Primary and secondary emissions of pellets, logwood, and oil residential heating appliances: emissions factors, secondary particle formation and particle effective density

A. Albinet, C. Degrendele, S. Collet, A. El Mais, B. Temime-Roussel, B. D'Anna and H. Wortham

alexandre.albinet@ineris.fr

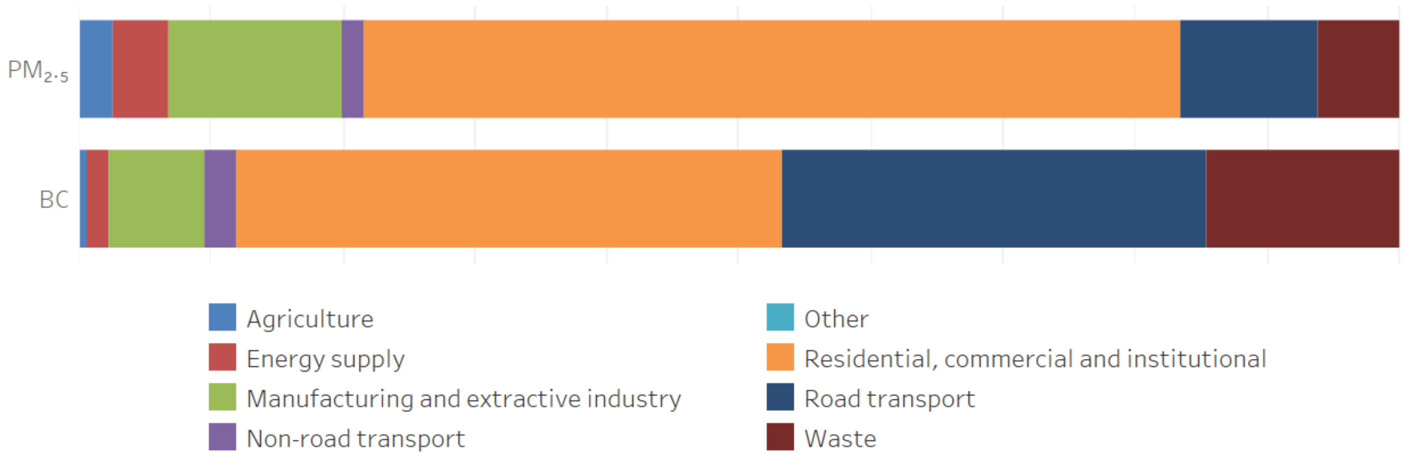
27th ETH Nanoparticles Conference, 12th June 2024

Biomass burning: a strong impact on air quality

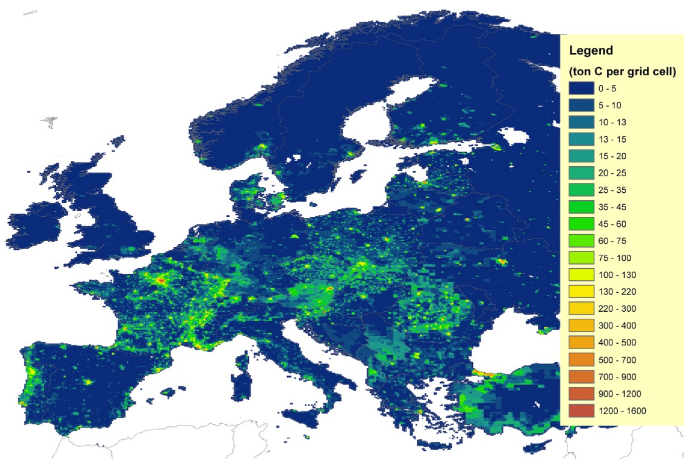
Residential wood combustion

⇒ Significant source of PM and black carbon (BC) in Europe

Sector contributions to PM emissions in Europe



(EEA, 2021)



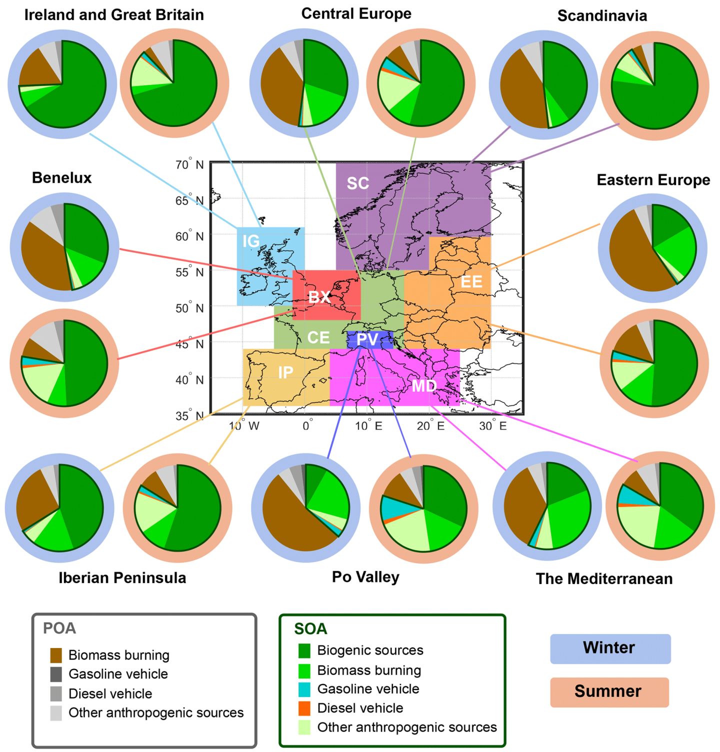
Spatial distribution of the OC emissions due to residential wood combustion in 2005

(Denier van der Gon, 2015)

Primary and secondary biomass burning emissions

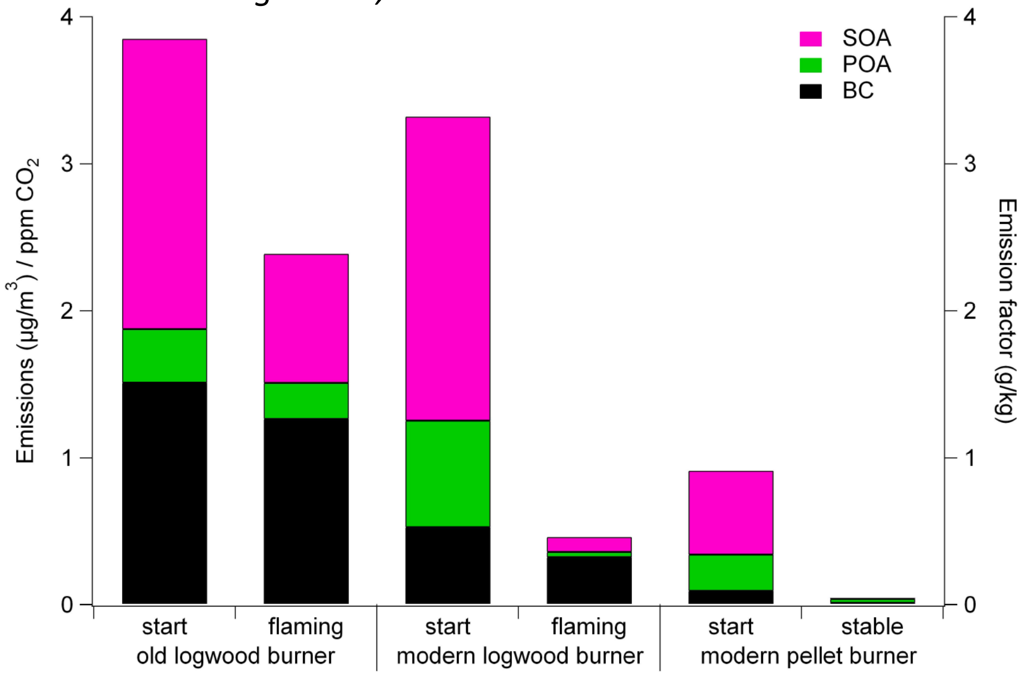
- ✓ Large emissions of BC and primary organic aerosols (POA)
- ✓ Large emissions of volatile and semi-volatile organic compounds (VOCs and SVOCs)

⇒ Significant formation of secondary OA (SOA)



Jiang et al., 2019

Heringa et al., 2011



RÉPUBLIQUE FRANÇAISE
Liberté
Égalité
Fraternité

INERIS
maîtriser le risque
pour un développement durable

Primary and secondary biomass burning emissions

✓ Large emissions of BC and primary organic aerosols (POA)

✓ Large emissions of volatile and semi-volatile organic compounds (VOCs and SVOCs)

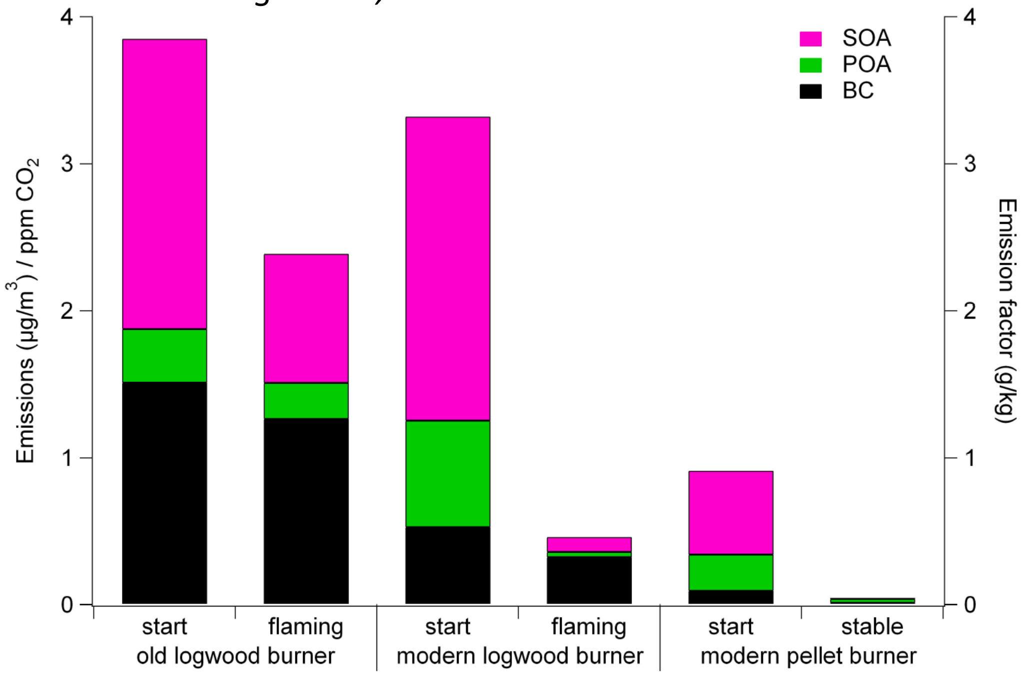
⇒ Significant formation of secondary OA (SOA)

❑ Large primary emissions and SOA formation for old logwood stoves

❑ Lower primary emissions and SOA formation for modern logwood and pellets stoves

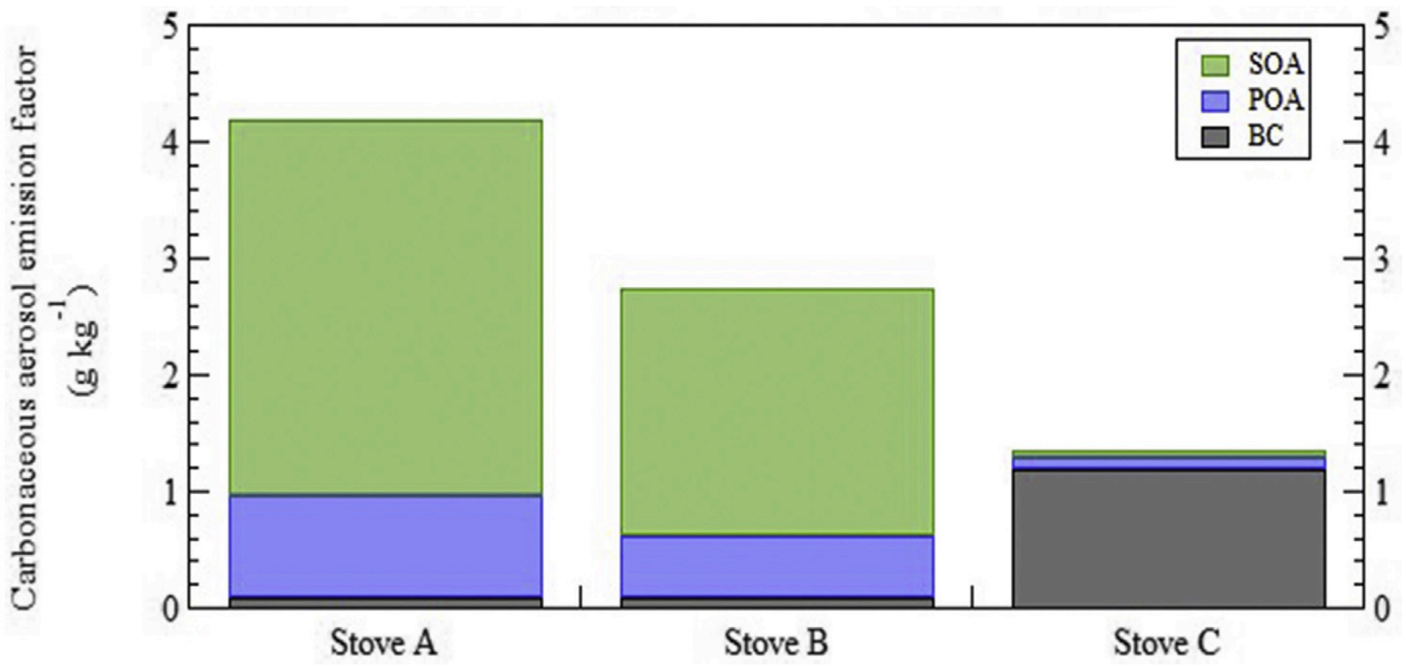
Only few studies about the secondary particle formation from pellets appliance emissions (n ≈ 8)

Heringa et al., 2011



Primary and secondary wood combustion emissions

Bertrand et al., 2017



Woodlog	Woodlog	Pellets
<2002	2010	2010

- ❑ Lower primary emissions and SOA formation for the modern logwood stove
- ❑ Low POA emission and low SOA formation but very high BC emissions from the pellets stove

Only few studies on the BC emissions from pellets appliances (n ≈ 6)

Main objectives

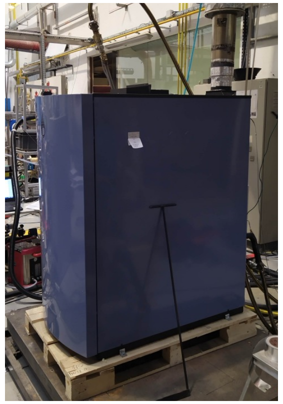
1) Evaluation of primary and secondary emissions from different modern pellets stoves and boilers
   a) Impact of working outputs
   b) Impact of pellets composition (softwood *vs* hardwood)

2) Comparison with primary and secondary emissions from residential heating appliances using other fuels (logs and oil)

Residential heating appliances tested

- 3 × Pellets Wood Stove (PWS1 - 8 kW; PWS2 - 8 kW; PWS3 - 9 kW)
- 3 × Pellets Wood Boiler (PWB1 - 24.5 kW; PWB2 - 22 kW; PWB3 - 21.7 kW)
- 1 × LogWood Stove (LWS - 7 kW)
- 1 × LogWood Boiler (LWB - 30.5 kW)
- 1 × Condensing Oil Boiler (OB - 24 kW)




Experimental conditions

Outputs

Pellets: 3 conditions (PWS1 and PWB1)

- Reduced (30 %)
- Intermediate (50 %)
- Nominal (100 %)

Logwood: 2 conditions

- Nominal
- Reduced

Oil: 1 condition

- Nominal

Fuels

Pellets: 2 types

- Softwood (conventional)
- Hardwood (GRAMIX project)
  (PWS3 and PWB2)

Logs

- Beech or Oak (LWS)
- Wooden charm (LWB)

Experimental conditions

Outputs

Pellets: 3 conditions (PWS1 and PWB1)

- ~~Reduced (30 %)~~
- Intermediate (50 %) (PWS2 and PWS3)
- Nominal (100 %)

Logwood: 2 conditions

- Nominal
- Reduced

Oil: 1 condition

- Nominal

Dysfunction observed

Fuels

Pellets: 2 types

- Softwood (conventional)
- Hardwood (GRAMIX project) (PWS3 and PWB2)

Logs

- Beech or Oak (LWS)
- Wooden charm (LWB)

Experimental conditions

Outputs

Pellets: 3 conditions (PWS1 and PWB1)

- Reduced (15 %) (PWB3)
- Intermediate (50 %)
- Nominal (100 %)

Logwood: 2 conditions

- Nominal ⇒ boiler
- ~~Reduced~~

Oil: 1 condition

- Nominal

Fuels

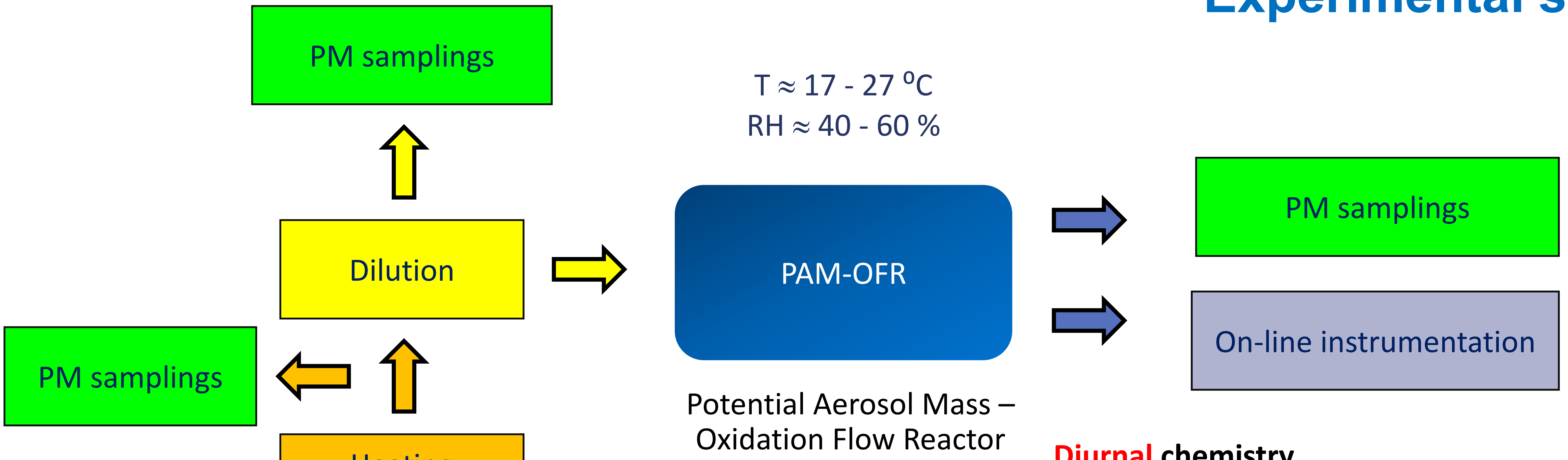
Pellets: 2 types

- Softwood (conventional)
- Hardwood (GRAMIX project) (PWS3 and PWB2)

Logs

- Beech or Oak (LWS)
- Wooden charm (LWB)

Experimental setup



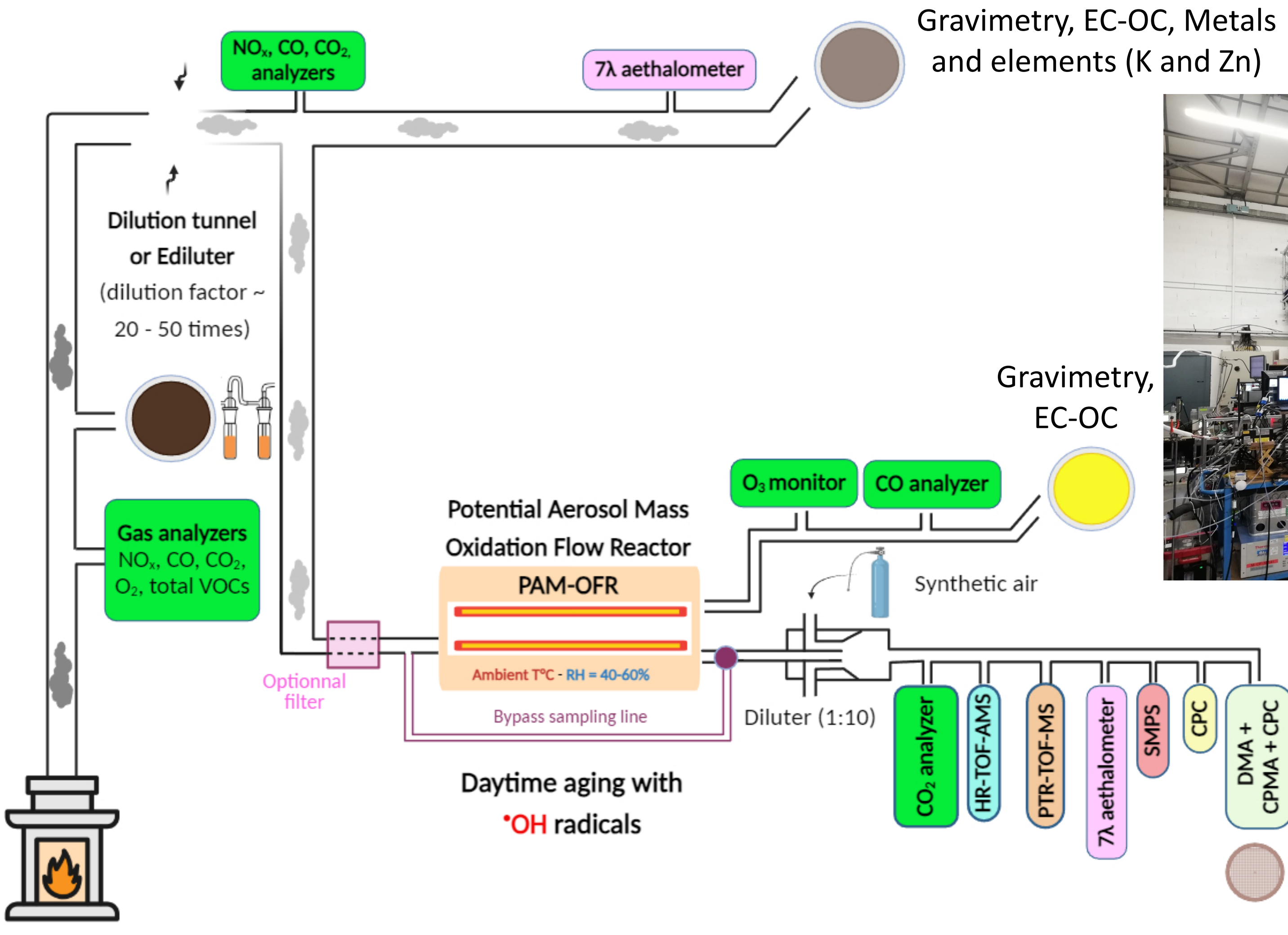
Diurnal chemistry

OH radicals

$O_2 + h\upsilon\ (185\ nm) \rightarrow 2O(^3P)$
$O(^3P) + O_2 \rightarrow O_3$
$O_3 + h\upsilon\ (254\ nm) \rightarrow O(^1D) + O_2$
$O(^1D) + H_2O \rightarrow 2OH$

OH exposure: 3.5×10^{11} - 1.2×10^{12} molecules cm^{-3} s (≈ 4.0 - 13.6 eq. aging days)

Experimental setup



INERIS

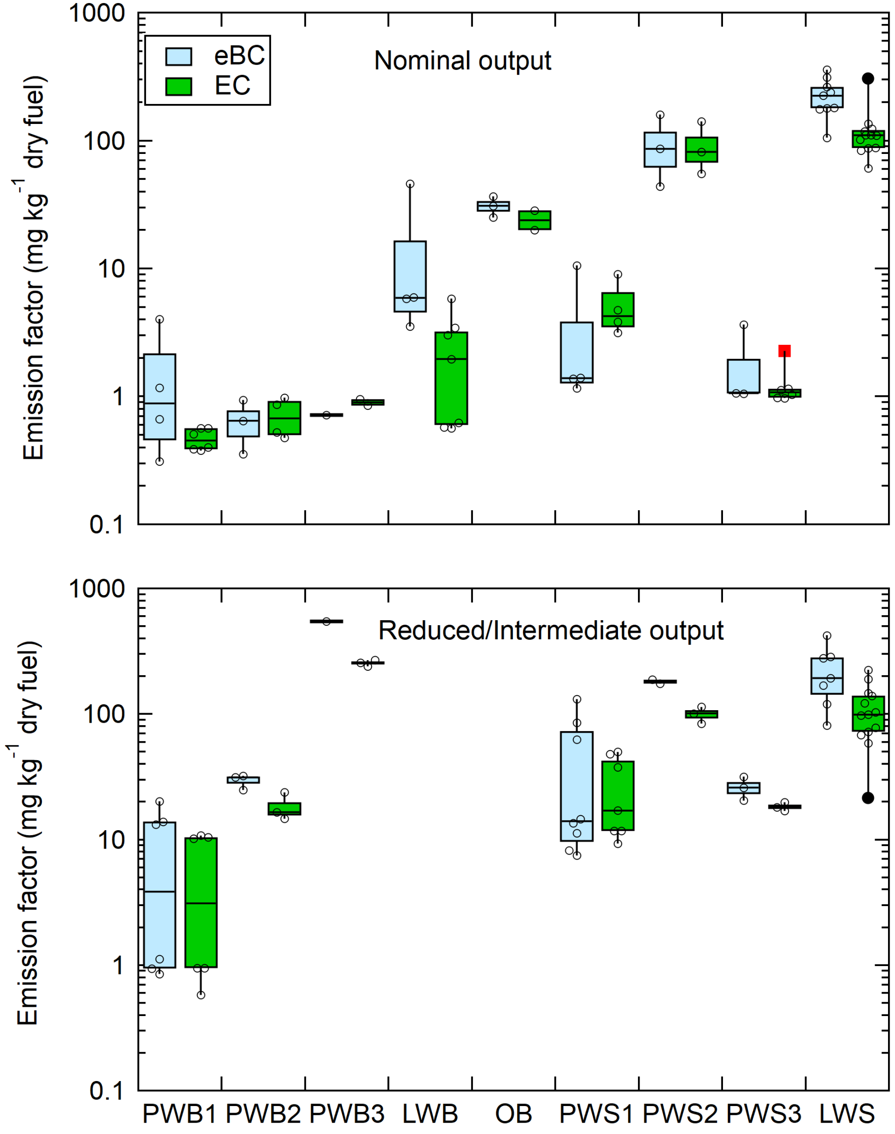
PWS, LWS
01-02/2021, 11-12/2021

CETIAT ensemble, innover et valider

PWB, LWB, OB
09-11/2021

n = 3-14 experiments by tested condition

BC emissions



<u>2 measurement methods</u>

- Equivalent BC (eBC) : aethalometer, absorbance at 880 nm
- EC : thermo-optical method following filter sampling

⇒ No possible direct comparison

OB, large emissions ⇒ dysfunction

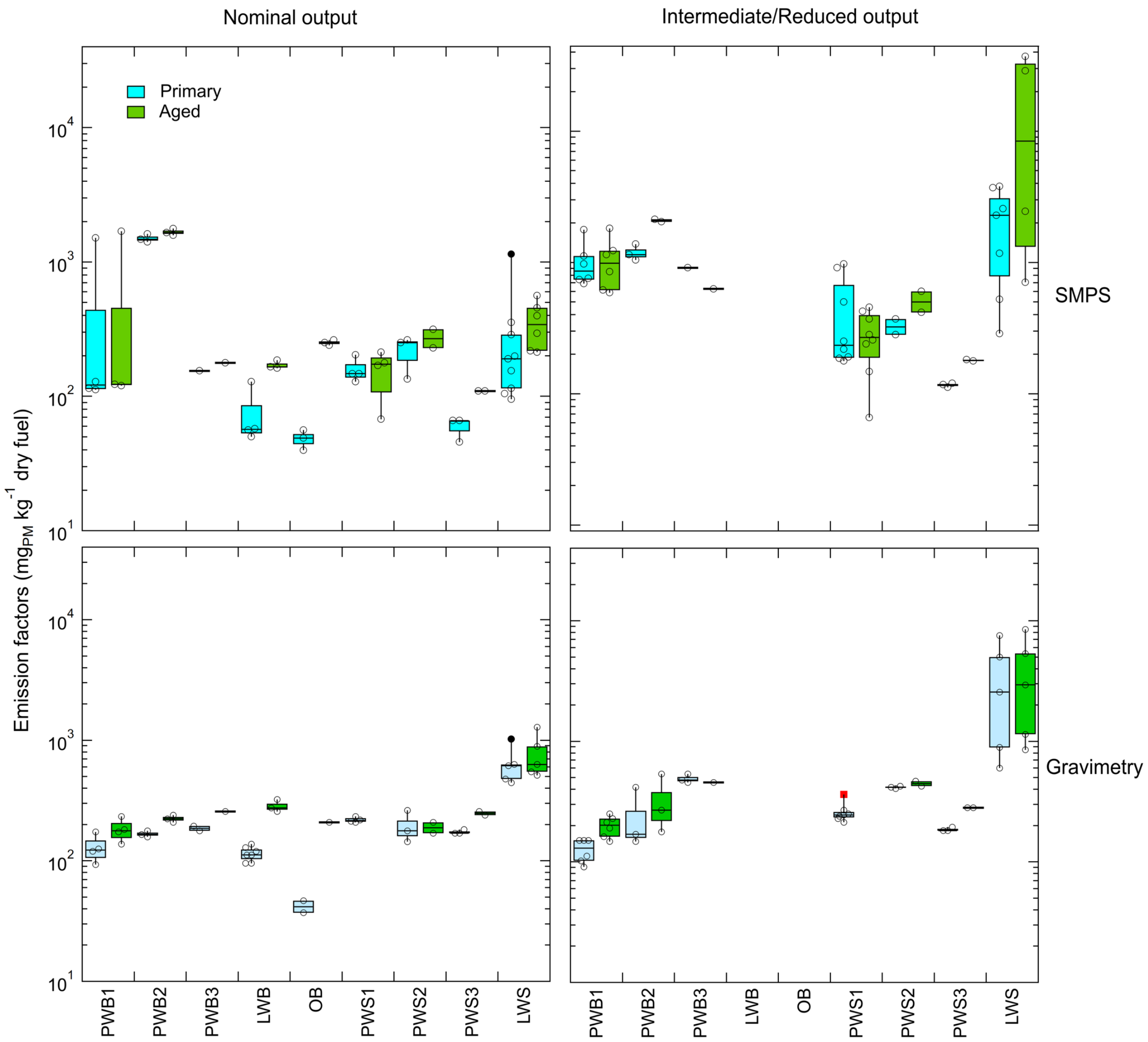
Large emissions for PWB3 (15%) and PWS2 ≈ LWS

PWB << LWB ≈ PWS (except PWS2) << LWS

Reduced/Intermediate output > Nominal output for pellets

Reduced output ≈ Nominal output for LWS

PM emission factors (EF): primary *vs* aged

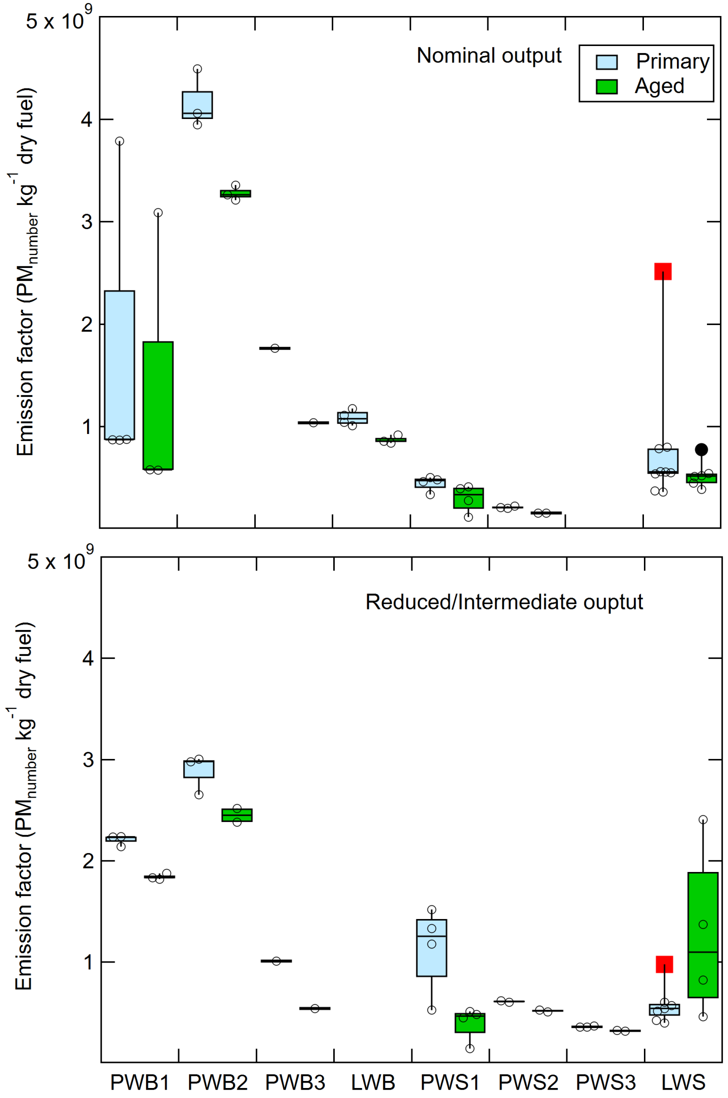


Aged PM > or >> Primary PM

Secondary particles formation

- ✓ OB: High formation
- ✓ Pellets appliances: Low formation
- ✓ LWB and LWS: Low to very high formation (reduced output)

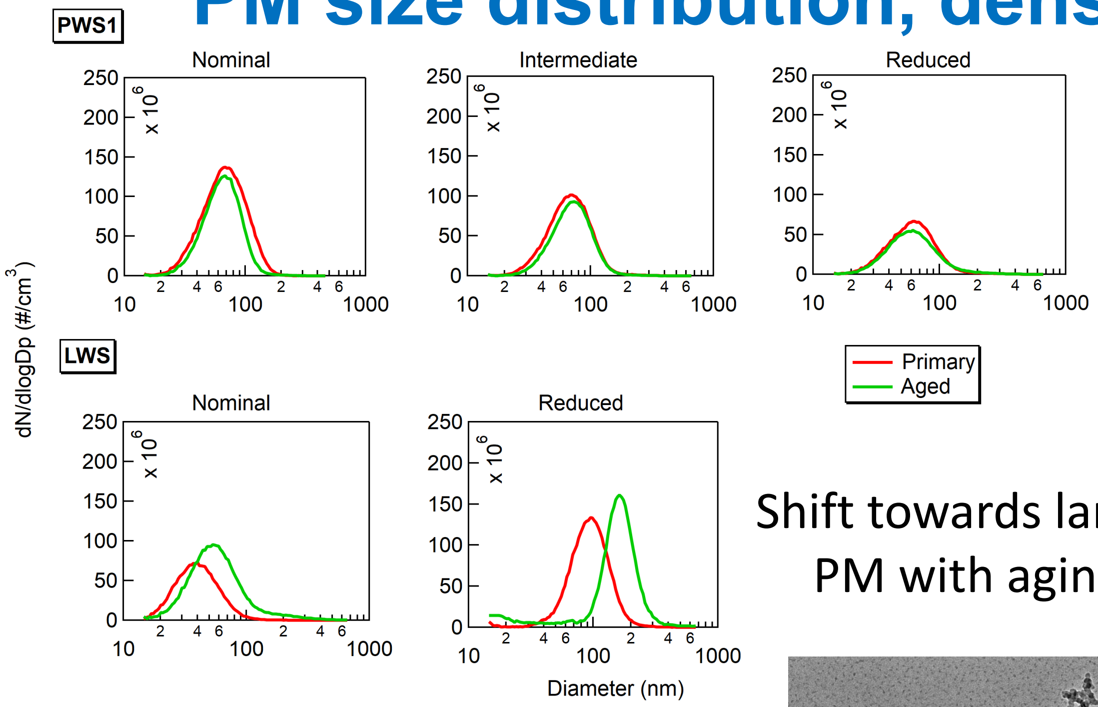
PM number emission factors (EF): primary *vs* aged



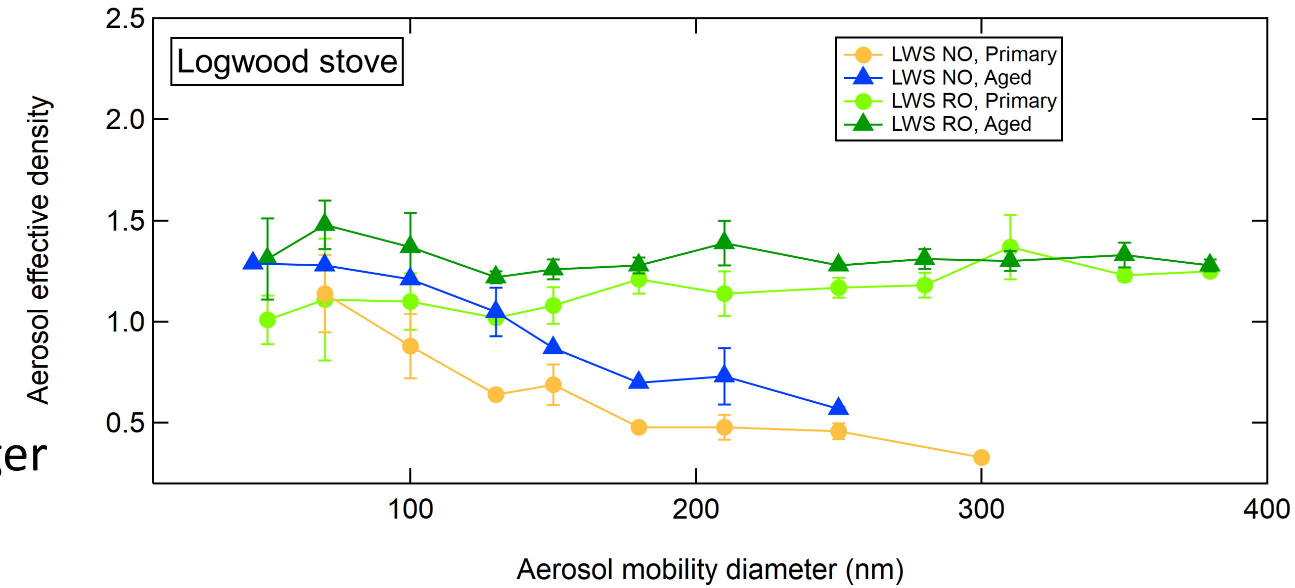
↘ of PM number with aging (except LWS in reduced output)

Condensation processes of semi-volatiles species and PM coagulation within the PAM-OFR

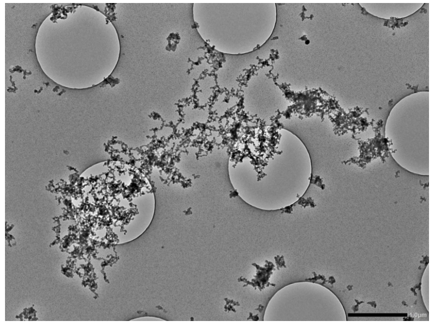
PM size distribution, density and morphology: primary *vs* aged



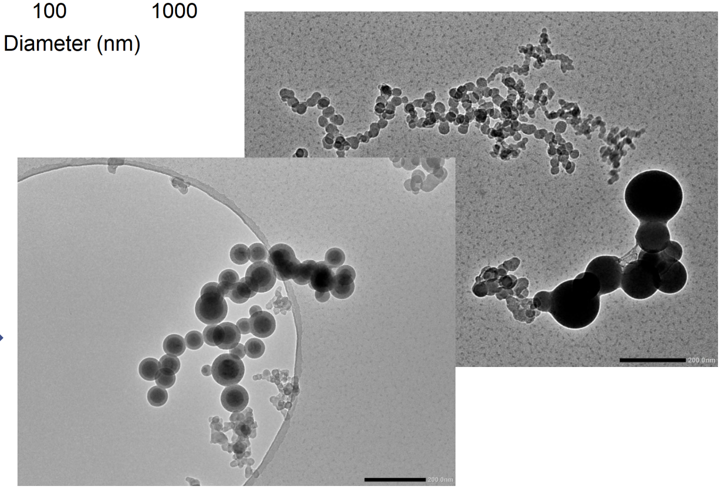
Shift towards larger PM with aging



↗ PM density with aging



Aging



Soot + few tarballs

Soot + Numerous individual tarballs or agregates

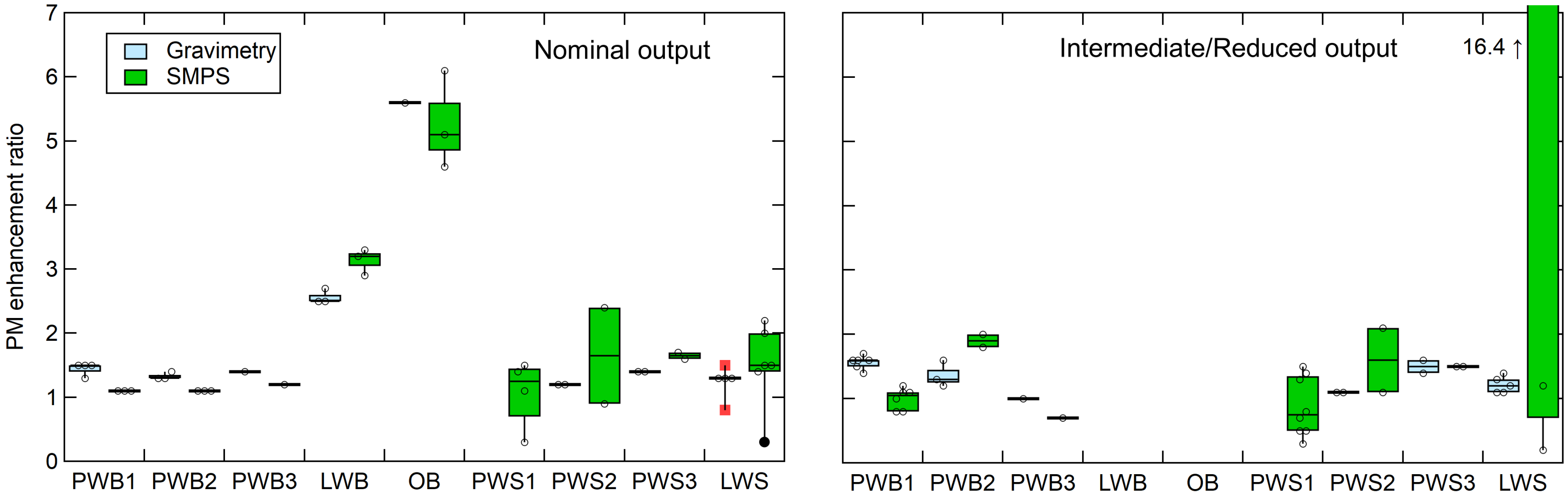
Output	*Primary PM*	*Aged PM*	*Secondary PM (nucleation)*
Logwood stove			
Nominal	0.5	0.8	1.5
Reduced	1.2	1.3	1.5
Pellets stoves			
Nominal/Intermediate	1.2	1.6	nd
Reduced	1.0	1.1	nd
Logwood boiler			
Nominal	1.0	1.4	1.5
Pellets boilers			
Nominal/Intermediate/Reduced	1.9	1.9	nd
Reduced (15 %)	1.0	1.0	nd
Oil boiler			
Nominal	0.8	1.2	1.5

RÉPUBLIQUE FRANÇAISE
Liberté
Égalité
Fraternité

INERIS
maîtriser le risque pour un développement durable

Secondary PM formation potential

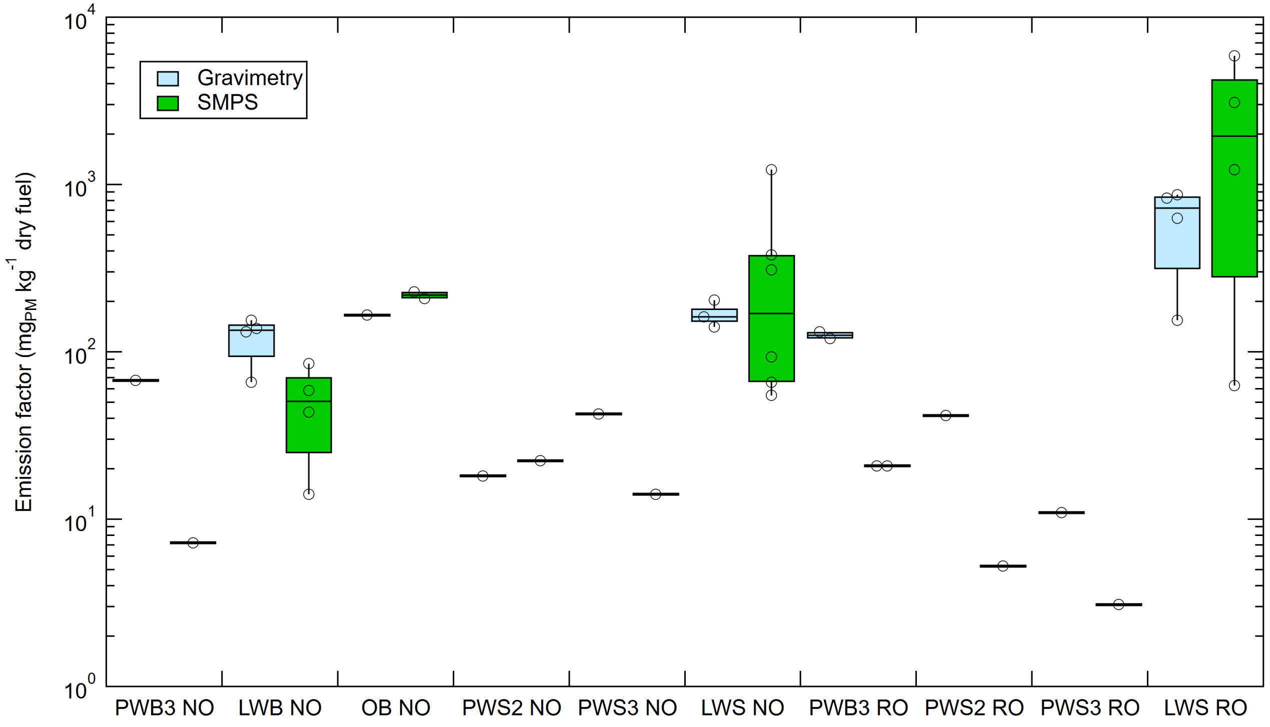
PM enhancement ratio = Aged PM / Primary PM



- ✓ LWS ≈ 1 - 16
- ✓ OB ≈ 5 - 6
- ✓ LWB ≈ 3
- ✓ Pellets appliances ≈ 1 - 2 (PWB and PWS)

LWS > OB > LWB > PWB and PWS

Secondary PM formation by nucleation processes



OB: High SO_4 formation (≈ 50 % of PM)

⇒ SO_2 converted in sulfate

Significant nitrate fraction for LWB while NO_x emissions ≈ other appliances ⇒ nitro organic species

EF nucleation processes ≈ EF primary emissions

EF nucleation processes < EF aged emissions (× 2 - 10)

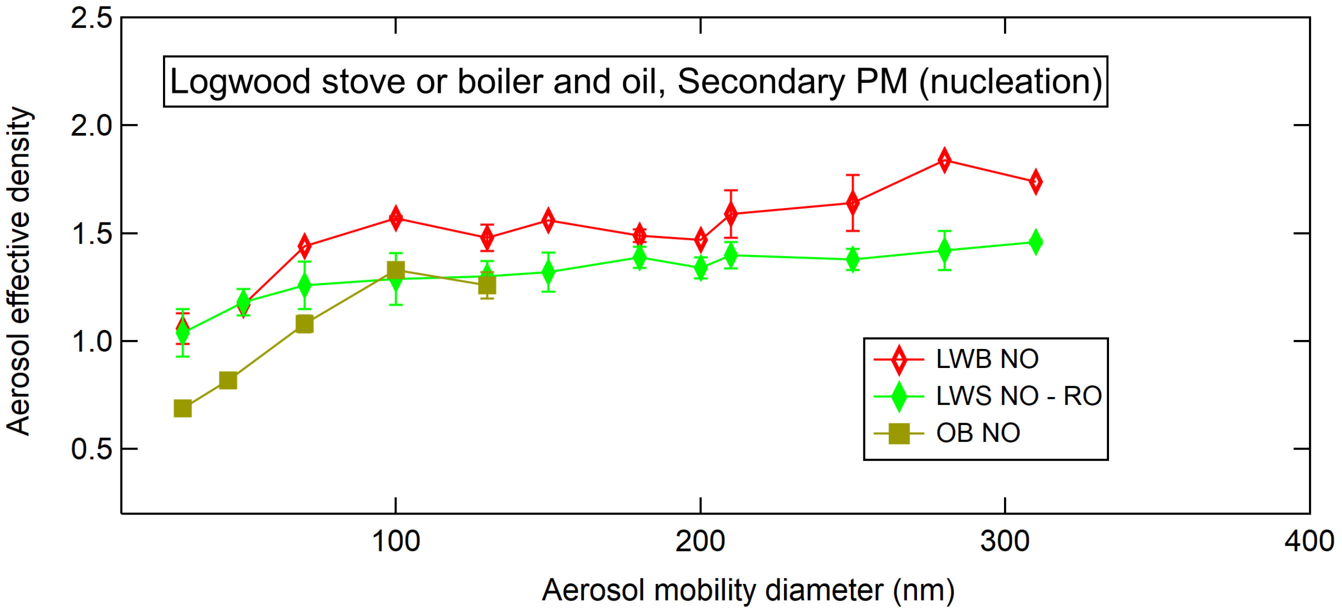
Significant heterogeneous reactivity processes (gas/particles)

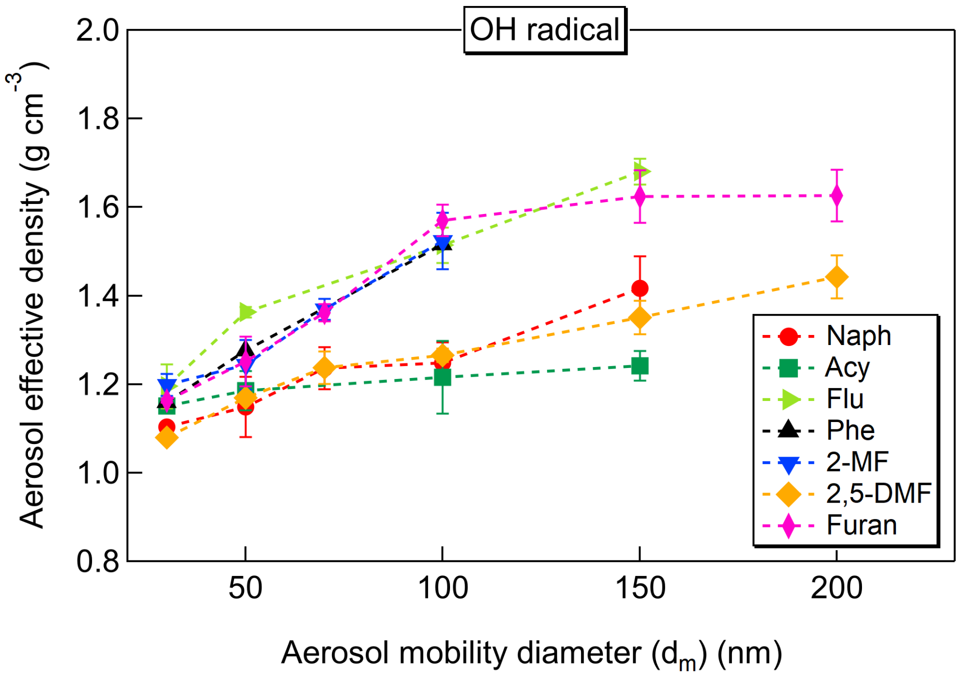
LWS > or >> PWB and PWS

LWS nominal ≈ OB

George et al., 2015

Secondary PM formation by nucleation processes: effective density





↗ Secondary PM density with PM size up to a "plateau" at 180 nm

Differences in terms of chemical composition and morphology with PM size

El Mais et al., 2023

Conclusions

Black carbon

Emissions of pellets appliances 10-100 times < logwood and oil appliances

Emissions +++ pellets boiler in very reduced output (15 %) ⇒ **need of restricting the operating range by the manufacturers**

1/3 pellets stoves ⇒ large emissions ≈ logwood stove ⇒ **high heterogeneity in the emissions ⇒ further works requested**

Secondary PM

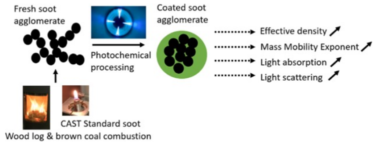
Pellets appliances ⇒ Low formation (× 1 - 2)

Logwood appliances (notably stove) ⇒ High formation in reduced output (up to × 16, OM), and comparale to pellets appliances in nominal output (× 1-3), larger emissions than pellets appliances anyway

Oil boiler ⇒ High formation (× 5 - 6, OM and SO_4) but low primary emissions

Softwood pellets

No impact on primary, secondary PM and BC but ↗ in CO and, in a lesser extend, in total VOCs and NO_x



PM effective density and morphology

Changes in the effective densities (↗) and morphologies (larger PM, soot ⇒ tarballs) of the PM due to aging

Leskinen et al., 2023

Thank you for your attention !

alexandre.albinet@ineris.fr

Abd El Rahman EL MAIS, Nicolas KAROSKI, Adrien DERMIGNY, Vincent FUVEL, Yannick DUPUIS, Ahmed ABIDA, Christophe RICHET, Medhi DIONIGI, Serge COLLET, Arnaud PAPIN, Ahmad El MASRI, Serguei STAVROVSKI, Farid AIT BEN AHMAD, Nathalie BIANCHINI, Valérie MINGUET, Rachel GEMAYEL, Robin AUJAY-PLOUZEAU, Céline FERRET, Alexandre ALBINET

Céline DEGRENDELE, Brice TEMIME-ROUSSEL, Grazia Maria LANZAFAME, Barbara D'ANNA, Henri WORTHAM

Nicolas DANTHONY, Pedro JORQUERA-FERRAT

Funding
(EPOCHAG project)

Isabelle AUGEVEN-BOUR, Manon VITEL +