RÉPUBLIQUE FRANÇAISE
Liberté
Égalité
Fraternité

INERIS
maîtriser le risque pour un développement durable

MEASUREMENT OF BLACK CARBON EMISSIONS: FEEDBACK ON MEASUREMENT METHODS FOR STATIONARY SOURCES AND STATE OF KNOWLEDGE ON BC EMISSIONS FROM PELLET STOVES

Isaline Fraboulet, Cécile Raventos,
Serge Collet
isaline.fraboulet@ineris.fr

-

Presentation plan

- Feedback on BC measurement methods for stationary sources, based on results obtained within the Carablack Ademe funded project

- State of knowledge on BC emissions from pellet stoves, based on results obtained from several Ademe funded projects carried out by Ineris and bibliography search

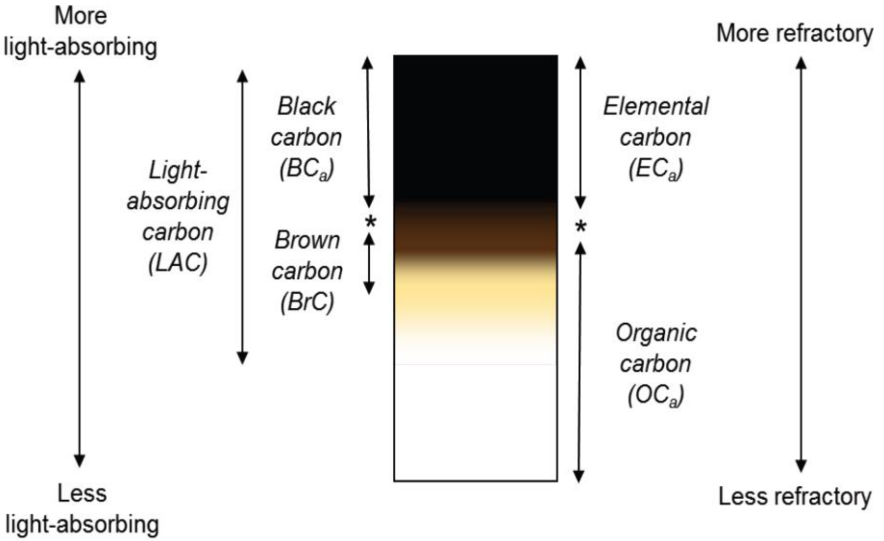
Some terms and definitions

The Scientific Advisory Group for Aerosols of the Global Atmosphere Watch program of the World Meteorological Organization (WMO) recommends:

Use BC as a qualitative term whatever the measurement method is, but use:

- ✓ **Equivalent Black Carbon (EBC) when optical methods are used.** The absorption rate of the aerosol is converted in EBC with the Mass Absorption Coefficient (MAC). The MAC should be always reported
- ✓ **EC Element Carbon when thermo-optical methods are used**

Optical Classification | **Thermal Classification**



« Report to congress on black carbon » US EPA - 2012

1. Feedback on measurement methods for stationary sources

Context

- Feedback based on the Carablack project (2015-2018)
  - ADEME (The French Environment and Energy Management Agency) funded project,
  - coordinated by INERIS (French National Institute For Industrial Environment And Risks) in partnership with CITEPA with the collaboration of LSCE (French Laboratory for Sciences of Climate and Environment) and industrial sites where trials were performed

- Feedback would need to be updated in terms of new studies carried out/ methods evaluated

- Characterisation of BC in ambient air as requested by Directive 2008/50/CE

  → One measurement method standardised in EU since 2017: EN 16909

- But absence of requirement for BC measurement at the emission of stationary sources; no emission limit value (ELVs)

  → Limited emission measurement data from stationary sources

  → Measurement methods rarely described: comparability of methods and results?

  → No standardised methods

Carablack, objectives and methods

- **To identify the measurement methods of black carbon in stacks:**
  - Already applied for the characterisation of stationary source emissions
  - Or those applied for ambient air but which could be adapted for emission

  ➔ State of the art on black carbon measurement methods

- **To assess the different measurement methods selected during a campaign of trials on INERIS bench test:**
  - Implementation
  - Performances
  - Comparison of results obtained during measurements on a same BC/PM matrix
  - Assessment of the size distribution of black carbon particles

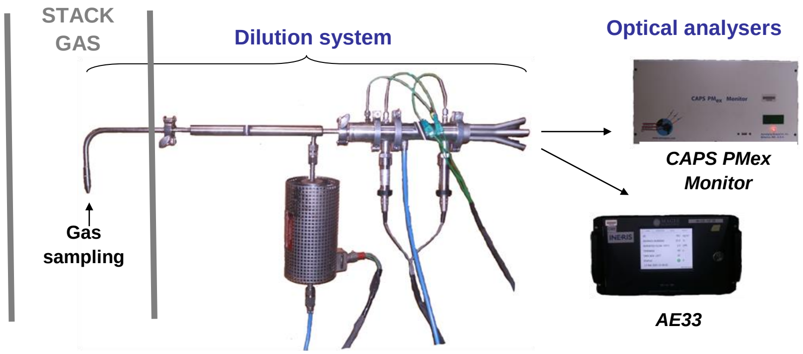
- **Measurement campaigns performed on 2 biomass boilers for heat and hot water production (capacity 3.5 and 5.5 MW): selected in a sector identified as an important contributor of black carbon emissions in France**

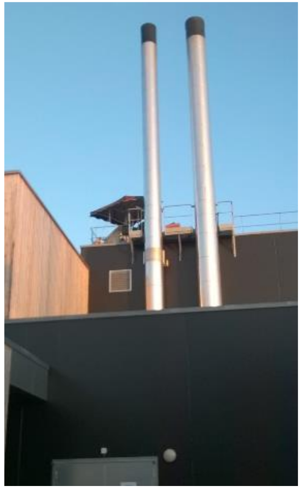
RÉPUBLIQUE FRANÇAISE
Liberté
Égalité
Fraternité

INERIS
maîtriser le risque
pour un développement durable

Carablack, measurement methods assessed

- Optical methods downstream diluter (automatic method)





- Thermal and thermo-optical methods
  - **Sampling systems implemented:**
    - Sampling system of total particles onto filter
    - Impactor to determine size distribution: fractions PM_{10} and $PM_{2.5}$
  - **Analycal methods tested:**
    - Thermal analysis: **method 2-Steps** developed by Cachier et al. (1989)
    - Thermo-optical analysis (method standardised for ambient air → EN 16909 since 2017)

Lessons learned from trials on bench test and on site

➢ Measurement methods assessed

- **Good repeatability of each method**
- **BUT ⚠ : significant differences between results obtained from optical/thermal/thermo-optical methods** (differences also observed in ambient air)

  ➔ Importance to specfiy the method, the protocol and the equipment used together with the results obtained

  Comparison of thermo-optical and optical methods

  Relatively good agreement during trials on bench tests:

  ➔ BC(optical method)/BC(thermo-optical method) = **between 0.6 and 2.8**

  But not so good on site:
  ➔ BC(optical method)/BC(thermo-optical method) = **between 0.1 and 6.2**

- **Difficulties due to lack of reference material**

Conclusions and perspectives

Conclusions

- No measurement tested method entirely satisfactory
- Results by the methods tested show differences
- No simple correction factor can be applied to the different methods
- Importance to specify the method, the protocol and the equipment used together with the results obtained
- Methods not to be used on a routine basis for regulatory purposes, more adapted to research activities

Perspectives

- Feedback necessary on new methods: hybrid methods (incandescence and photo acoustic methods)
- More comparisons on emission matrices needed
- Development of a reference material required
- Selection of one method necessary to harmonize the practices
- Once a method is selected, validation/pre-standardization/standardization work preferable prior to setting up ELVs

2. State of knowledge on BC emissions from pellet stoves and boilers

Formation of Black Carbon in Combustion Processes

Formation at High Temperature (above 900°C) in Hydrocarbon Flames

Formed from polycyclic aromatic hydrocarbons (PAHs) with high molecular weights, ranging from 276 to 520 g/mol. These PAHs transform into solid particles called crystallites. When they aggregate, these crystallites form spherical particles with diameters ranging from 10 to 50 nm (in their pure state).

In an environment rich in soot particles, these particles coagulate with each other to form larger spherical particles. Further along in the flame, they adhere together to form large aggregates.

Initially, they contain only carbon and hydrogen. The presence of oxygen and nitrogen appears later on more mature soot particles, which have the capacity to adsorb other molecules.

Formation of Black Carbon in a flame

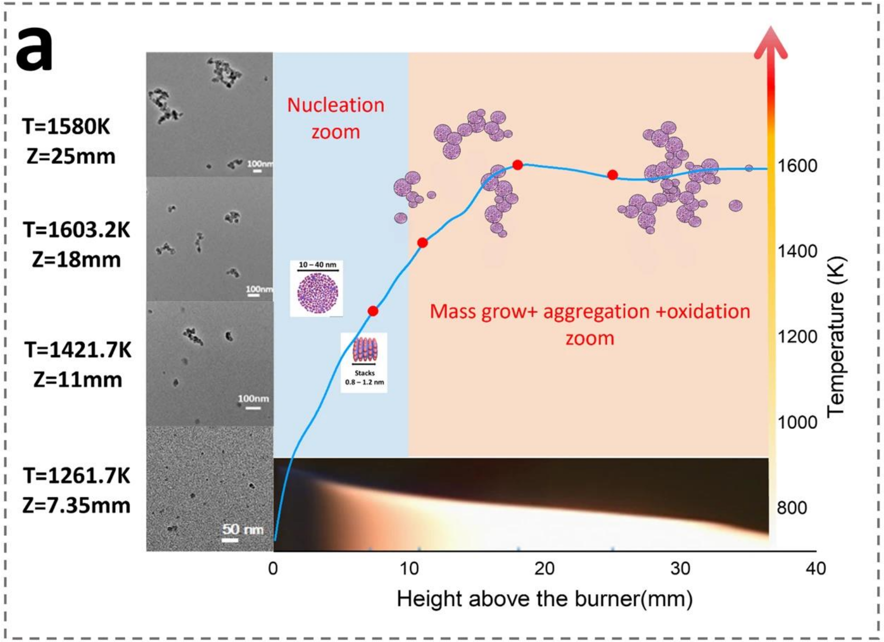
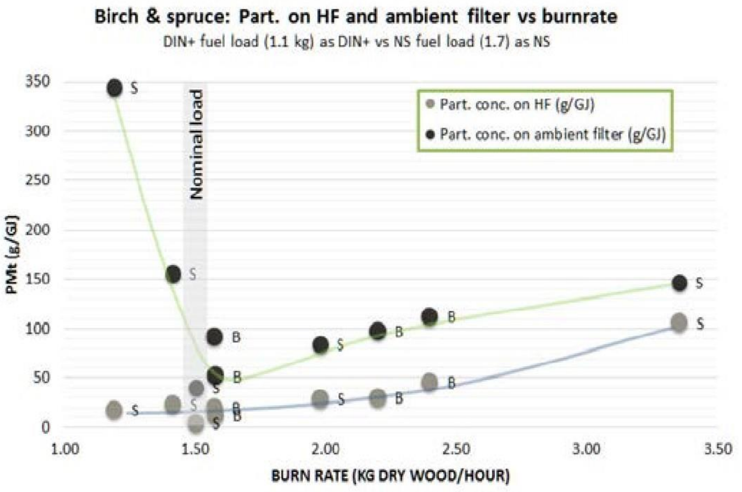


Figure 16 : blue line (a) corresponds to the flame temperature profile and the red dots to the sampling positions. TEM images of the samples taken at each sampling point are shown. (Shao et al. 2023).

Factors Influencing Emissions

Air Deficiency at High Temperature (overload)

Poor mixing of pyrolysis gas and oxidant (O_2) leads to a local air deficiency.



Seljeskog et al. 2017, values represented in the form of a graph by Nussbaumer 2017

RÉPUBLIQUE FRANÇAISE
Liberté
Égalité
Fraternité

INERIS
maîtriser le risque
pour un développement durable

Particle composition

Modern appliances	Stoves and boilers (< 50 kW)
Fuel	Pellets
Particle emission factor	≈ 30 g/GJ
Organic matter	25-50%
BC	5-20%
Inorganic compounds (potassium salts, sodium salts, metals, etc.)	30-60%

Emission factors

Average Emission Factor for pellet stoves: eBC ≈ 2 g/GJ (≈ 30 g/GJ for particles)
eBC (optical method) is weakly correlated with BC (thermo-optical method)

For automated appliances, BC emissions are very low.

For pellet and wood chip boilers (< 500 kW), BC < 15% of total particles (< 2% with optimized combustion).

However, a significant number of pellet stoves experience malfunctions (local air deficiency), leading to sharp increases in BC: example 67 g/GJ, Bertrand et al. 2017 (similar results found in other studies: Epochag 2023, etc.).

RÉPUBLIQUE FRANÇAISE
Liberté
Égalité
Fraternité

INERIS
maîtriser le risque
pour un développement durable

Bibliography

Recent studies:

- **PerfPAG 2022 (pellet stoves, https://librairie.ademe.fr/)**

- **Epochag 2023 (pellet stoves and boilers, https://librairie.ademe.fr/)**

- Bertrand A., Stefenelli G., Bruns E.A., Pieber S.M., Temime-Roussel B., Slowik J.G., Prévôt A.S., Wortham H., El Haddad I., Marchand N. (2017). Primary emissions and secondary aerosol production potential from woodstoves for residential heating: Influence of the stove technology and combustion efficiency. Atmos. Environ. 169, 65-79, https ://doi.org/10.1016/j.atmosenv.2017.09.005.
- Nussbaumer T. (2017). Aerosols from biomass combustion. Technical report on behalf of the IEA Bioenergy Task 32.
- Seljeskog M., Alexis Sevault A., Ostnorb A., Skreiberg O. (2017). Variables affecting emission measurements from domestic wood combustion. Energy Procedia 105, 596-603.
- Shao C., Wang Q., Zhang W. et al. (2023). Elucidating the polycyclic aromatic hydrocarbons involved in soot inception. Commun. Chem. 6, 223, https://doi.org/10.1038/s42004-023-01017-x.

Thank you for your attention