Primary and Secondary Aerosol Emissions from Modern Small-scale Wood Combustion Appliances with Advanced Secondary Air Supply

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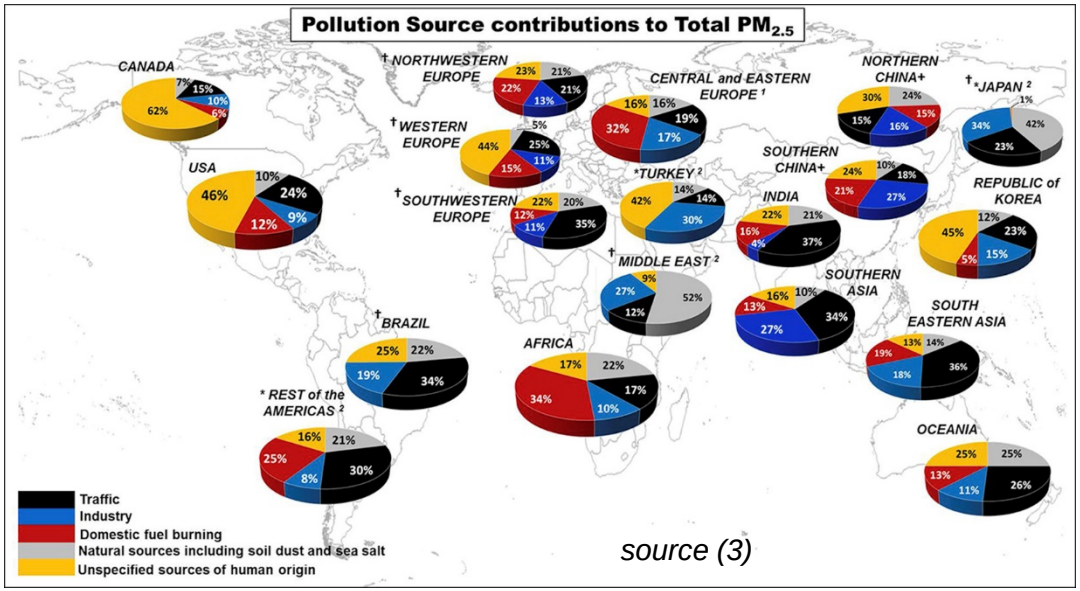
Background: residential wood combustion

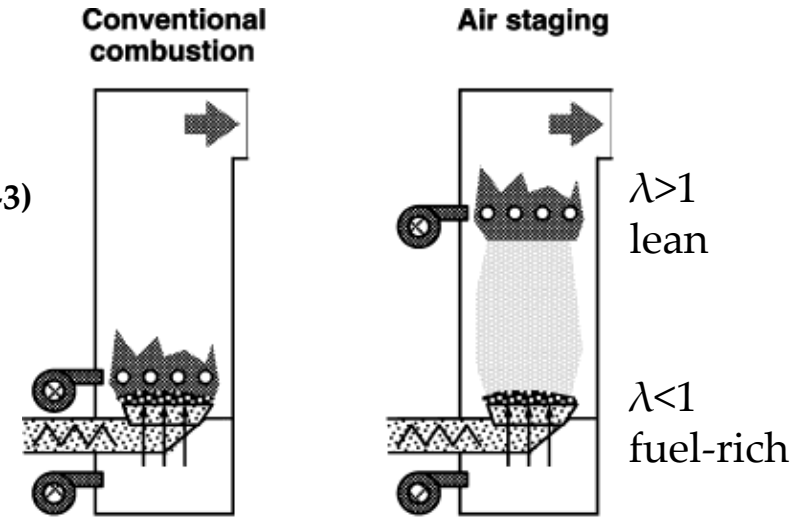
- **increased firewood** usage: political, ecological, economical reasons
- **major source of air pollution** in Europe areas, esp. during winter[1-3]
- indication of **substantial SOA** formation potential[4]
- wood combustion aerosol: **harmful effects on human health**[5-6]



[1]*Genberg 2013 ACP,* [2]*Pastorello 2011 Atm Environ,* [3]*Karagulian 2015 Atm Environ,* [4]*Grieshop 2009 ACP,* [5]*Jalava 2012 Atm Environ,* [6]*Sehlstedt 2010 Particle Fibre Toxicol*

Advanced Combustion Technology

- **air staging**, fuel staging and flue gas recirculation
- **substantial reduction in CO, VOC and PM**[1-3]
- low primary air: volatilisation, fuel-rich gas
- excess secondary air: efficient combustion



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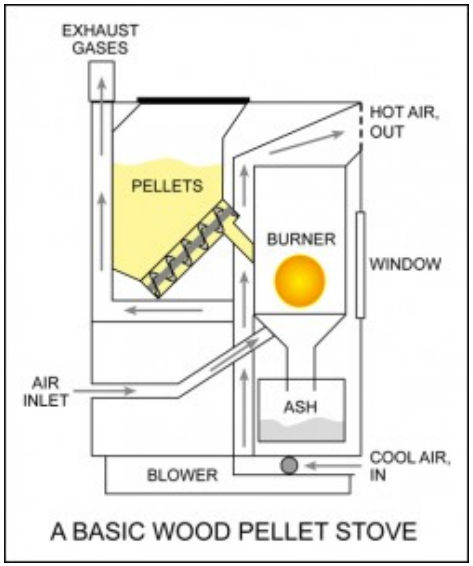
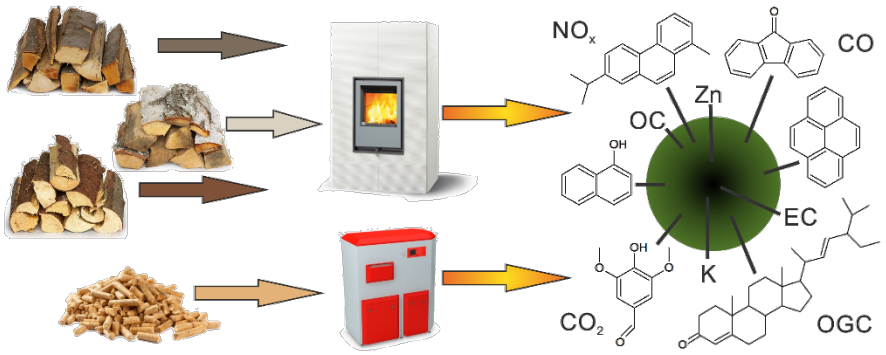


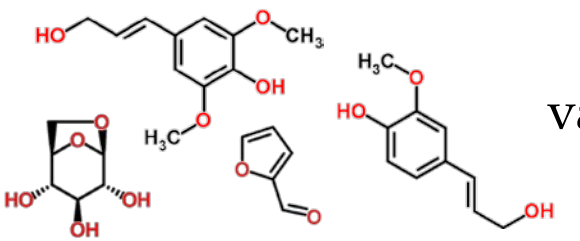
- automatically-fired appliances:
  - pellet stove (room-space heating)
  - pellet boiler (for warm water supply)
- **generally lower emissions than logwood stoves**[4,5]
- strong dependence on pellet quality / raw material and start-stop or continuous operation[6-8]

[1]Khodaei 2017 Fuel, [2]Nussbaumer 2003 Energy&Fuels , [3]Nuutinen 2014 Biomass&Bioenergy [4]Orasche 2011 Energy&Fuels, [5]Schmidl 2011 Atm Env, [6]Lamberg 2013 Energy&Fuels, [7]Chandrasekaran 2013 Energy&Fuels, [8]Win 2014 Energy&Fuels

Purpose

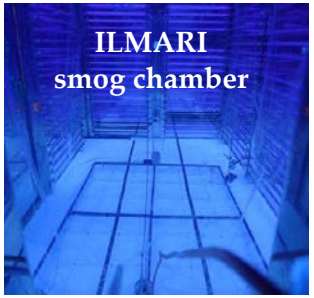
emissions factors

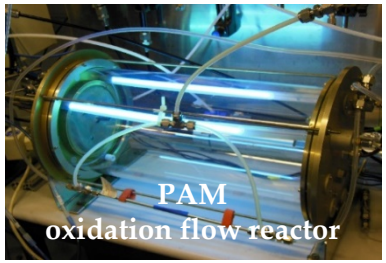




validity of molecular markers

SOA formation potential





Campaign at UEF

modern masonry heater

- *Hiisi 4*, Tulikivi Ltd. (Finland), **air staging**
- massive soapstone, slow heat release
- beech, birch and spruce logwood
- 6 consecutive batches (2.5 kg) à 35 min with 30min char-burning (∑time=4h)

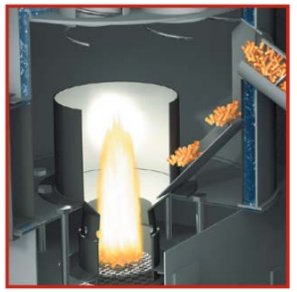
beech

spruce

birch

pellet boiler

- PZ-RL Biotech (Energietechnik GmbH, Austria)
- 25 kW nominal load, **air staging**
- combustion scenarios:
  - boiler starting phase (BSP)
  - optimised combustion at nominal load (**OPT**)
  - ≈30% reduced secondary air (**RSA**)
    → proxy for old-type boilers

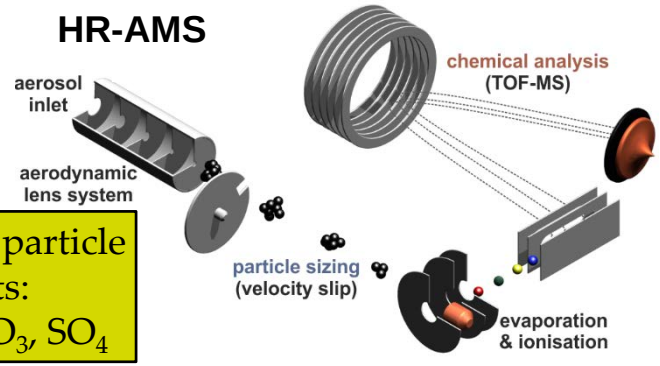


Fuel feed into burner flame

softwood pellets

Instruments

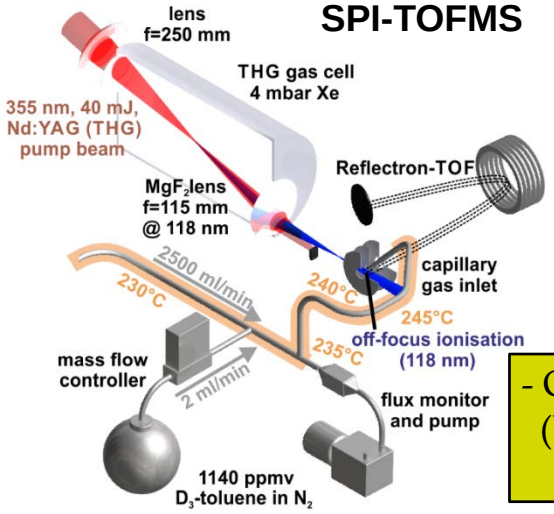
HR-AMS



- Refractory particle constituents:
  → OM, NO_3, SO_4

DeCarlo, Kimmel, Trimborn, et al., *Anal Chem*, 74 (2006), 8281–8289

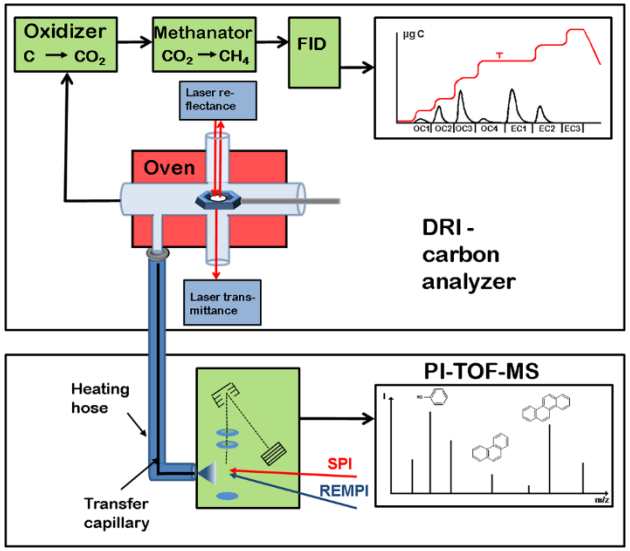
SPI-TOFMS

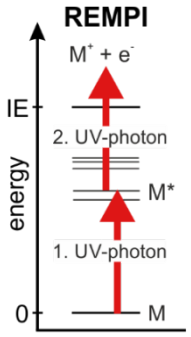


- Organic vapours (VOC to SVOC, untargeted)

Czech, Sippula, Kortelainen, et al., *Fuel*, 17 (2016), 334-342

TOCA-REMPI-TOFMS

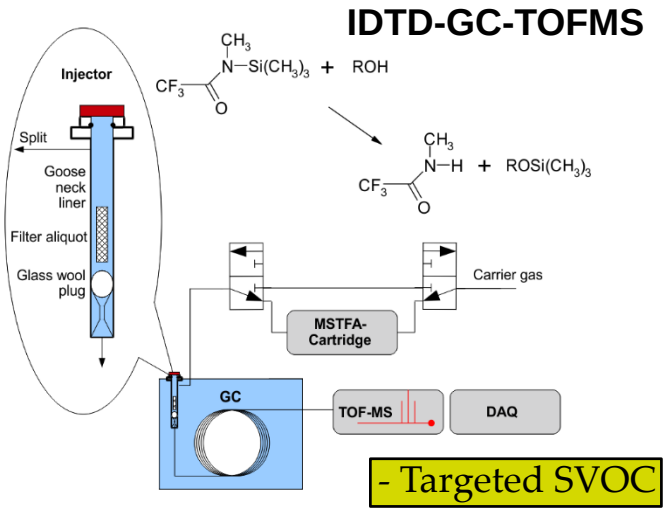




- ECOC
- Untargeted aromatic profile of SVOC/LVOC

Diab, Sreibel, Cavalli, et al., *Atm Meas Tech*, 8 (2015), 3337–33353

IDTD-GC-TOFMS

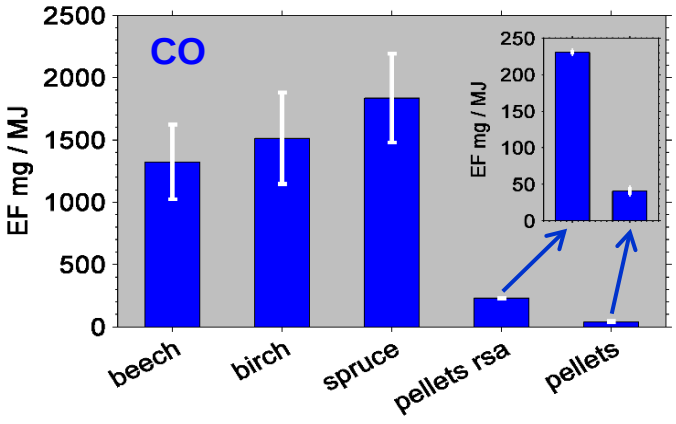


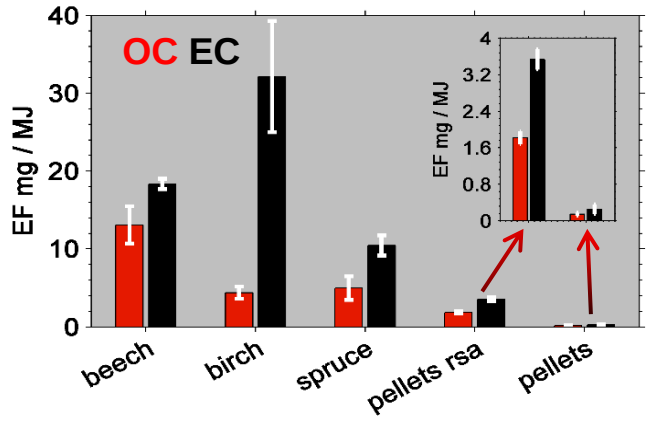
- Targeted SVOC

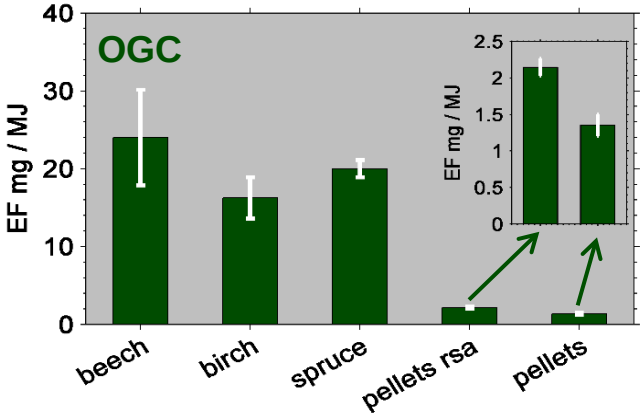
Orasche, Schnelle-Kreis, Abbaszade, Zimmermann, *Atm Chem Phys*, 11 (2011), 8977–8993

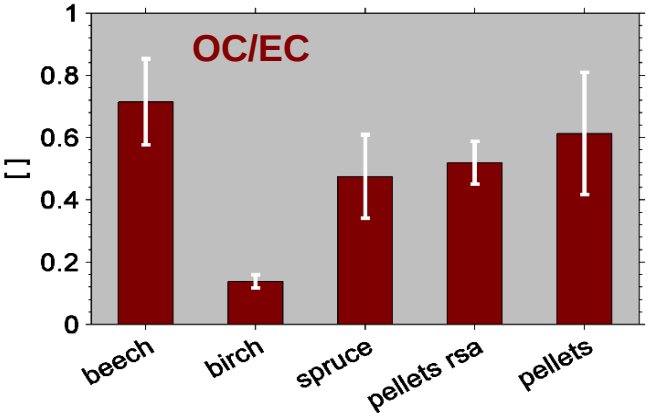
Carbonaceous Emissions

Gaseous Carbon









Particle-bound Carbon

- Insignificant effect of wood type on emissions except EC
  - Essential oil-rich / resinous woods (birch, pine,…): EC↑ → OC/EC↓ (<1)
- Significant effect of combustion technology: logwood > pellet rsa > pellet

Czech, Miersch, Orasche et al., *Sci Tot Environ*, 612 (2018), 636-648

Combustion Condition: Slow vs Proper Ignition

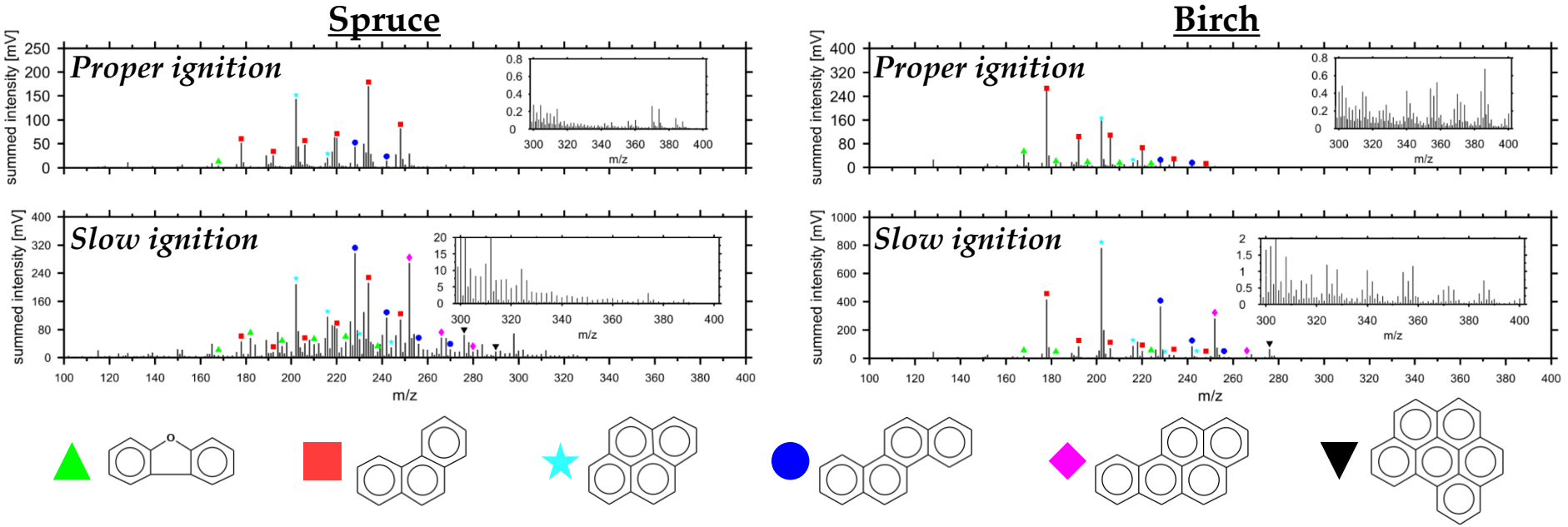
Spruce

- **primary decomposition products:** levoglucosan and methoxy-phenols
- PAH and OGC
- **extractives:** resin components

Birch

- PAH in general
- shift to **larger PAHs**
- no change in extractives

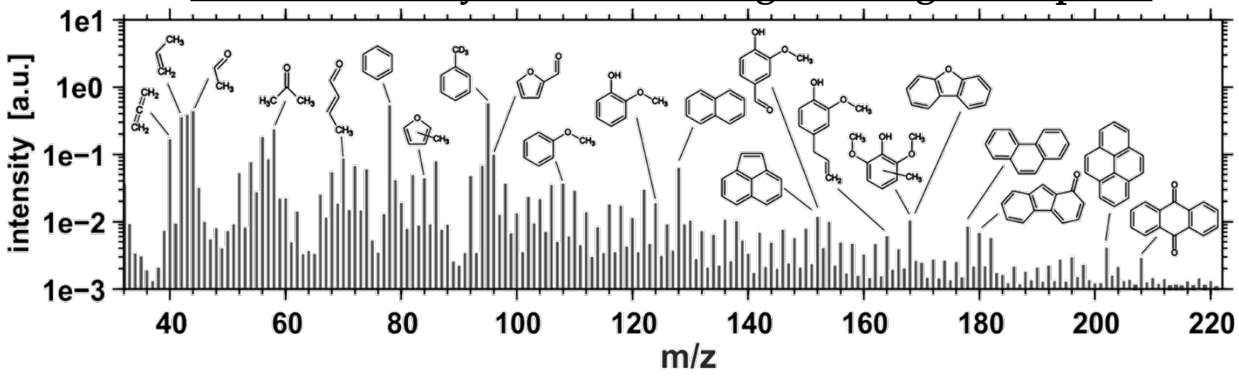
REMPI Mass Spectra, Thermodesorption 25-280°C



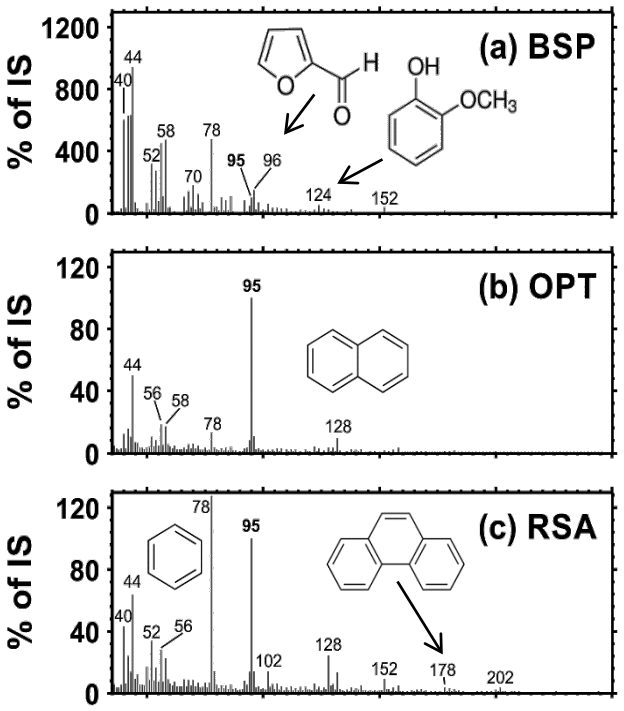
Czech, Miersch, Orasche et al., *Sci Tot Environ*, 612 (2018), 636-648

Molecular Markers for Wood Combustion

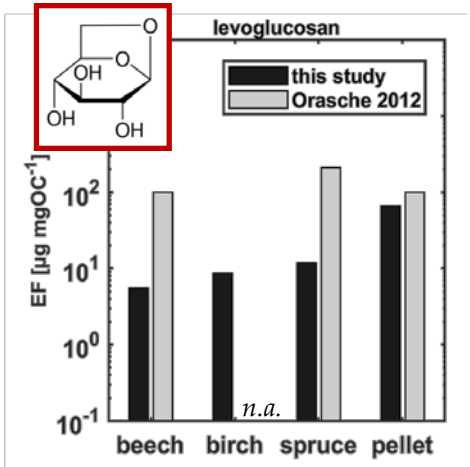
modern masonry heater: beech logwood organic vapours



pellet boiler organic vapours



- compared to conventional stoves **substantial reduction** of established wood / biomass combustion **markers** per emitted OC

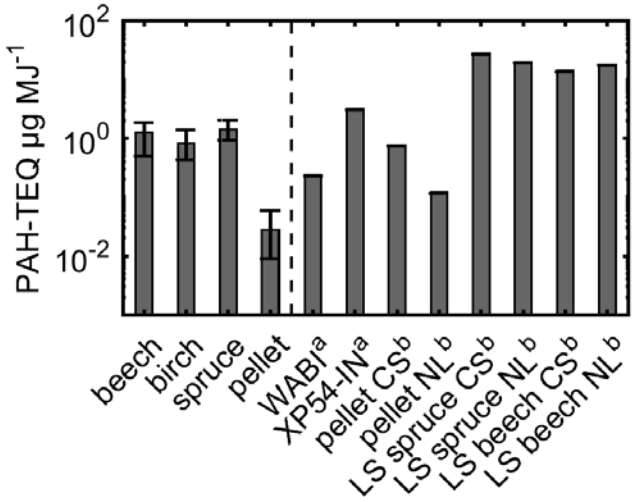


- **levoglucosan** (cellulose)
- **furans** (carbohydrates)
- **phenols** (lignin)

BSP… boiler starting phase
OPT… optimised combustion
RSA… reduced secondary air
IS... internal standard D3-toluene of 91ppb

Czech, Miersch, Orasche et al., *Sci Tot Environ*, 612 (2018), 636-648
Czech, Pieber, Tiitta et al., *Atm Environ*, 158 (2017), 236-245
Czech, Sippula, Kortelainen et al., *Fuel*, 177 (2016), 334-342

Aerosol Toxicity: beech logwood vs pellets



Emission factcor of total phenolic species

	Beech		Birch		Spruce		Pellet	
	mg/MJ	mg/gOC	mg/MJ	mg/gOC	mg/MJ	mg/gOC	mg/MJ	mg/gOC
Czech 2018	72	3.7	20	3.4	8.4	1.4	0.59	3.4
Orasche 2012[a]	2220	150	-	-	690	62	18	26

[a]Tschamber Energy & Fuels 2016 (air staging wood stove)
[b]Orasche Energy & Fuels 2012 (conventional stove)

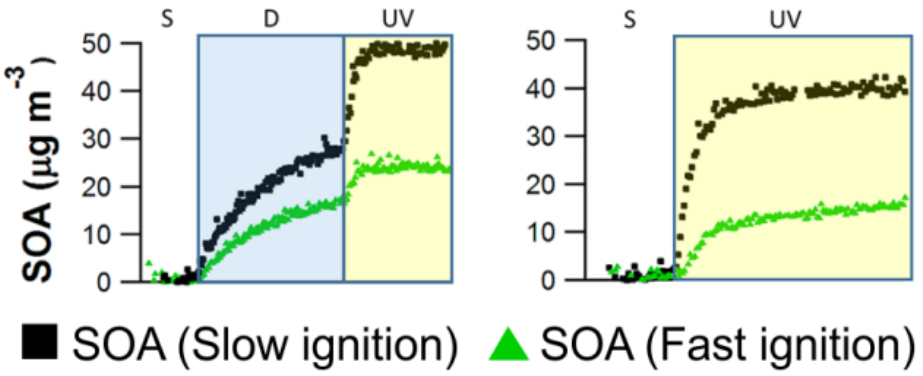
- PAH-TEQ suggest lower carcinogenity with combustion technology
- Beech logwood (73 ngPM cm^{-2}) vs Softwood Pellets (27 ngPM cm^{-2}): transcriptome and proteome analysis of A549 cells in Air-Liquid-Interface
  - dose: OC_{beech}=8xOC_{pellet}, **$Phenols_{beech}$=8x$Phenols_{pellet}$**, 3xZn_{beech}=Zn_{pellet}
  - significant **DNA damage** for both exposures
  - addressed to absence of **antioxidants** in pellet combustion aerosol

Czech, Miersch, Orasche et al., *Sci Tot Environ*, 612 (2018), 636-648
Kanashova, Sippula, Oeder, et al., *J Clin Mol Med*, 1 (2018), 23-35

SOA Formation from Modern Wood Combustion

Smog Chamber: spruce logwood

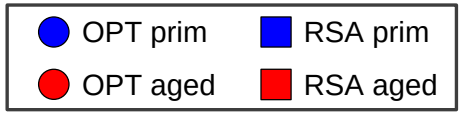
- SOA formation doubled POA after 8-12 h photochemical age
- higher SOA for slow ignition
- slightly lower ER_{OA} than conventional wood stoves
- enhancement ratio of OA lower for dark ageing, but substantial increase after additional UV exposure

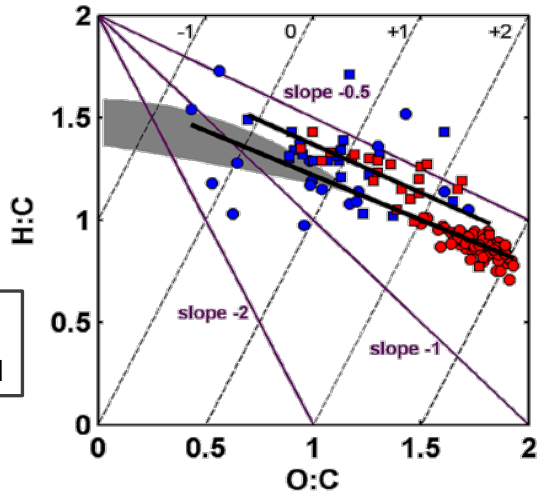


Slightly lower ER_{OA} + lower EF → lower SOA-EF

PAM: softwood pellets

- **no significant increase in OA** for pellet boiler OPT despite decrease in H:C and increase in O:C
- doubling of POA after 18 h photochemical age for RSA → high SOA yields from effective aromatic precursors

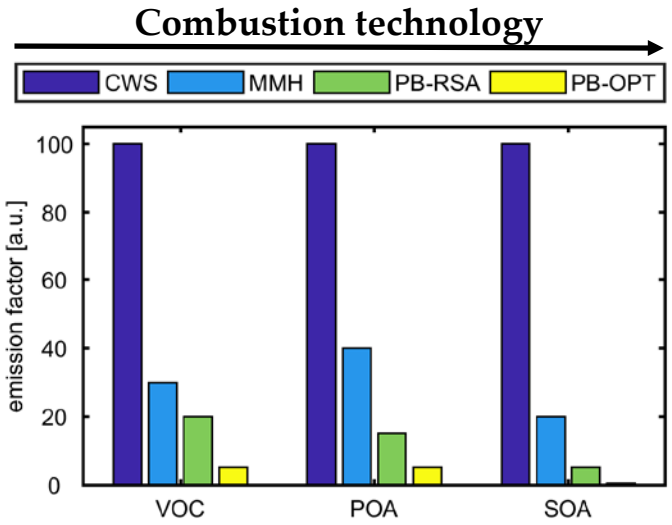




Tiitta, Leskinen, Hao et al., *Atm Chem Phys*, 16 (2016), 13251-13269
Czech, Pieber, Tiitta et al., *Atm Environ*, 158 (2017), 236-245

Summary:

- Reduction of emissions compared to stoves / pellet boilers without advanced air staging technology
- Reduction of SOA at similar level as POA and VOC
- SOA-formation for pellet boiler under optimised conditions even not detectable



- decreasing importance of wood type at higher quality of combustion
- significantly lower emissions of molecular markers of wood combustion
  → complicates identification in source apportionment
- efficient combustion reduces antioxidant content (phenolics)
  → indication of missing scavenging effect of intermediates in PAH metabolism
  → comparable level of DNA damage for logwood pellet combustion

Thank you for your interest!

Field Campaign 2013 at UEF, Kuopio

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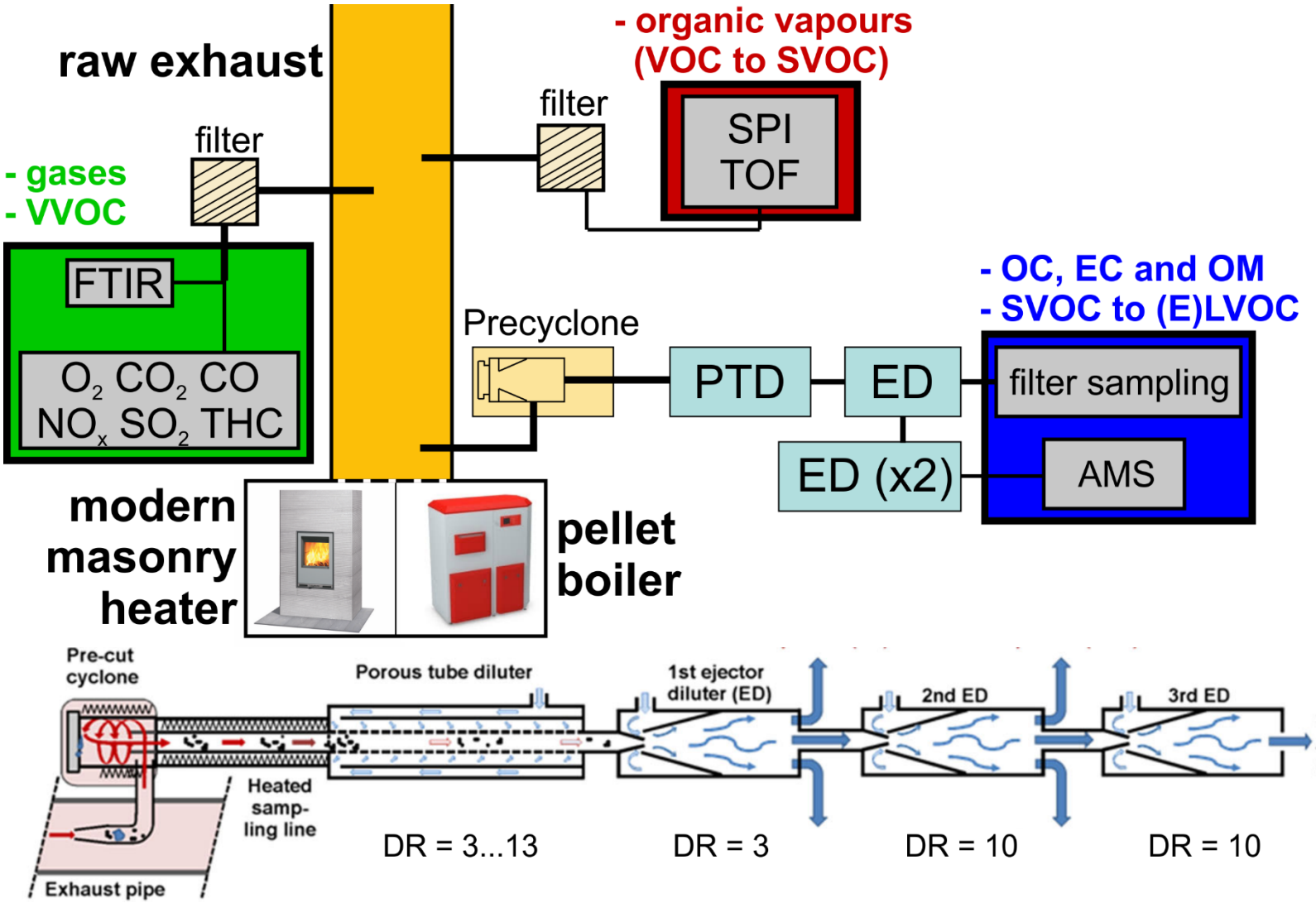
FONDO NAZIONALE SVIZZERO
SWISS NATIONAL SCIENCE FOUNDATION

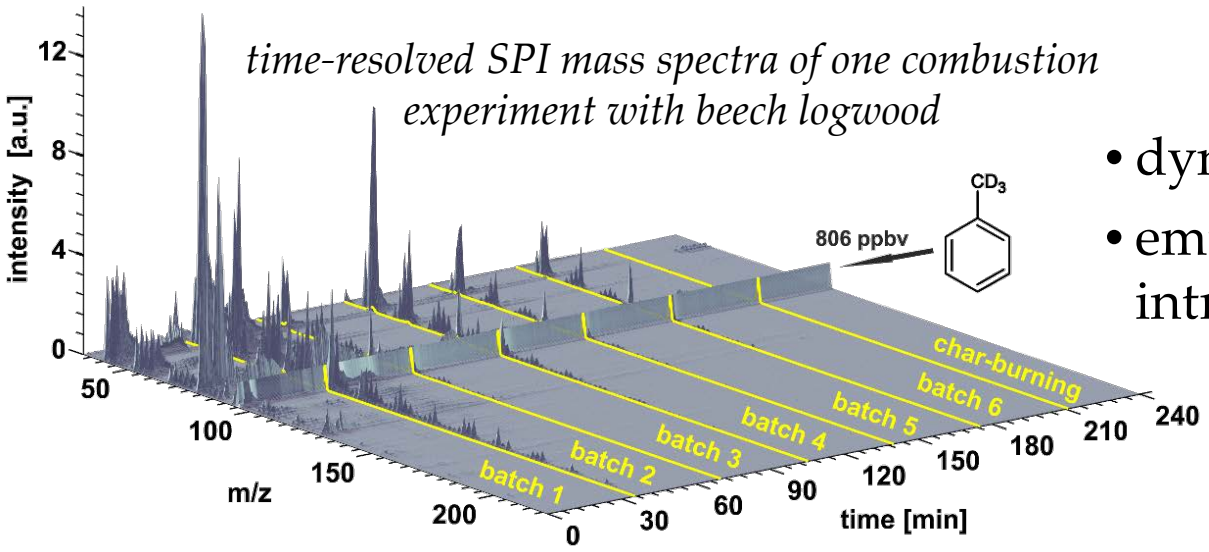
Supplemental slides

Basic Experimental Setup



Reda, Czech, Jakobi, et al., *Energy & Fuels*, 29(6) (2015), 3897–3907

Dynamic Emissions from Logwood Combustion



- dynamic emission pattern
- emission maximum at introduction of new batch

- **>50% of emissions during first 70min** (first two batches)
- **within one batch concentrations** of single species change over **2-3 orders of magnitude**

  → consideration **possible exceedance of threshold values**, e.g. for aldehydes, in exposure studies

Czech, Sippula, Kortelainen et al., *Fuel*, 177 (2016), 334-342