

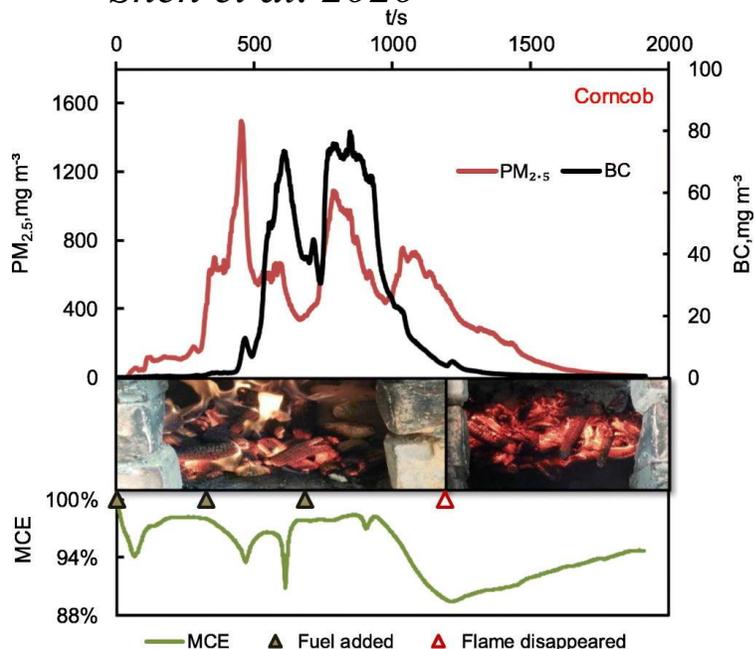
TFTEI

Under the Convention on Long Range Transboundary Air Pollution

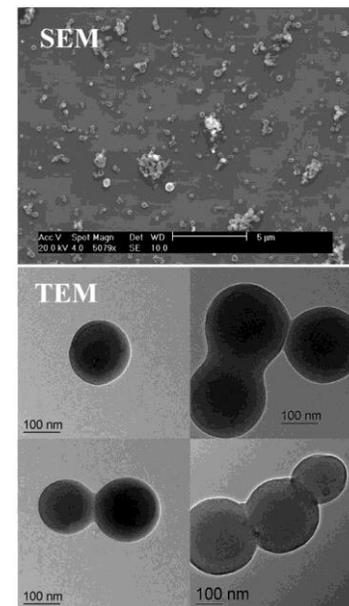
Technologies for PM and Black Carbon emission reductions (Annex X of GP)

TFTEI technical secretariat
Bertrand Bessagnet (Citepa)

Shen et al. 2020



Acknowledgments to Isaline Fraboulet and Cécile Raventos (INERIS) for fruitful discussions



Preparation of the work programme for the review of the Gothenburg Protocol by the Gothenburg Protocol Review Group (GPG) in cooperation with the Chair of the Working Group on Strategies and Review and the Chair of the Executive Body

In the scope of questions addressed on BC and how to reduce its emissions, technical work undertaken by TFTEI to:

- Review impact of current PM obligations and measures on BC and PAH emissions
- Evaluate BC measures

This technical work is complementary to the draft “guidance document on **prioritizing reductions of particulate matter** so to also achieve reduction of black carbon” developed by TFIAM

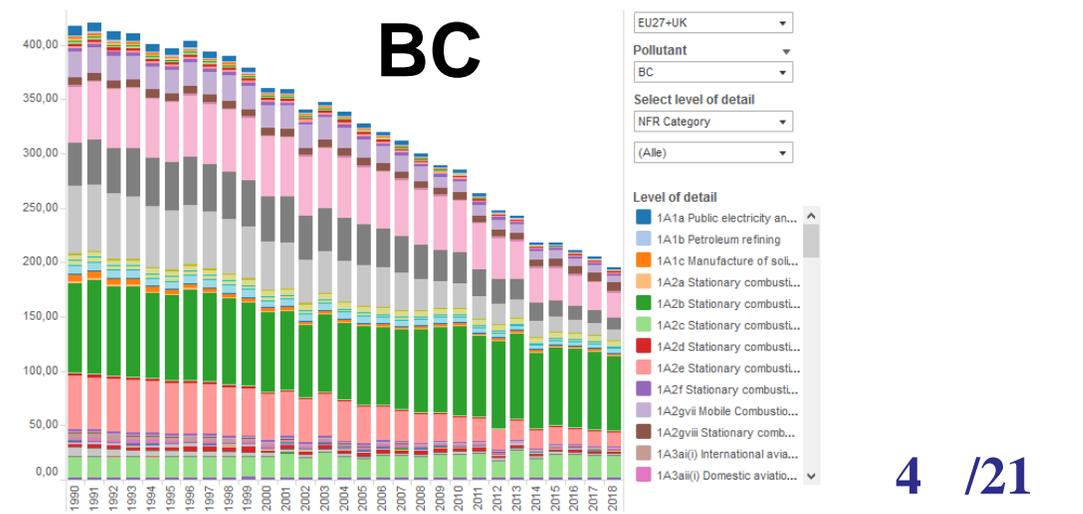
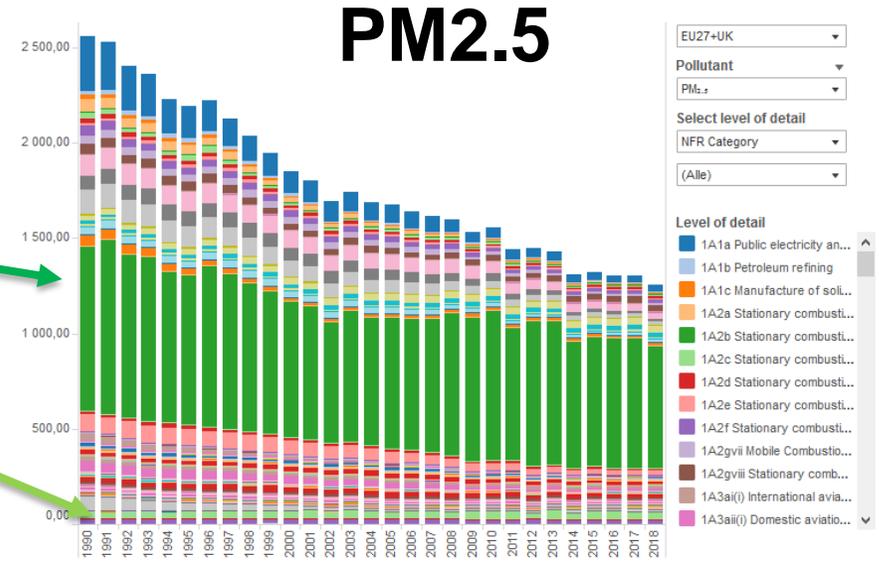
The document prepared will address main sources of BC: residential heating, road transport, non road machinery, flares, ...

Outline

- Focus on small scale residential emissions (So far)
- Some recent key findings picked-up in the recent literature on wood/biomass burning appliances for **BC, PAH, UFP** emission reductions
- Conclusions - Key messages

Main target sectors for EU27+UK

- 2018, PM2.5
 - 1A4bi Residential: Stationary
 - 5C2 Open burning of waste
 - 1A1a Public electricity and heat production
- 2018, BC
 - 1A4bi Residential: Stationary
 - 5C2 Open burning of waste
 - 1A4cii Agriculture/Forestry/Fishing: Off-road vehicles and other machinery
 - Road transport....
- Road transport: an issue for EECCA countries



The Gothenburg Protocol

- *1999 Protocol to Abate Acidification, Eutrophication and Ground-level Ozone to the Convention on Long range Transboundary Air Pollution, as amended on 4 May 2012 (ECE/EB.AIR/114)*
 - “dust” is supposed to be TSP (ELV expressed under standard conditions)
 - Limit values for emissions of particulate matter from stationary sources
 - **Recommended** limit values for dust emissions released from small combustion sources

Recommended limit values for dust emissions released from boilers and process heaters with a rated thermal input of 100 kWth–1 MWth

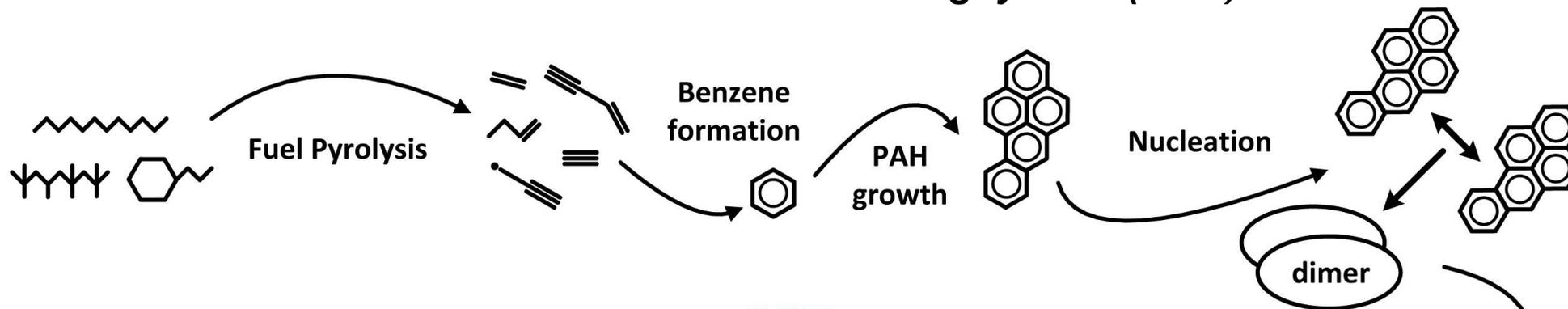
| | | <i>Dust (mg/m³)</i> |
|-------------------------------|------------------------|--------------------------------|
| Solid fuels 100 kWth–500 kWth | New installations | 50 |
| | Existing installations | 150 |
| Solid fuels 500 kWth–1 MWth | New installations | 50 |
| | Existing installations | 150 |

Recommended limit values for dust emissions released from new solid fuel combustion installations with a rated thermal input < 500 kWth to be used with product standards

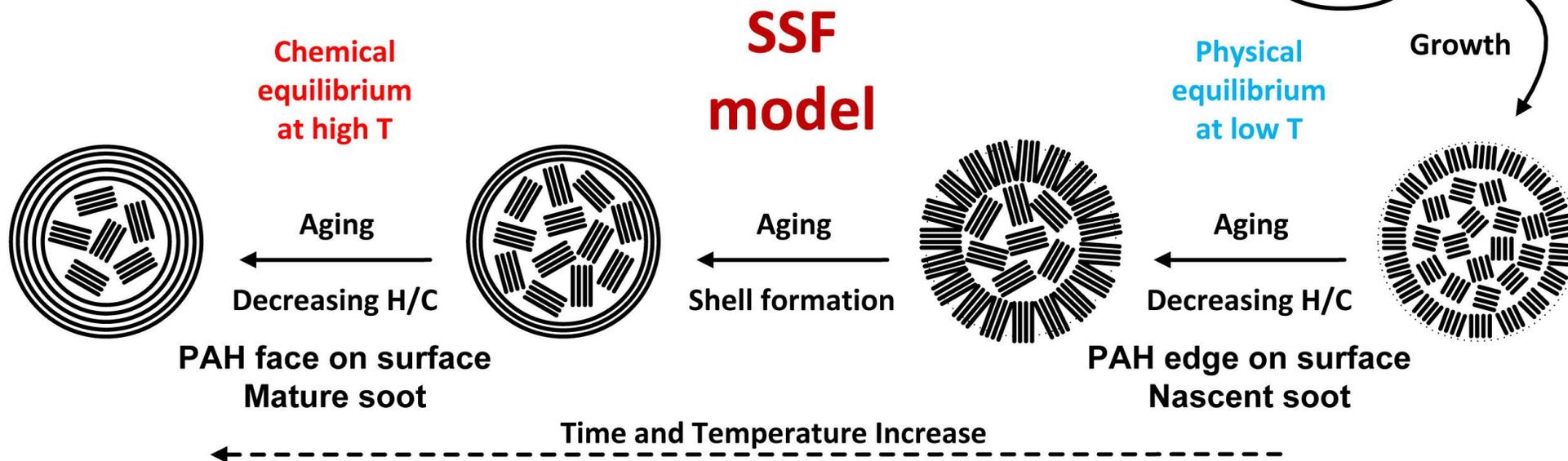
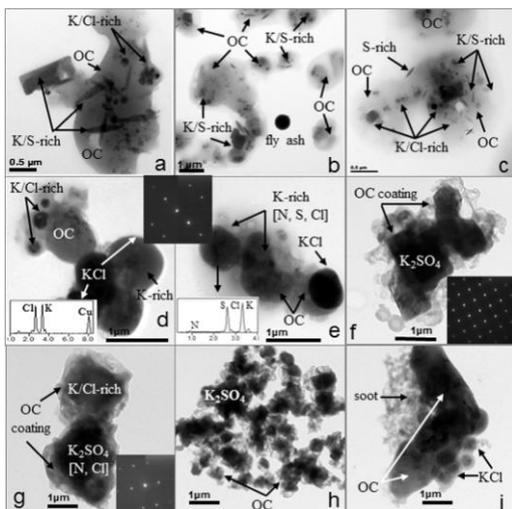
| | <i>Dust (mg/m³)</i> |
|--|--------------------------------|
| Open/closed fireplaces and stoves using wood | 75 |
| Log wood boilers (with heat storage tank) | 40 |
| Pellet stoves and boilers | 50 |
| Stoves and boilers using other solid fuels than wood | 50 |
| Automatic combustion installations | 50 |

Soot and ash formation pathway

Kholghy et al. (2016)



Chen et al. (2017)



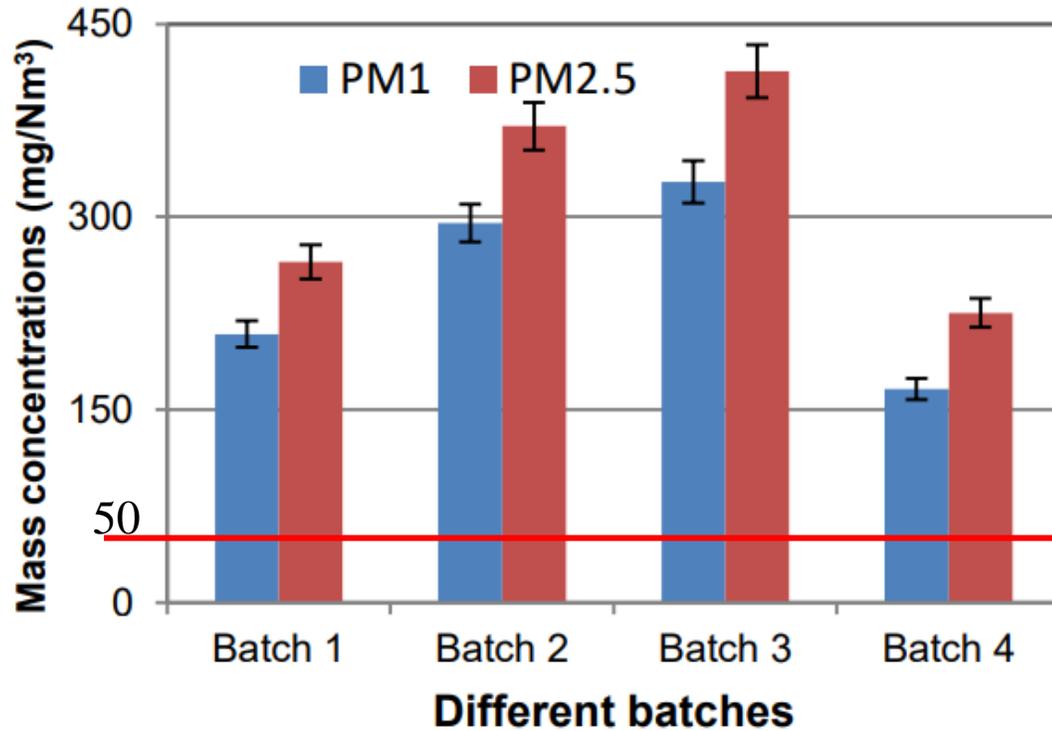
SSF: soot surface shell formation

EF for carbonaceous species

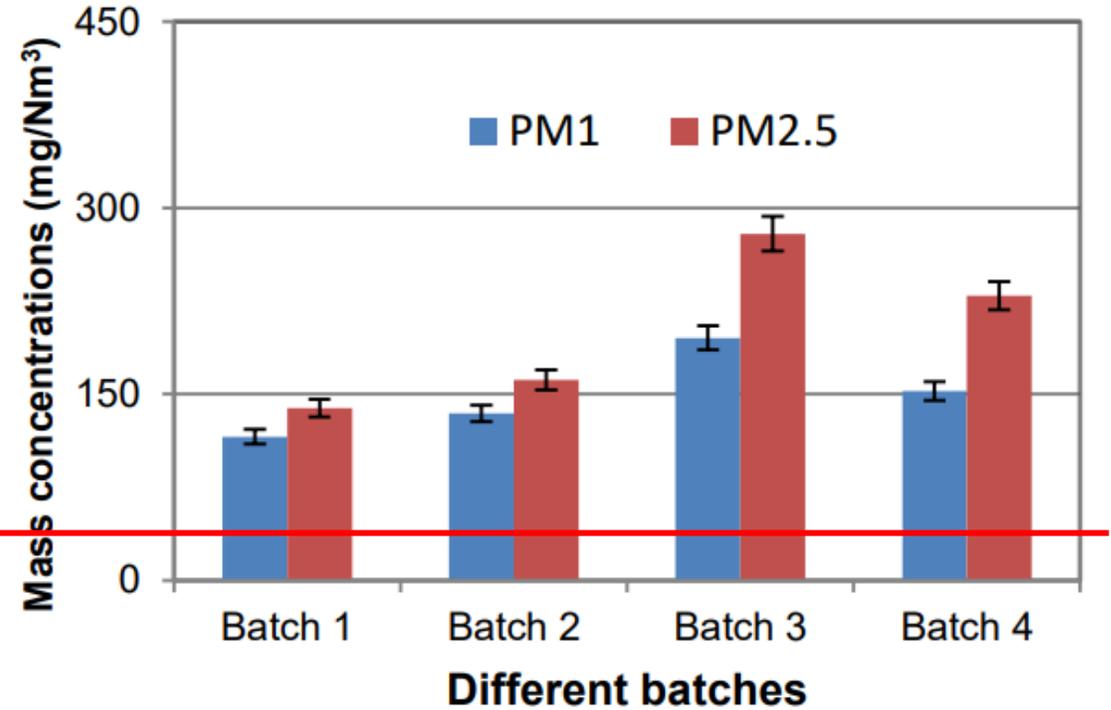
Seay et al. (2020)

| Appliance (efficiency) ^a | Description | Aerosol wood combustion EFs ^c | | | |
|---|---|--|---------------------------|---------------------------|------------------------|
| | | PM _{2.5} , g/GJ ^b | BC, g/GJ (%) ^c | OC, g/GJ (%) ^c | SO _x , g/GJ |
| Open fireplaces (20%) | Simplest combustion device, including a basic combustion chamber directly connected to a chimney and a large opening to the fire bed. Devices are characterized by high, non-adjustable excess of combustion air. | 820 | 57.4 (7) | 352.6 (43) | 11 |
| Conventional stoves (45%) | Includes both closed fireplaces and conventional radiative stoves. Closed fireplaces are equipped with front doors and have air flow control systems. Because of their design and combustion principles, they more closely resemble and are grouped with conventional stoves rather than open fireplaces. Conventional radiative stoves include both downburning and upburning techniques with poorly organized combustion processes. | 740 | 74 (10) | 333 (45) | 11 |
| Conventional boilers < 50 kW (60%) | Devices that heat water for indirect heating. Over-fire boilers are characterized by non-optimal supply of combustion air caused by natural draft, which causes incomplete combustion. Under-fire boilers include manual feed systems, stationary grates, and a two-part combustion chamber. | 470 | 75.2 (16) | 178.6 (38) ^d | 11 |
| High-efficiency stoves (65%) | Covers traditional stoves with improved utilization of secondary air in the combustion chamber. Also includes catalytic converter stoves, which reduce emissions from incomplete combustion. | 370 | 59.2 (16) | 140.6 (38) ^d | 11 |
| Advanced / ecolabelled stoves and boilers (70%) | Characterized by multiple air inlets and pre-heating of secondary combustion air by heat exchange with hot flue gases. Ecolabelling schemes assign a standard for improved efficiency and reduced emissions and are largely based around European standards. Also includes state of the art downdraft multi-chamber boilers. | 93 | 26.04 (28) | 28.83 (31) | 11 |
| Pellet stoves and boilers (85%) | An advanced stove that uses an automatic feed for pelletized fuels, which are distributed to the combustion chamber by a fuel feeder. These stoves are often equipped with active control systems for combustion air supply. Category also includes automatic pellet-fired boilers, which include fully automatic systems for feeding fuel and for supply of combustion air. | 60 | 9 (15) | 7.8 (13) | 11 |

PM concentrations in flue gas from two Belgian modern wood stoves



(a)



(b)

Fig. 4. PM₁ and PM_{2.5} emissions from different batches, (a) for the 10 kW stove, (b) for the 20 kW stove

A large range of EF

- From the EMEP/EEA guidebook:
 - PM2.5 : **740** g/Gj
 - PM10 : 760 g/Gj
 - TSP=800 g/Gj
 - BC as a fraction of PM2.5 (2-20% taken as 10%: **74** g/Gj)
- BC EF certainly underestimated according to a french study (CARABLACK, Raventos et al., 2018)
- BC by thermo-optical methods difficult to estimate if OM is high

GAINS Emission factors ranges

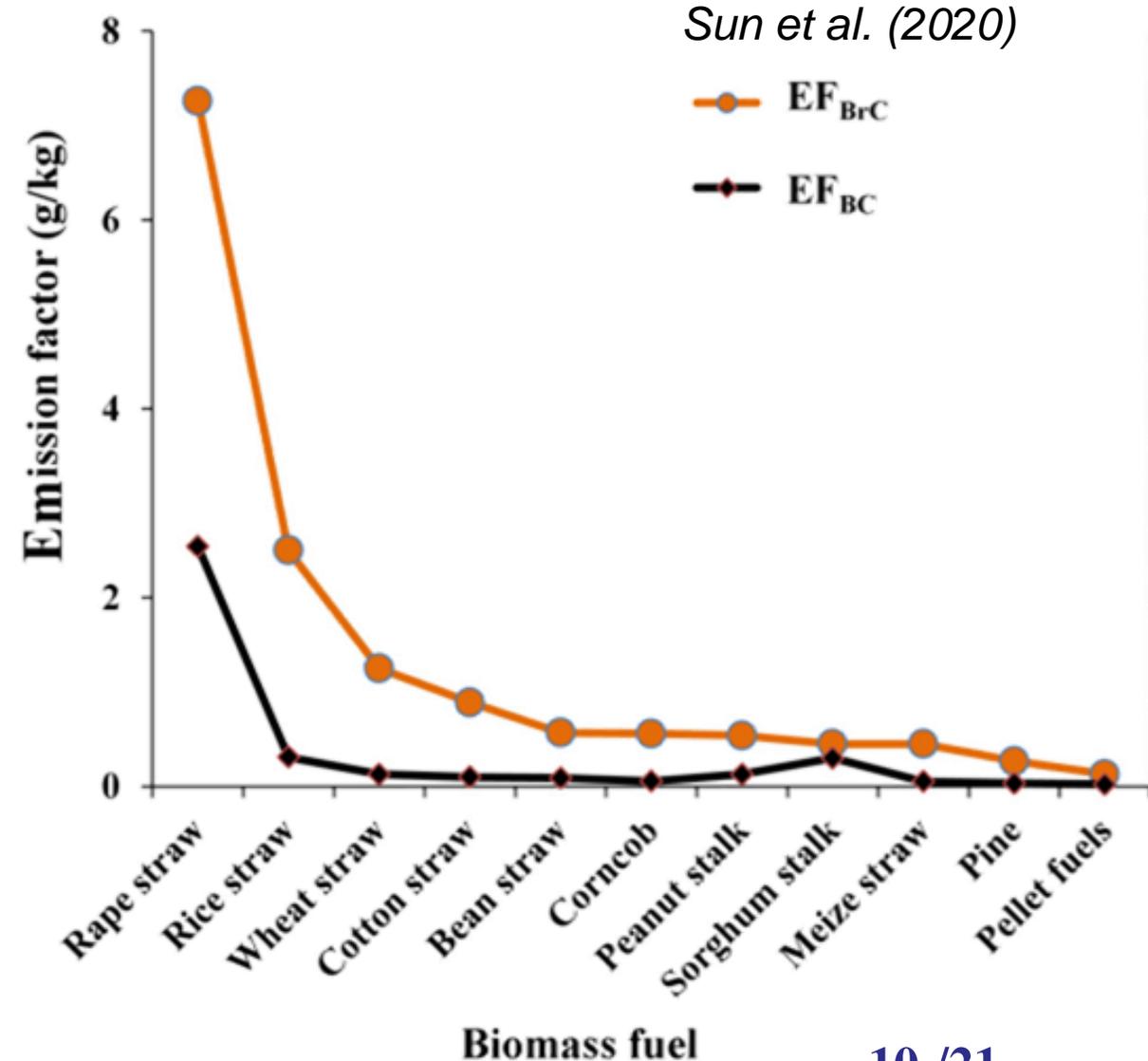
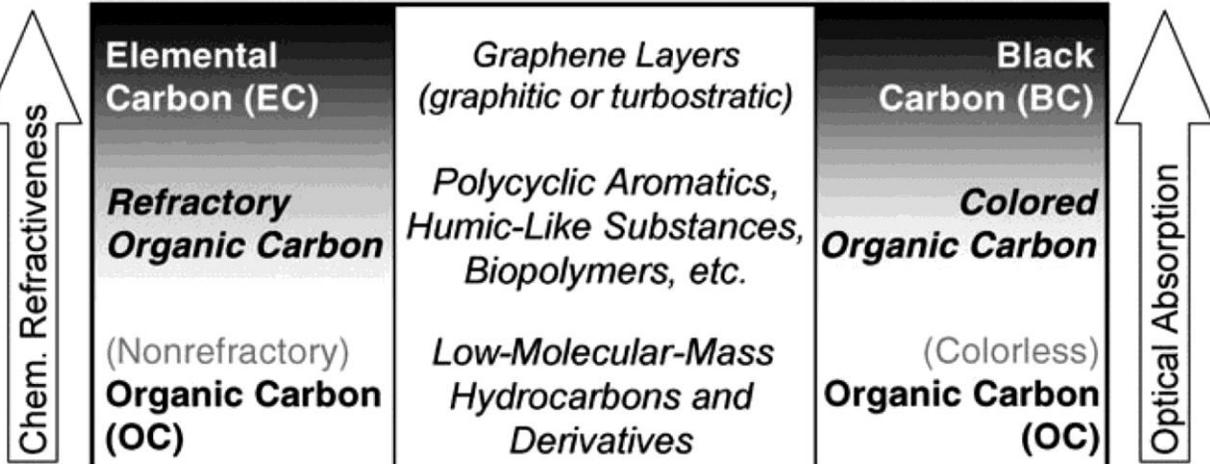
| Stoves | BC (g/GJ) | BC(%PM) |
|--------------|-----------|---------|
| Traditionnal | 32-100 | 4-22 |
| Improved | 30-95 | 25-55 |
| New | 9-30 | 18-30 |
| Pellet | 1.3-4.0 | 10-17 |

Klimont et al. (2017)

Black/Elemental/Brown Carbon

- Black or Brown Carbon?
- BC carbon c/should include BrC and then a part of Organic Matter
 - Possible double counting
 - EC is relevant for health issues
 - BC more relevant for climate impacts

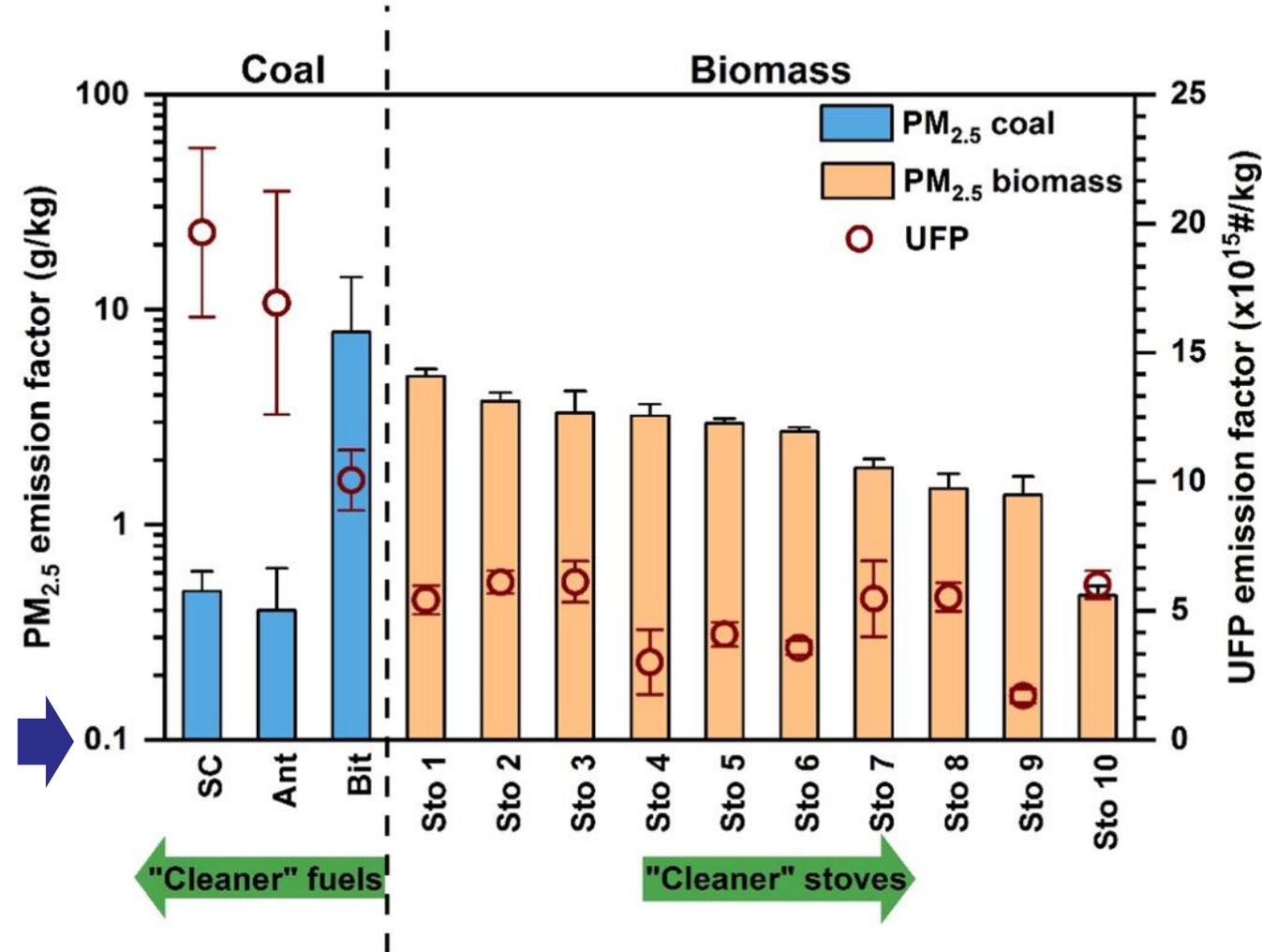
| Thermochemical Classification | Molecular Structures | Optical Classification |
|-------------------------------------|--|---------------------------------|
| Elemental Carbon (EC) | Graphene Layers (graphitic or turbostratic) | Black Carbon (BC) |
| Refractory Organic Carbon | Polycyclic Aromatics, Humic-Like Substances, Biopolymers, etc. | Colored Organic Carbon |
| (Nonrefractory) Organic Carbon (OC) | Low-Molecular-Mass Hydrocarbons and Derivatives | (Colorless) Organic Carbon (OC) |



Laskin et al. (2015)

BC and UFP

- High emissions of **particle mass** concentration often occur at the beginning of the combustion (i.e. the first **30 min** after fire start) while high emissions of **particle number** concentration occur in a later combustion period (60–150 min).
- Notable **antagonism** between reducing PM_{2.5} mass based emissions and reducing ultrafine particle number based emissions among various control strategies that were proposed for reducing pollution from residential combustion
 - Competing processes
nucleation/absorption/coagulation



Wang et al. (2020)

Effect of catalytic combustor

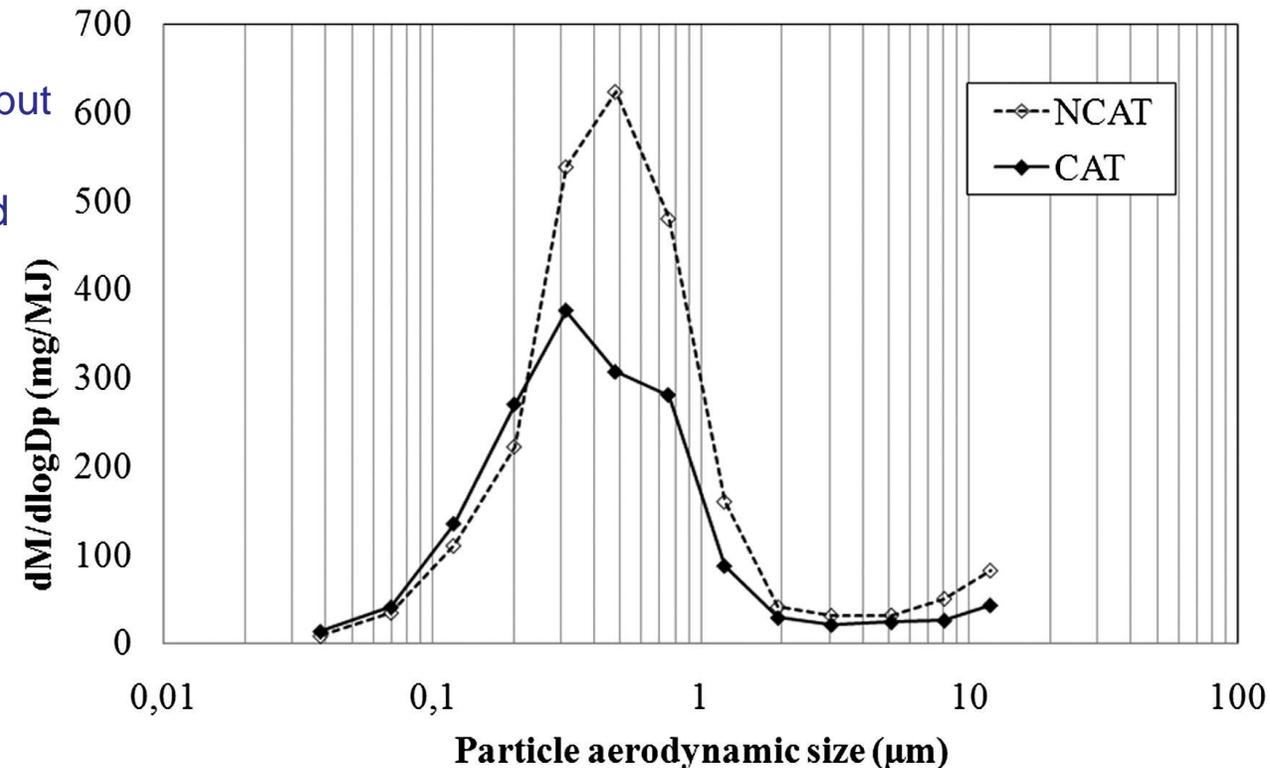
- Reduction of gaseous and particulate emissions from small-scale wood combustion with a catalytic combustor (Sauna stove in Finland)
- **PM1** (particle mass below aerodynamic size of 1 μm) was **reduced by 30%** during the whole combustion cycle.
- During gasification, a **44% reduction of PM1** was achieved but there was no reduction during burn out.
- The **organic and elemental carbon** analyzed from PM1 had reduced also only during gasification by **56% and 37%**, respectively.

However....

- The usage of catalytic converters in RWC is controversial.
- A catalytic converter reduced the adverse products of incomplete combustion such as CO, OGC and **PAHs** on average 26%, 24%, and **24%**, respectively.
- On the other hand, there is a clear **increase** in the concentrations of **PCDD/Fs** (8.7-fold) when the catalytic converter was used

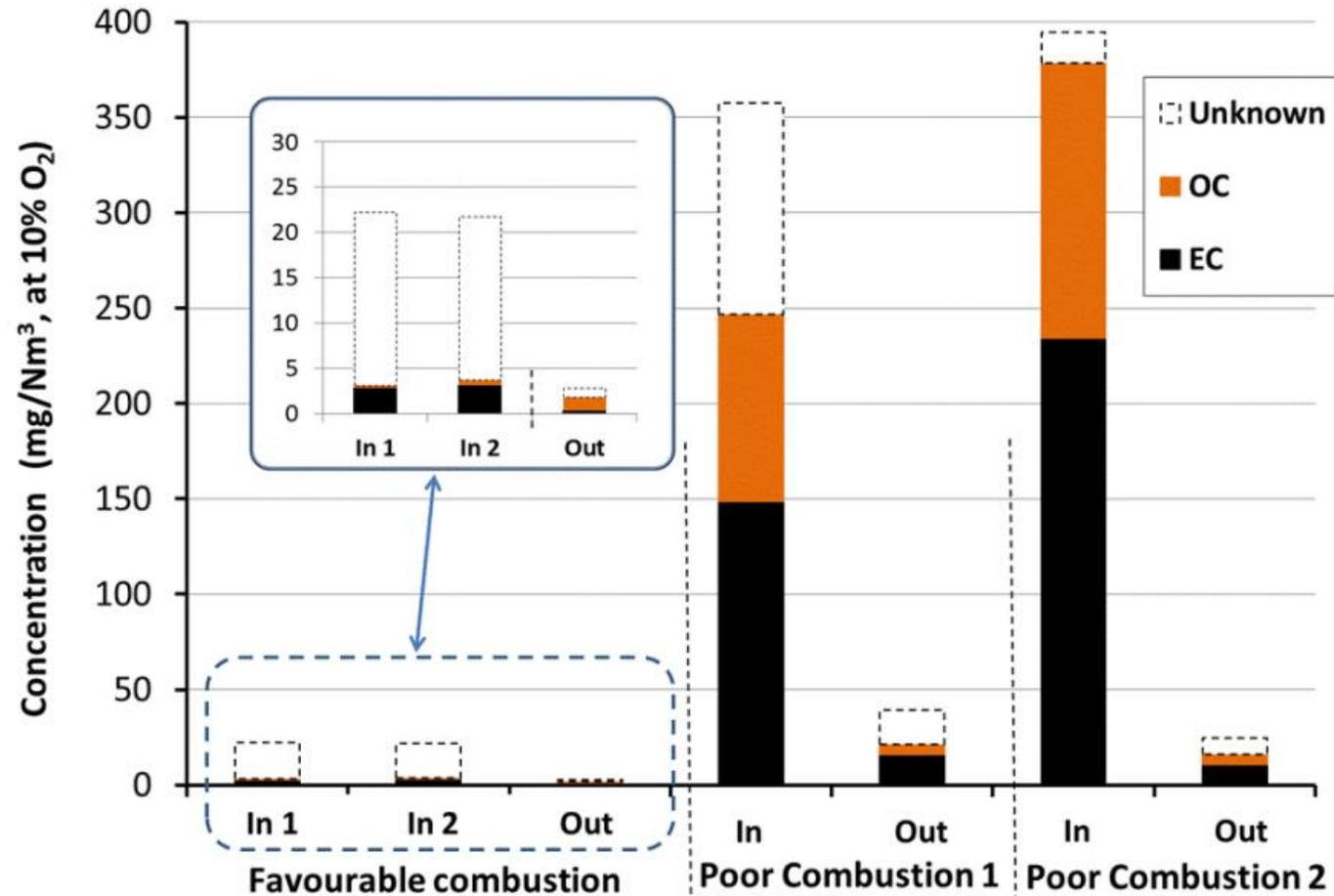
(Kaivosoja et al., 2018)

Hukkanen et al. (2012)



Bäfver et al. (2012)

- Tests on wood Pellet Boiler under favorable and poor combustion
- Reduction efficiency > 90%
- Possible formation of condensable organic matter in the ESP in case of Temperature drop

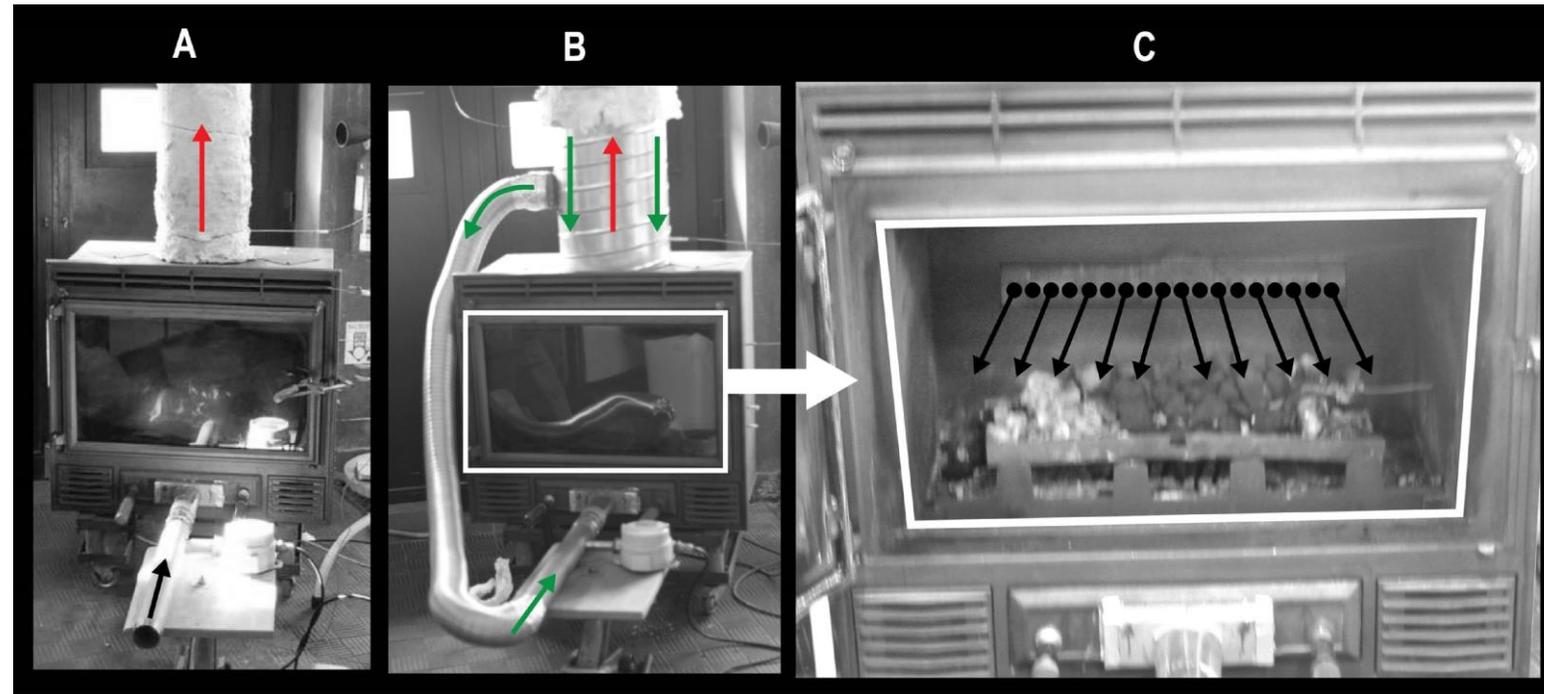


Confirmed by industrial combustion processes (Mertens et al., 2020)

A low cost retrofit solution for conventional wood stoves

- PM EF decrease from 8.9 to 6.9 g/kg_{fuel} but far from the reference limit
- Energy savings involved 30% in cost saving on annuities
- Expected similar performances on BC

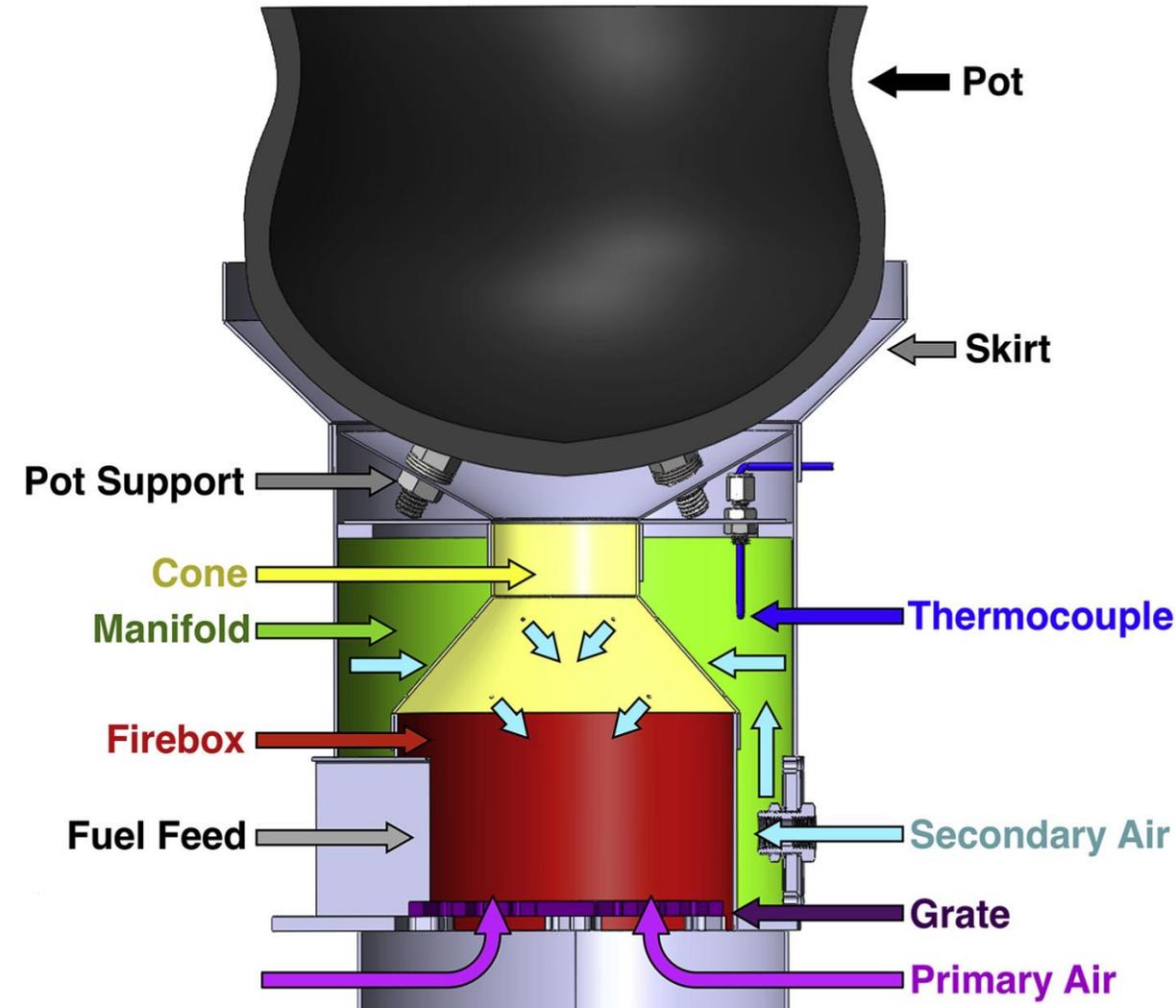
Carvalho et al. (2018)



→ EXHAUST COMBUSTION FLUE GAS → PREHEATED ATMOSPHERIC AIR → ATMOSPHERIC AIR

Secondary air injection in wood-burning cookstoves

- Low-cost (<\$10) fans and blowers are available to drive the secondary flow, and can be independently powered using an inexpensive **thermoelectric generator** mounted nearby.
- The size-resolved PM measurements show that secondary air injection inhibits particle growth, but the total **number of particles generated remains relatively unaffected or can increase.**
- Reduction of mass emissions of particulate matter (PM), carbon monoxide (CO), and black carbon (BC) by at least 90% relative to a traditional cooking fire
- Possible improvements for UFP:
 - Better calibration of air staging and fuel feeding
 - Turbulent conditions



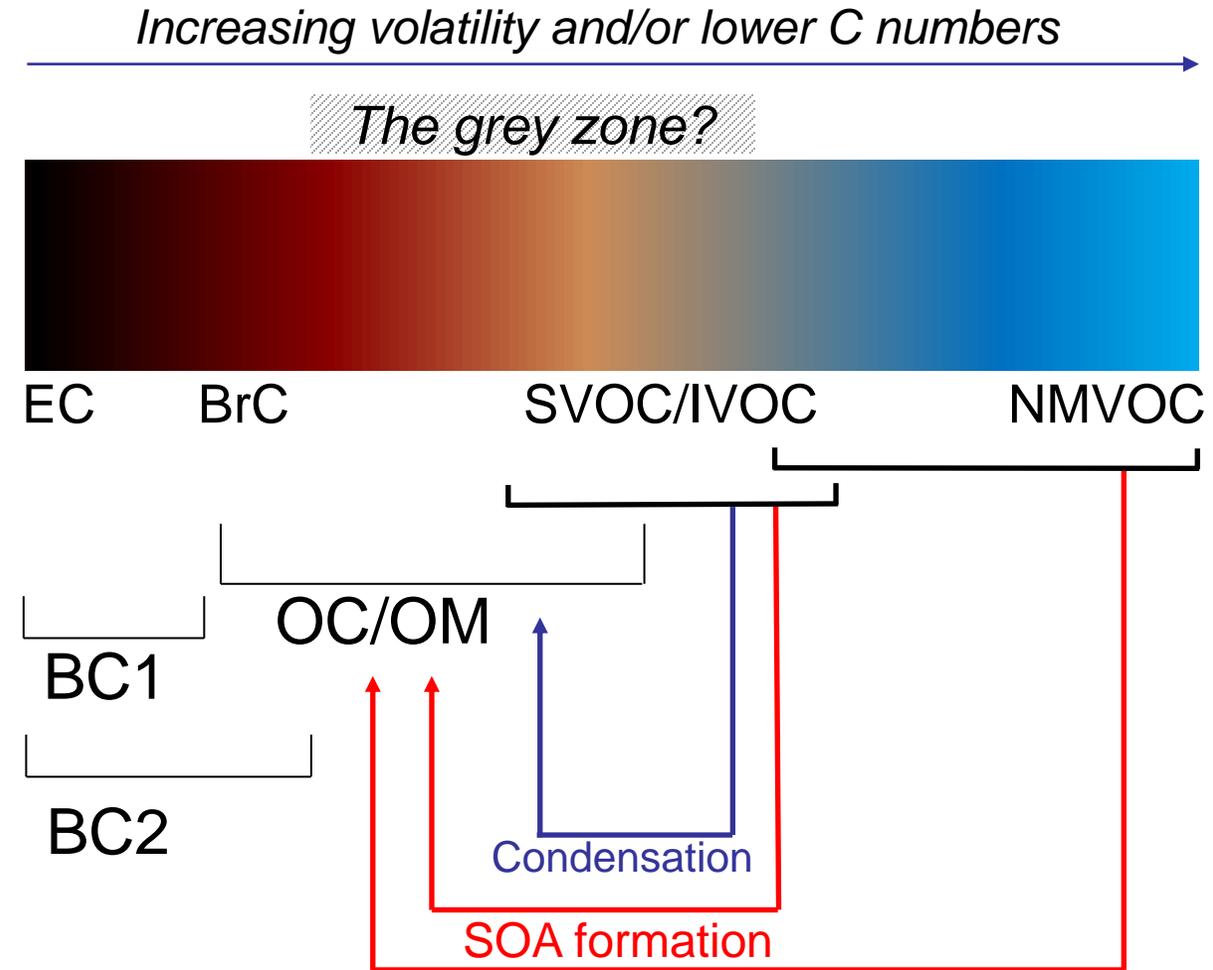
Caupel et al. (2020)

Conclusions I/III



- Large range of BC Emission Factors
- Most BC emissions occur during the ignition stage
- Unsteady state combustion increase all emissions (particularly for short time uses)
- Dry wood better, avoid coniferous type woods for PAH and BC
- Use of pellets decreases PAH but could increase PCDD/F species
- Modern stoves:
 - Use of catalytic converters stoves (mostly used in US, more useful for the condensable part, and limit the SOA formation later)
 - Advanced combustion systems with secondary injections are more common in EU
 - Air staging and fuel feeding could be better calibrated
 - Use of pellet stoves should be encouraged to reduce emissions
 - Use of ESP: useful for the solid fraction of PM (BC), less on the condensable part
 - Retrofit low cost solutions to improve efficiency of more traditional stoves
 - UFP number could not be reduced by advanced technologies abating PM

- **The main critical issue: How to manage BrC in Emission Inventories??**
 - BrC as primary or secondary organic condensable fraction
 - Not included in BC if EF based on thermo-optical methods
 - Partly included in OC or OM
- **Include BrC in BC is relevant for CC policies and would show more co-benefits between AQ and CC policies**
- **The ratio BrC/BC is relevant to know for a good assessment of emission reduction strategies on CC and AQ**
 - Impact of PM abatement technologies on SOA formation and SVOC/IVOCs
 - A crucial point for residential wood burning
 - Implication for CC and AQ mitigation strategies
- **Is BrC a species to be considered at the emission or is it a modelling issue?**
 - The characteristic time of formation is certainly the key to attribute the species to « Emissions » or « AQ model outputs »



BC1 by thermo-optical or pure thermal methods

BC2 by pure optical methods

Conclusions III/III

Key messages

- BC/PAH emission reduced by modern devices
- Not clear for UFP
- A remaining issue on Organic Matter
 - Possible formation of OM in the flue gas
 - BrC-SVOC/IVOC need to be better estimated in emission inventory
 - Emission factor
 - Volatility split (Dave's presentation)
 - Important to better emphasize synergies between CC and AQ policies

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Thank you very much
for your attention!
Questions?

TFTEI Technical Secretariat

