Reconciling particulate emissions with ambient measurements for biomass combustion

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Wood burning represents a renewable and CO_2-neutral energy source. However, as a combustion processes, it emits particulate matter that has an impact in climate, visibility and human health. Until very recently, the contribution from wood burning to the ambient particulate matter concentration was thought to be minor and neglected in favour of reducing pollution from other sources. This situation has changed: recent source appointment studies point to wood burning as one of the main sources of particulate matter pollution. Nevertheless, this form of combustion presents a completely new challenge because an important portion of the atmospheric particulate matter related to wood burning is initially emitted in the gas phase. Theses are hydrocarbon molecules, also referred to as organic gaseous carbon (OGC), that once in the atmosphere are transformed into particles called secondary organic aerosols (SOA).

In this paper, we show the emission side of this situation. We present the emission-factors of different residential biomass combustion devices, with an emphasis on the organic matter emitted in the condensed- as well as in the gas-phase. The relevance of SOA as a component of ambient PM becomes evident when comparing these two phases. Our measurements show that organic matter amounts only to a relatively small fraction of the directly emitted particulate mass. Typical organic-matter-to-black-carbon ratios (OM/BC) are of the order of 1.3 for a pellet boiler and all the way down to 0.2 for a Logwood stove. This is in clear opposition to the high concentration of organic matter measured in the atmosphere, where the organic-matter-to-elemental-carbon ratio related to wood combustion can be as high as 20 (see e.g. Szidat, 2006, and reference therein). The difference is caused by the organic matter of the SOA originating from OGC emissions.

This raises the question of how to quantify the quality of the combustion and its potential impact. For instance, a modern logwood stove and an automatic pellet boiler can have similar particulate matter emission factors but their OGC emissions are completely different (see fig. 1). Specially during the stable phase, an automatic pellet boiler emits almost no OGC. Other studies (e.g. Chirico, 2010) confirm that pellet boilers have a small SOA production potential. Furthermore, it has been shown that for relatively small variations on the PM emissions factors (i.e. less than one order of magnitude), the emission factors for OGC can vary over about three orders of magnitude (Johansson, 2004).

Current standards include only with the solid fraction of the emissions, ignoring the gas phase and, more importantly, its SOA production potential. This results in a big discrepancy between the particle mass measured on the side of the emissions and the actual atmospheric concentrations. The discrepancy directly affects other studies based on measured emission factors. For instance, risk assessments and environmental impact studies have an incomplete set of data that does not account for SOA, and properties like toxicity and warming potential are very different between primary and secondary aerosols due of their different chemical composition. For this reason, it is of extreme importance to include the OGC as a standard to characterize a burner. Particulate mass by itself, regardless of the metric, is not enough.

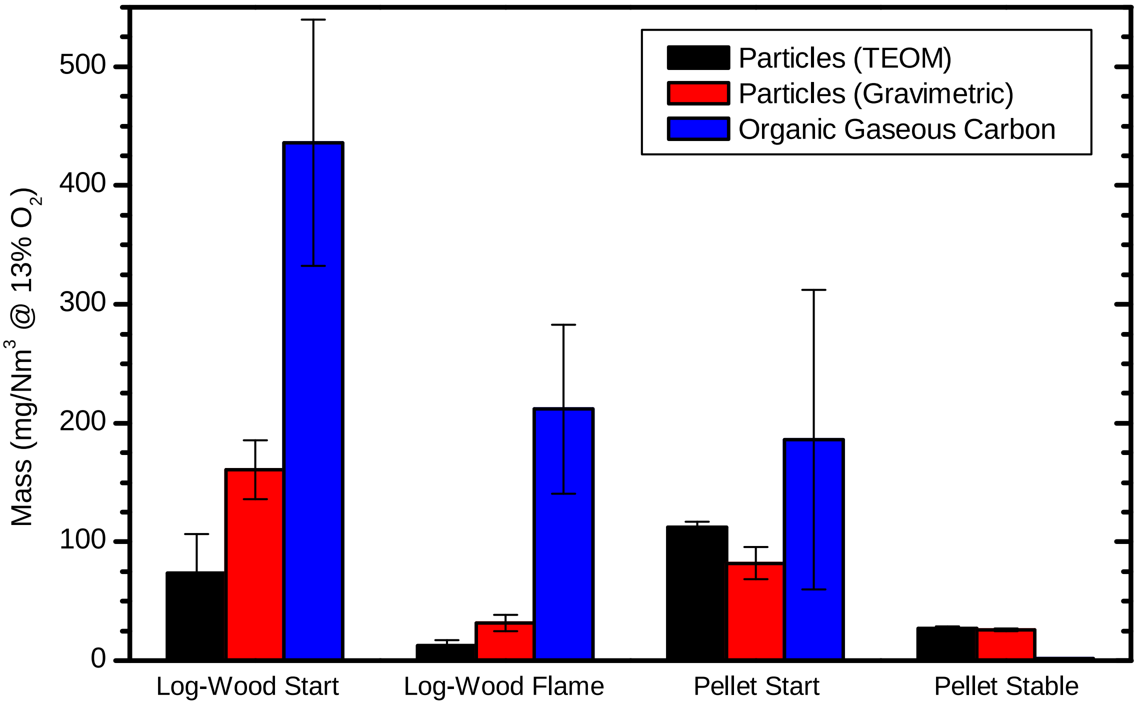

Figure 1. Particulate matter and organic gaseous carbon emission factors for a logwood stove (nominal power 8 kW) and an automatic pellet boiler (nominal power 25 kW) for different combustion conditions as measured by a tapered element microbalance (TEOM), the reference method (gravimetric) and, for the organic gaseous carbon, a flame ionization detector (FID).

References

Szidat, S., T.M. Jenk, H.-A. Synal, M. Kalberer, L. Wacker, I. Hajdas, A. Kasper-Giebl, U. Baltensperger (2006). *Contributions of fossil fuel, biomass-burning, and biogenic emissions to carbonaceous aerosols in Zurich as traced by ^{14}C*, J. Geophys. Res. **111**, D07206, doi:10.1029/2005JD006590.

Chirico, R. *et al.* (2010). *Primary emission and secondary formation of organic aerosol from vehicles* in 14th ETH Conference on Combustion Generated Particles, June 26th -29th, 2011, ETH Zurich, Zurich, Switzerland.

Johansson, L.S., B. Leckner, L. Gustavsson, D. Cooper, C. Tullin, A. Potter, (2004). *Emission characteristics of modern and old-type residential boilers fired with wood logs and wood pellets*, Atmos. Environ. **38**, pp. 4183–4195.

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Biomass Combustion: PM emissions vs. ambient measurements

ETH NPC 2010

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IMBALANCE

Fachhochschule Nordwestschweiz
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IMpact of Biomass burning AerosoL on Air quality aNd ClimatE
Financed: ETH Competence Center for Environment and Sustainability
http://www.cces.ethz.ch/projects/clench/imbalance

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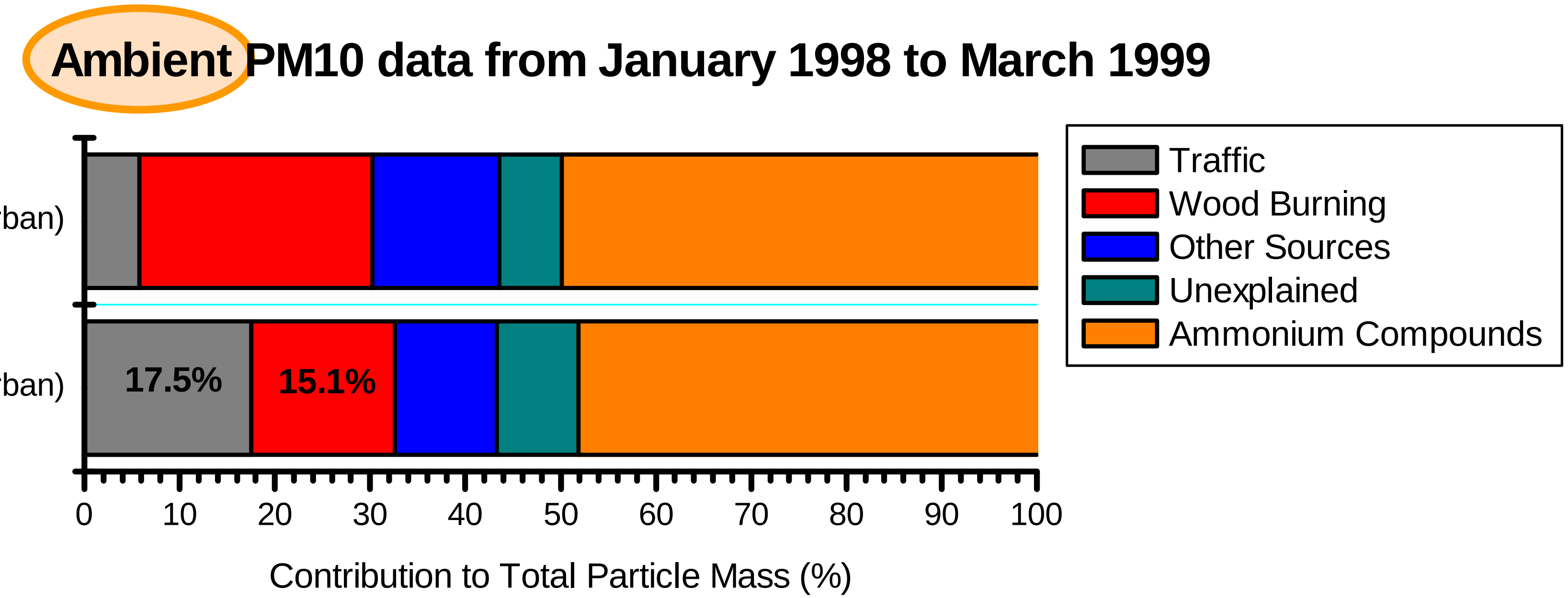
Reason behind the interest



- *Particulate matter from wood burning is comparable to traffic-PM even in urban sites like Zurich (on a mass-based metric).*
- *In suburban locations, PM from Wood can surpass PM from traffic.*

preliminary data, IMBALANCE Project (EMPA)

Official view on Wood Burning

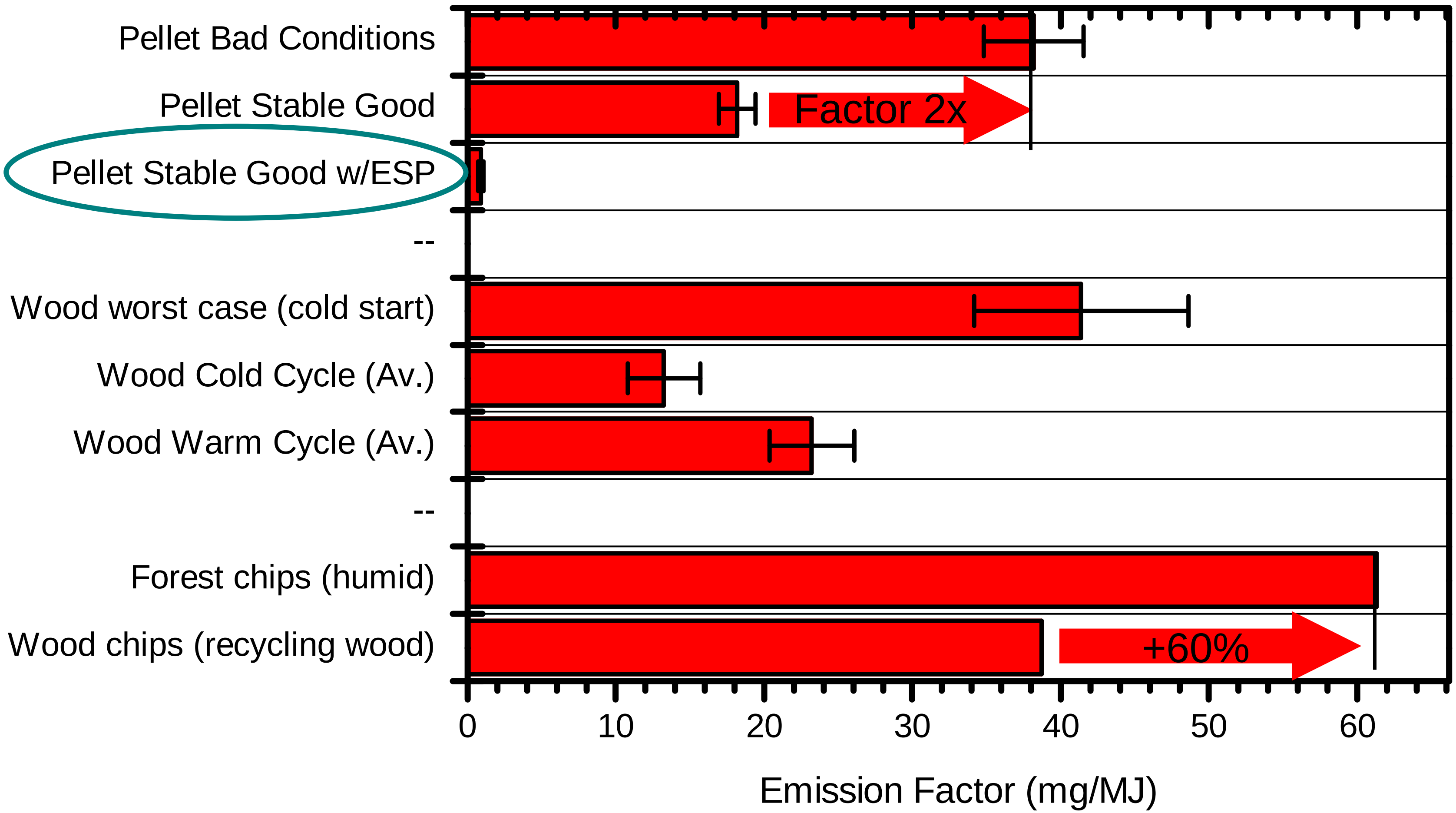
- The Swiss Government recognized that PM from wood burning is problem, and that the percentage of PM originating from wood burning may duplicate within the next decade.
- As a reaction to this, lower emission standards will be introduced starting 2011.
- Emission limits for installations over 70kW will be even stricter.

Type of combustion installation (below 70 kW)	CO mg/m^3 @ 13% O_2	PM mg/m^3 @ 13% O_2
Log Wood Boilers	800	60 -> 50
Automatic Operation Boilers	400	90 -> 60
Pellet Boilers	300	60 -> 40
…		
…		

What we measured

- Different types of fuels and burners (pellets, log wood)
- Different burning conditions (normal operation, reduced O_2, cold and warm start, bad fuel quality, etc)
- Different parts of the cycle. Most parameters time resolved (as opposed to integrated values)
- Particle mass (TEOM, gravimetric), size and number concentration (SMPS, FMPS, DiSC), particle length (DiSC).
- Particle-phase organics (AMS), Black Carbon (MAAP).
- CO, CO_2, gas phase hydrocarbons (FID), etc…

PM Emission Factors



Energy content in fuel around 15 to 18 MJ/kg

Primary/Secondary Emissions

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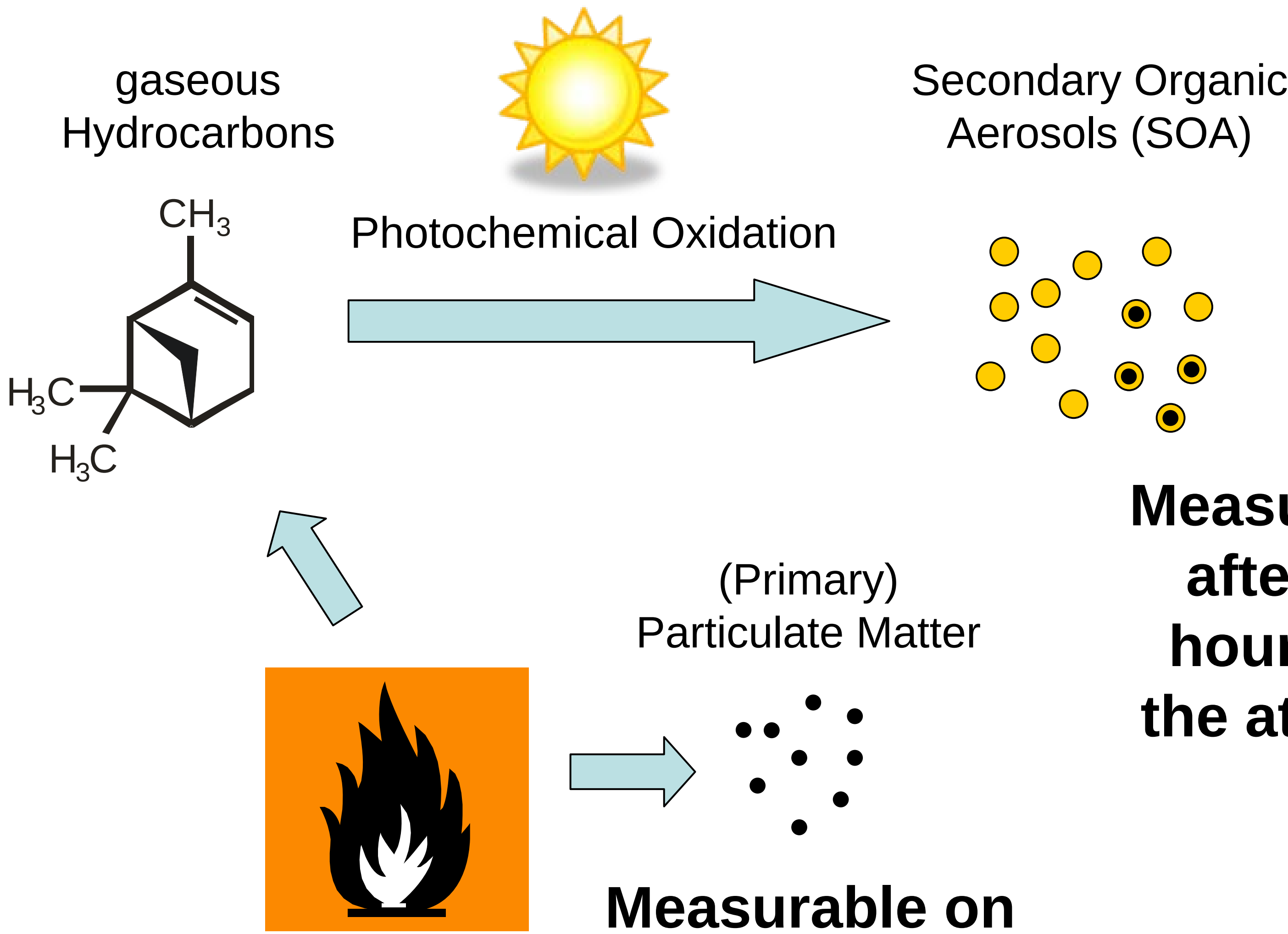


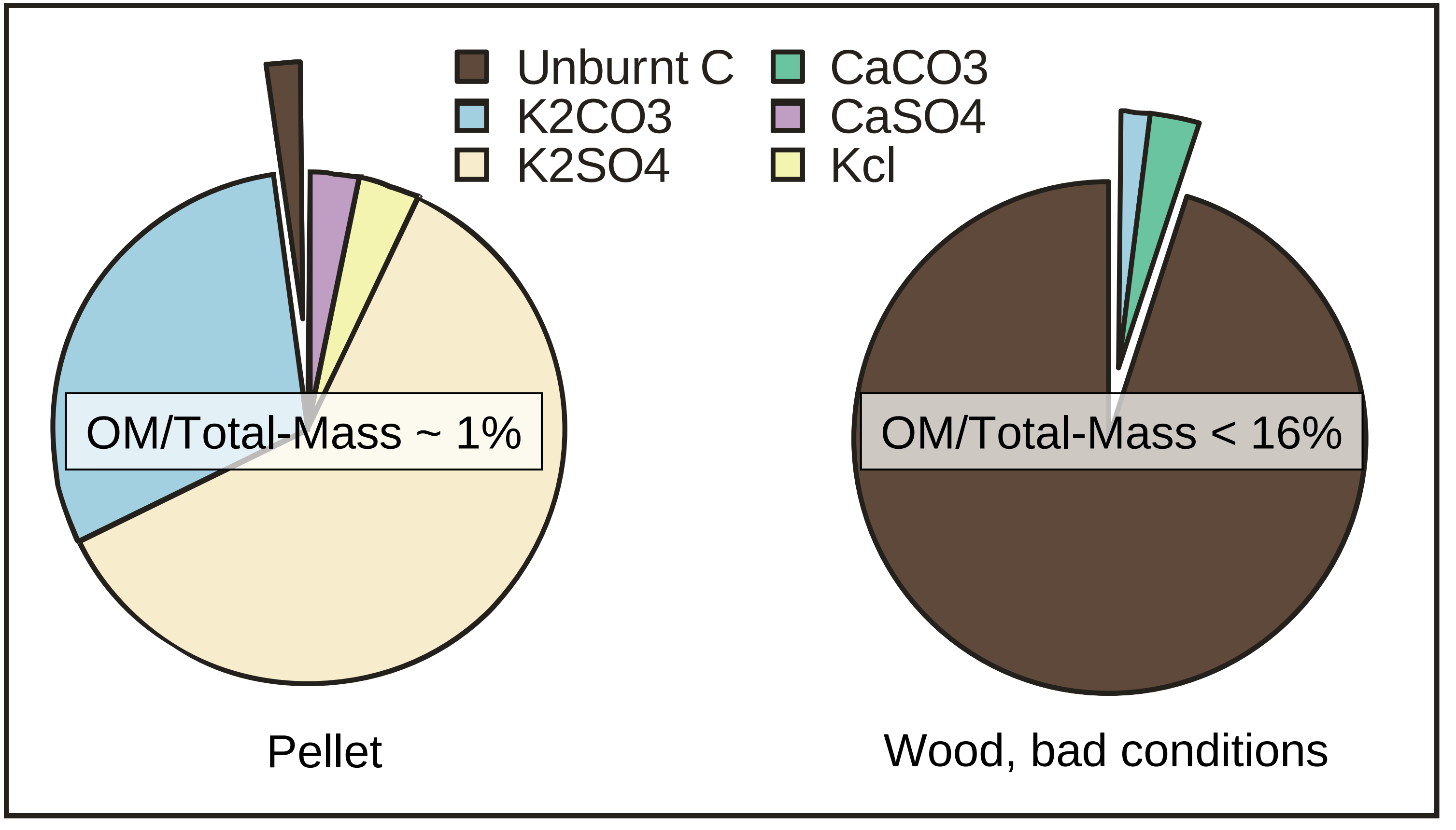
Measurable only after several hours/days in the atmosphere

Measurable on emission site

Particle Composition

From our AMS and MAAP data (Paul Scherrer Institute)

Pellet: Organic-Mass/Black-Carbon (OM/BC) = 1.3 (Good Combustion)
Wood: Average OM/BC = 0.2



from Nussbaumer et al., 2008

Ambient Organic Mass

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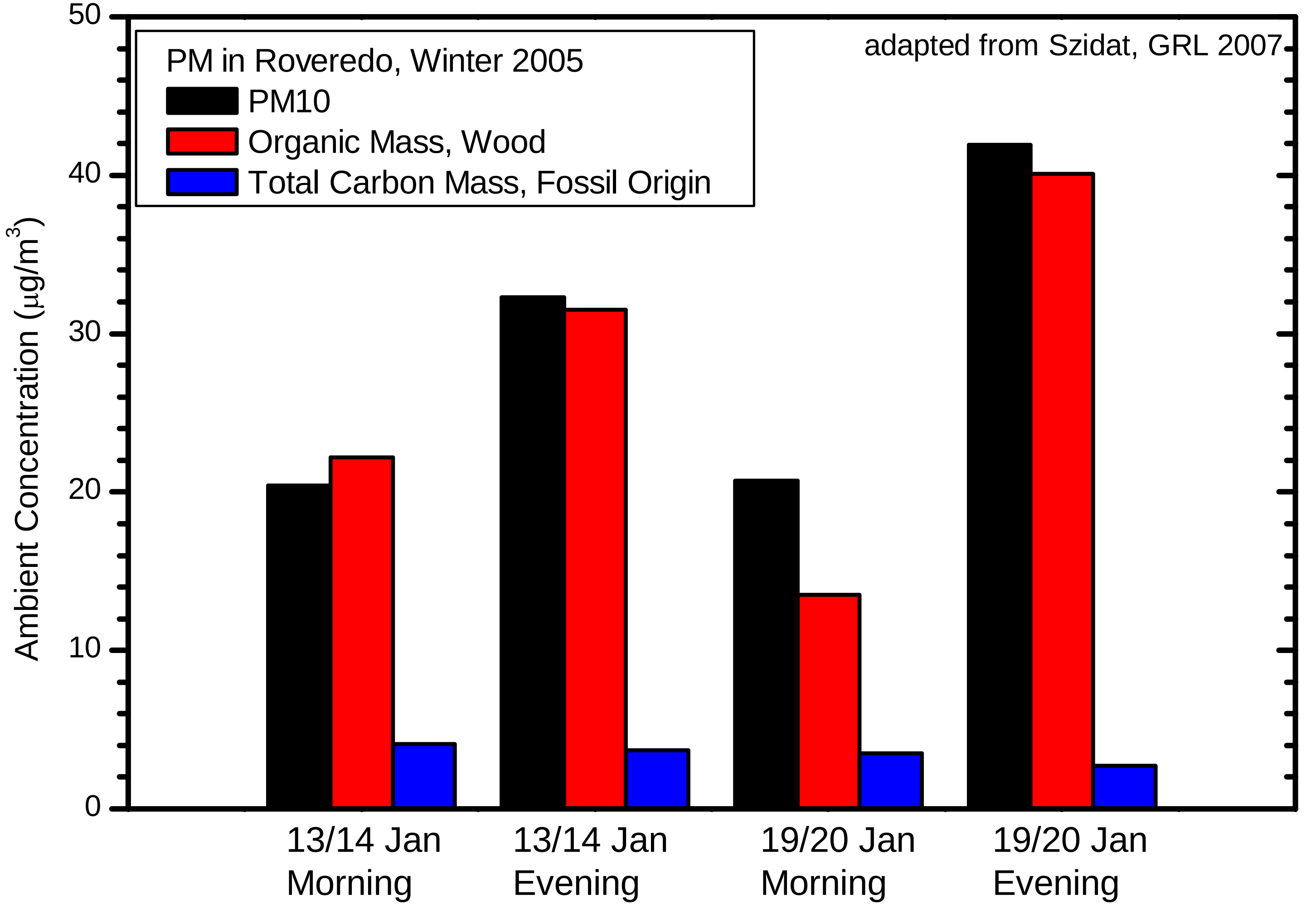
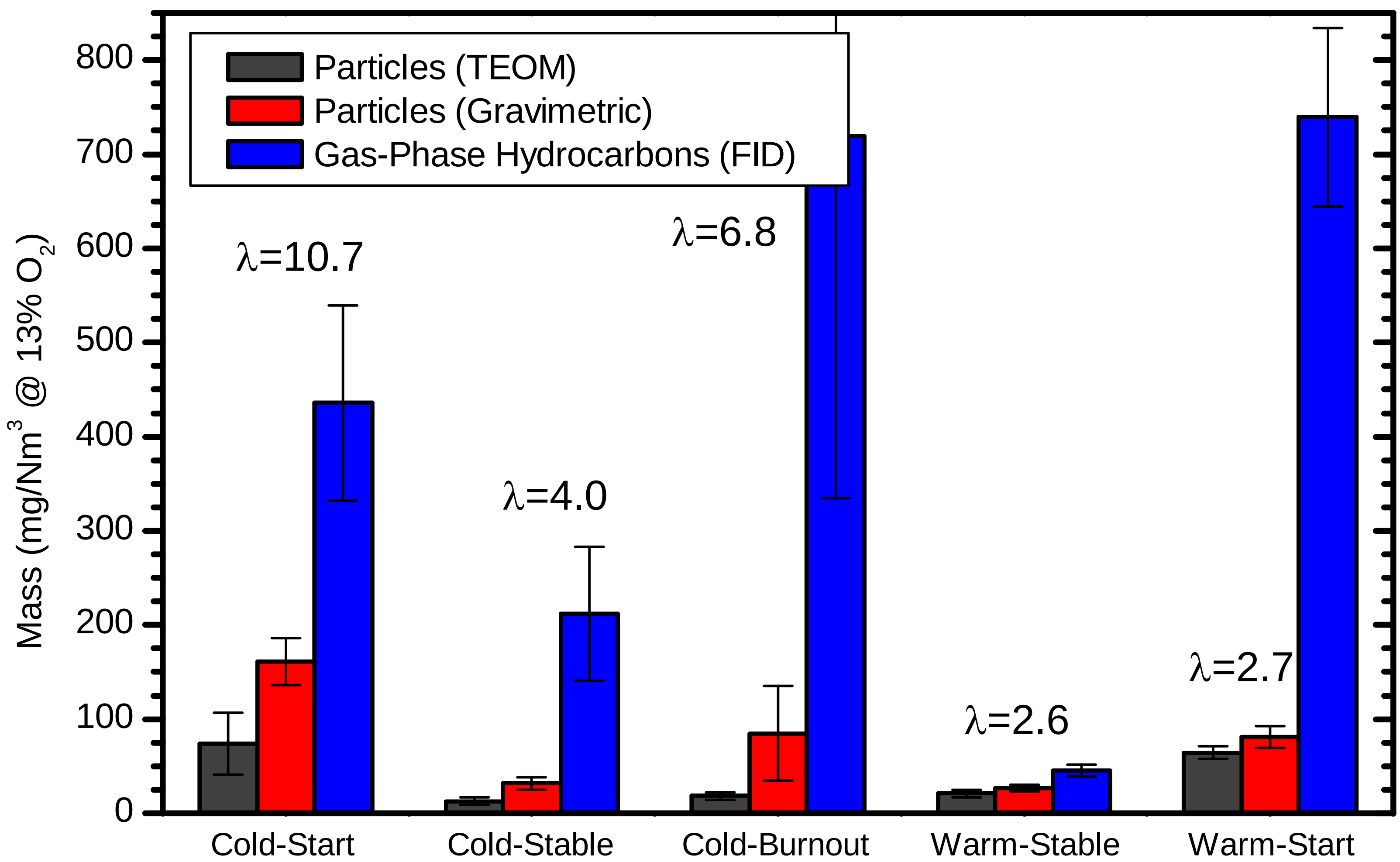
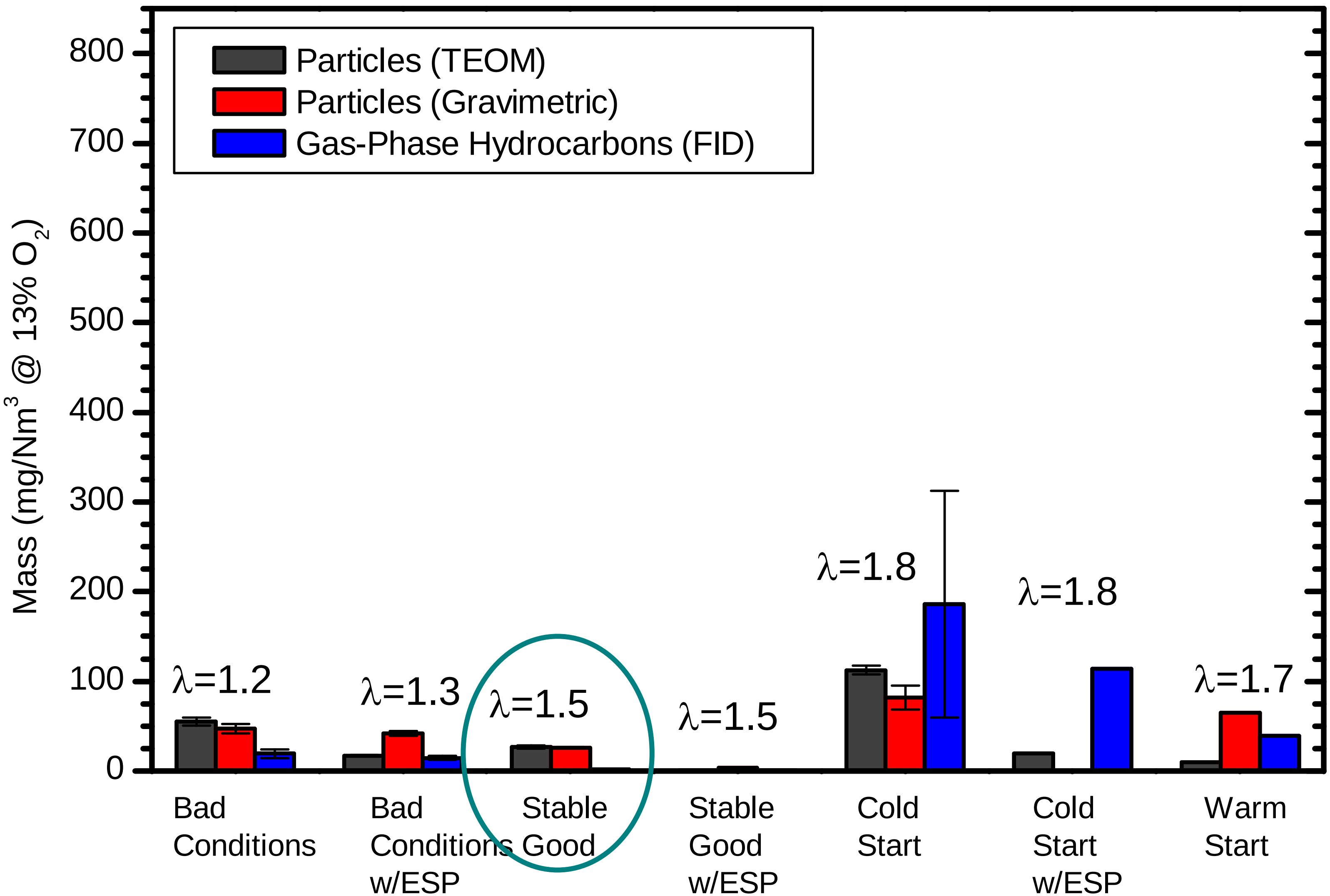


- For the whole study, Organic Mass to Elemental Carbon ratio was OM/EC=7.6
- **For wood particles OM/EC=20.7**

PM and Hydrocarbons - Wood

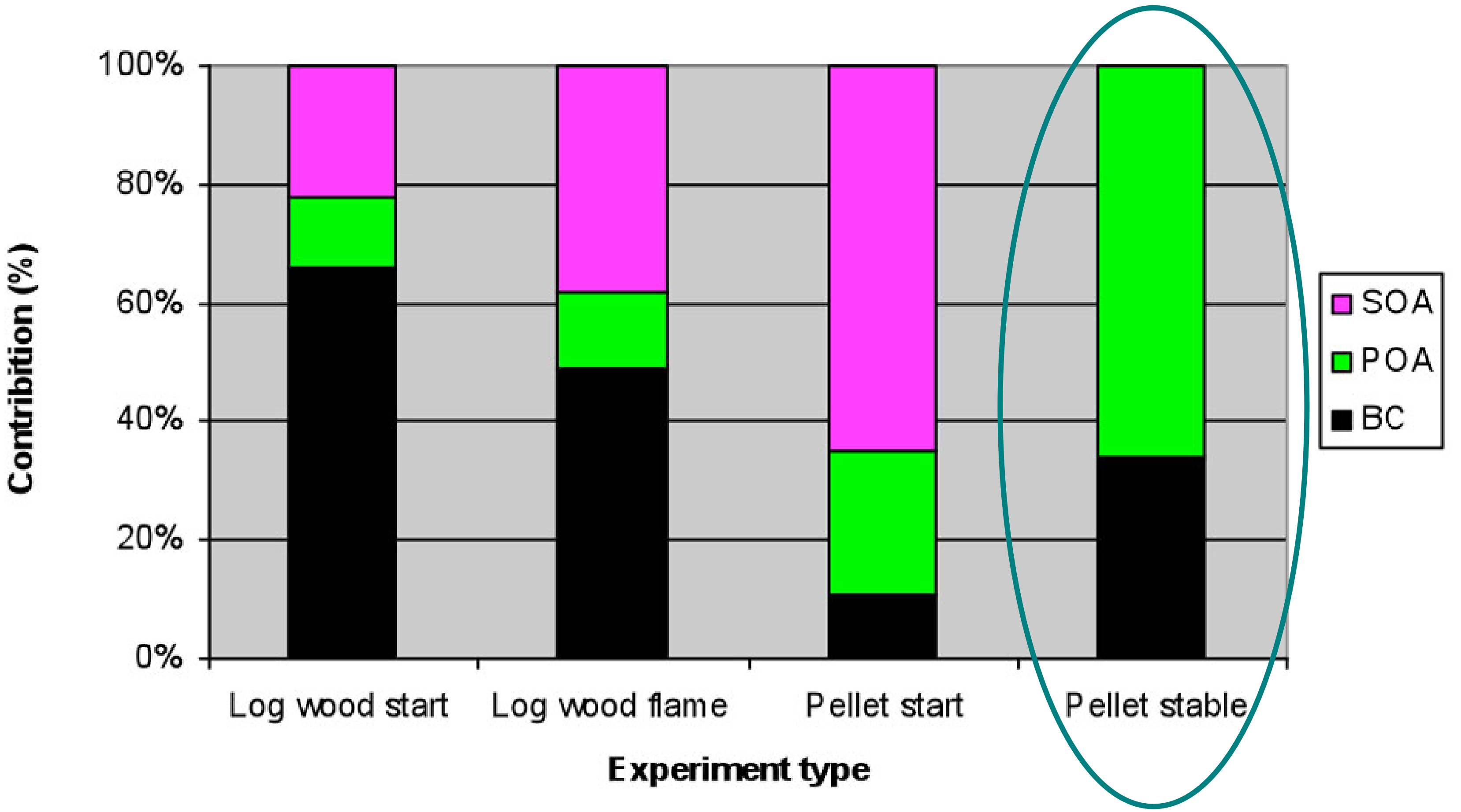


PM and Hydrocarbons - Pellet



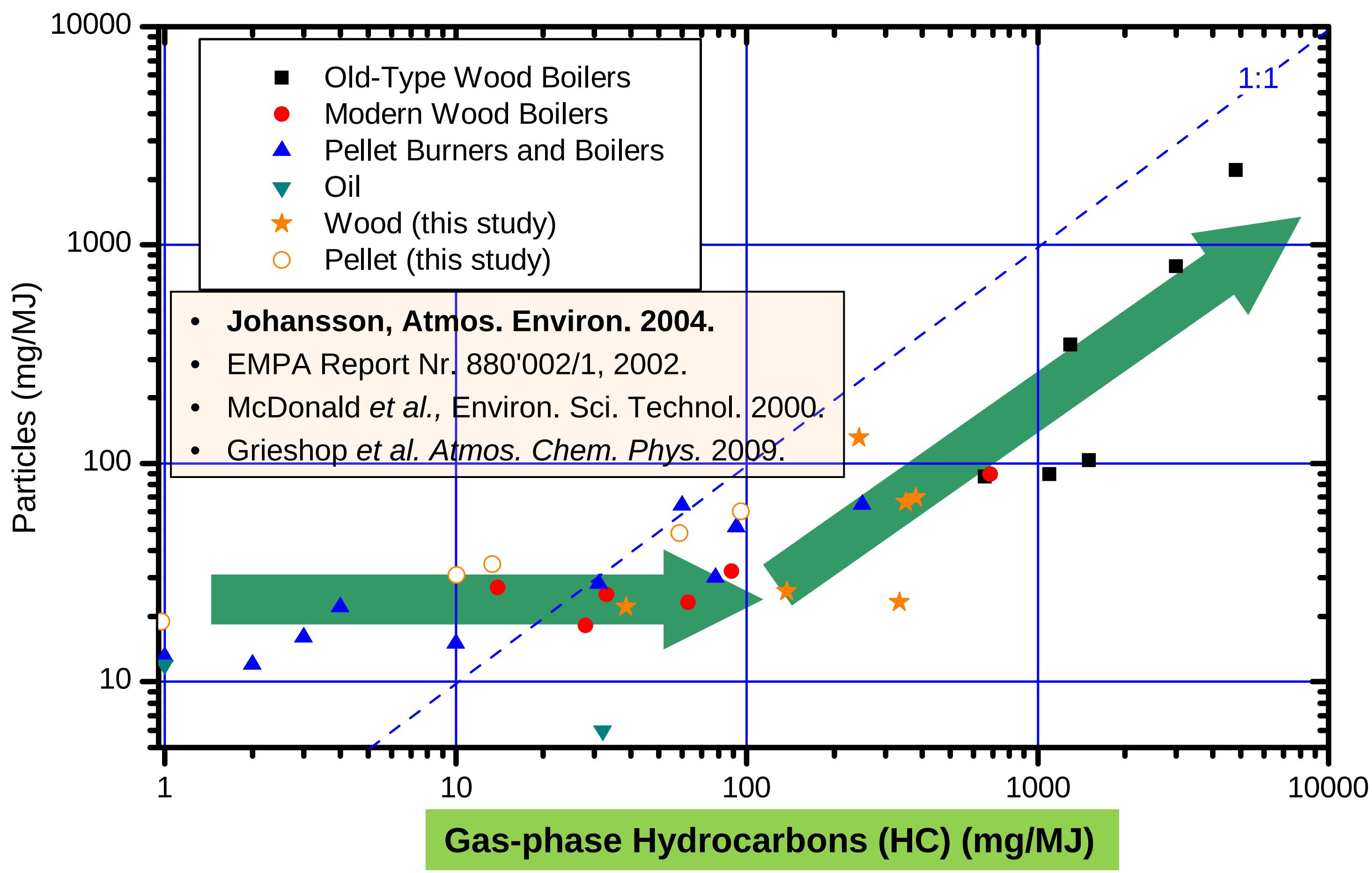
SOA formation potential

Average contribution of black carbon, primary organic aerosol (POA) and secondary organic aerosol (SOA) after 5 hours of aging in the smog-chamber.



From Prevôt et al. ETH-NPC 2010

HC & combustion quality



Other names for Gas-Phase Hydrocarbons:
Hydrocarbons (HC), Total Organic Compounds (TOC), Organic Carbon (OC), Organic Gaseous Compounds (OGC), VOCs, SVOCs, etc…

Legislation on HC-Emissions

European Norms that <u>do not</u> explicitly contemplate Hydrocarbon emissions:

- EN 303-5, EN 12809, EN 13240, EN 14785, EN 12815, EN 13229
- These norms cover all authorized types of wood-like-biomass combustion but only provide emission limits for CO and PM.

Exceptions:

- Austrian Norm for Heating installations with solid or liquid fuels imposes an OGC limit in mg/MJ.
- Swiss Norm LRV 814.318.142.1 limits OGC, but <u>only for wood burning installations larger than 10MW</u>.
- Implicit limitation on norm EN 303-5 for heating boilers.

Summary

- Emissions from Wood Burning are an important source of ambient PM.
- On the side of the emissions, PM is important but gives an incomplete picture.
- In the atmosphere, SOA mass can exceed primary PM. Gas-Phase Hydrocarbons are the source of SOA.
- **Future legislation should contemplate these HC and, ideally, their Secondary Organic Aerosol Production Potential…**

Roveredo (GR)

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Photo: Paul Scherrer Institute

Secondary Organic Aerosol can constitute the majority of the ambient particulate mass!