16th ETH-Conference on Combustion Generated Nanoparticles
June 24th – 27th 2012

Paper/Poster-Abstract Form

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Title:
First results on primary emissions and secondary organic aerosol formation from gasoline and diesel vehicles as well as scooters

Abstract: (min. 300 – max. 500 words)

The abstracts for papers and posters must contain unpublished information on your research subject: background, investigation methods, results and conclusions. Graphs and references are very welcome. Acronyms should be avoided. Abstracts with < 300 words can not be considered. General information on products which are already commercially available can not be accepted as presentations for the conference but are very welcome at the exhibition of particle filter systems and nanoparticle measurement instruments.

A large fraction of ambient particulate matter (PM) consists of organic aerosol (OA), either primary (POA), from direct emissions, or secondary (SOA), formed from gaseous precursors. Therefore any attempt to mitigate the effect of on-road vehicle emissions on public health and the environment should consider not only primary aerosol emissions but also the secondary aerosol production potential (SAPP). However, only the primary aerosol emissions from on-road vehicles are currently subject to direct vehicle legislation.
We determined emission factors (EFs) (g / kg fuel) for primary PM and for SOA, from two- and four-stroke scooters, a gasoline car and a truck, all complying with the latest European emissions standards. We also quantified the effect of ambient temperature on emissions and SOA formation from gasoline cars and from trucks. Vehicle exhaust was introduced into the new PSI mobile smog chamber during regulatory driving cycles on chassis dynamometers at the European Joint Research Centre Ispra, Italy. A high-resolution time-of-flight aerosol mass spectrometer equipped with a PM 2.5 lens was used to quantify OA. Black carbon was measured using a multi angle absorption photometer and

an Aethalometer (AE 33 prototype). A high resolution proton transfer time-of-flight mass spectrometer was used to investigate volatile organic compounds gases including the determination of the OH clock using deuterated n9-butanol (Barmet et al., 2012). Primary exhausts of 2-stroke scooter emissions could be shown to be very high whereas the SAPP versus primary organic emissions of gasoline cars and 4-stroke scooters were strongly enhanced. A summary of the results will be presented at the conference.

This work is supported by the Swiss Federal Office for the Environment, the Federal Roads Office, the French environmental agency and the EUROSTARS project E!4825 "FCAeth".

Barmet et al. (2012) Atmos. Meas. Tech. 5, 647-656.

Short CV:
- Physics at ETH Zürich
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- Postdoc in Boulder, USA and Paul Scherrer Institute, Switzerland
- Since 2000: Head of gasphase and aerosol chemistry group in the Laboratory of Atmospheric Chemistry, Paul Scherrer Institute, Switzerland

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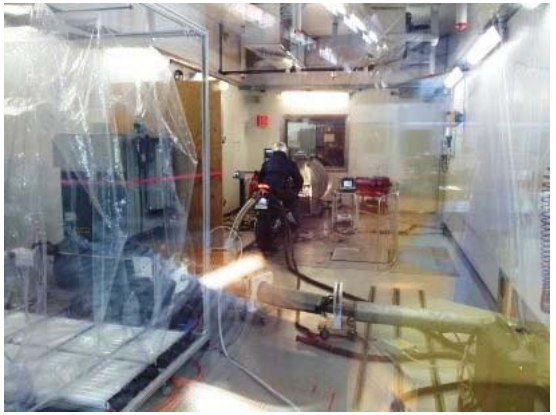
Primary and Secondary Organic Aerosol from Road Vehicles

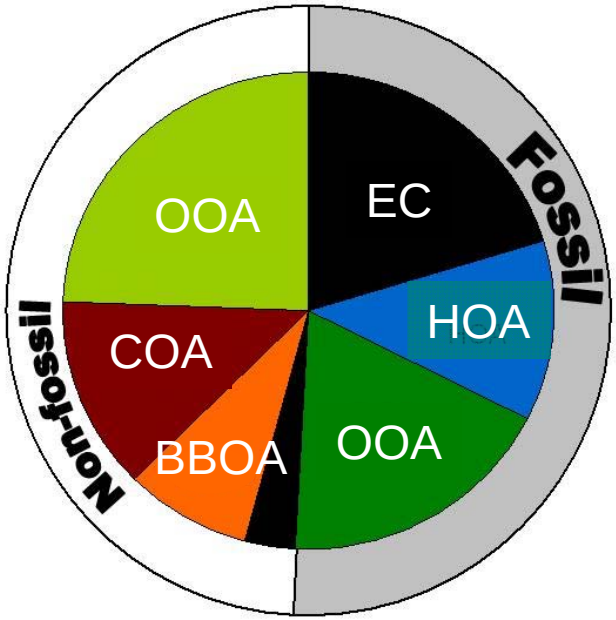
A.S.H. Prévôt, **S. M. Platt**, I. El Haddad, A.A. Zardini, M. Clairotte, C. Astorga, R. Wolf, J. G. Slowik, B. Temime, N. Marchand, R. Richter, P. Barmet, I. Jezec, L. Drinovec, G Mocnik, U. Baltensperger



Motivation

Aerosol affects climate, damages health, and reduces visibility

A major source in urban areas is road vehicles:



Elemental Carbon (EC), Organic aerosol OA: Hydrocarbon like (HOA) Primary, Oxygenated (OOA) Secondary, Biomass burning (BBOA), Cooking (COA)

Urban top-down (source apportionment: ^{14}C, PMF), up to 50% of urban background (March, Barcelona)

Primary = Direct emission
Secondary= From precursors

Minguillón et al., ACP, 2011

Motivation

Globally secondary organic aerosol SOA is significant

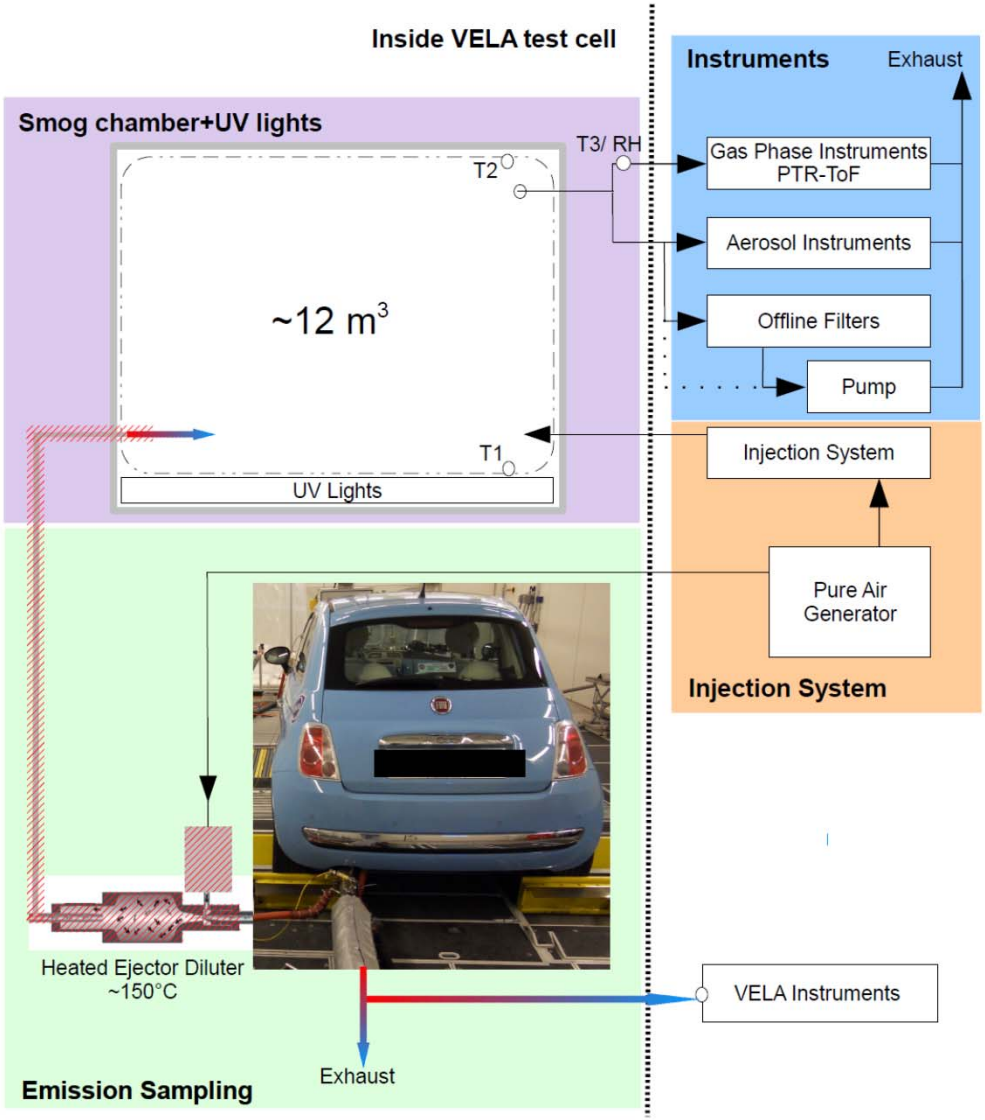
No secondary aerosol production potential (SAPP) emission factors for many large primary sources

Therefore our aims:

- Primary aerosol emission factors for different vehicle types
- Determine SAPP emission factors,
- Understanding characteristics of emissions leading to SOA formation

What happens next?

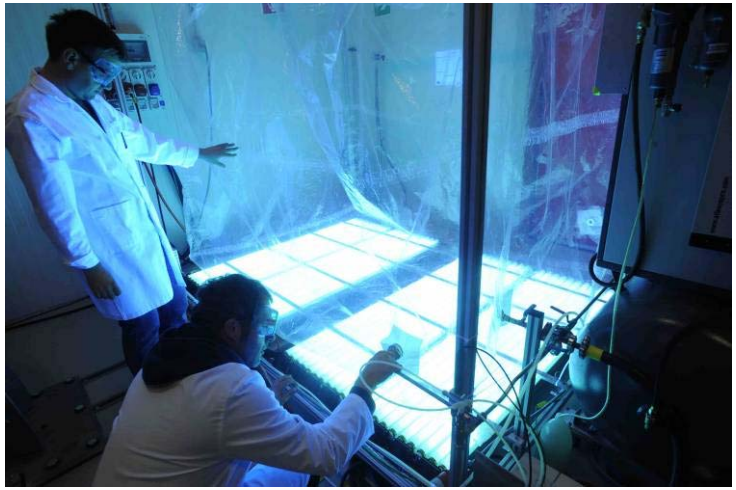
Experimental Set Up



Ispra 2011 campaign: combined PSI mobile chamber + chassis dynamometer (JRC Ispra)

Hot exhaust sampled from driving cycles

UV lights to simulate sun



Instrumentation

Smog Chamber		VELA		
Aerosol	Gas Phase	Aerosol	Gas Phase	Other
HR-ToF-AMS ($PM_{2.5}$ lens)	THC Analyser	Filter Collection (CVS)	FID	On board diagnostics
MAAP	NOx high		FTIR	T Sensors
Aethalometer	NOx low		CO/CO_2	
Prototype Aethalometer	O_3		NOx	
CPC (3775)	CO_2 Licor		GC-FID	
SMPS (Calibration unit)	CO			
EC/OC Filters	Picarro (CH_4, H_2O CO, CO_2)			
	PTR-ToF-MS			
	T, RH Sensors			

Overview

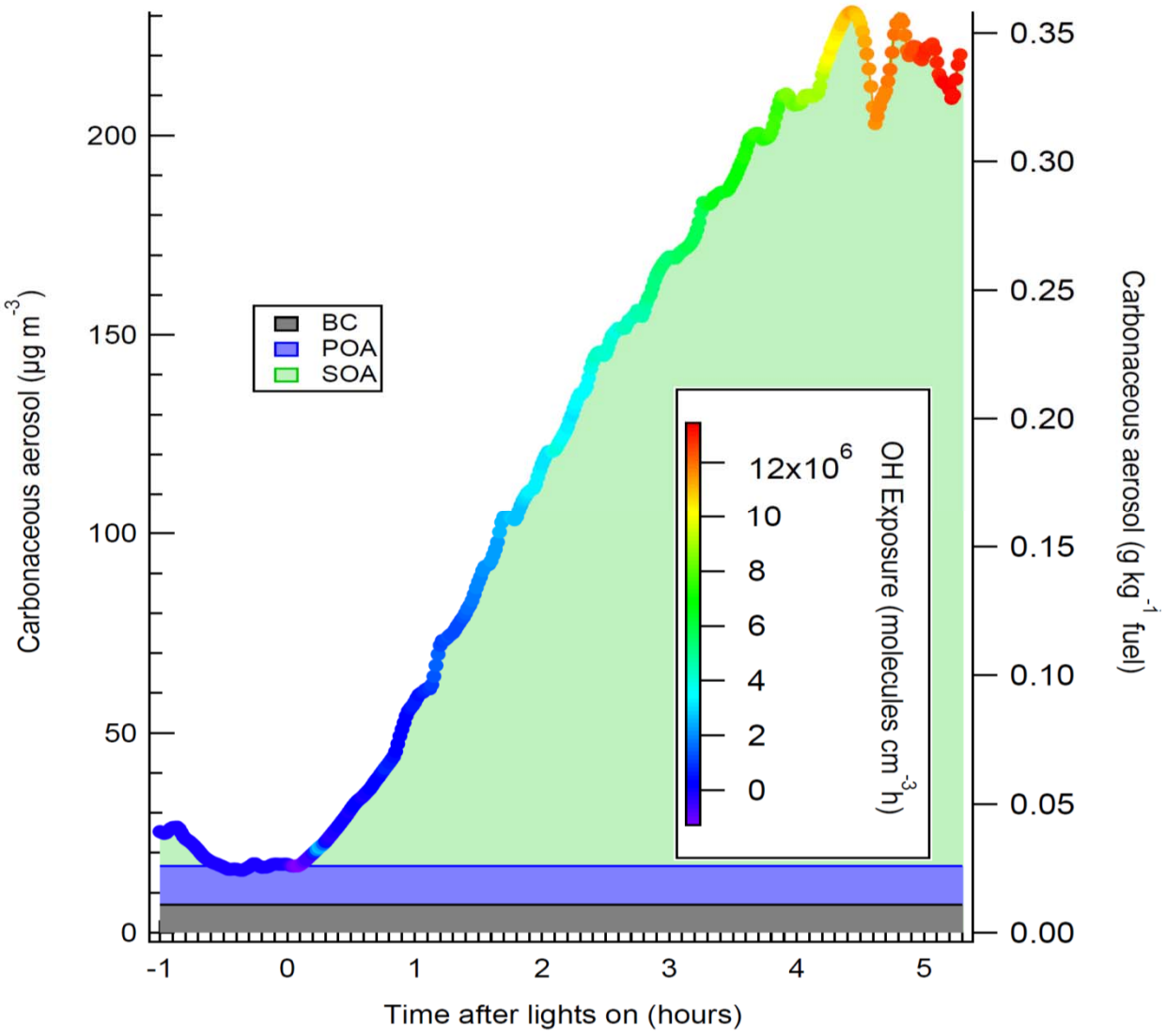
Primary emission and SOA formation for 2- and 4-Stroke Scooters, gasoline car, heavy duty diesel

Vehicles complying with latest European emissions regulations

Other parameters were varied e.g. fuel, temperature, testing phase

Vehicle	Standard	Additional Parameters Investigated	Number of Tests
2-Stroke Scooter	Euro 2	Ultra clean fuel/ lubricant, Test phase	11
4-Stroke Scooter	Euro 2	Ultra clean fuel	4
Gasoline Passenger Car	Euro 5	Temperature 22/-7℃	4
Heavy Duty Diesel	Euro 5	LPG Flexi fuel, Temperature 22/-7℃	6

Example: Gasoline emissions aging



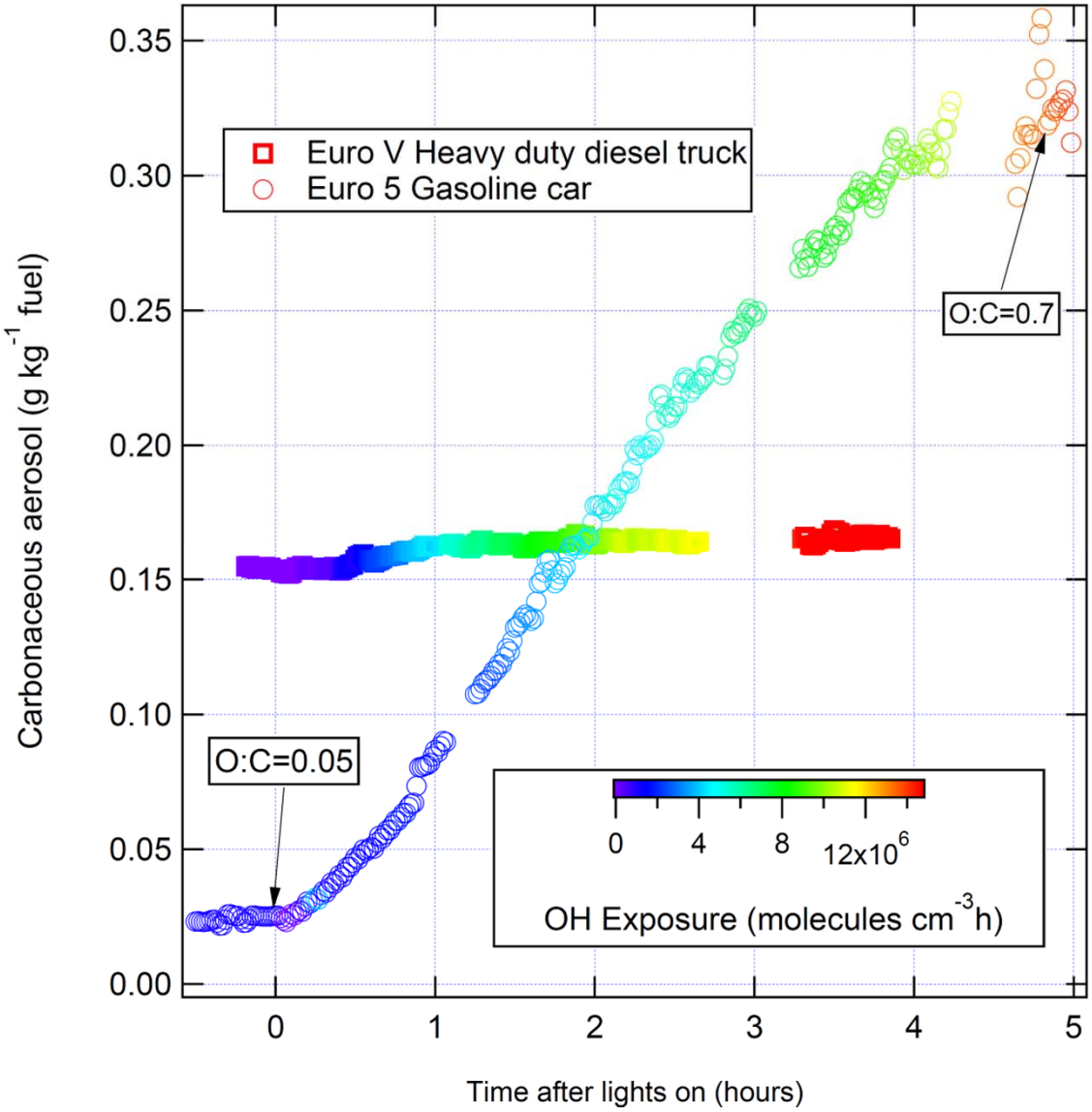
Wall loss corrected AMS + Aethalometer data

Data from smog can be used for emission factor calculation

Chamber aging related to atmospheric aging via [OH] determination

C=Carbon, from CO_2, CO, Hydrocarbon (HC), primary organic aerosol (OA), WC=fuel carbon content

Comparison: Diesel and Gasoline



Higher primary emissions from diesel vehicle

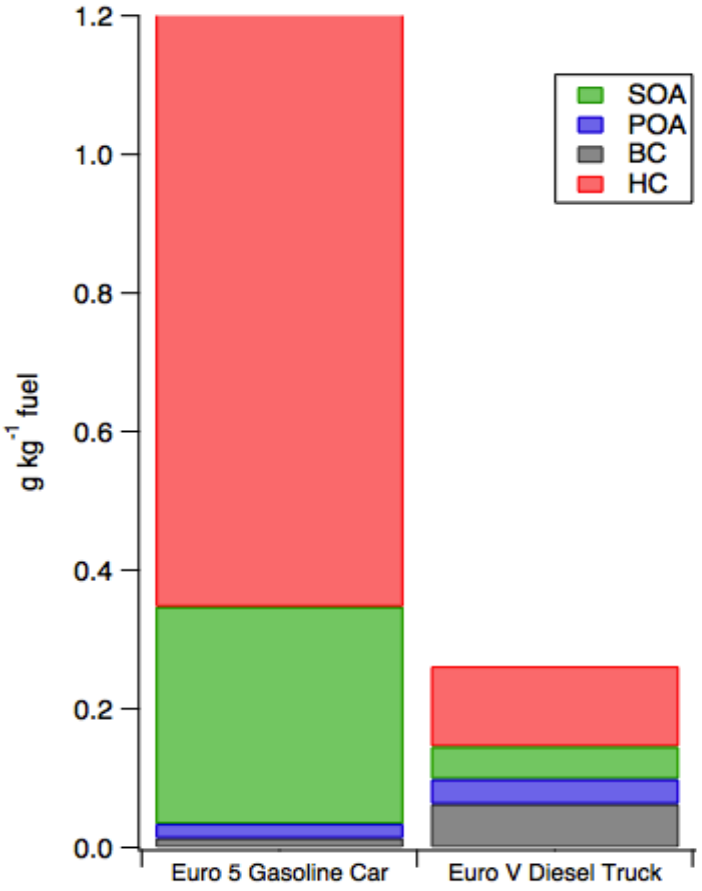
But SOA formation from gasoline emissions is higher (even including BC)

Largest emissions from gasoline (12 hours in the atmosphere)

Gasoline emissions become highly oxidised: unique feature

Comparison: Diesel and Gasoline

Average composition after aging:



BC dominates diesel PM emission

POA is higher for diesel

SOA highest for gasoline (compared at 12h OH)

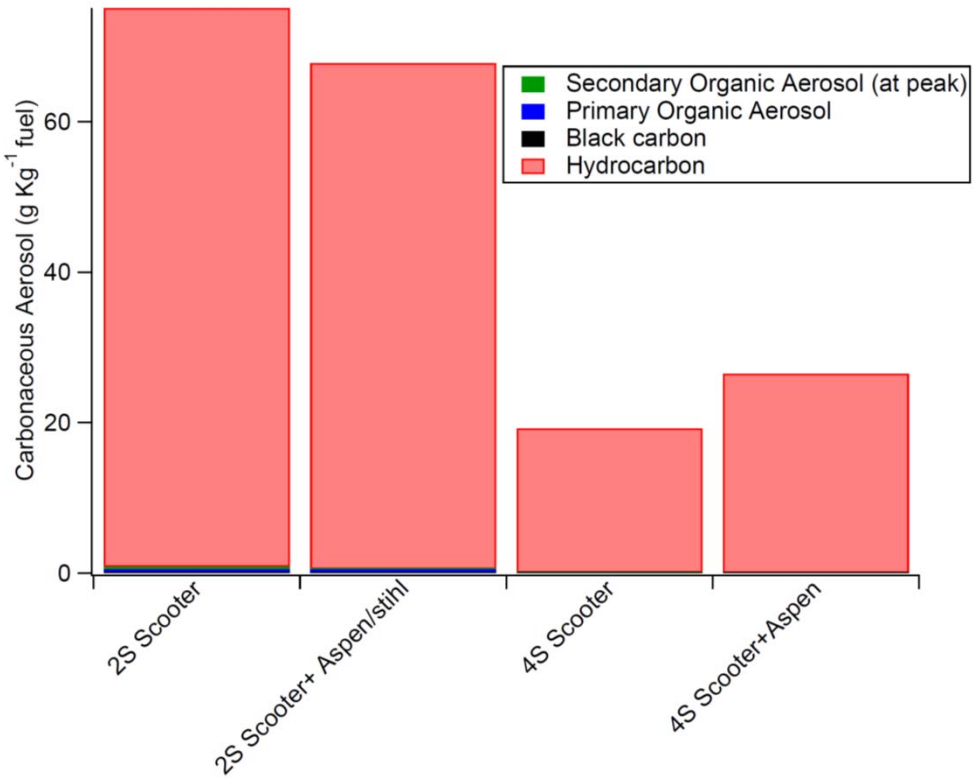
Euro 3 car shows similar profile, overall higher per kg emissions

Result of higher hydrocarbon emission

*Chirico et al., 2010, 60km/h steady state, SOA 5 hours

2- and 4-Stroke Scooters

Note scale change!



Composition (vol%)	Standard fuel	Alkylate fuel
Aromatics	29.6	0.1
Olefins	4.2	0.1
Benzene	0.2	0.01

High emissions from 2-Stroke scooters due to properties of engine cycle

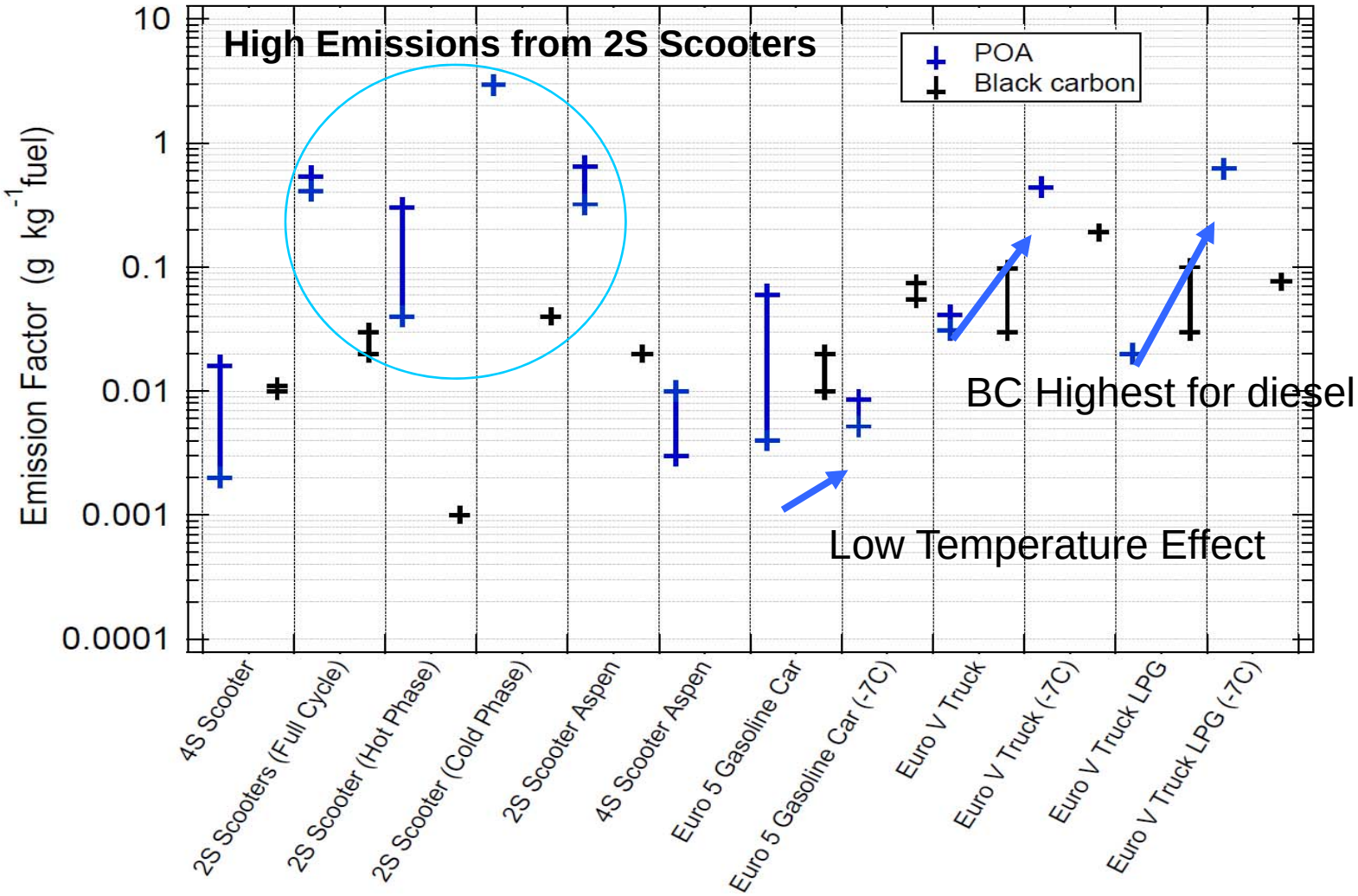
Considerable reductions from a switch to 4-Stroke engines

Considerable SOA reduction when using alkylate (e.g. Aspen) fuels

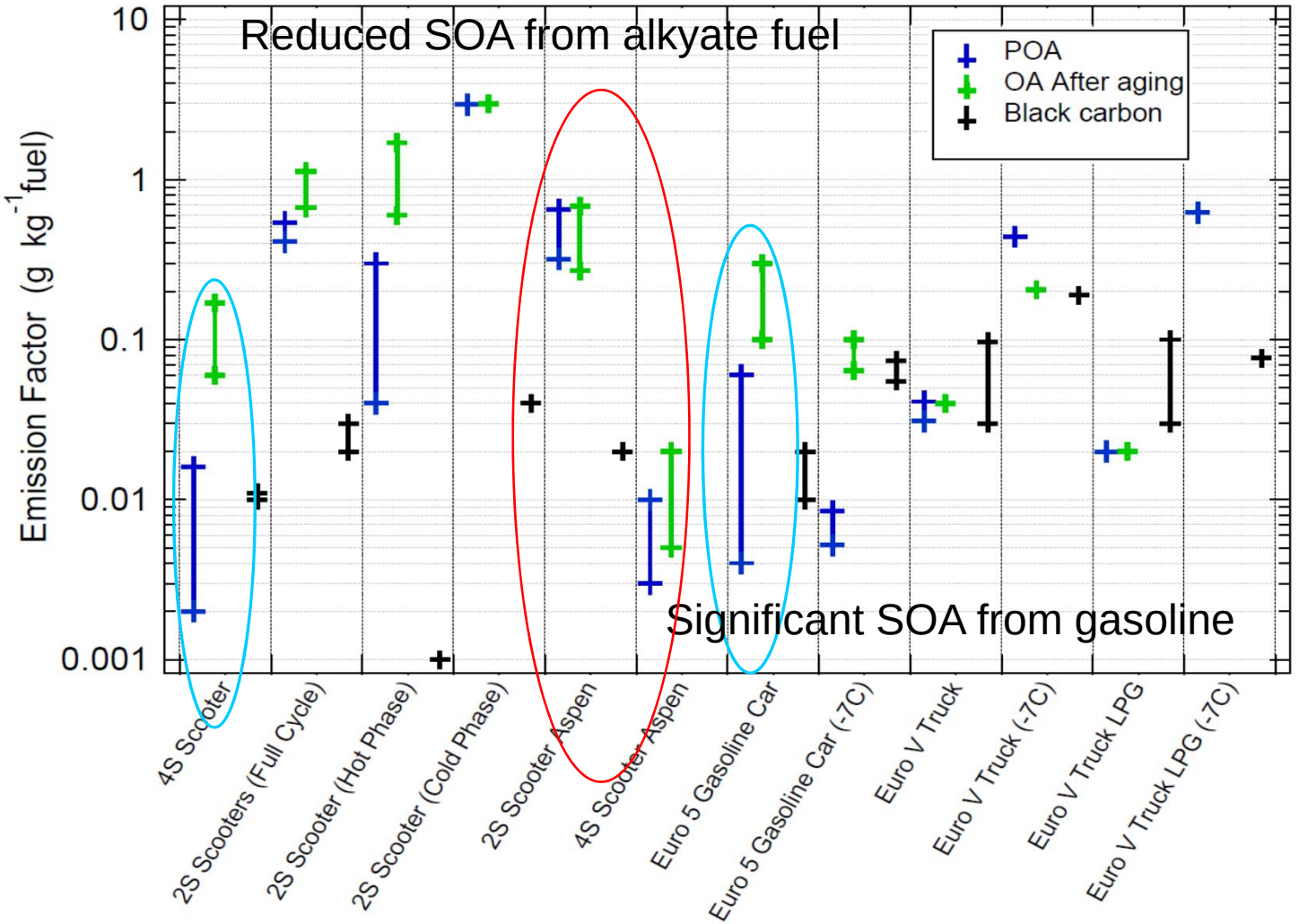
Removal of aromatic precursors: no change in HC when using alkylate

Zardini et al., Poster

Primary Emissions Summary



Secondary Formation Summary



Conclusions

High SAPP from gasoline highlights the need to consider SOA when trying to understand contribution of vehicles to ambient PM

Gasoline emissions are unique due to rapid formation of highly aged SOA: similar to the OOA observed in ambient. Data analyses in Los Angeles indicate gasoline cars to be the most important source of SOA (Bahreini et al., 2012)

High emissions from scooters show that there can be orders of magnitude between emissions from different vehicles

Vehicle type, engine technology, fuel type and temperature all affect SAPP

This study shows the utility of the mobile chamber developed at PSI

Results are based on very few vehicles. More work needs to be done. The smogchamber might be replaced by a flow tube reactor (e.g. Alejandro Keller yesterday) that needs to be evaluated by comparison with aging in smogchamber

Outlook

More vehicles and conditions 2013 campaign, possibly:

- More vehicles to get better statistics
- Flexi-fuel vehicle: gasoline/ethanol
- Diesel at high speeds,
- Heavy construction machinery
- Low temperature aging

Further analysis of chemistry of aging: what role does the composition of primary emission play in SOA formation?

Acknowledgements

Special thanks to our sponsors

- Swiss Federal Roads Office
- Bundesamt für Umwelt

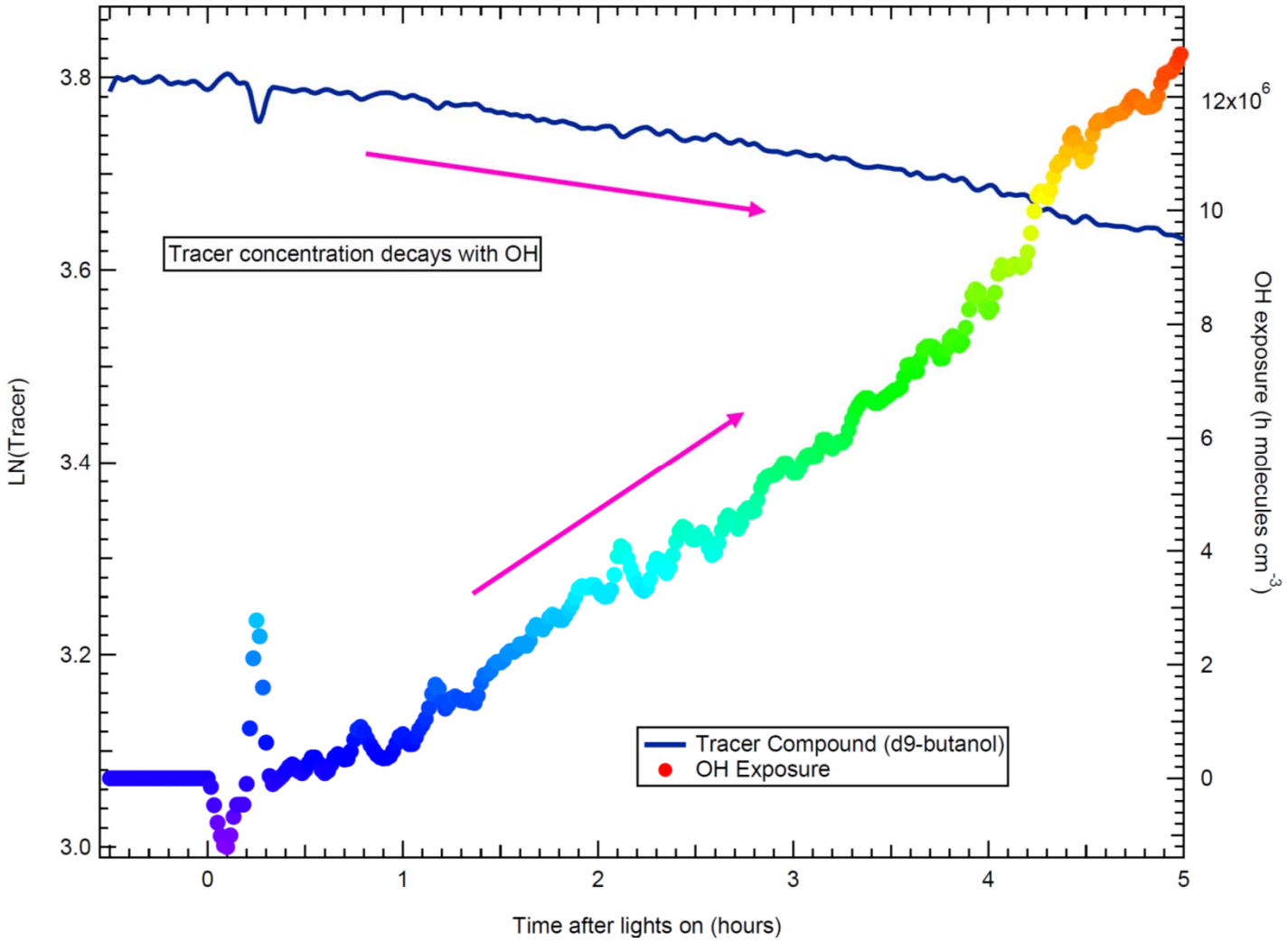
Schweizerische Eidgenossenschaft
Confédération suisse
Confederazione Svizzera
Confederaziun svizra

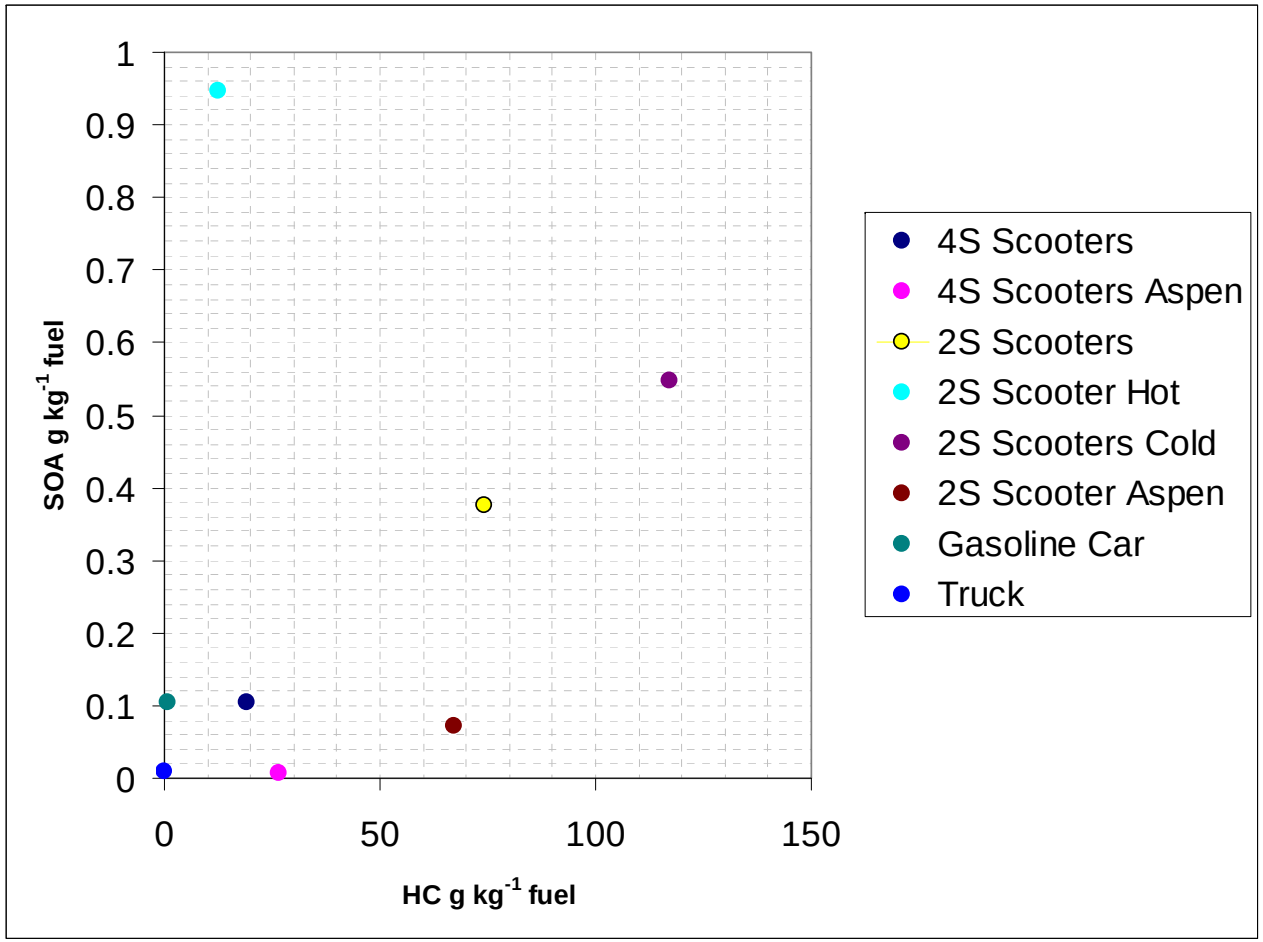
Swiss Confederation

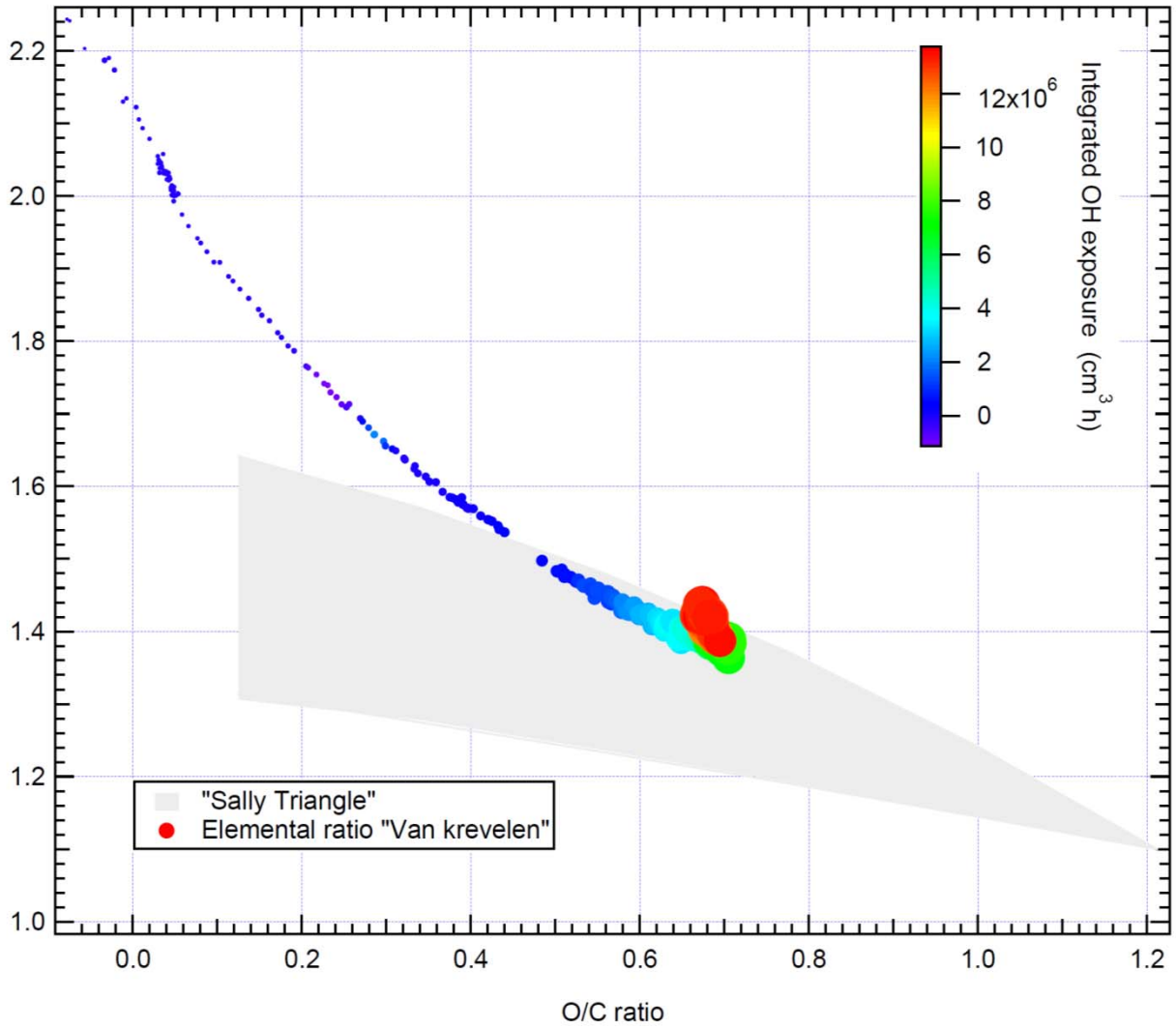
Federal Office for the Environment FOEN

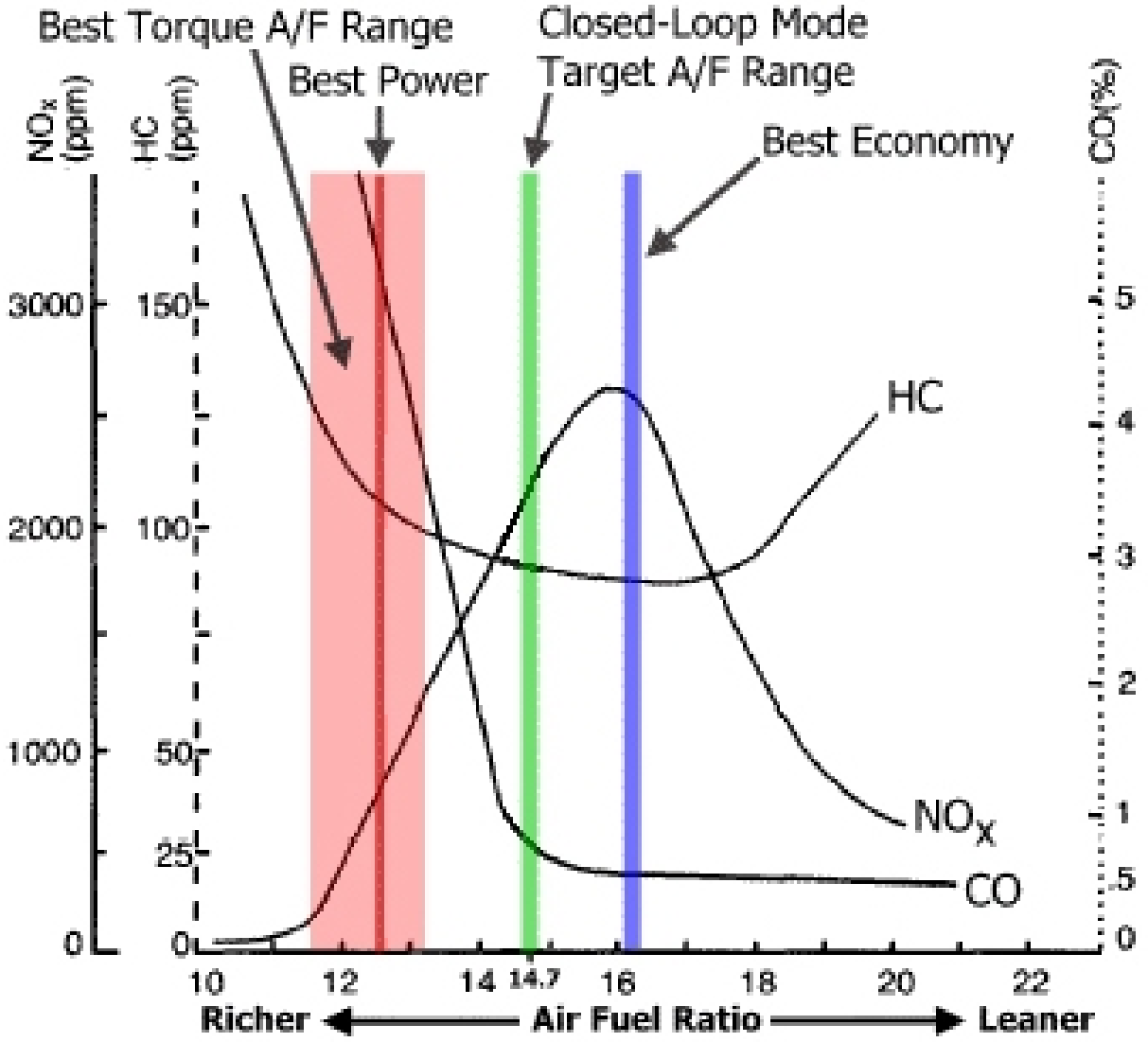
Thank you for your attention!

Back up slides


Tracer concentration decays with OH
Tracer Compound (d9-butanol)
OH Exposure
LN(Tracer)
OH exposure (h molecules cm-3)
Time after lights on (hours)
3.8
3.6
3.4
3.2
3.0
12x10^6
10
8
6
4
2
0
0
1
2
3
4
5

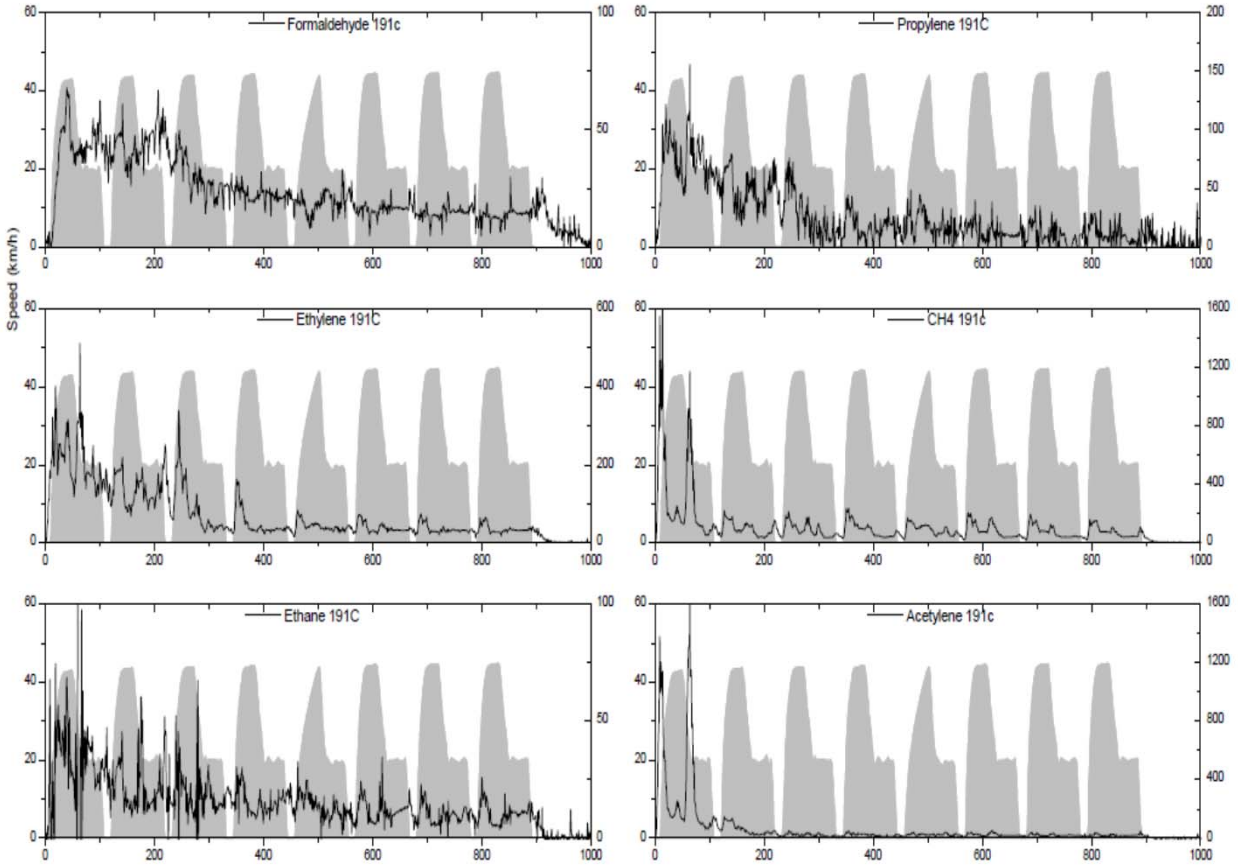

1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
SOA g kg-1 fuel
0
50
100
150
HC g kg-1 fuel
4S Scooters
4S Scooters Aspen
2S Scooters
2S Scooter Hot
2S Scooters Cold
2S Scooter Aspen
Gasoline Car
Truck


2.2
2.0
1.8
1.6
1.4
1.2
1.0
0.0
0.2
0.4
0.6
0.8
1.0
1.2
O/C ratio
12x10^6
10
8
6
4
2
0
Integrated OH exposure (cm^3 h)
"Sally Triangle"
Elemental ratio "Van krevelen"


Best Torque A/F Range
Best Power
Closed-Loop Mode
Target A/F Range
Best Economy
NOx (ppm)
HC (ppm)
CO(%)
3000
2000
1000
0
150
100
50
25
0
5
4
3
2
1
.5
0
HC
NOx
CO
10
12
14
14.7
16
18
20
22
Richer
Air Fuel Ratio
Leaner

FTIR Exhaust Measurements

4S Scooter (22.09.2011)



- ECE 47 test used for all
- Most experiments with the full cycle
- “Cold phase” not regulated
- Important feature: higher HC during cold phase
- HC increase with acceleration

- A. Zardini