Wood combustion: Emissions and contribution to Secondary organic aerosol budget

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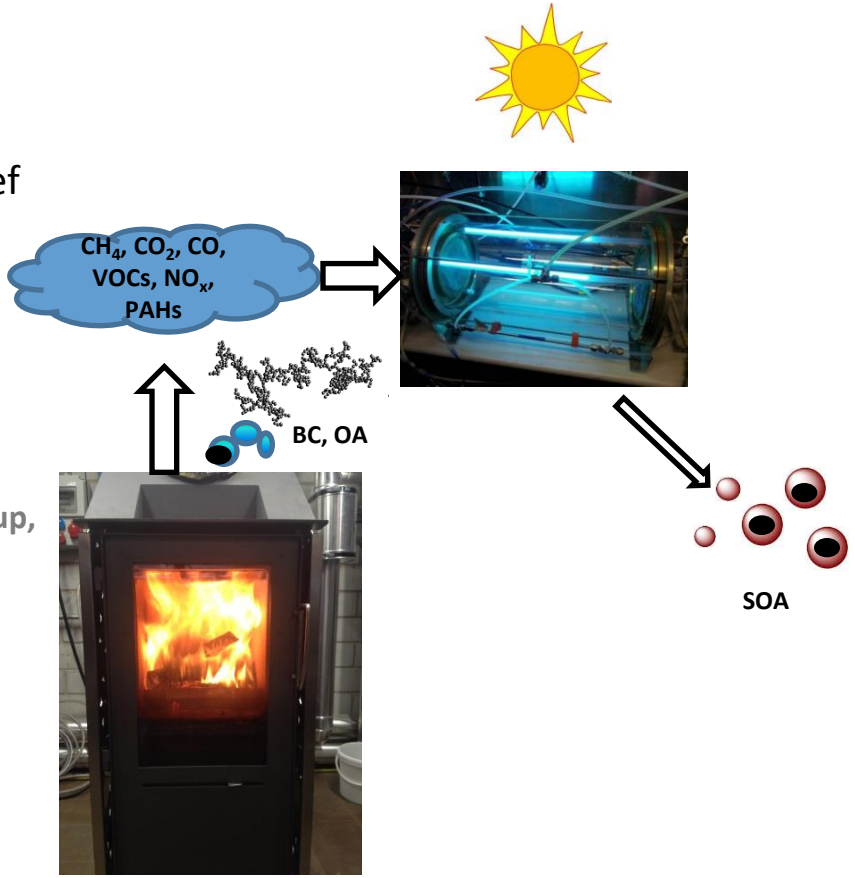
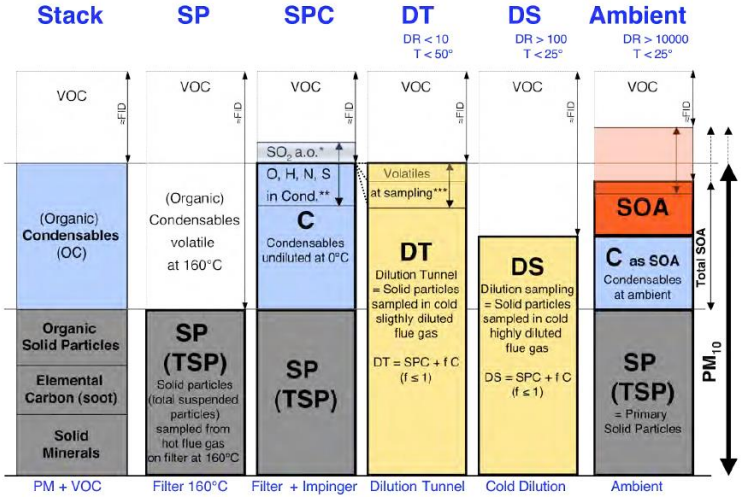
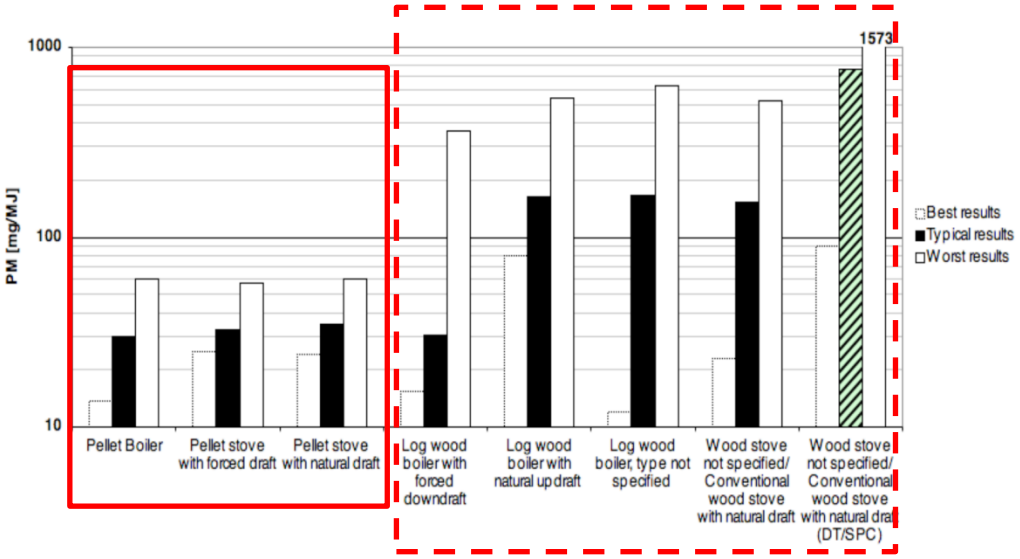


Introduction

- ~3 million people use small scale appliances for domestic heating (World Energy Council, 2007)
- Contributes ~90% of total POA mass from combustion and enhanced SOA mass (Bond et al., 2004, Alfarra et al., 2007)
- >30% contribution to carbonaceous matter in Europe and dominant source in Swiss Plateau and alpine region (Sandradewi et al., 2008; Lanz et al., 2010)



Comparison of different sampling techniques with total PM in flue gas (Nussbaumer et al., 2008a, b)

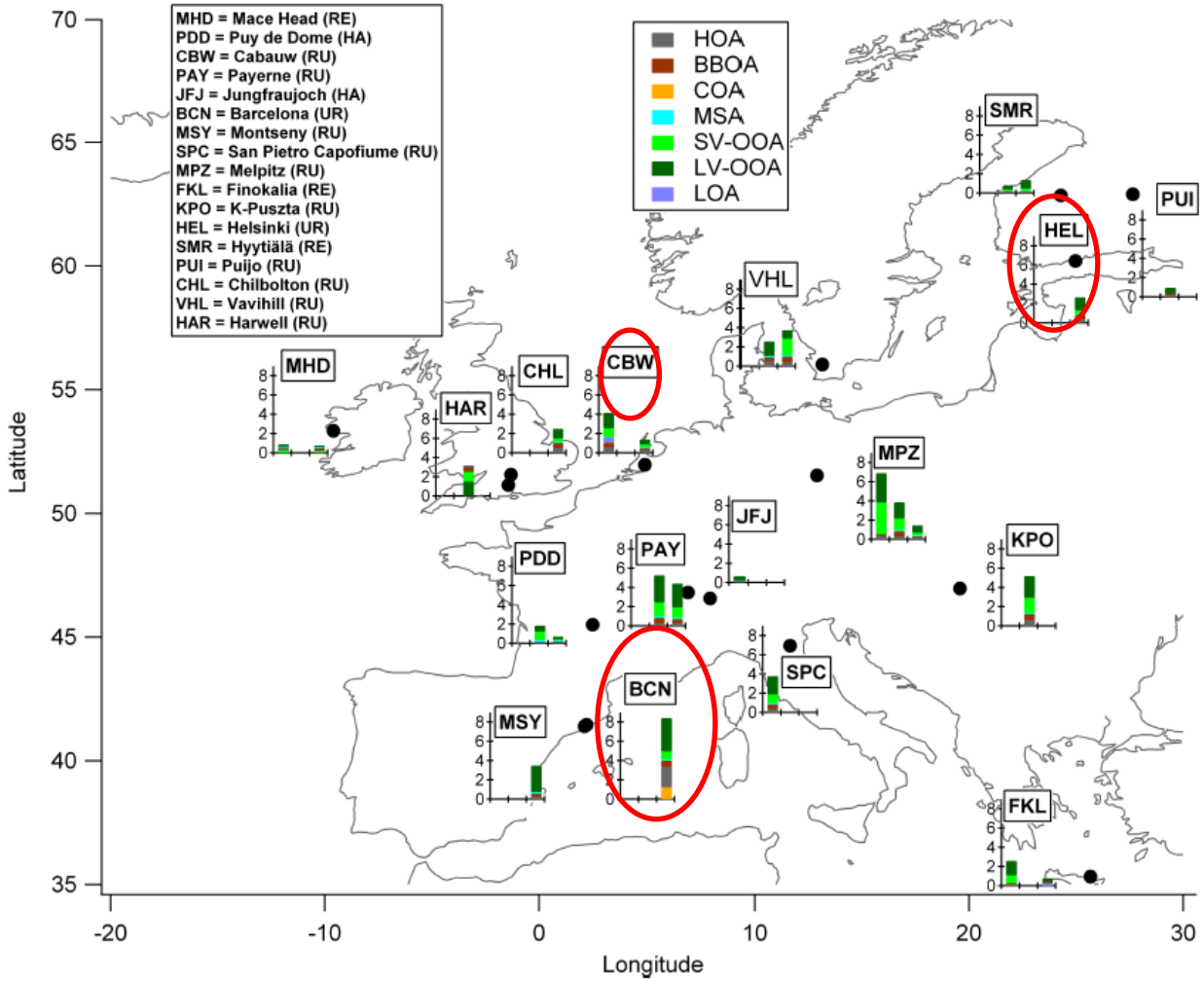


Average emission factors from different wood combustion appliances (Nussbaumer et al., 2008a, b)

Appliance	DT (gGJ^{-1})	SP (gGJ^{-1})
Traditional stove	800 (290-1932)	150 (49-650)
Fire place	900	260 (23-450)
Medium boiler	80 (30-350)	70 (30-350)

Average DT (dilution tunnel) and SP (Solid particles) particle emission factors in Europe in 2005 (Denier van der Gon et al., 2015)

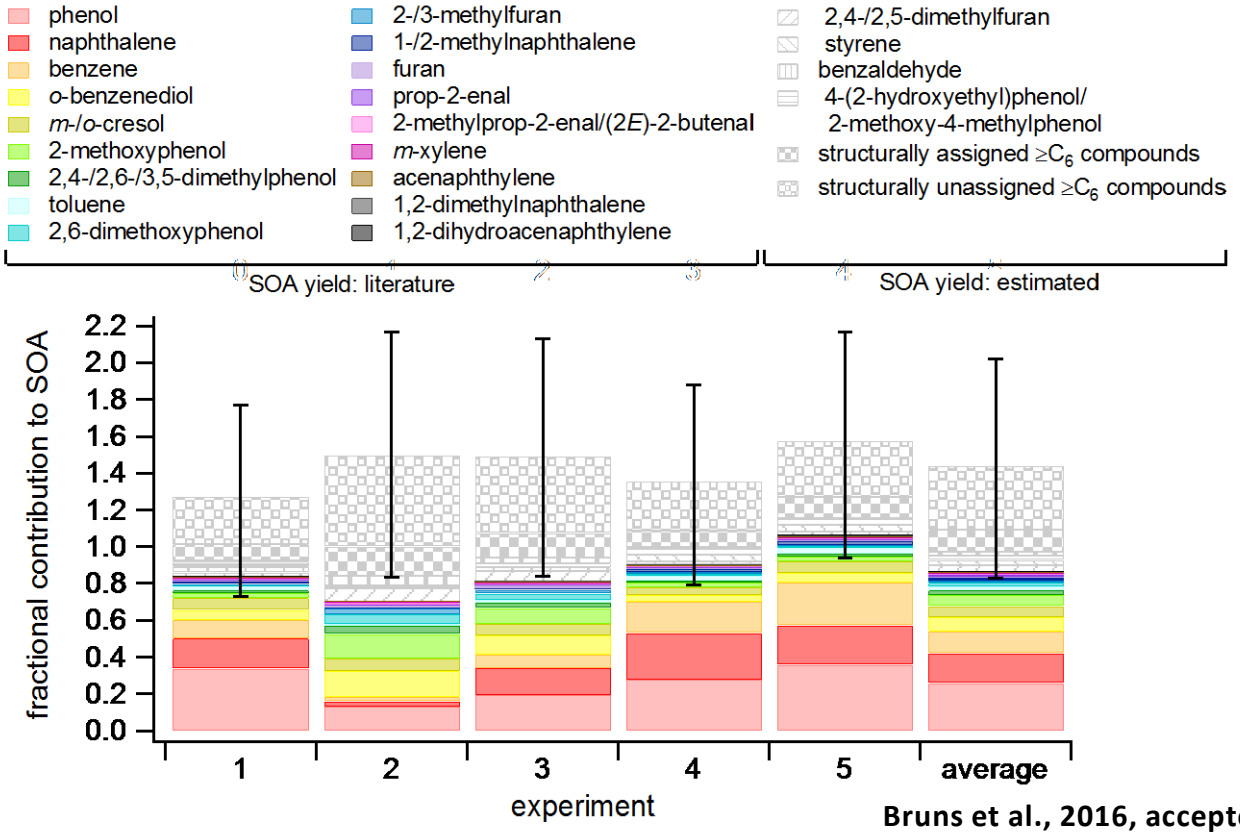
Spatial distribution over Europe



Measurements from intensive field campaigns in 2008 and 2009 (EUCAARI: European Integrated Project on Aerosol Cloud Climate and Air Quality Interactions and EMEP: European Monitoring and Evaluation Programme) (Crippa et al., 2014)

Secondary organic aerosol precursors

- Large differences in recent global estimates of SOA production from large measurement datasets and current emission inventories
- Traditional SOA precursors: benzene, toluene and α-pinene (accounts <20% SOA formed)
- Inclusion of SVOC's IVOCs : Volatility Basis Set Approach

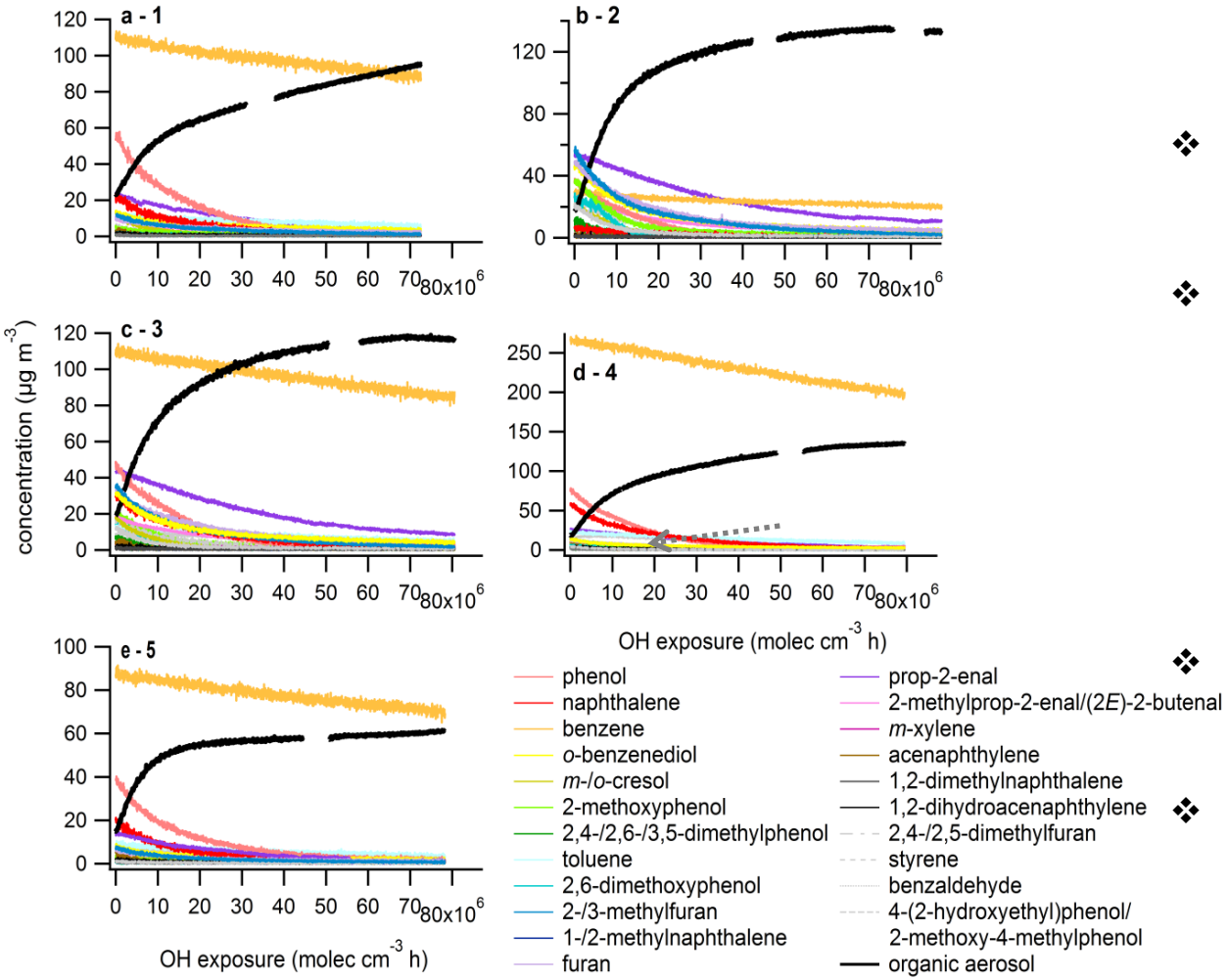


Bruns et al., 2016, accepted in Nature *Sci. Reports*

Fractional contribution of 22 individual NMOGs and two lumped NMOG categories to observed SOA

- 84-116% of the observed SOA is accounted for by these 22 NMOGs

Evolution of gas phase SOA precursors

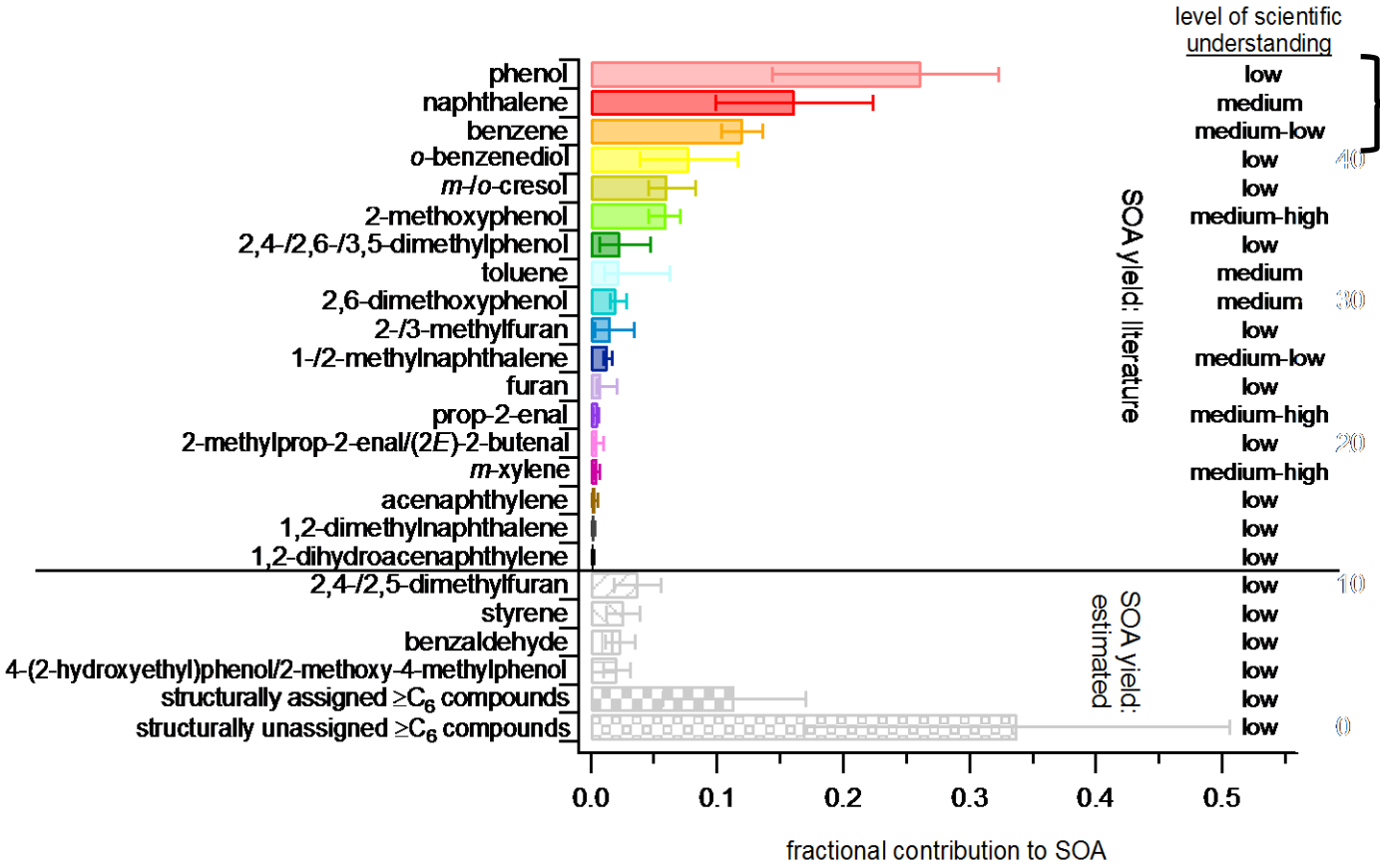


Evolution of 22 individual NMOGs and organic aerosol with aging

- **3-7 times more SOA than primarily emitted (~3 days of aging)**
- Previously, wood burning chamber experiments showed SOA:POA of 1.5 or more and showed an OA enhancement of 3 and 1.6 for high and low loading in modern masonry heater (Bruns et al., 2015, Heringa et al., 2011).
- Nearly 70% of the mass is structurally identified.
- In addition, high monoterpenes emission from soft wood combustion should be considered in models.

Bruns et al., 2016, accepted in Nature *Sci. Reports*

Contribution of non-traditional SOA precursors



~80% contribution to observed SOA

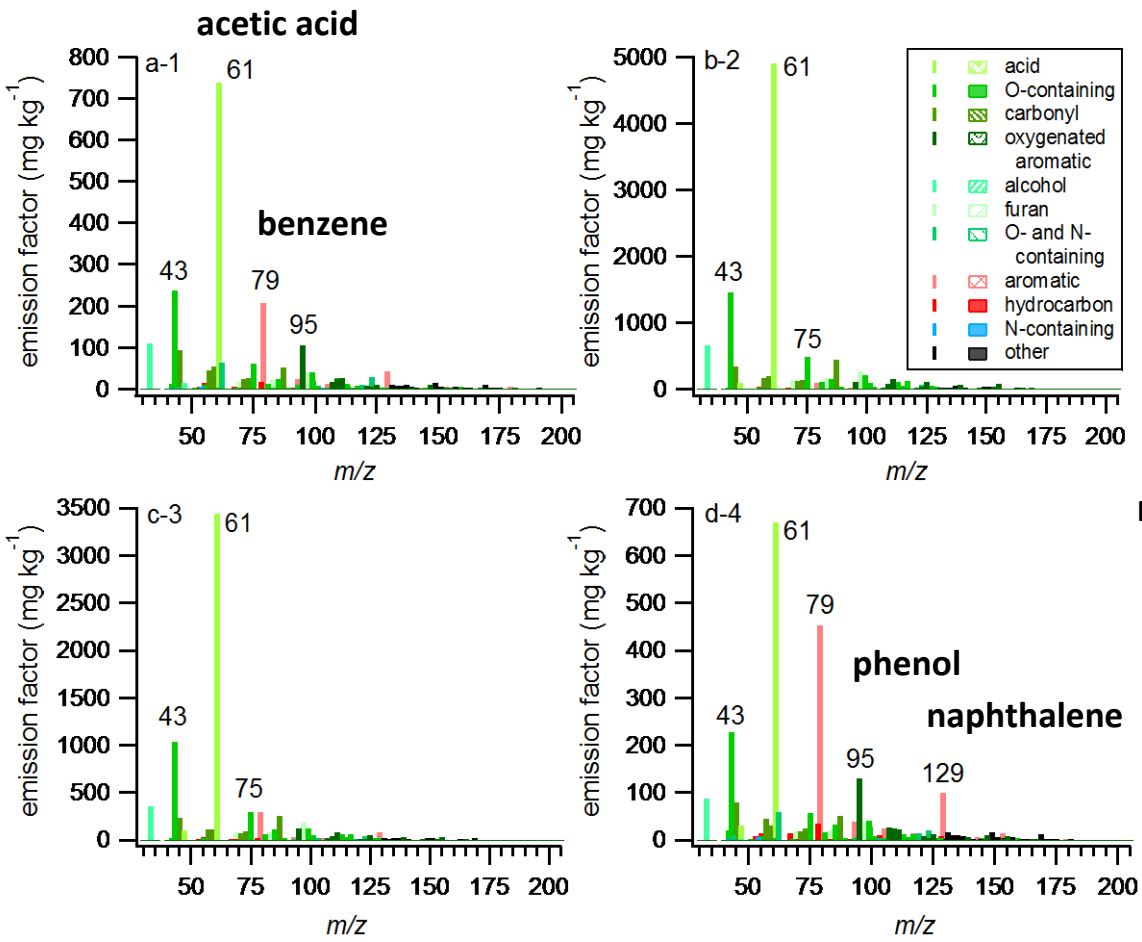
Traditional NMOGs contributed up to 27%

Average fractional contribution to observed SOA and level of scientific understanding of 22 individual NMOGs and two lumped NMOG categories

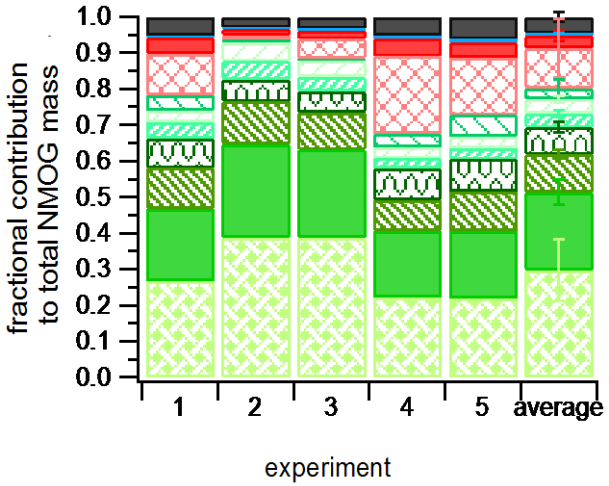
Bruns et al., 2016, accepted in Nature *Sci. Reports*

- **Inclusion of these identified NMOGs and terpenes in Chemical transport models is required for modelling residential wood combustion SOA**

Primary gas phase emissions



Mass spectra of primary emissions from Beech wood measured by PTR-ToF-MS and categorized by functional groups

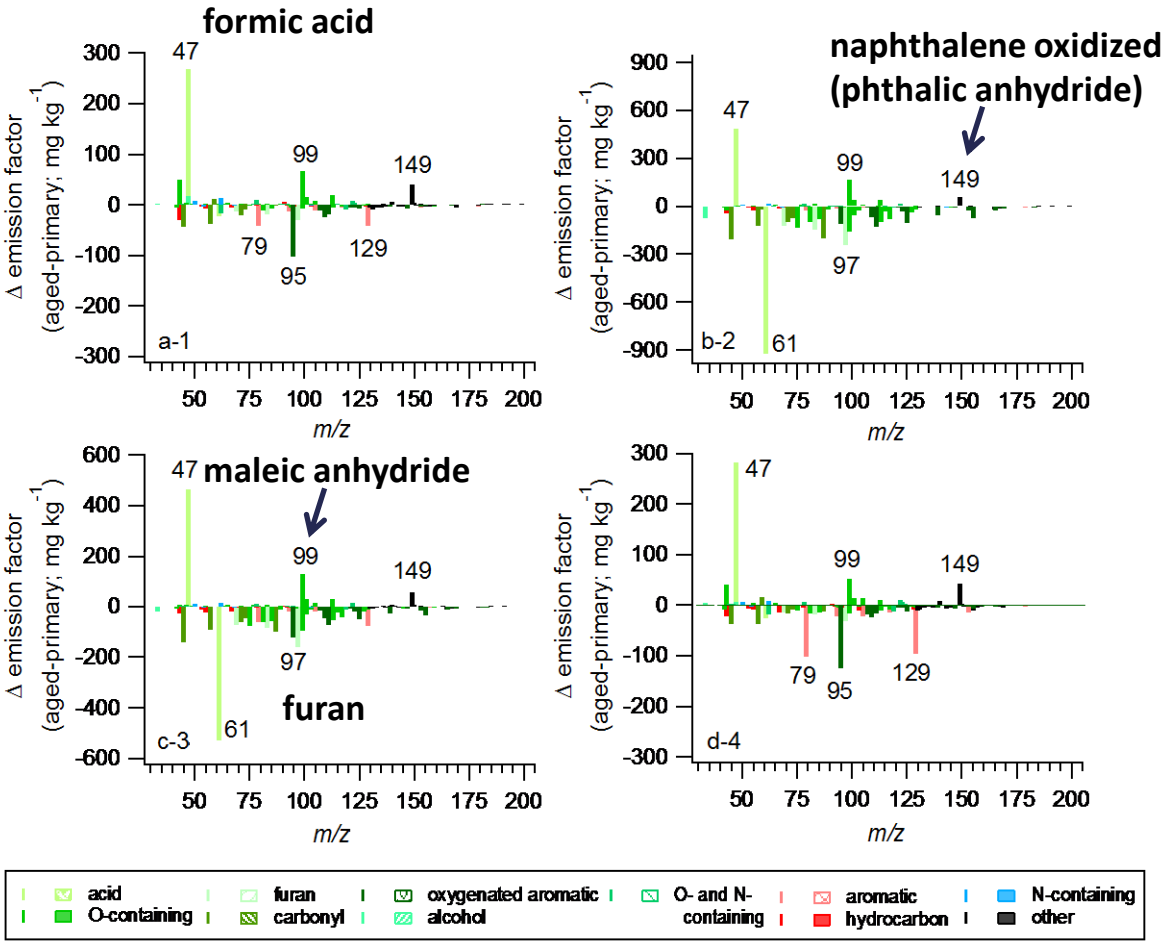


Fractional contribution of each functional group to total NMOGs

- 68-94% contribution from oxygenated species (mainly acetic acid)
- Major contributors in aromatics class: phenol, benzene and naphthalene.

Bruns et al., submitted

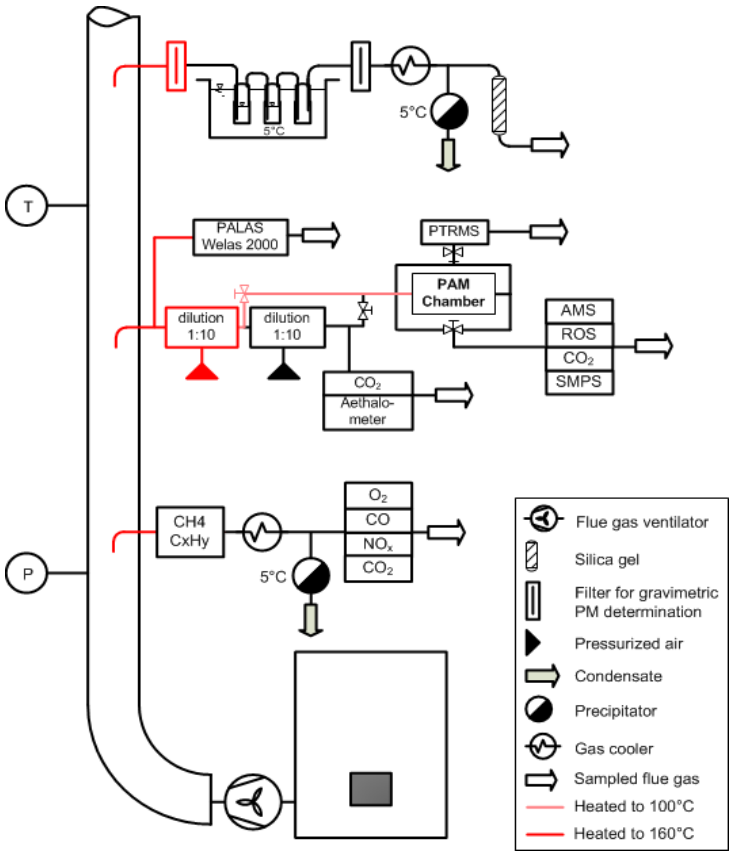
Gas phase evolution with aging



Absolute difference of aged and primary EF mass spectra

Bruns et al., submitted

Experimental details



Schematic of the experimental setup

Combustion devices

Log wood stove

Pellet boiler

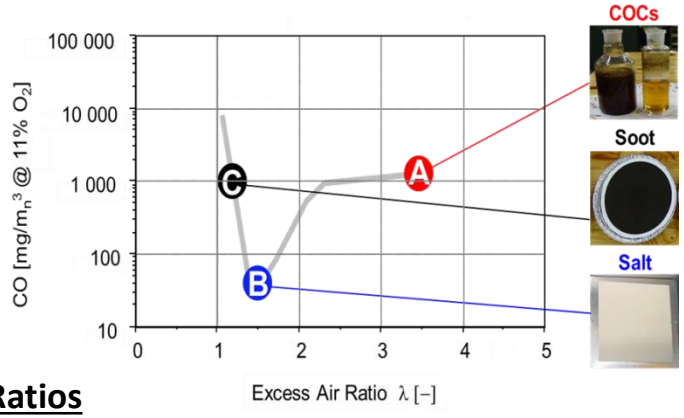
Types of fuel

Log wood

Wood pellets

Wood chips

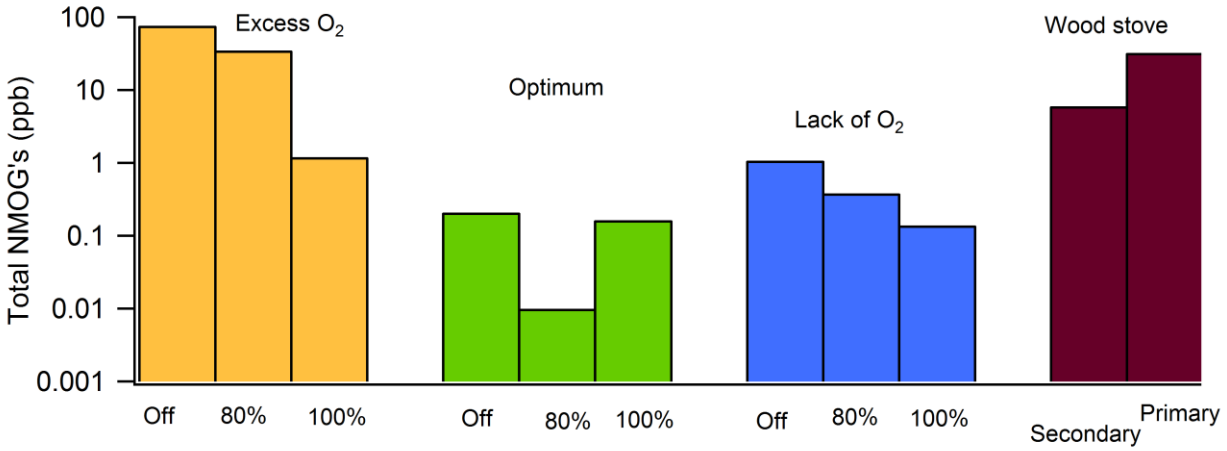
Burning regimes



Excess Air Ratios

Pellet boiler (lack of O_2) : 1.3-1.35
Pellet boiler (Excess of O_2) : 2.67-3.38
Pellet boiler (optimum) : 1.54-1.64
Log wood stove(Flaming): 2.7-10.9
(Burn out): 6.67-10.72

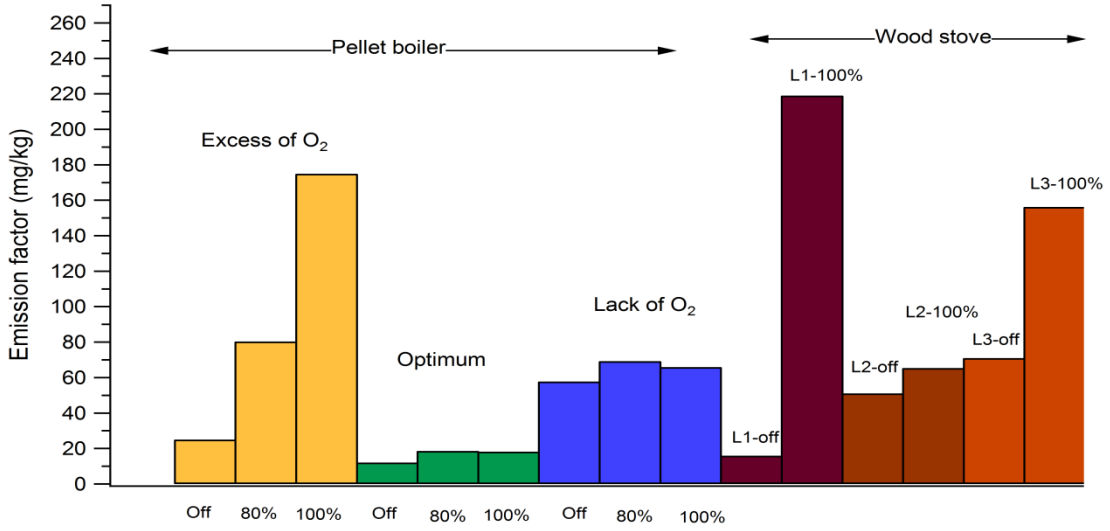
Emissions from different combustion devices



- Primary emissions were nearly same in flaming conditions for both pellet boiler (Excess O_2 condition) and log wood stove but highly variable in the latter.

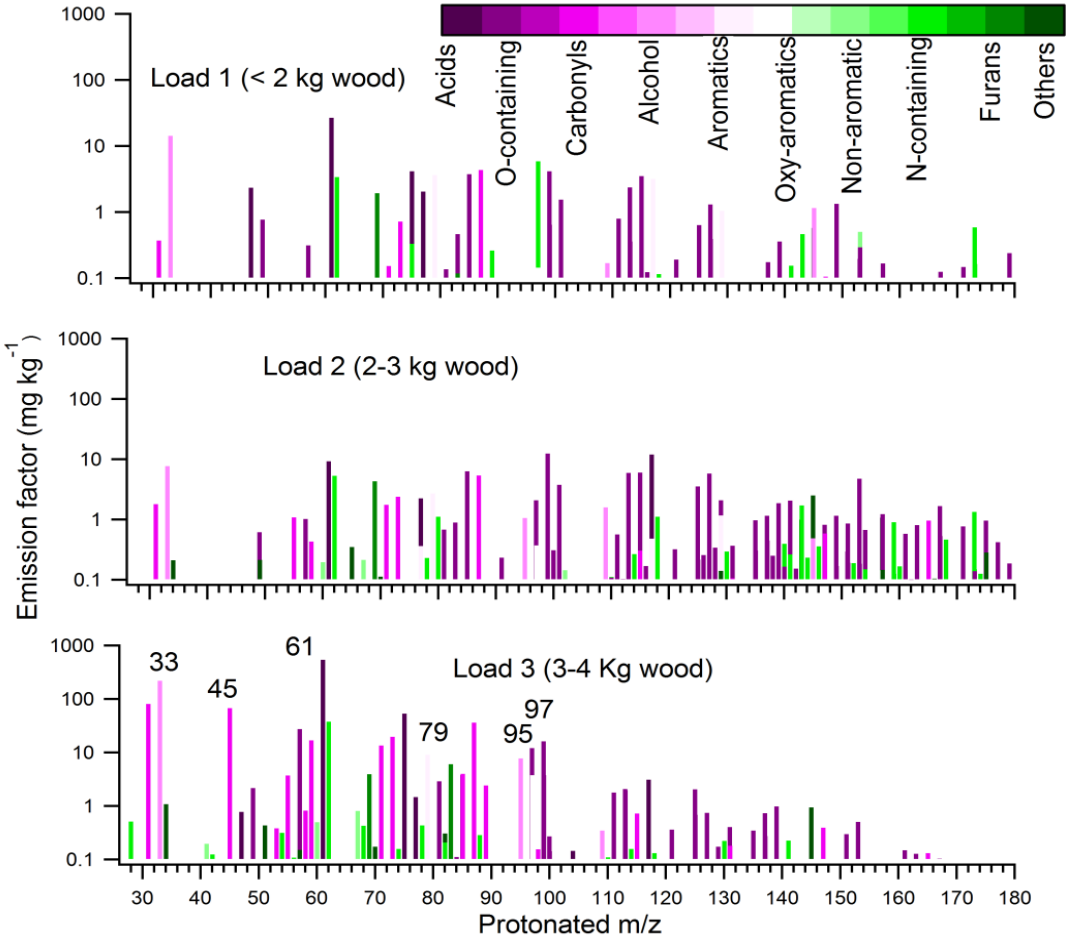
Total NMOG's emitted from different combustion devices under flaming conditions and different Air-to-fuel ratios

- SOA: POA ratio in log wood stove is 3 times higher as that of pellet boiler (Excess O_2 conditions) and highly variable with operating conditions



OA emission factors from different combustion devices under flaming conditions and different Air-to-fuel ratios

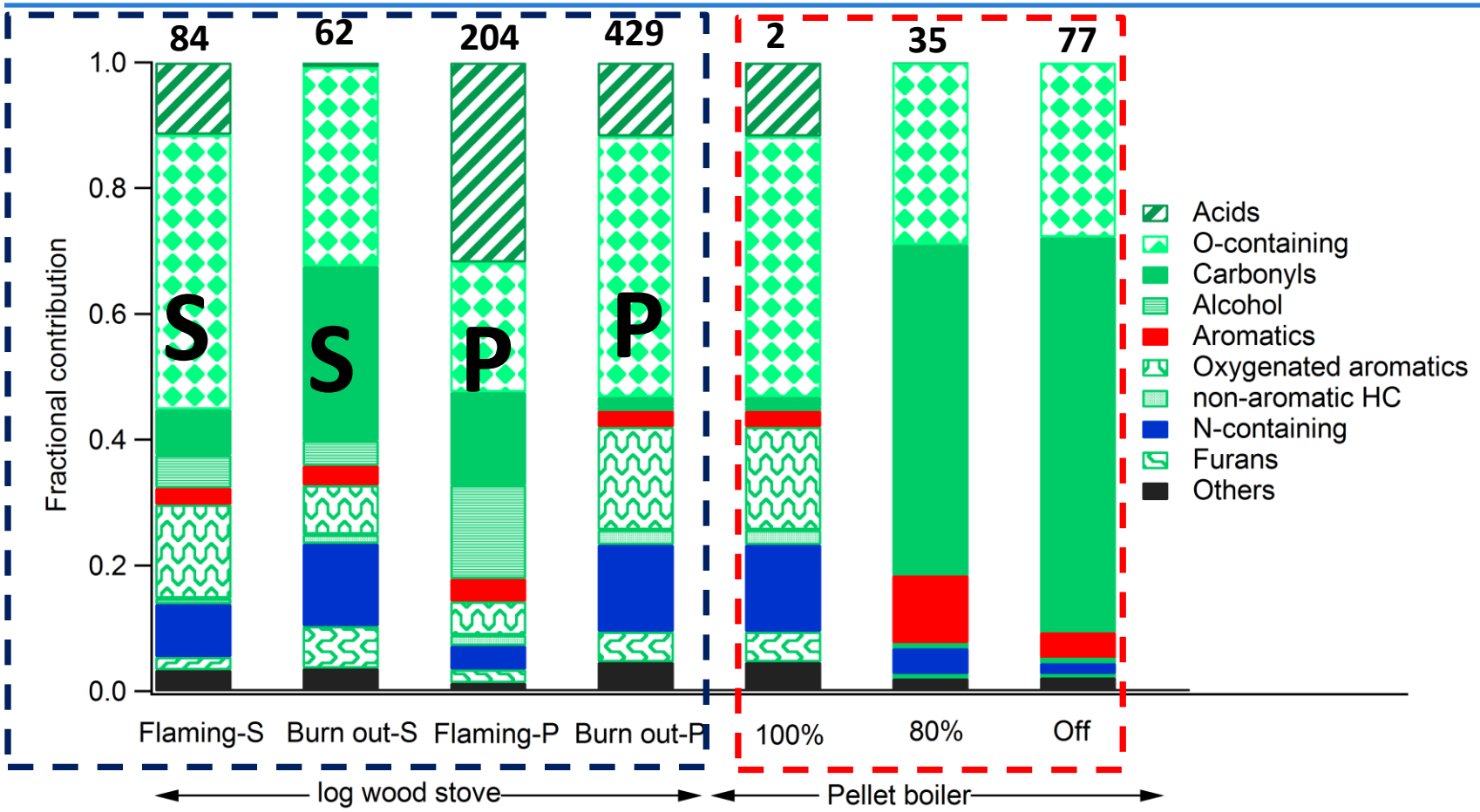
Emissions from different load conditions



Fractional contribution of different functional groups according to the wood load in log wood stove

- Methanol, acetic acid and formaldehyde increased by a factor of ~ 10 with increase in load
- Aromatics emissions increased by a factor of up to 2-3

Contribution to Primary and Secondary NMOGs



Fractional contribution of different functional groups during different combustion conditions and combustion devices (Pellet boiler (flaming with Excess O_2) and log wood stove (burn out and flaming)

- Pellet boiler emissions are dominated by carbonyls among which equal portion is shared by formaldehyde and acetaldehyde (~ 43-45%)
- Primary emission of acids, carbonyls, alcohols is more in flaming phase (log wood stove) whereas more O-containing, oxygenated aromatics, N-containing species and furans are present in burn out phase

Conclusions

- 22 non-traditional NMOGs identified account for ~84-115% of SOA mass
- Evolution of different NMOGs from residential wood combustion with aging is due to their highly reactive nature.
- Due to inter-burn variability, constraining the EF's is difficult.
- Highest emissions from log wood stove with variability in NMOGs, primary organic aerosol (POA) and BC depending on the operation conditions.
- Formaldehyde (43%) and acetaldehyde (45%) are the major contributors in Pellet boiler emissions whereas in log wood stove acids and alcohols are additionally emitted primarily.
- As NMOGs are related to SOA formation and might be used as an SOA indicator

Acknowledgements

- Swiss National Science Foundation (NRP 70 “Energy Turnaround”; WOOSHI grant 140590 and starting grant BSSGI0_155846)
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- Competence Center Environment and Sustainability (CCES) (project OPTIWARES)

Fore more details, please visit poster entitled:

(1) Wood combustion for energy in buildings (Poster no. 26, Poster Session 2: Biomass Combustion)

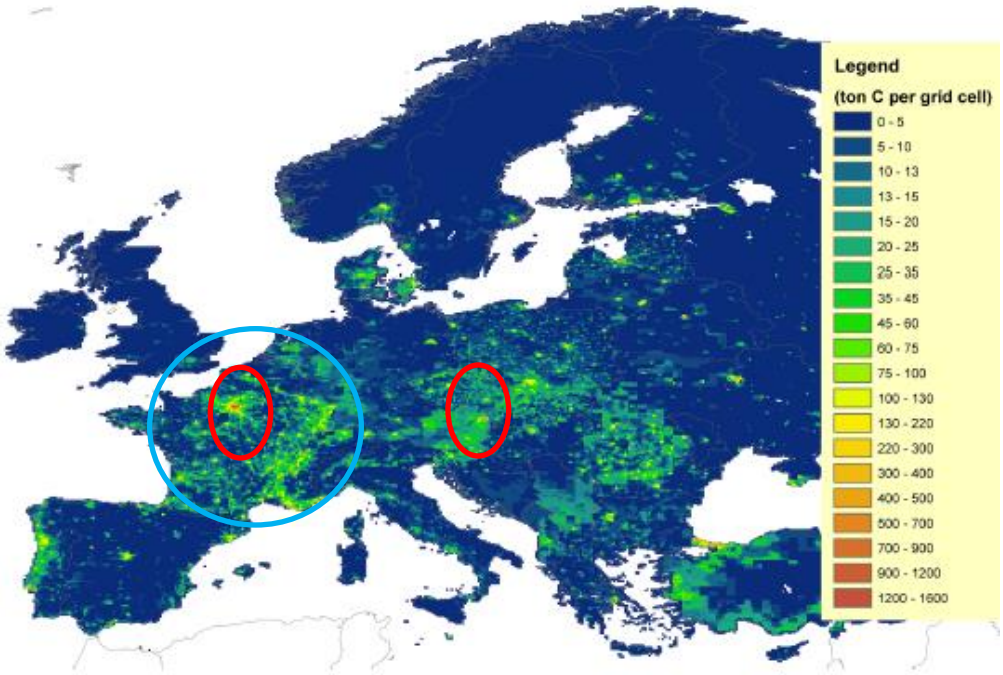
P. Zotter, D. Bhattu, J. Zhou, G. Stefenelli, A. Lauber, I. El Haddad, J. Slowik, A. S. H. Prévôt, J. Dommen, & T. Nussbaumer

(2) Toxicity assessment of wood combustion emissions from different burning and aged conditions by using a reactive oxygen species (ROS) analyzer (Poster no. 78, Poster Session 6: Health)

J. Zhou, E. A. Bruns, P. Zotter, G. Stefenelli, D. Bhattu, T. Nussbaumer, J. G. Slowik, U. Baltensperger, A. S. H. Prévôt, I. El Haddad, & J. Dommen

Thank you for your attention!

Revised emission estimates

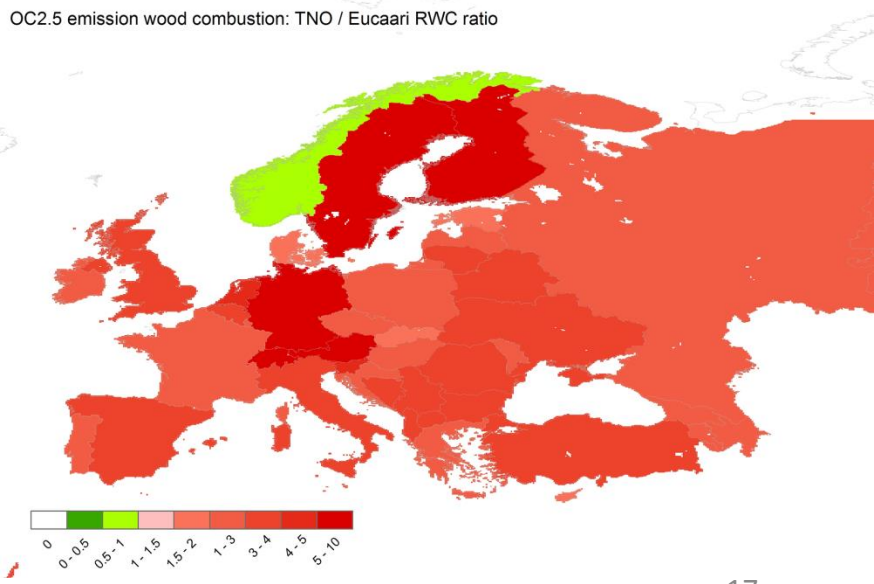


Spatial distribution of OC emissions (as per TNO-new RWC inventory) (Denier van der Gon et al., 2015)

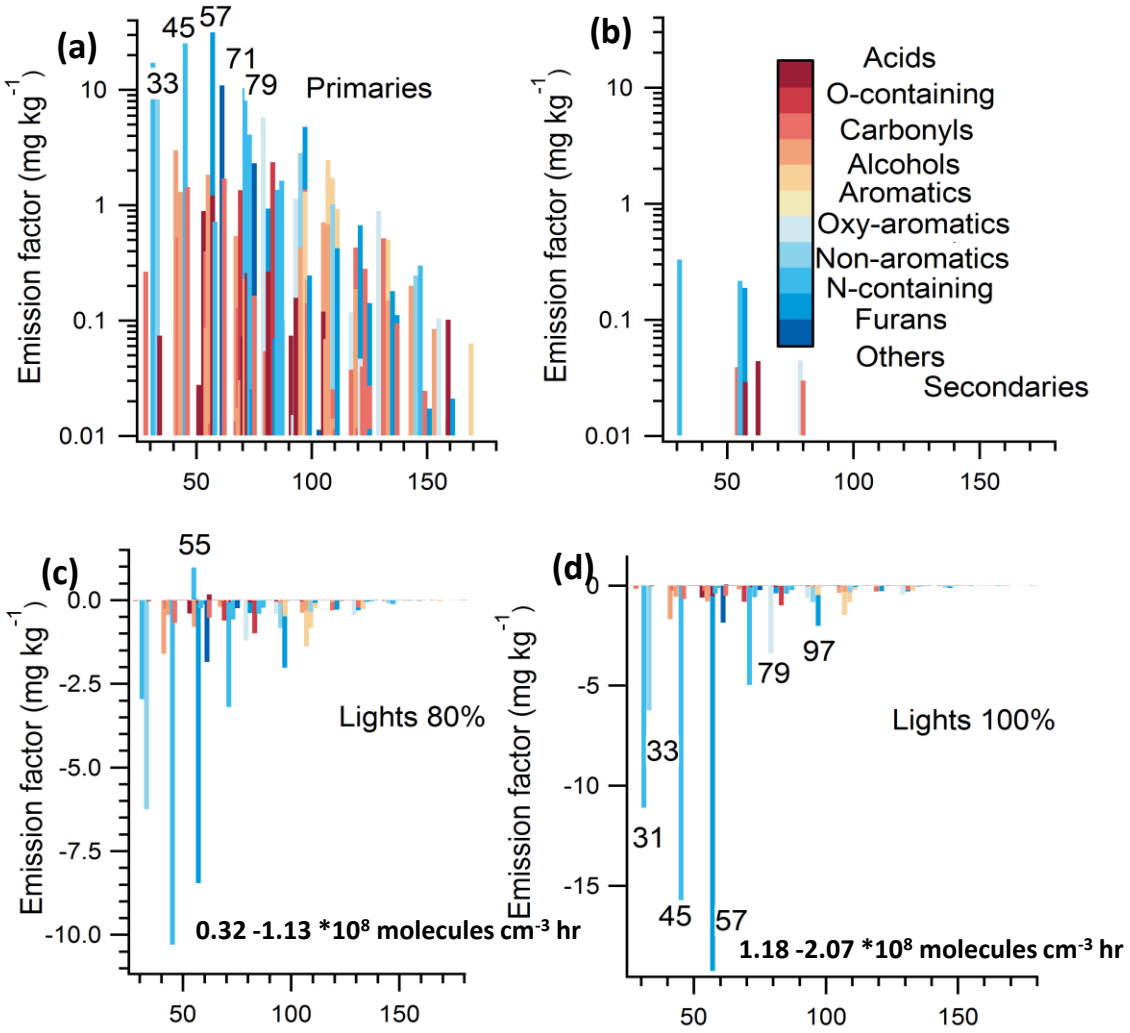
Totally neglecting the secondary aerosol formation from small scale appliances can lead to less effective current EU emission policies and actions (EU commission regulation 2015/1185) (Leskinen et al., 2016)

- Implementation of revised bottom-up emission inventory for anthropogenic carbon aerosols (mainly RWC) showed better agreements of models results with observations.
- Revised RWC emissions are 2-3 times higher than previous inventory which further leads to 20% increase in European $PM_{2.5}$ emissions.

Ratio of revised TNO-new RWC inventory relative to the previous EUCAARI OC emission inventory (Denier van der Gon et al., 2015)

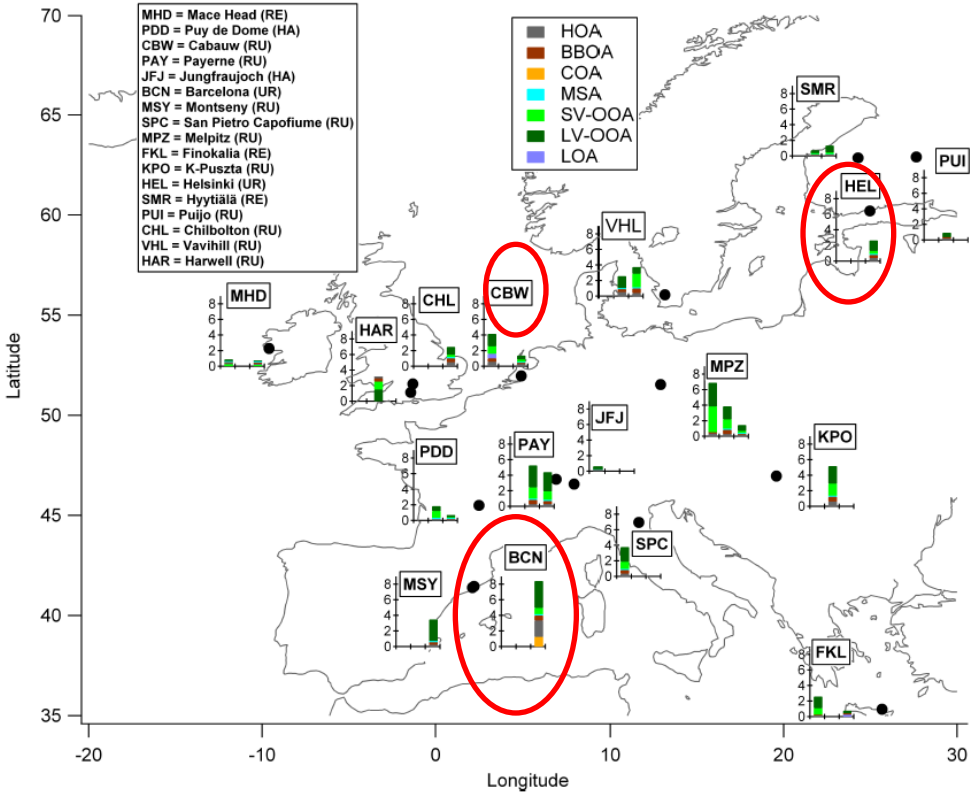


Evolution of primary emissions (pellet boiler)



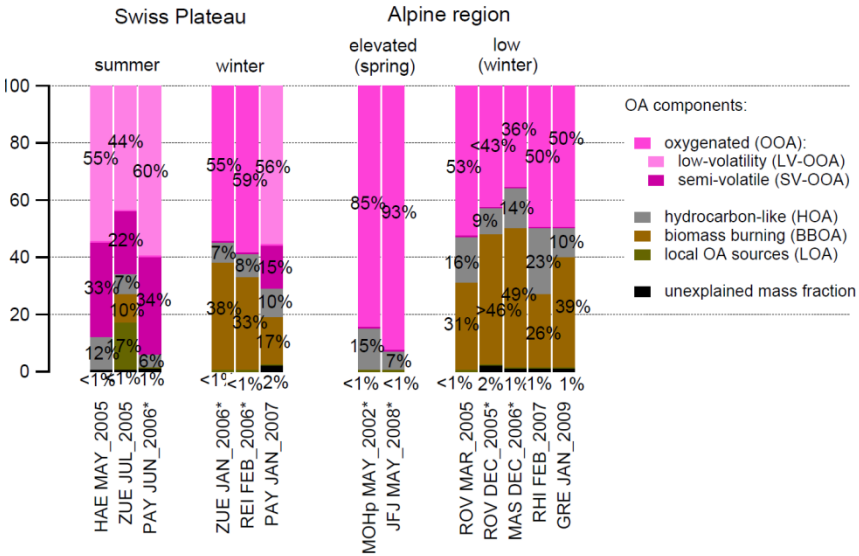
(a) Primary and (b) secondary emission factors mass spectra from pellet boiler under flaming conditions (Excess O_2). (c and d) Absolute differences of primary and secondary emission factors mass spectra under different OH exposure conditions

Spatial distribution over Europe



Measurements from intensive field campaigns in 2008 and 2009 (EUCAARI: European Integrated Project on Aerosol Cloud Climate and Air Quality Interactions and EMEP: European Monitoring and Evaluation Programme) (Crippa et al., 2014)

Aerosol composition in Central Europe



Relative contribution of OA components over two sites (Swiss plateau and Alpine region: low and high altitudes) in summer and winter (Lanz et al., 2010)