

Characterisation of particles from wood combustion with respect to health relevance and electrostatic precipitation

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Biomass combustion is a relevant source of inhalable primary aerosols in the size range smaller than 10 microns (Particulate Matter PM₁₀) formed in basically three different pathways:

1. An incomplete combustion at low temperature can lead to Condensable Organic Compounds (COC) as pyrolysis products and also called tar, which form primary organic aerosols and contribute to brown carbon (C_{brown}) in the ambient air.
2. An incomplete combustion at high flame temperature but with local lack of oxygen or flame quenching can lead to soot formation thus resulting in nearly Elemental Carbon (EC) finally contributing to Black Carbon (BC) in the ambient.
3. Mineral matter including alkali metals and chlorine in the fuel leads to the formation of inorganic fly ash particles mainly consisting of salts such as chlorides and oxides. In addition, other metals may also be available in certain concentrations.

Beside primary aerosols, biomass combustion also contributes to Secondary Organic Aerosols (SOA) and Secondary Inorganic Aerosols (SIA). Volatile Organic Compounds (VOC) resulting from incomplete combustion act as precursors for SOA, while NO_x and SO_x from nitrogen and sulphur in the fuel act as precursors for nitrates and sulfates. Recent findings in the ambient air show that organic matter in PM₁₀ results dominantly from SOA with a minor contribution of POA. The three different particle classes exhibit completely different chemical and physical properties. Consequently, health effects as well as behaviour of aerosols from biomass combustion in flue gas cleaning systems are expected to vary significantly depending on fuel type and combustion regime. Therefore the health effects and the electrical properties of different combustion particles have been investigated.

Diesel soot, soot and COC from incomplete combustion of wood, and mainly inorganic particles from a nearly complete combustion of wood have been compared. The samples were used for biological tests on cell toxicity and on chromosome defects with lung cells from the chinese hamster. In addition, Polycyclic Aromatic Hydrocarbons (PAH) were analysed.

The results show, that Diesel soot exhibits a medium level of toxicity and chromosome defects, while particles from the automatic wood combustion exhibits more than 5 times lower toxicity. However, particles from the badly operated wood stove exhibit more than 10 times higher toxicity and accordingly also levels of PAH than Diesel soot with the onset of chromosome defects also found at more than 10 times lower particle concentration in the cell medium. Particles from well operated wood stoves showed similar or slightly less toxicity than Diesel soot.

The particles from biomass combustion are collected in a laboratory electrostatic precipitator (ESP). Three different combustion regimes are maintained by a modified pellet boiler, i.e., high temperature and sufficient oxygen, high temperature and local lack of oxygen, and low temperature. The resulting particles are classified as salts, soot, and Condensable Organic Compounds (COC) based on the particle type expected from the theory of particle formation. The chemical and electrical properties were analysed and confirm the classification:

While salts exhibit a low carbon content, soot and COC are high in carbon. Soot and COC can be distinguished by significantly different molar C/H-ratio being 6.44 for soot and 1.24 for COC. The electrical conductivity, which is a key parameter for the precipitation and dust layer build-up in the ESP, is measured at different temperatures and humidities. Significant differences in conductivity are found for salts, soot, and COC, and in addition, a strong influence of the humidity of the flue gas is observed. Salt is confirmed to be ideal for ESP, while soot reveals high conductivity leading to re-entrainment of agglomerated particles, and COC exhibit low conductivity leading to back-corona which can be limiting at low humidity. The presented particle properties can be applied as guideline for ESP design and operation.

Acknowledgments: Swiss Federal Office of Energy and Federal Office for the Environment

1. Particles from biomass combustion

1.1 Relevance of Particulate Matter (PM) in the ambient air

Particulate matter smaller than 10 microns (PM₁₀) is associated with diverse health effects and regarded as one of the most relevant parameters in air pollution [1, 2]. Limit values for both, particle emissions and immissions are indicated as mass concentrations and hence do not distinguish between strongly toxic and less harmful substances. However, combustion particles are generally regarded as potentially harmful since they may consist of or be carrier of toxic and carcinogenic substances such as polycyclic aromatic hydrocarbons (PAH). In addition, the particles are often found in the size range smaller than 1 micron and often even smaller than 0.1 microns. These type of nano-particles are most relevant due to its large specific surface and the potentially increased mobility.

1.2 Particle types from fuel constituents and from incomplete combustion

Biomass combustion is related to three basic types of primary particles, which are summarized as 'salts', 'soot', and Condensable Organic Compounds 'COC', and exhibit completely different chemical and physical properties:

- COC are formed directly from biomass pyrolysis and in subsequent reactions in two pathways:
 - At low temperature volatile or condensed organic compounds are formed from wood pyrolysis with characteristic compounds and properties depending on residence time, heating rate, temperature and other operation parameters.
 - At moderate temperatures and local lack of oxygen, organic compounds can be converted to secondary and tertiary tars including polycyclic aromatic hydrocarbons (PAH), which can appear as condensables.
- Soot is formed from organic precursors in zones of high temperatures and lack of oxygen, where volatiles and primary tars react to secondary and tertiary tars and form PAH, which consequently can form soot particles by further synthesis reaction and agglomeration with release of hydrogen.
- Inorganic particles, basically salts, are formed from minerals (i.e., ash constituents) in the fuel. These particles are dominant at near-complete combustion and thus can only be partially avoided by fuel conversion in the primary zone at low temperature and low oxygen content (low-particle combustion, [3, 4])

The formation mechanisms are described in Figure 1. The combustion conditions and the resulting particles strongly depend on fuel type, combustion technology and type of operation [5, 6, 7]. In automatic wood combustion, nearly complete combustion can be achieved and hence salts are dominant as particles. However, during start-up, and in phases of inappropriate operation, condensables or soot can also be emitted from automatic plants.

Incomplete combustion is often found in manual wood combustion, whereby soot or condensables can be the dominant part of the total particulate matter released to the atmosphere. Due to the different temperature regimes and the different influence of the residence time for soot and COC formation, usually either one of the two particle type dominates the particle ensemble.

1.3 Conversion of carbon in biomass combustion

Figure 2 shows the main mechanisms during carbon conversion in combustion and subsequent reactions in the atmosphere. Methane acts as greenhouse gas but without major health impacts, while brown carbon resulting from COC and SOA is most relevant for health. Black Carbon (BC) is relevant for both, health and climate.

Due to these different effects, the partitioning of the carbon into different pollutants in the atmosphere is most relevant. Conversion factors are introduced for the partitioning of VOC, NMVOC, and NCNMVOC and described by α , ε , γ , and δ . The shown figures result from measurements in different residential wood combustion devices and are further described in [8], where emission factors for different combustion technologies are summarized.

The emission factors show, that non-methane VOC are most relevant as precursors for PM in the ambient air for residential biomass combustion, as their contribution to PM exceeds the contribution of primary solid aerosols (also called solid particles, SP), although SP include both, soot and inorganic particles. On the other hand, properly operated automatic wood combustion devices exhibit small fractions of carbonaceous emissions and consequently mainly contribute to inorganic PM in the ambient air.

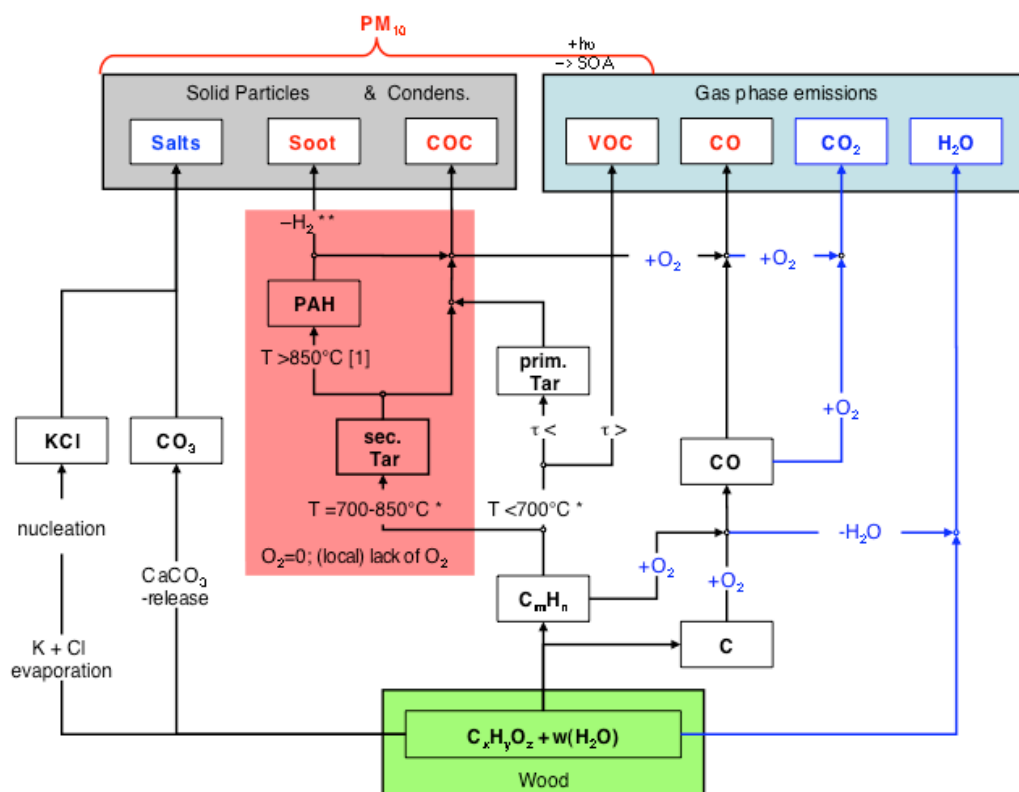


Figure 1: Mechanisms of aerosol formation in biomass combustion.

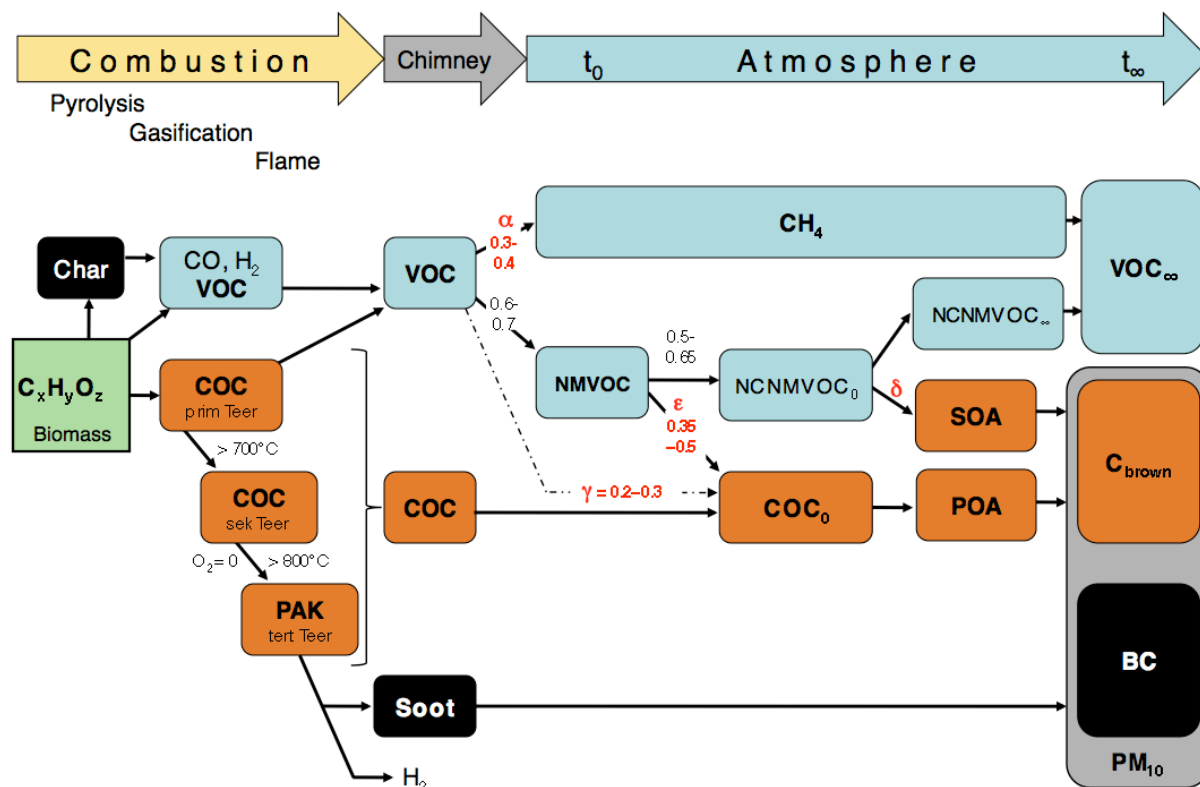


Figure 2: Conversion of carbon during combustion and subsequent reactions in the atmosphere. The compounds in the chimney are measured during emission measurement tests, however, influenced by the sampling.

2. Health effects of particles from biomass combustion

2.1 Method

Particle emissions were monitored by sampling from the hot flue gas on a heated filter probe. Furthermore, the contribution of condensable organic matter in the flue gas sampled by quenching according was assessed under different combustion conditions and compared to the solid particulate matter. In addition to these measurements, the time dependence and size distribution of the particle emissions was monitored in the size range from 20 nm to 10 μm by a combination of a Scanning Mobility Particle Sizer (SMPS) and an Optical Particle Counter (OPC). Furthermore, chemical analyses on organic carbon and polycyclic aromatic hydrocarbons (PAH) of solid particles and condensable matter were performed as described in [9, 10].

The composition of the particles is an important parameter for the interaction with the human body and resulting health effects. The toxicity of the particles was tested by a standard test method with lung cells of the Chinese hamster. This cell line allows also a standard detection of chromosome defects, which are an indicator for the carcinogenic potential of the applied particles. The health relevance was assessed by the cell toxicity and the chromosome aberration described earlier. The present publication presents results on particles from incomplete wood combustion and on the influence of the stove operation. Diesel soot was sampled during stationary operation of a modern Diesel passenger car on a test-bench. Similar sampling was applied for an automatic wood boiler and for wood stoves.

2.2 Results

A comparison of three different particle types, i.e., Diesel soot, mainly inorganic particles from a well operated automatic wood boiler, and particles from a badly operated wood stove, was performed and reveals the following ranking (Fig. 3 and 4):

The particles from automatic wood furnaces exhibit the lowest toxicity which corresponds to the effects found with pure potassium chloride. The chromosome defects of these particles were below the detection limit.

Diesel soot exhibits a medium toxicity (i.e. 5 times higher than inorganic particles in the selected tests) and detectable chromosome defects.

Particles from incomplete combustion of wood in a badly operated stove result in the highest toxicity and chromosome defects, i.e., app. 15 times higher than for Diesel soot. Furthermore, the chemical analyses reveal substantially higher concentrations of PAH, which are assumed to contribute to the increased toxicity. The high carcinogenic potential of wood particles from incomplete combustion is in line with results from investigations which reveal comparable lung cancer pathogenesis from smoking and cooking with wood.

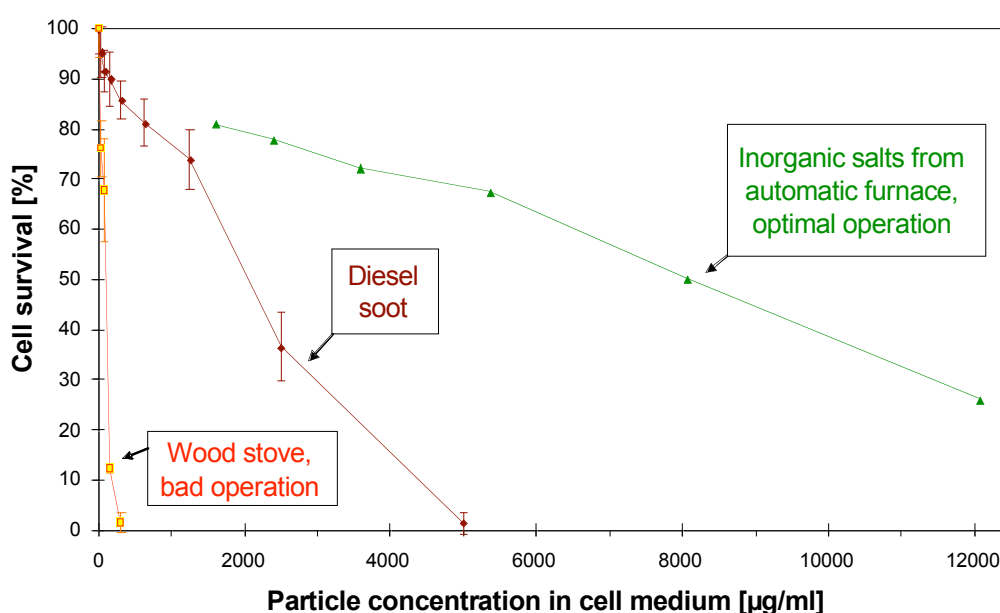


Figure 3: Cell survival as a function of particle concentration for Diesel soot, mainly inorganic particles from automatic wood combustion, and wood particles from incomplete combustion in a badly operated stove [10].

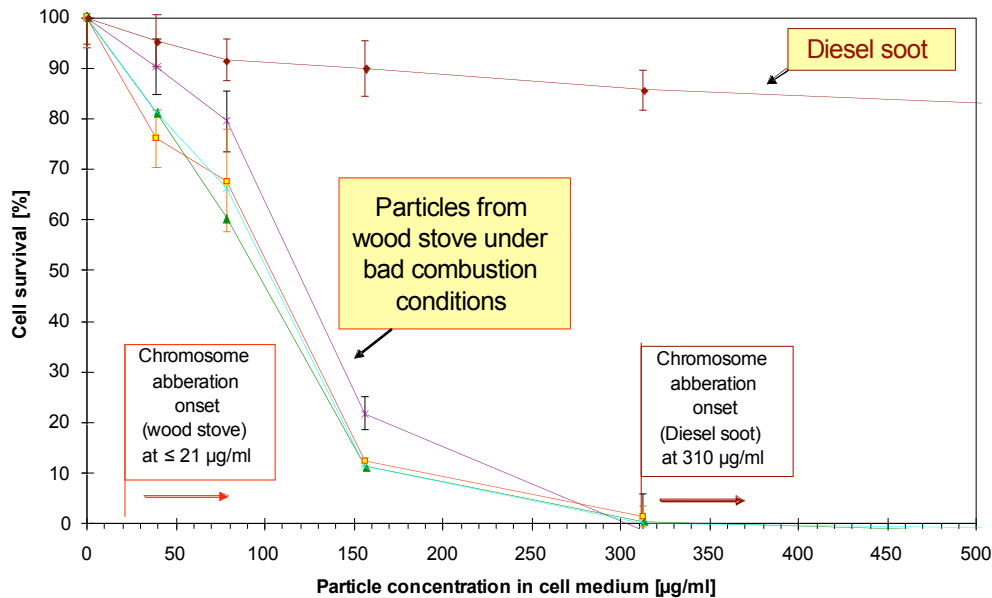


Figure 4: Cell survival as a function of particle concentration for Diesel soot and wood particles from incomplete wood combustion (data as in fig. 2 but in magnified scale). In addition, the level of chromosome aberration onset is indicated [10].

3. Physical properties of particles from biomass combustion with respect to precipitation in electrostatic precipitators (ESP)

3.1 Method

The principles for electrostatic precipitators (ESP) are well known since decades [11]. However, design parameters are well known for coal particles and for power plants which are operated at nearly stationary conditions, while data for small-scale biomass combustion are scarce. For this reason, particle parameters were investigated in an experimental investigation. For this purpose, an electrostatic lab-scale precipitator was designed as tube type ESP with a maximum voltage of $U_{\max} = -65$ kV and connected to a pellet boiler. The pellet boiler was modified to enable stationary operation at specific combustion conditions, which normally exist only during transient phases such as during start-up. The ESP was designed to enable precipitation efficiencies as typically found in commercial small and medium scale ESP, i.e., safely $> 90\%$ for all particle sizes and $> 95\%$ as average precipitation efficiency for typical particle collectives found in biomass combustion. Electrical conductivity was analysed acc. to IEEE Std 548-1984 (due to missing valid standards, the old standard is used) [12].

3.2 Results

In biomass combustion, three combustion regimes can be distinguished which – among other parameters – are related to the level of excess air available in the combustion chamber. Figure 5 shows the particles found in the present laboratory device for different excess air ratios:

At low excess air ratio (regime C), soot is formed in hot zones in the flame as a synthesis product through the release of hydrocarbons containing primary tars from wood pyrolysis, formation of secondary tars in an atmosphere with lack of oxygen, PAH formation, and finally release of hydrogen during particle growth thus resulting in a high C/H ratio.

At optimum excess air (regime B), near complete combustion is achieved, if good mixing of combustible gases with air is guaranteed and quenching of the flame is avoided. Consequently, carbonaceous matter in solid and liquid phase is emitted in very small concentrations, while inorganic particles formed from ash constituents are available as particulate matter (PM) in the flue gas and predominantly found as salts.

At high overall excess air (regime A), the combustion temperature decreases, resulting in incomplete combustion. Due to low temperature, the formation of soot and the release of hydrogen is suppressed, resulting in high concentrations of primary and secondary tars formed during pyrolysis consequently leading to condensable organic compounds (COC) in the flue gas with low C/H ratio.

Since ESP operation is ideal at operation at optimum excess air, nowadays applications are often limited to such combustion conditions, while the ESP is often shut-off during unideal combustion conditions.

The results on the electrical conductivity are shown in Fig. 6 for two different atmospheres which refer to relatively dry flue gases and to flue gas with high humidity. The two investigated humidities cover the typical range from biomass combustion. Dry flue gases are possible in a wood stove, while high humidity is found in an automatic boiler in case of wet biomass.

The three particle types exhibit significantly different conductivities. In addition, the humidity strongly influences the conductivity. In dry flue gas, optimum conditions are only found for salt, while COC leads to back-corona and soot leads to re-entrainment. At high humidity, operation with salt particle is only favourable at temperatures above 120°C. COC exhibits optimum conductivity, but remains critical to remove in case of dry ESP.

These results lead to the following conclusions:

- Three different particle types from wood combustion have been identified which correspond to different combustion regimes, i.e.,
 - soot resulting from combustion at high temperature but with low excess air and consequently local lack of oxygen,
 - particles which consist dominantly of mineral matter such as salts found at high combustion temperature and with sufficient local excess air,
 - condensable organic compounds (COC) resulting from low temperature combustion conditions at high excess air.
- The three particle types exhibit completely different physical and chemical properties, among which the electrical conductivity is most relevant for ESP operation. The identified properties confirm the particle type and the particle properties as expected from the proposed theory of the particle formation mechanisms.
- Particles from good combustion (mainly inorganic compounds such as salts) exhibit ideal conductivity for ESP.
- Soot reveals high conductivity thus enabling high precipitation efficiency but severe re-entrainment of agglomerated particles
- Condensable organic compounds (COC) exhibit low conductivity thus leading to back-corona which limits ESP operation
- ESP operation for good and stationary conditions during wood combustion with mainly inorganic particles enables uncritical operation, while ESP operation can be critical e.g.
 - during start-up due to COC from low temperatures - during throttled air, either due to COC released at low temperatures or due to soot formed at high temperatures in zones with lack of oxygen.

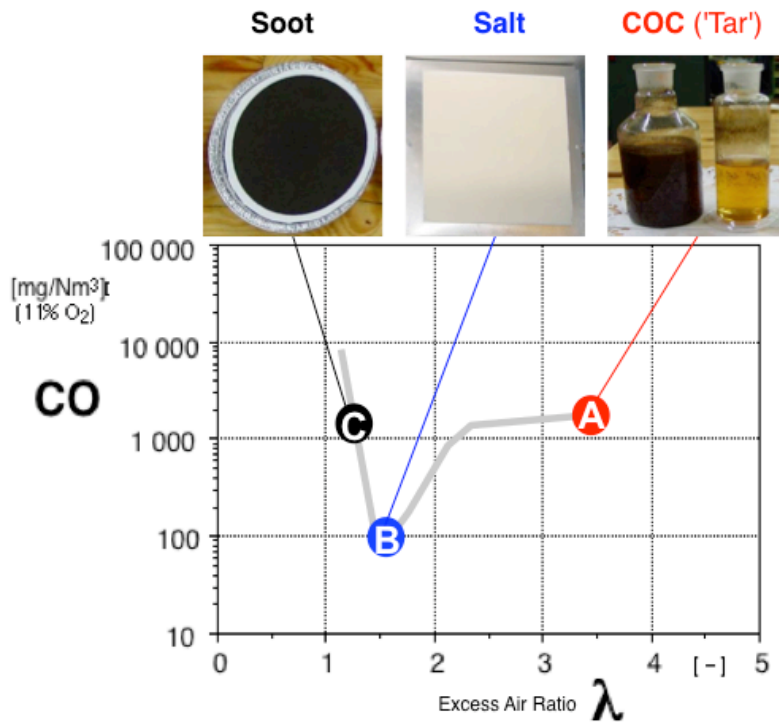
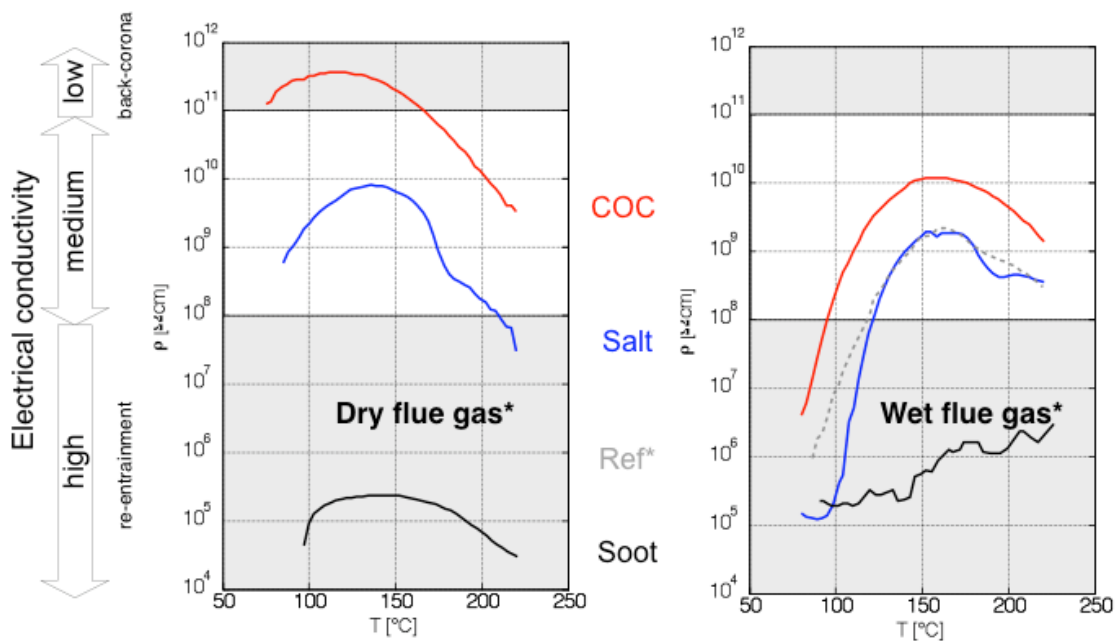


Figure 5: Three regimes of biomass combustion shown in the diagram CO as function of excess air λ . The three combustion regimes are related to three different types of combustion particles, i.e., soot, salts, and COC, depending on the level of excess air and other parameters [13].



*Dry: 5 vol.-% H_2O e.g. excess air ratio 3 & wood moisture content 5%
 *Wet: 20 vol.-% H_2O e.g. excess air ratio 1.2 & wood moisture content 50%
 *Ref: 13 vol.-% H_2O : excess air ratio 1.5 & wood moisture content around 30%

Figure 6: Electrical conductivity measured for soot, salts, and COC sampled in the laboratory equipment during different combustion conditions [14]. In addition, results from dust sampled in a commercial ESP operated with natural wood chips in an automatic combustion plant are shown and indicated as "Ref". The optimum range for precipitation in ESP is indicated according to Parker [15, 16].

4. Literature

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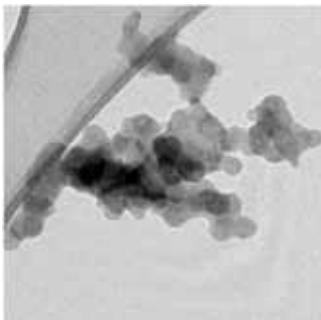
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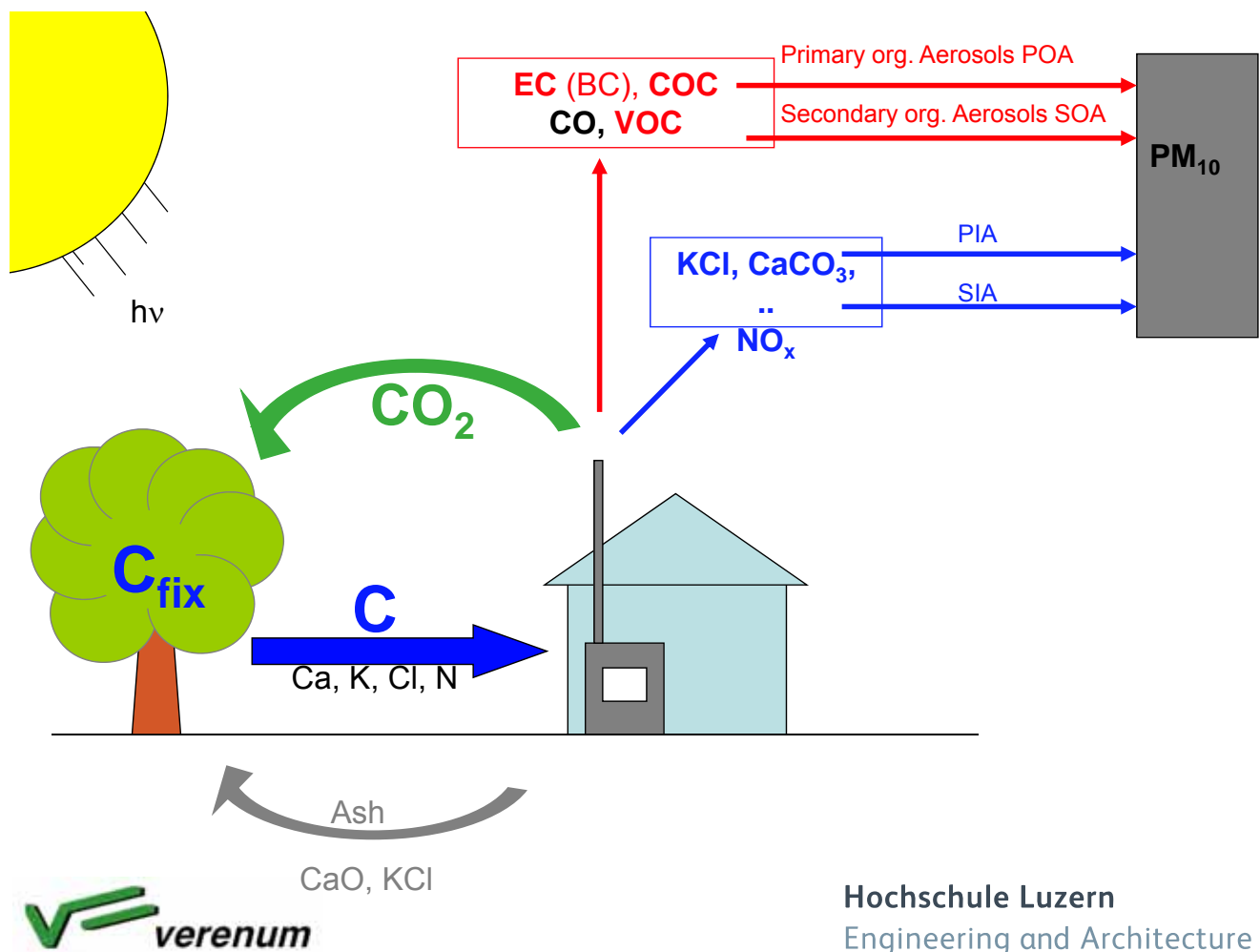


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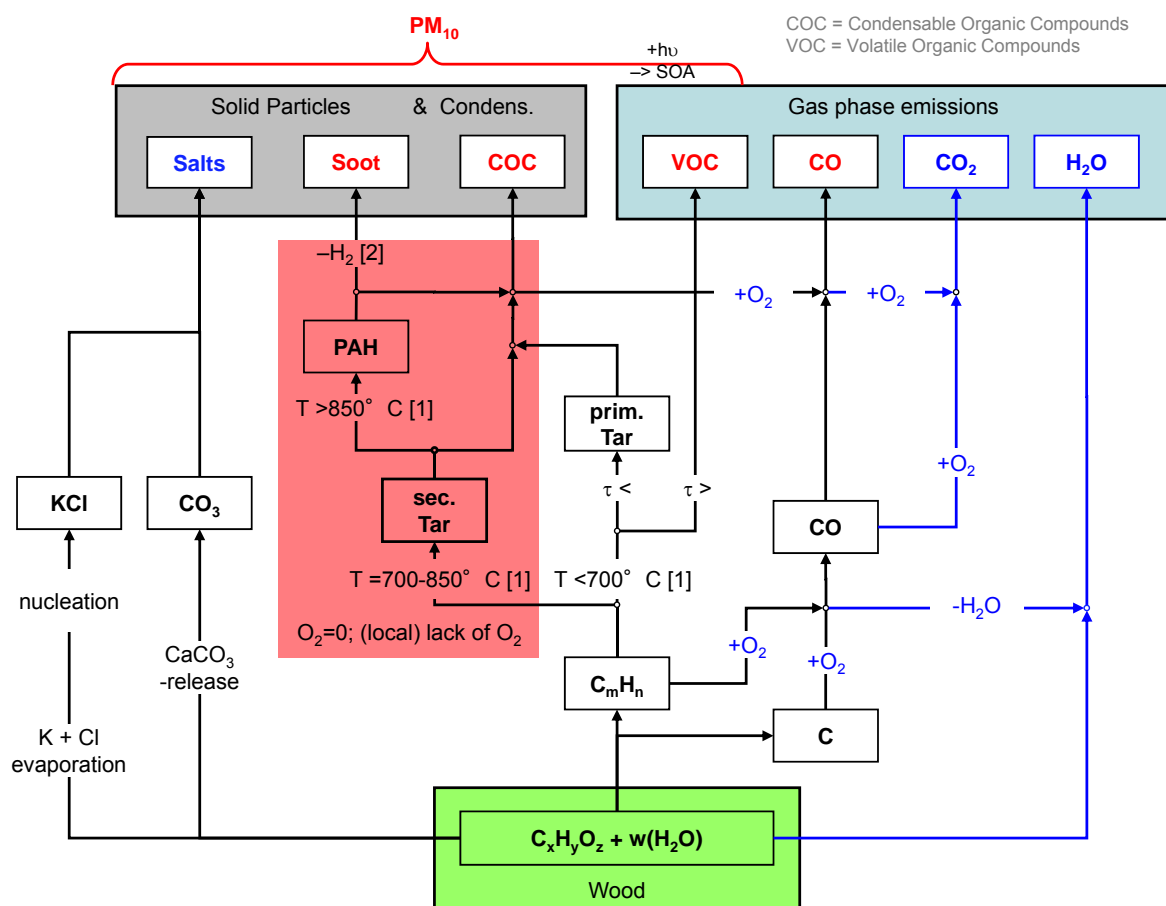
- ➡ 1. Particle formation
- 2. Health effects
- 3. Properties in ESP
- 4. Conclusions



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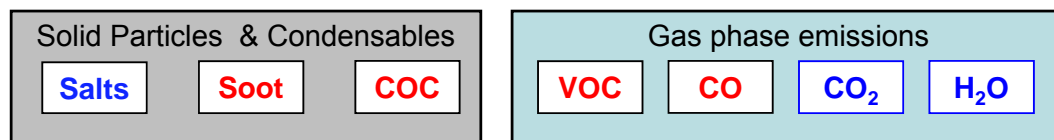


Particle Formation



[1]: Evans and Milne, 1987
[2]: Jess, 1996

Particle Sources



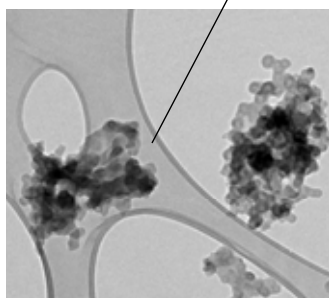
[Schmid]



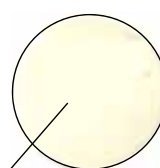
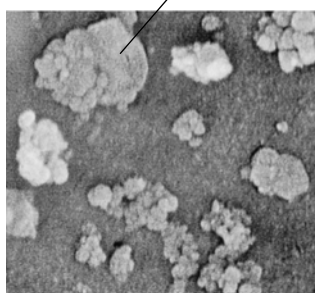
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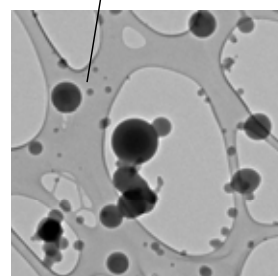
Soot





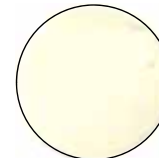

Salts

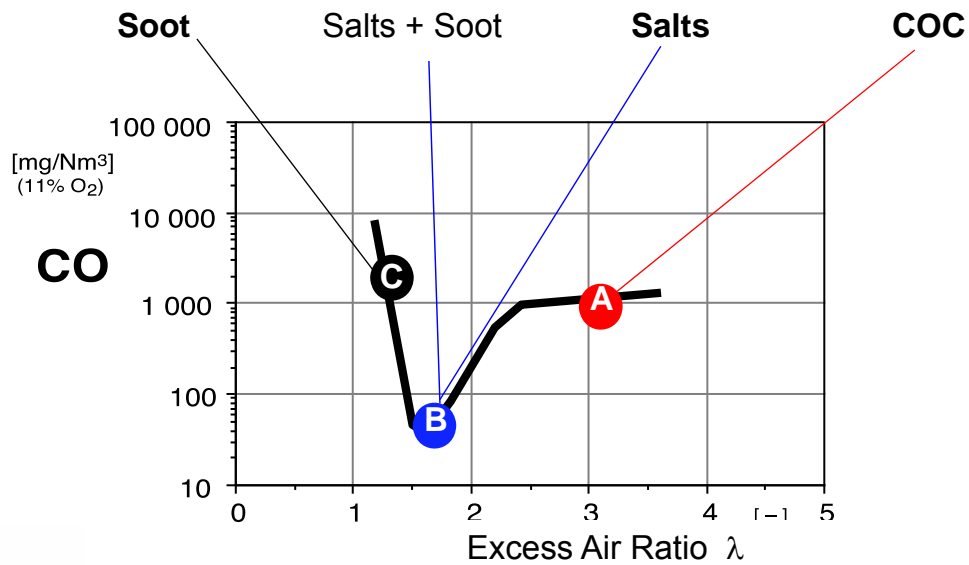


COC



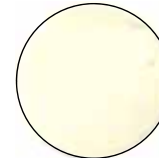



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Flaming Combustion			Pyrolysis
lack of O ₂ in the flame	– Mix –	T and O ₂ good	T low at start or high O ₂ O ₂ lack gas. or fl. ext.
			



[T. Nussbaumer, *Energy & Fuels* 2003, 17]

Flaming Combustion			Pyrolysis
lack of O ₂ in the flame	– Mix –	T and O ₂ good	T low at start or high O ₂ O ₂ lack gas. or fl. ext.
			

	Soot	Salts + Soot	Salts	COC
C / H	> 6 – 8			≈1 (< 2)
Electrical conductivity	high		medium	low (insulating)
Suitability for ESP	re-entrainment		ideal	back-corona



Analysis



VOC
NMVOC



Particles

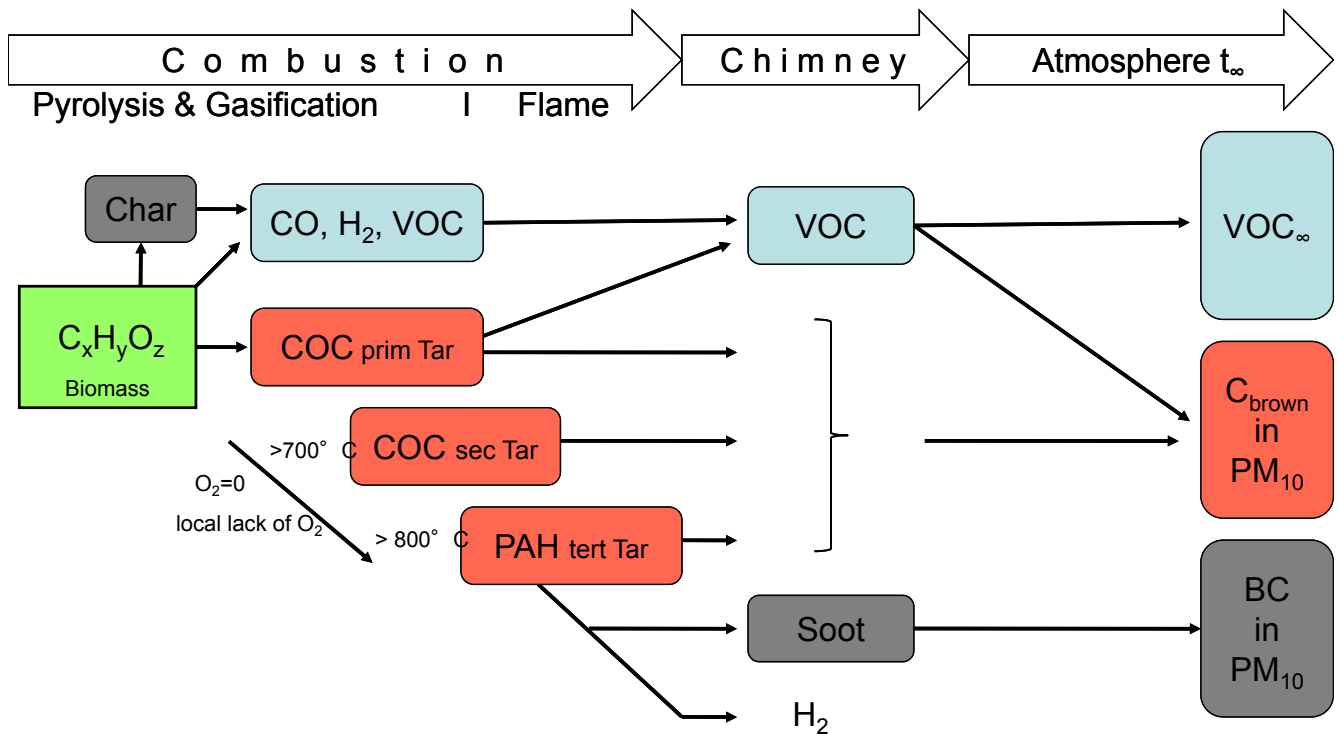


COC



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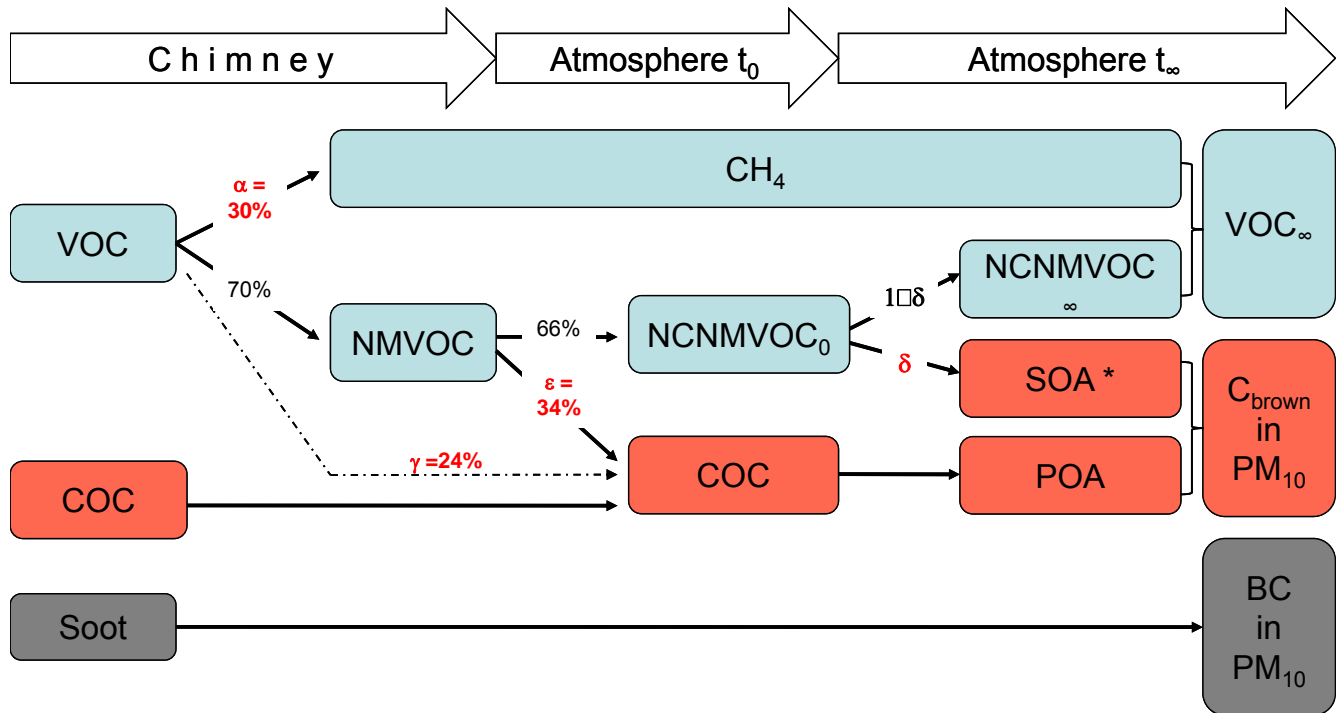
Conversion of Carbon during Combustion and consecutive Partitioning of VOC



[Nussbaumer 2010]

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Conversion of Carbon during Combustion and consecutive Partitioning of VOC



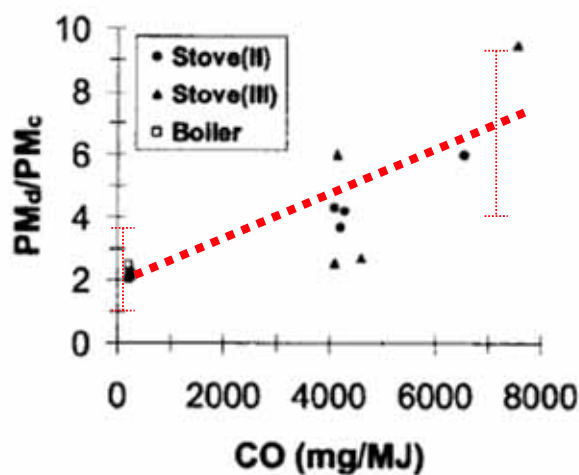
* $m(\text{SOA}) = f \delta m(\text{NCNMVOC}_0)$
 $f = 1.6 \pm 0.2$ for urban aerosol
 $f = 2.1 \pm 0.2$ for non-urban aerosol
 $f = 2.2\text{--}2.6$ (2.4) for wood smoke
 [Turpin, B.; Lim, J., Aerosol Science and Technology 35: 602–610 (2001)]



[Nussbaumer 2010]

Contribution of biomass to BC and Brown carbon?

$\frac{\text{Salt} + \text{Soot} + \text{COC}}{\text{Salt} + \text{Soot}}$



near-complete combustion: COC = 0

incomplete combustion: COC = 1 to 10 x (Salt + Soot)



[Johansson et al., World Bioen. 2008]

Emission factors	NO ₂ [mg/MJ]	CO [mg/MJ]	VOC [mg/MJ]	CH ₄ [mg/MJ]	NMVOC [mg/MJ]	PM [mg/MJ]
Open Chimneys	80	3000	600	240	360	> 100
Roomheaters	80	3000	600	240	360	> 100
Roomheaters with pellets	60	300	50	20	30	< 60
Hearths/cooker	70	5000	800	320	480	> 200
Modern log wood boilers	80	1500	50	20	30	< 50
Old log wood boilers	70	5000	800	320	480	> 200
Autom. boilers < 50 kW	120	600	30	12	18	< 100
Pellet boilers < 50 kW	60	200	15	6	9	< 50
Autom. boilers < 500 kW	120	500	15	6	9	< 100
Pellet boilers 50–500 kW	60	150	15	6	9	< 50
Autom. boilers > 500 kW	150	300	10	4	6	< 100
Pelletkessel > 500 kW	70	150	10	4	6	< 50



[Nussbaumer 2010]

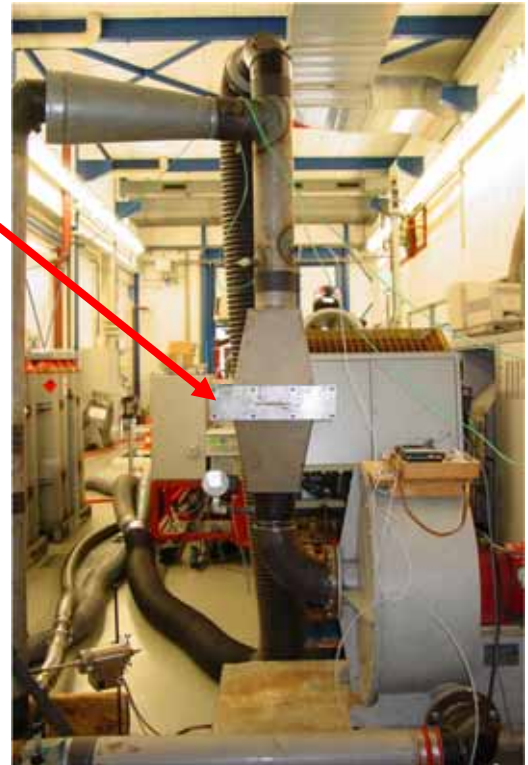
Content



1. Particle formation
2. Health effects
3. Properties in ESP
4. Conclusions

Source 1: Euro 3 Diesel engine without DPF

Filter

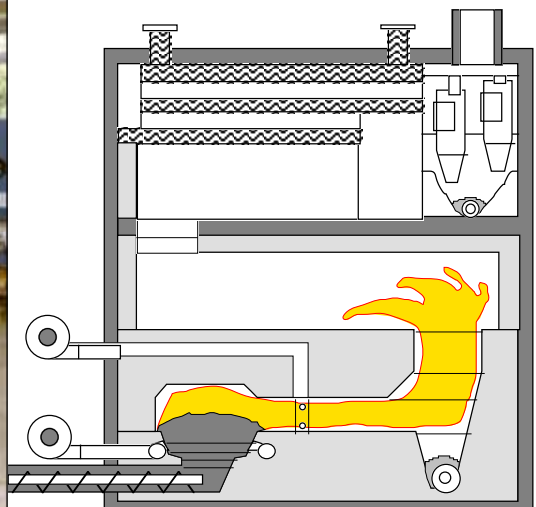


Test-bench at EMPA Dübendorf

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Source 2: Automatic wood combustion plant



[Verenum / EMPA / Müller AG] Hochschule Luzern
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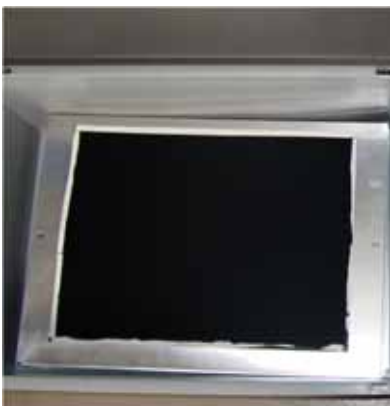
Source 3: Wood Stoves



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Samples

Diesel soot



Automatic wood boiler

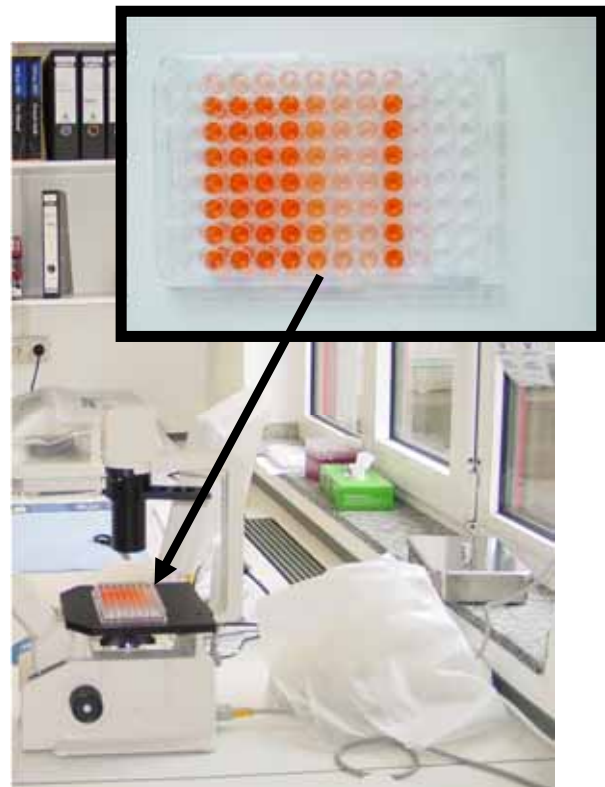


Wood stove



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Investigation of cell cultures

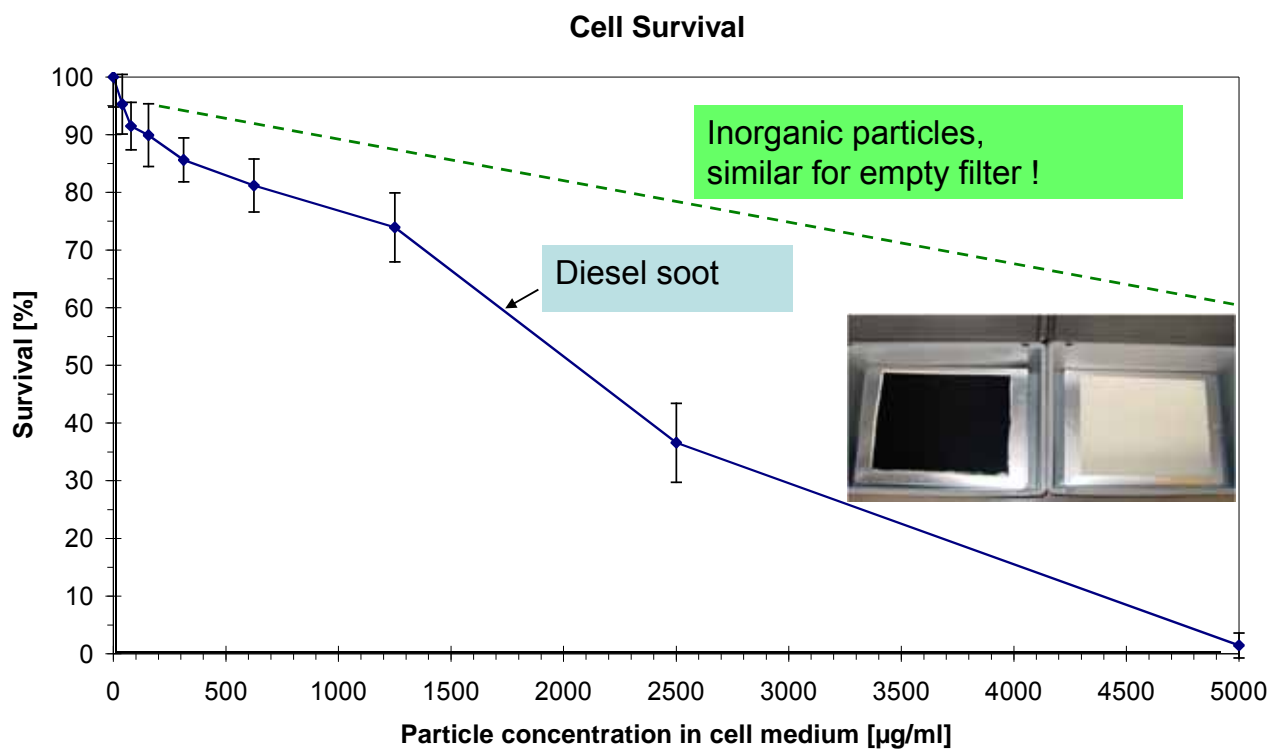


RCC Cytotest Cell Research

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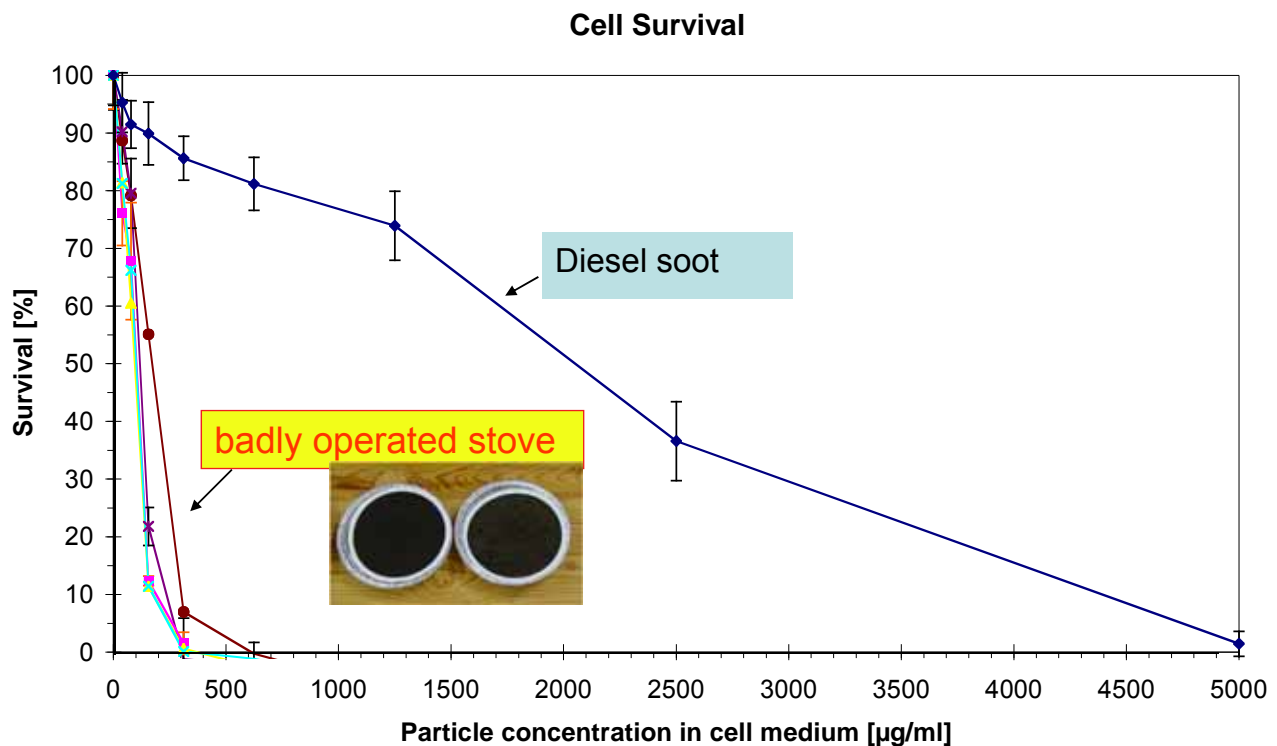
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Cytotoxicity tests on lung cells of Chinese hamster

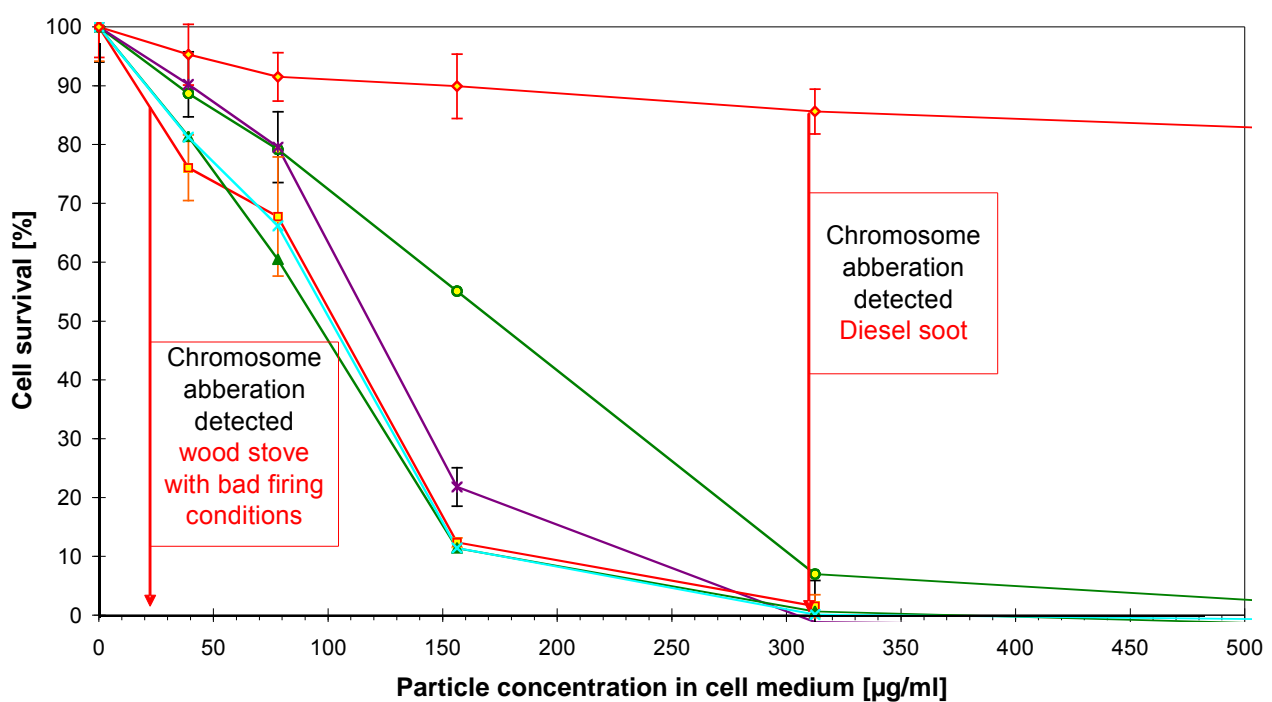


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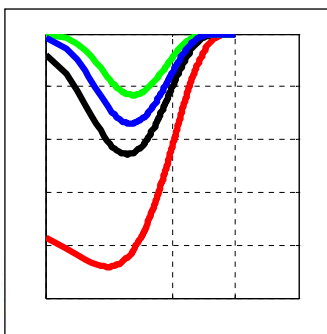


Carcinogenic potential estimated by chromosome defects



Content

1. Particle formation
2. Health effects
- ➔ 3. Properties in ESP
4. Conclusions



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ESP for Wood Combustion

- Design parameters for coal at stationary operation [White, 1963]
- Scarce information on wood particle from heating (on/off) and small-scale

for wood boilers
from 200 kW – 1MW



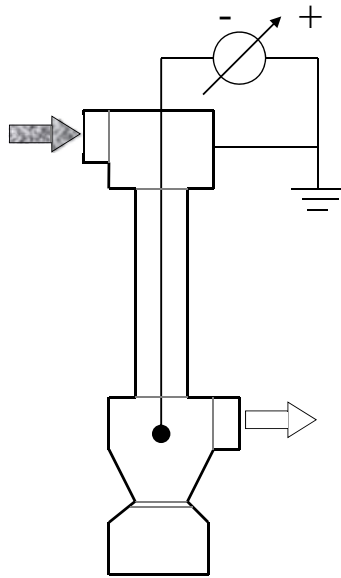
[Scheuch]

for residential heating
from 5 kW – 70 kW



[Oekotube]

Electrostatic Precipitators (ESP)



Precipitation Efficiency by Deutsch:

$$\eta = 1 - e^{-\frac{w}{SCA}}$$

w = migration velocity [m/s]
 = f (el. field strength, D , T , H_2O ,...)

SCA = spec. collection area
 [$m^2/(m^3/s)$] = [s/m]

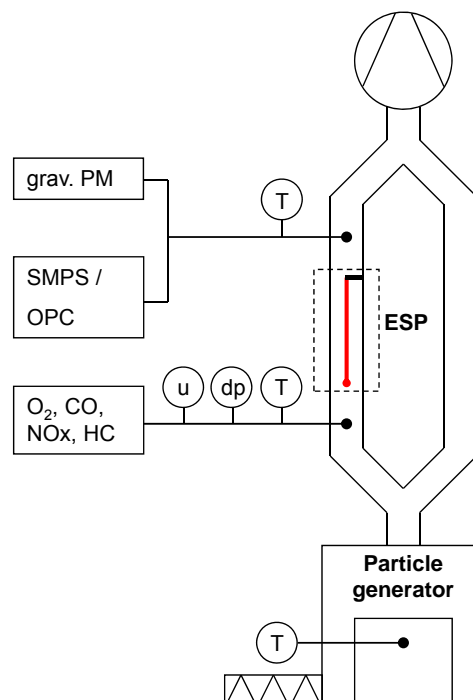
Experimental Setup

ESP:

L 1000 [mm]
 D 100 [mm]
 v 1 [m/s]
 SCA 45 [s/m]
 U_{max} -65kV

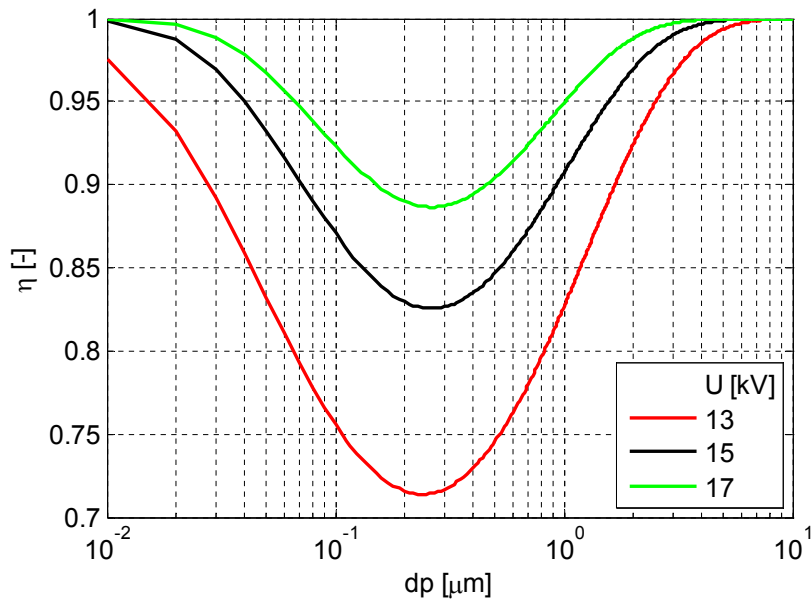
Particle generator:

Pellet boiler
 modified
 Q 15kW



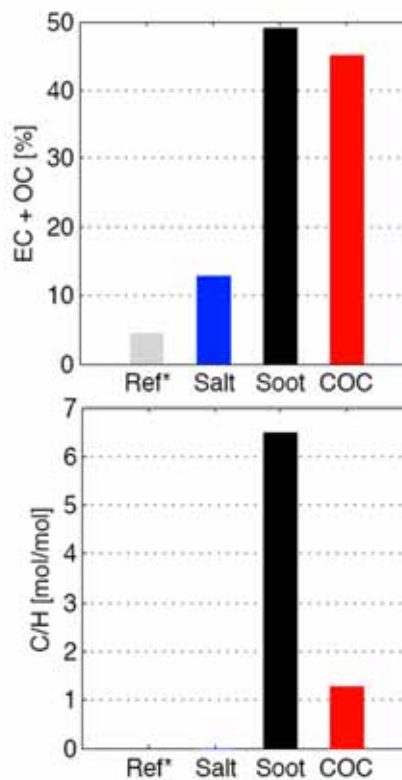
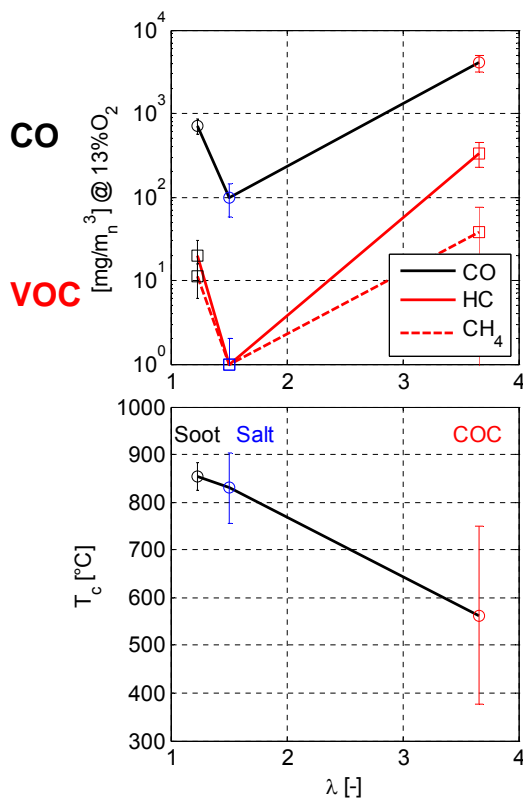
Experimental Setup

ESP efficiency as function of electric field strength



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Gas and Chemical Analysis



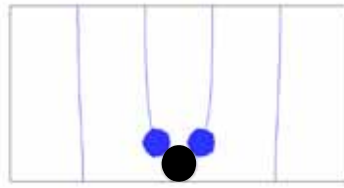
*Ref: 1MW AWC



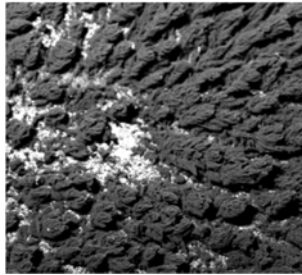
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Dust Layer Build-up

Conductiv particles:
→ 'dendritic' build-up

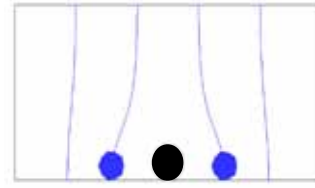


Soot



weak adhesion /
re-entrainment

Normal or isolating particles:
→ homogeneous build-up



Salt

COC



stable layer



sticky layer



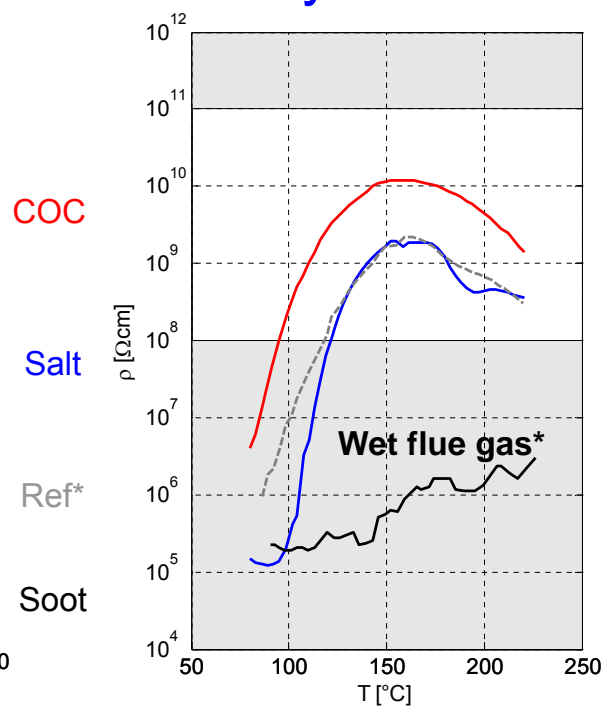
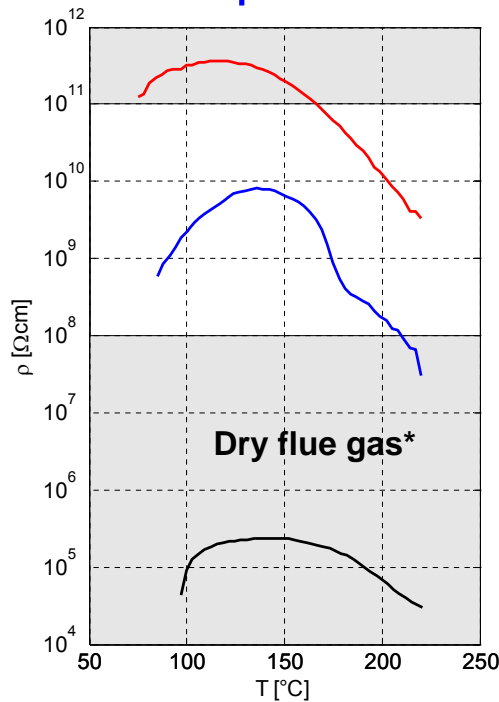
[Blanchard et. al., 2002]

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Specific Dust Resistivity

Electrical conductivity [3]
low back-corona
medium
high re-entrainment



*Dry: 5 vol.-% H_2O e.g. excess air ratio 3 & wood moisture content 5%

*Wet: 20 vol.-% H_2O e.g. excess air ratio 1.2 & wood moisture content 50%

*Ref: 13 vol.-% H_2O : excess air ratio 1.5 & wood moisture content around 30%



[3]: Parker, 1997

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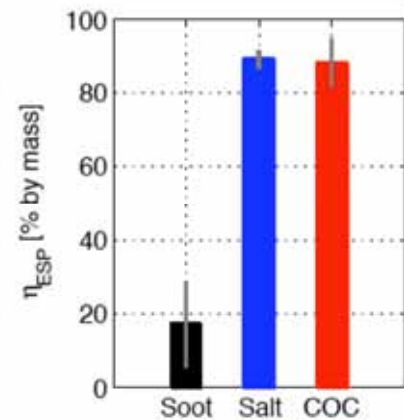
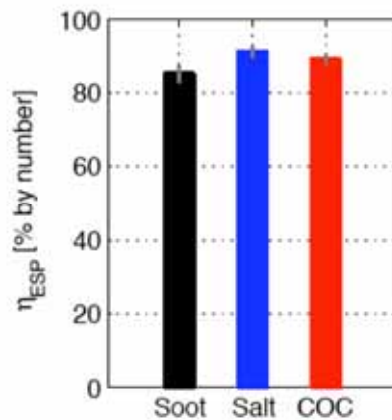
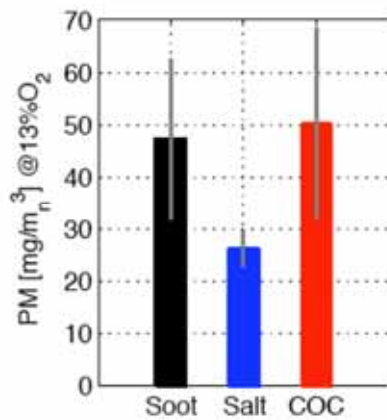
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Raw gas PM I

ESP efficiency I

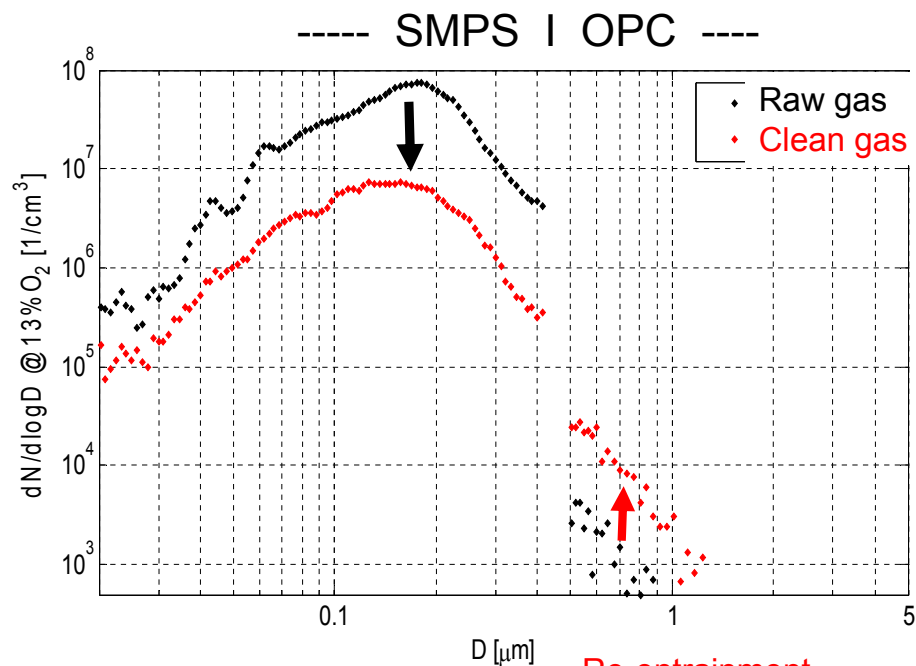
by number

by mass



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Particle Size Distribution for Soot

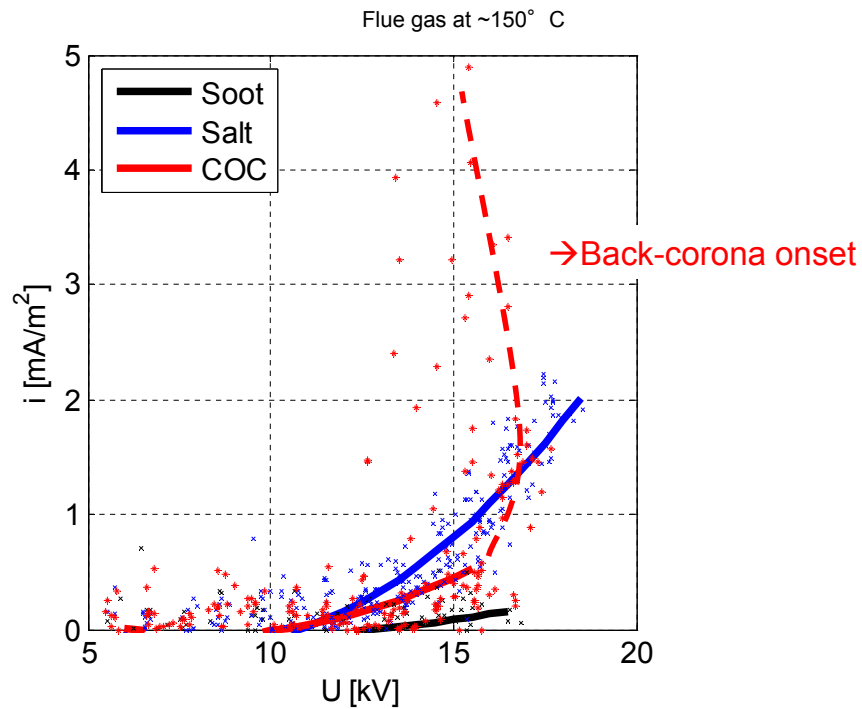


Re-entrainment
of agglomerated
soot particles

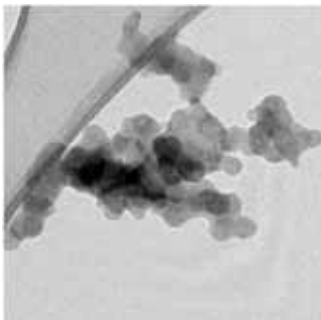


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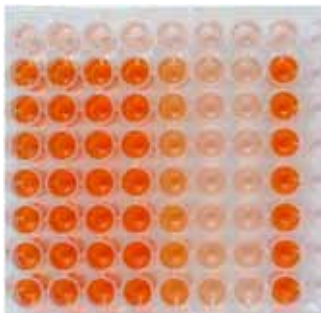
IU Characteristic



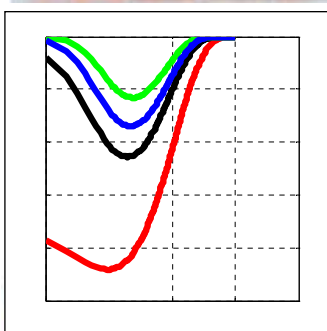
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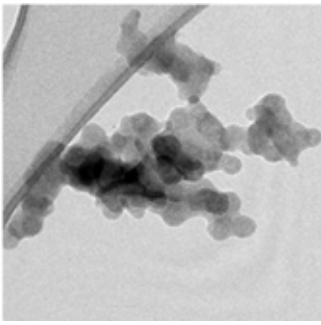
Content



1. Particle formation
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Conclusions 1. Particle formation

1. Biomass produces **salt, soot, and COC/VOC**.
The three types of aerosols correspond to different combustion regimes
2. There is high priority to reduce
 - a) **COC/VOC** as precursors of POA and SOA, and
 - b) **Soot** as directly emitted primary aerosol
3. There is no target conflict between organics and soot, **but**:
 - a) The formation mech. & counter measures are **different**
 - b) There is a need of at least **two measurements** to trap primary and secondary aerosols e.g. **particles and VOC**
4. The role of VOC for POA and SOA can be described by the partitioning factors to NMVOC (70%) and COC (24%)



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2. Health effects

1. Diesel soot and wood particles exhibit similar size around $0.1\mu\text{m}$, are inhalable and mostly PM₁
2. Salts from near-complete wood combustion exhibit one order of magnitude lower cell toxicity and chromosome defects than Diesel soot
3. Particles from incomplete wood combustion consist of soot and COC including PAH and exhibit far higher cell toxicity and chromosome defects than Diesel soot
4. Nowadays PM emission limits do not distinguish between particle types and hot sampling does not trap COC, which need to be considered



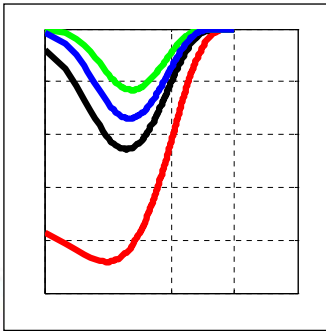
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3. Properties in ESP

1. Salt, soot, and COC exhibit completely different properties, incl. the el. conductivity crucial for ESP:

- Salts are ideal for ESP
- Soot is conductive and leads to re-entrainment
- COC is el. insulating leading to back-corona
In addition, COC causes sticky dust layers

2. ESP operation is critical during transient periods, hence information on the combustion and the particles is needed to optimise ESP operation



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