood combustion in the size range of 500 kW to 5 MW applied in under-stoker and moving grate boilers and greater 5 MW in fluidised bed boilers reveal typical PM in the raw flue gas of more than 100 mg/MJ, however with a carbon content of less than 5 wt%. Consequently, particle separation by electrostatic precipitation (ESP) or by fabric filter is commonly applied enabling particle emissions to the ambient of less than 5 mg/MJ.

Birch & spruce: Part. on HF and ambient filter vs burnrate
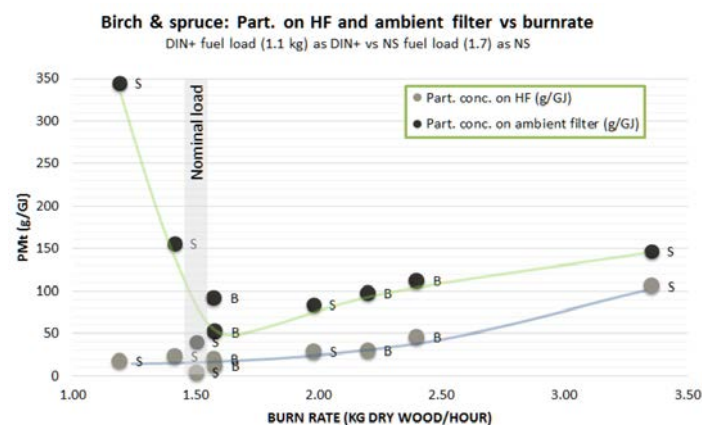DIN+ fuel load (1.1 kg) as DIN+ vs NS fuel load (1.7) as NS

Figure 13 PM emissions as function of burn rate in a wood stove and comparison between PM measured on hot filter (HF) and ambient filter thus also trapping condensable compounds [25].
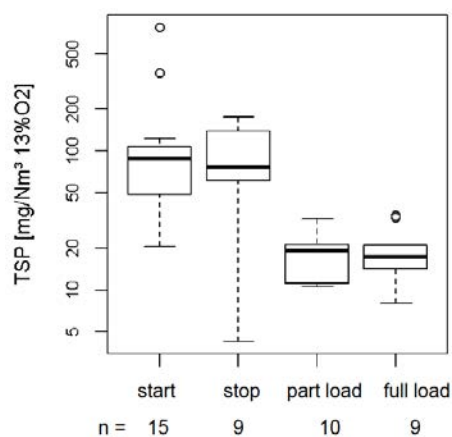

Figure 14 Influence of combustion phase on total suspended particles (TSP) [26].
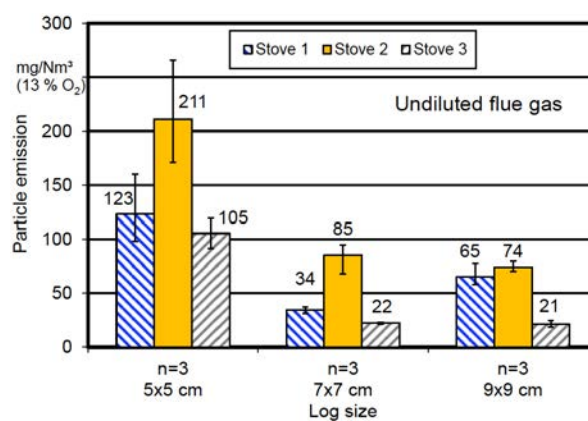

Figure 15 Influence of log size on particle emissions in three wood stoves [27].

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3 Health relevance of combustion aerosols

Many studies show strong evidence that airborne particles are related to adverse health effects including increased mortality [44]. Especially, submicron particles (PM_1) are insufficiently filtered by nose and bronchia and consequently can penetrate into the lung inducing several negative effects (Figure 16). Since biomass combustion exhibits a high contribution to air pollution (in the cold season it can be the dominating source of PM_{10} [45]) and combustion particles are generally regarded as potentially harmful, several studies investigated potential health effects of biomass combustion emissions using the following methods:

1. Epidemiologic studies (humans only)

2. *In vivo* exposure studies (on animals and on humans)

3. *In vitro* exposure studies (on animal and human lung cells).

The three methods have different limitations and advantages as described in Table 1.

Table 1   Advantages and limitations of experimental models used in studies on health impacts from wood combustion emissions.

Method	Advantages	Limitations
Epidemiology	- Realistic exposure and effects - Long term exposure - Effect of SOA is captured	- The causality is limited and effects cannot be confidently related to one single event - Exposure is not highly specific and hard to quantify - Effects not necessarily caused by exposure
In vitro	- Exposure can cover large ranges of concentrations and duration - Well-controlled environment and reproducibility	- Exposure may not be realistic - Not representative of entire body - When animal cells were used, results may not be applicable to humans - Sampling and collecting PM may alter its particle characteristics - Investigation of SOA is difficult
In vivo	- Strong link between effects and exposure - Realistic air to lung exposure is possible	- Exposure duration and concentration are limited - Test subjects are highly variable - Investigation of SOA is difficult - If animals are tested, results may not be applicable to humans

Nevertheless, *in vivo* and in *in vitro* tests show, that biomass combustion particles have adverse health effects with evidence of causing cytotoxicity, oxidative stress, inflammation, physiological effects, genotoxicity, and carcinogenicity [19], [46]–[58]. In addition, a dose-response effect for particle exposure exists except in cases of overloaded biological systems where any concentration yields the maximum effect (e.g. all cells are dead). Epidemiological studies also found evidence for adverse health effects from biomass combustion PM, however, specific health effects remain difficult to identify by this method [47]. It was shown that combustion type and condition is an important factor influencing specific health effects. For example, incomplete combustion or low oxygen combustion in simple combustion devices exhibits a stronger impact on cell toxicity and cell functions than particles from near-complete combustion (Figure 17 [50]). Moreover, biomass combustion flue gases contain reactive

oxygen species (ROS), which are responsible for oxidative stress and therefore are used as markers for potential health effects [59]. The ROS content strongly depends on combustion temperature and consequently varies for different combustion devices and conditions. Moreover, an investigation with exposure of filter extracts and COC samples to cells reveals that PM from different biomass combustion devices and conditions shows decreasing cytotoxicity (Figure 17) and carcinogenicity in the following order [54]: COC, soot, and salt particles. Consequently, the negative health effects of wood combustion emissions can exceed the ones of traffic emissions as confirmed in another investigation in Figure 18 [49].

Other investigations (Figure 19) where cells where exposed to particle filter extracts reveal that old or manual combustion devices can induce higher cytotoxicity and carcinogenicity compared to new or automatic devices [51], [52]. Figure 20 shows results from cytotoxicity tests in sampling medium from different combustion conditions. The cytotoxicity indicated as the cell viability as function of the flue gas volume (left graph) indicates that automated combustion exhibits negligible toxicity. The pellet boiler at optimum conditions also causes very low toxicity. At non-ideal conditions as e.g. high excess of combustion air, the toxicity increases significantly. For the wood stove, the warm start caused about ten times higher cytotoxicity per flue gas volume (which is proportional to the energy input) compared to the flaming phase. The graph on the right shows the same results, however as function of the total organic content (TOC) found in the sample. This illustration indicates that the sampled particles exhibit comparable toxicity except for the automated boiler, where no significant toxicity is found. In addition, TOC can presumably be seen as a relevant indicator for the cell toxicity for the investigated samples. Similar results were found in [53] where the reduction of two pro-inflammatory cytokines exhibit a significant positive linear correlation with increasing amount of organic carbon as shown in Figure 21.
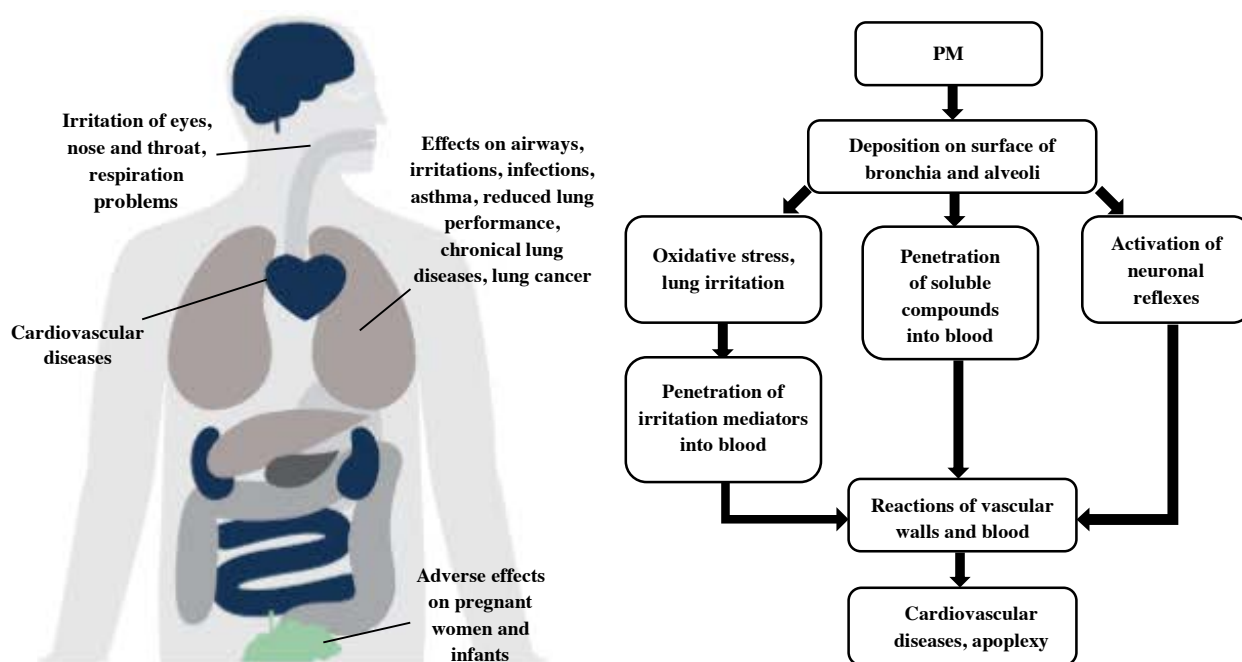

Figure 16   Adverse health effects of PM (left panel) and mechanisms of noxious effects of PM inside the human body ([48], adapted).
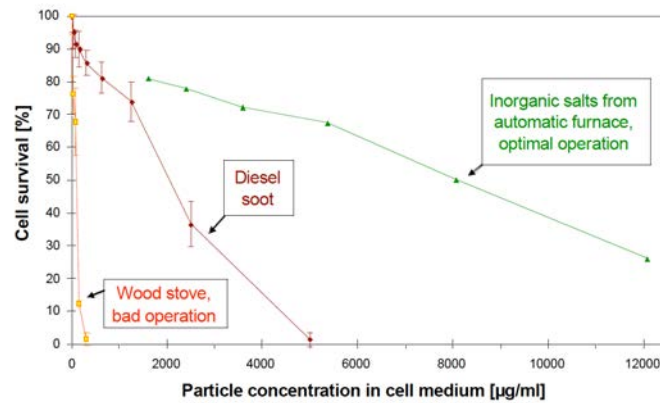
Figure 17   Cell survival rate as function of the particle concentration in the cell medium for Diesel soot, mainly inorganic particles from automated wood combustion, and particles from incomplete wood combustion in a badly operated stove [40].
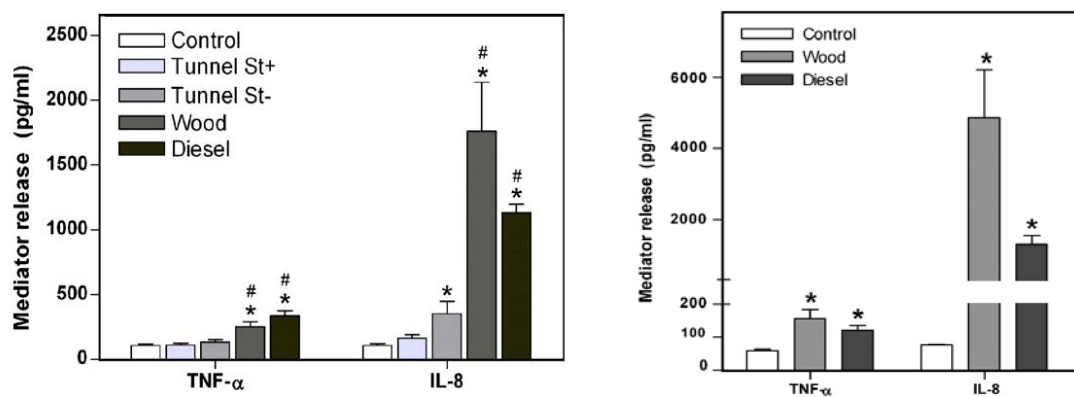

Figure 18   The release of TNF-α and IL-8 after 12 h exposure to equal concentrations of organic extracts (50 μg/well, equivalent to 30 μg/ml) (left panel) and of washed particles (140 μg/ml). Bars represent mean± S.E.M. of six experiments. Asterisks indicate significant increase compared to control, # indicates significantly higher release compared to Tunnel St+ and St– [49].
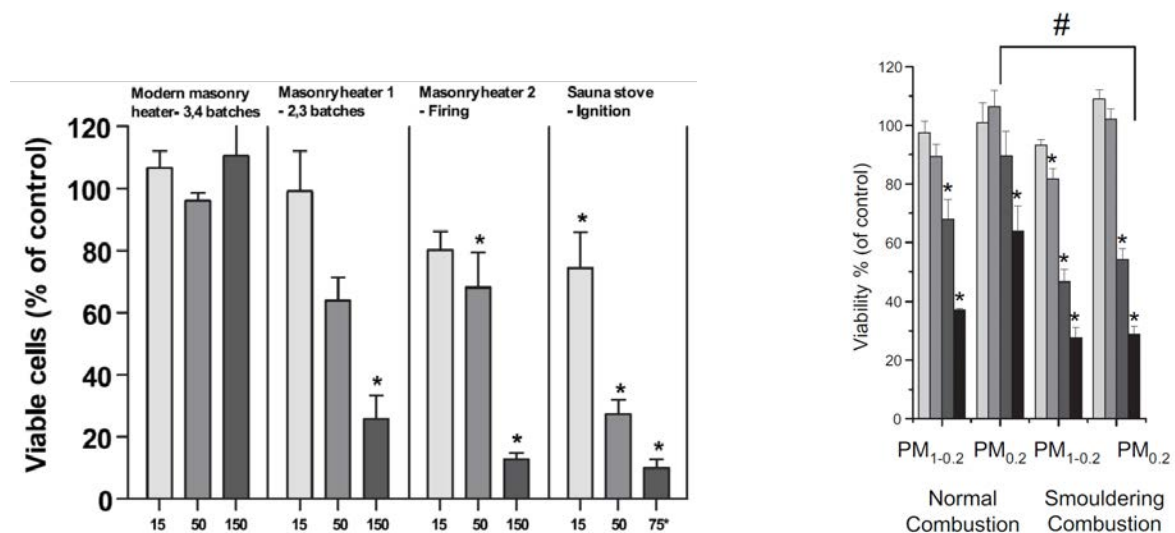
Figure 19   Left: Viability of human BEAS-2B bronchial epithelial cells after exposure to three doses (15, 50 and 150 μg/ml) of particles from four log wood combustion devices [52].

Right: Viability of RAW 264.7 macrophages after exposure to four doses (15, 50, 150 and 300 μg/ml) of $PM_{1-0.2}$ and $PM_{0.2}$ samples from normal and smoldering combustion [50]. Asterisk indicates statistically significant response compared to control cells. Statistical difference between each concentration of the samples in two combustion conditions is indicated with #.

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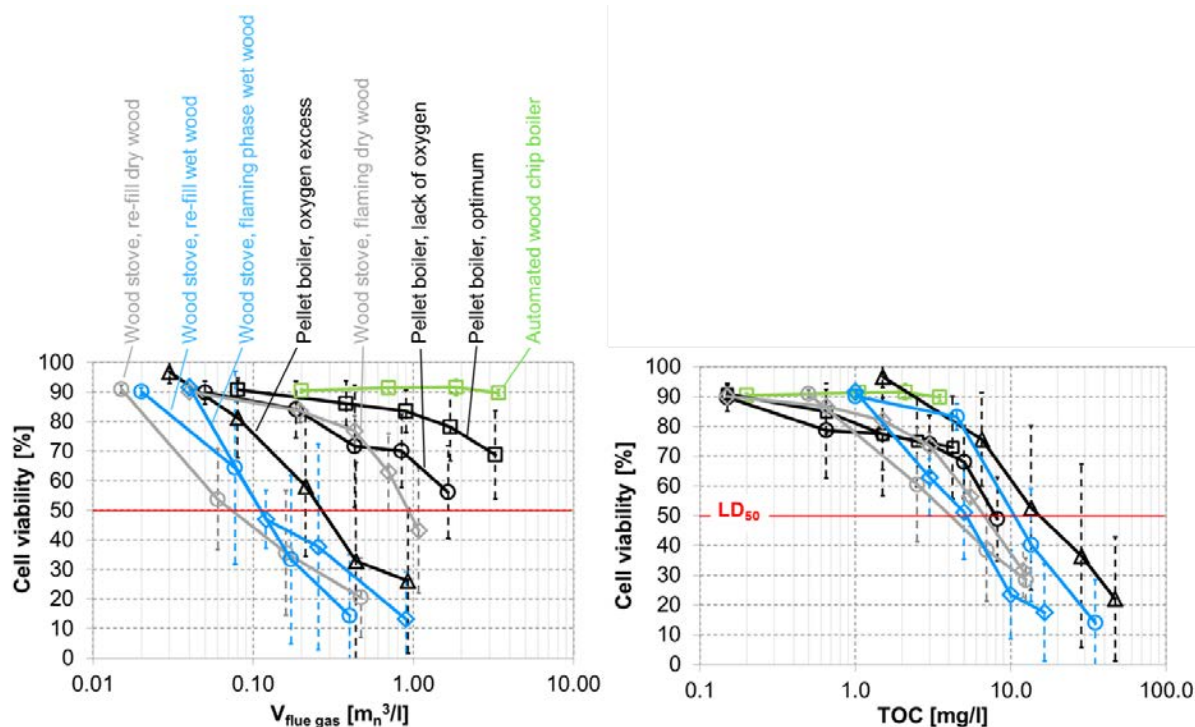
Figure 20   Cell viability for different wood combustion conditions as function of the specific flue gas volume at norm conditions (n) per litre sampling medium (left) and as function of TOC in sampling medium (right) [56].
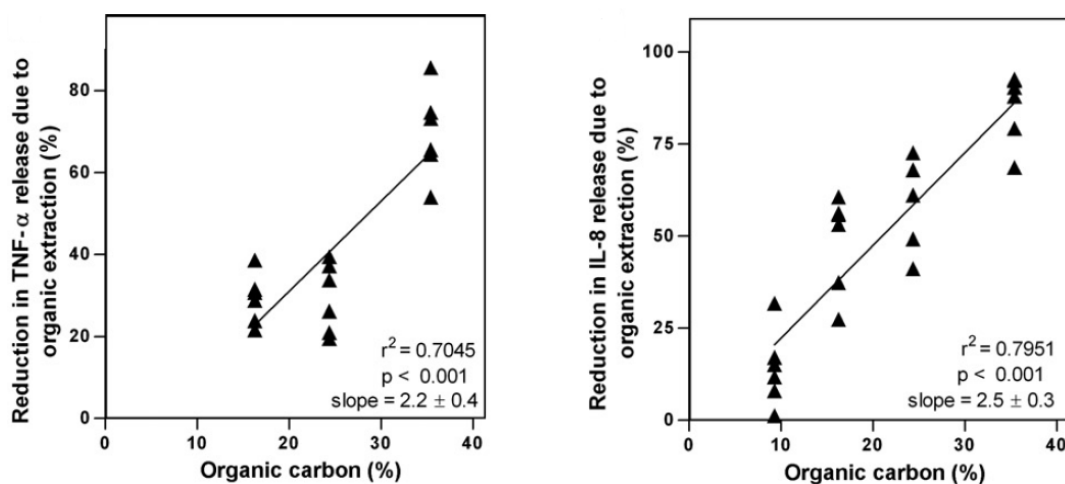
Figure 21   Reduction of pro-inflammatory cytokines TNF-α (tumour necrosis factor) and interleukin 8 (IL-8) of the monocytic cell line THP-1 as a function of the organic content of extracts of tunnel, diesel and wood smoke particles [53].

4   Conclusions

- Particulate Matter (PM) from combustion processes in ambient air causes adverse health effects.

- Biomass combustion contributes to ambient PM with three main types of particles found in the flue gases, i.e. carbonaceous solid material denoted as soot, condensable organic compounds denoted as tar, and inorganic particles from ash constituents mainly found as salts.

- In addition to this primary aerosol, secondary aerosol can be formed in the atmosphere, in particular secondary organic aerosol (SOA) formed from volatile organic compounds (VOC).

- Organic particles and condensables as found from manual combustion under non-ideal conditions exhibit a high cell toxicity, while inorganic particles as found in flue gases from properly operated automated biomass combustion systems reveal far lower or even undetectable cell toxicity under the investigated conditions.

- The contribution to organic aerosol is strongly influenced by the combustion conditions. Low organic emissions can be achieved at near-complete combustion in well designed and properly operated combustion systems. To guarantee such conditions, automated combustion is advantageous. For manually operated devices, an ideal start-up is crucial beside other needs such as the use of dry wood in reasonable size and avoiding air throttling. In case of unsuited fuel or inappropriate operation, high emissions of primary organic aerosol and precursors of SOA can result due to incomplete combustion.

- In case of near-complete combustion, inorganic particles can be found in the raw flue gas in significant concentrations in particular for automated biomass combustion systems. This type of particles can, however, be effectively separated in particle precipitators.

- Consequently, biomass combustion can reveal a high impact on the local ambient air quality in case of inappropriate use. On the other hand, properly operated state-of-the art combustion systems exhibit a low environmental impact and enable to effectively substitute fossil fuels and to mitigate the atmospheric CO_2 increase. Beside the use of modern technologies, a proper plant operation and a stringent enforcement of emissions regulations need to be ascertained.

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