

Primary and secondary aerosol from modern road vehicles

S. M. Platt¹, S. M. Pieber¹, I. El Haddad¹, A. A. Zardini², R. Suarez-Bertoa², J. [1]G. Slowik¹, R. J. Huang¹, S. Hellebust³, B. Temime-roussel³, N. Marchand³, L. Drinovec⁴, G. Mocnik⁴, S. J. Fuller⁵, M. Kalberer⁵, U. Baltensperger¹, C. Astorga² and A. S. H. Prévôt¹

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Introduction

Airborne particulate matter (PM) damages health [2, 3] and affects climate [4]. A significant fraction of the total ambient aerosol mass is secondary organic aerosol (SOA), formed via the reactions of precursors [5]. These precursors comprise gas phase volatile organic compounds (VOCs) or condensed phase matter which partitions at least to some extent to the gas phase [6, 7]. Therefore any source of VOCs or primary, directly emitted, organic aerosol (POA) may be associated with the production of SOA. In this context vehicular exhaust is an important source of primary PM and VOCs, particularly in urban areas where the health implications of pollutants are greater due to higher population density [8]. Unfortunately, little or no information on SOA formation from vehicle emissions exists in the literature. Furthermore, how SOA production varies by vehicle type (e.g. diesel or gasoline, vehicle legislative standard etc.) and thus the relative contribution of different vehicle classes to ambient PM, remains poorly constrained.

Methodology

Emissions factors (EF) (g kg^{-1} fuel), of POA and SOA were determined for several different vehicles: 2 and 4-stroke scooters, gasoline and diesel passenger cars and heavy duty diesel trucks all complying with the latest European emission standards. Emissions generated during regulatory driving cycles at the vehicle Emissions Laboratory (VELA) of the European Commission Joint Research Center (JRC), Italy, were introduced into the Paul Scherrer Institute mobile smog chamber [9], via a heated (150°C) injection system. After injection UV lighting was used to initiate

photochemistry inside the smog chamber. Nitrous acid (600 ppbv) was continually injected into the smog chamber to act as a source of OH radicals. An Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) was used to quantify organic aerosol (OA) in the smog chamber (for information concerning this instrument, see [10]), while black carbon emissions were quantified using an aethalometer (AE33). A suite of gas phase instruments was deployed to quantify carbonaceous emissions (hydrocarbons, CO and CO₂). Measurements NH₃, O₃, and NO_x concentrations were also performed. Relative humidity inside the smog chamber could be varied to represent high (90%) and low (40%) ambient relative humidity, while temperature could be varied between -7 and 22°C.

In addition to measurements during driving cycles, a series of experiments was performed on idling 2-stroke scooter emissions (Euro 1 and Euro 2 standard) using the same methodology as above, at the Paul Scherrer Institute smog chamber [11]. During the idling smog chamber experiments, online quantification of particle bound reactive oxygen species (ROS), a proxy for human health effects, was performed using an online methodology, detailed in [12].

Figure 1 shows a time series of a smog chamber experiment on idling scooter exhaust emissions. After an initial spike in OA concentration following sample injection, a time of twenty minutes was allowed for equilibration of the exhaust in the chamber and the concentration of OA at this point was regarded as the initial primary organic aerosol (POA) emission. After background measurements with the AMS to correct for concentrations of gas phase species, the lights were switched on to initiate photo-oxidation and secondary organic aerosol (SOA) formation. The first idling scooter experiment showed that SOA formation was sensitive to the presence of NO_x (as would normally be present in the ambient atmosphere) and subsequent scooter experiments were carried out with a steady injection of NO whereby NO_x was maintained at around 20 ppb. NO was not added in the case of passenger cars and trucks.

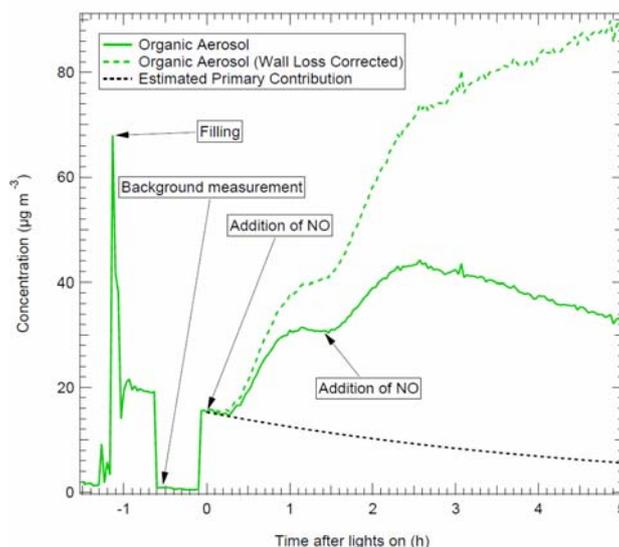


Figure 1: Time series of a typical scooter smog chamber experiment. The green line indicates the concentration of organic aerosol in the smog chamber with respect to time after lights on as measured by the aerosol mass spectrometer. The dashed green line is the wall loss corrected aerosol mass concentration based on the loss rate of the primary organic aerosol (see text), shown in black.

The data on OA concentration was corrected for wall loss in the chamber using the decay rate of the initial POA and used along with concentrations of gas phase hydrocarbons, CO, and CO₂ to calculate emission factors.

Results

2-stroke scooter EFs determined at both JRC and PSI are several orders of magnitude higher than those of other vehicle classes, including diesel and gasoline cars and heavy duty trucks. Consequently 2-stroke scooters can contribute significantly to urban PM, even at a low proportion of the total fleet. For example at 10% of a total vehicle fleet (seen in some Asian cities, such as Bangkok) 2S scooters would contribute 60-97% of roadside POA. Furthermore, idling emissions contain large quantities of volatile hydrocarbons and toxic aromatic hydrocarbons, as reflected by the fact that SOA comprised the main fraction of the OA after only a few hours. Online ROS measurements show that ROS concentrations are linked to the formation of SOA.

Gasoline vehicles generally exhibited high SOA formation, with SOA contributing the main fraction of PM mass. When SOA formation is included, overall PM pollution may be higher from gasoline cars than modern diesel cars equipped with diesel particle filters (DPF). Higher relative humidity significantly increases SOA formation, by a factor of three, when driving conditions are kept constant, suggesting that physical chemical processes occurring in the smog chamber (and thus the atmosphere) are affected.

Conclusions

We conclude that emission of OA by mopeds may be a major public health consideration in many urban areas and may represent a large contribution to the burden of organic aerosol globally. Gasoline vehicles may produce more PM than diesel vehicles when SOA formation is accounted for.

Acknowledgements

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References

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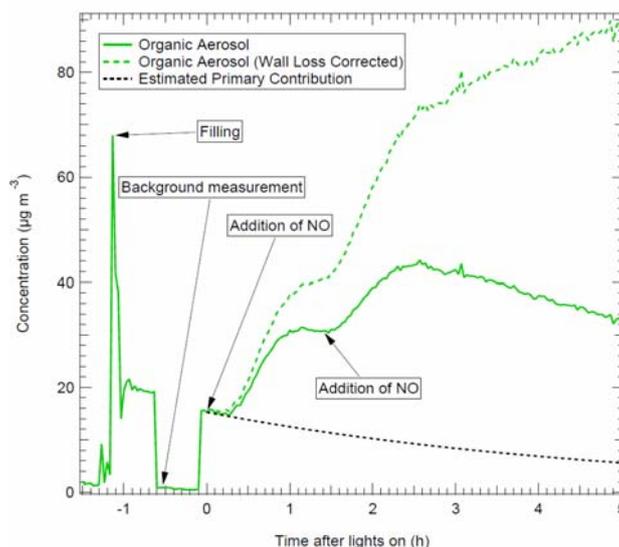


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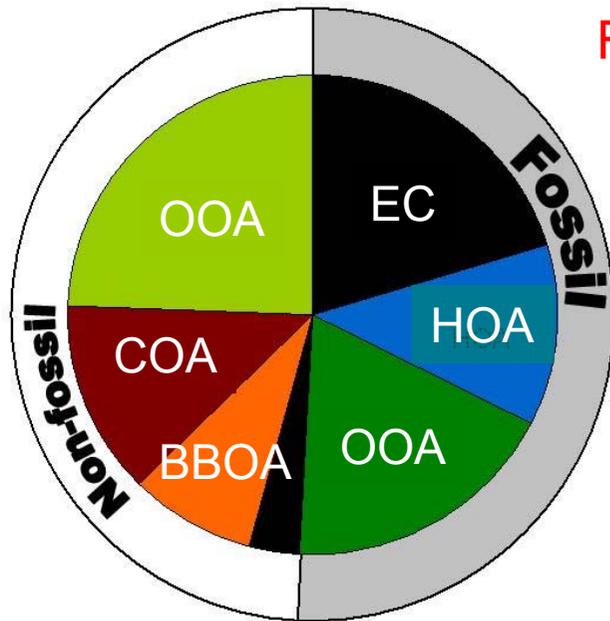
Aging of emissions from road vehicles

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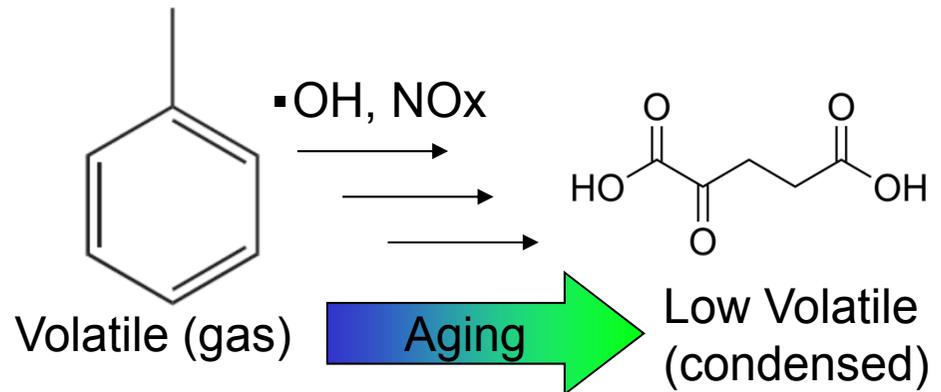


Aerosol affects climate, damages health, and reduces visibility

A major source in urban areas is road vehicles:



Fresh: primary PM (HOA+EC), VOCs



Aged: primary PM (HOA+EC), secondary PM (OOA), oxidised VOC

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

-Barcelona, March, Adapted from Minguillón et al., 2011

Overview

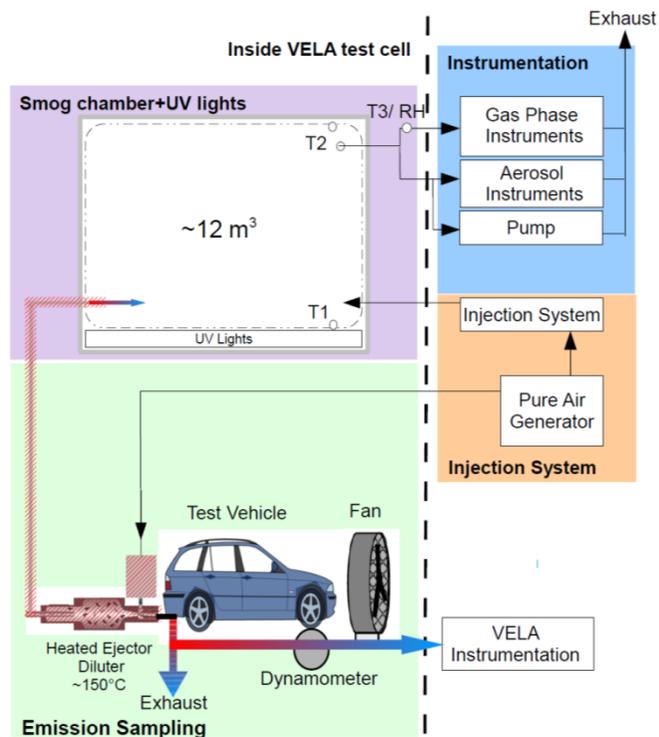
Campaign	Vehicle	Standard /Technology	Driving mode				Ambient condition				Alternative Fuel
			Idle	Full Cycle	Hot	Cold	22C	-7C	30-50%	80-90%	
Mopeds 2010	2S Scooter A	Euro 1/ OC	3				3		3		
	2S Scooter B	Euro 2/ OC	3				3		3		
	4S Scooter A	Euro 2/ OC	2				2		2		
	Total		8								
Ispra 2011	2S Scooter C	Euro 2/ OC		5	3	1	9		9		6 (Aspen)
	4S Scooter B	Euro 2/ OC		2	2		4		4		2 (Aspen)
	Gas. car A	Euro 5/ TWC, MPI		4			2	2	4		
	Truck A	Euro V/ SCR		6			4	2	6		3 (LPG)
	Total		23								
Ispra 2013	Gas. car B	Euro 5/ TWC, DI		9			6	3	7	2	
	Gas. car C	Euro 5/ TWC, DI		4			2	2			
	Diesel car A	Euro 5/ DPF, SCR		6			4	2	6		
	Diesel car B	Euro 5/ DPF, SCR		6			5	1	4	2	
	Truck B	Euro V/ DPF, SCR		6	2		4	2	6		
	Flexi car E85	Euro 5/ TWC(?)		6			4	2	4	2	6 (EtOH)
Total		39									
VACES 2012	Gas.car D	Euro 5/ TWC, DI	9				4		4		
Total			9								

OC=Oxidation catalyst, TWC=Three-way catalyst, MPI=Multi-point injection, DI=Direct injection, DPF=Diesel particle filter
SCR=Selective catalytic reduction, LPG=Liquid petroleum gas

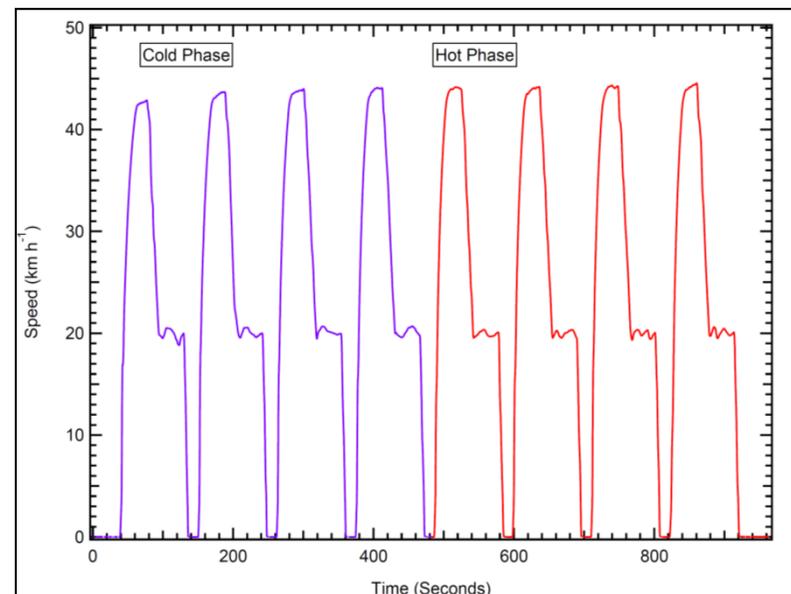
Different vehicles: scooters, cars (diesel and gasoline), and trucks

Different driving and experimental conditions

Experimental set up

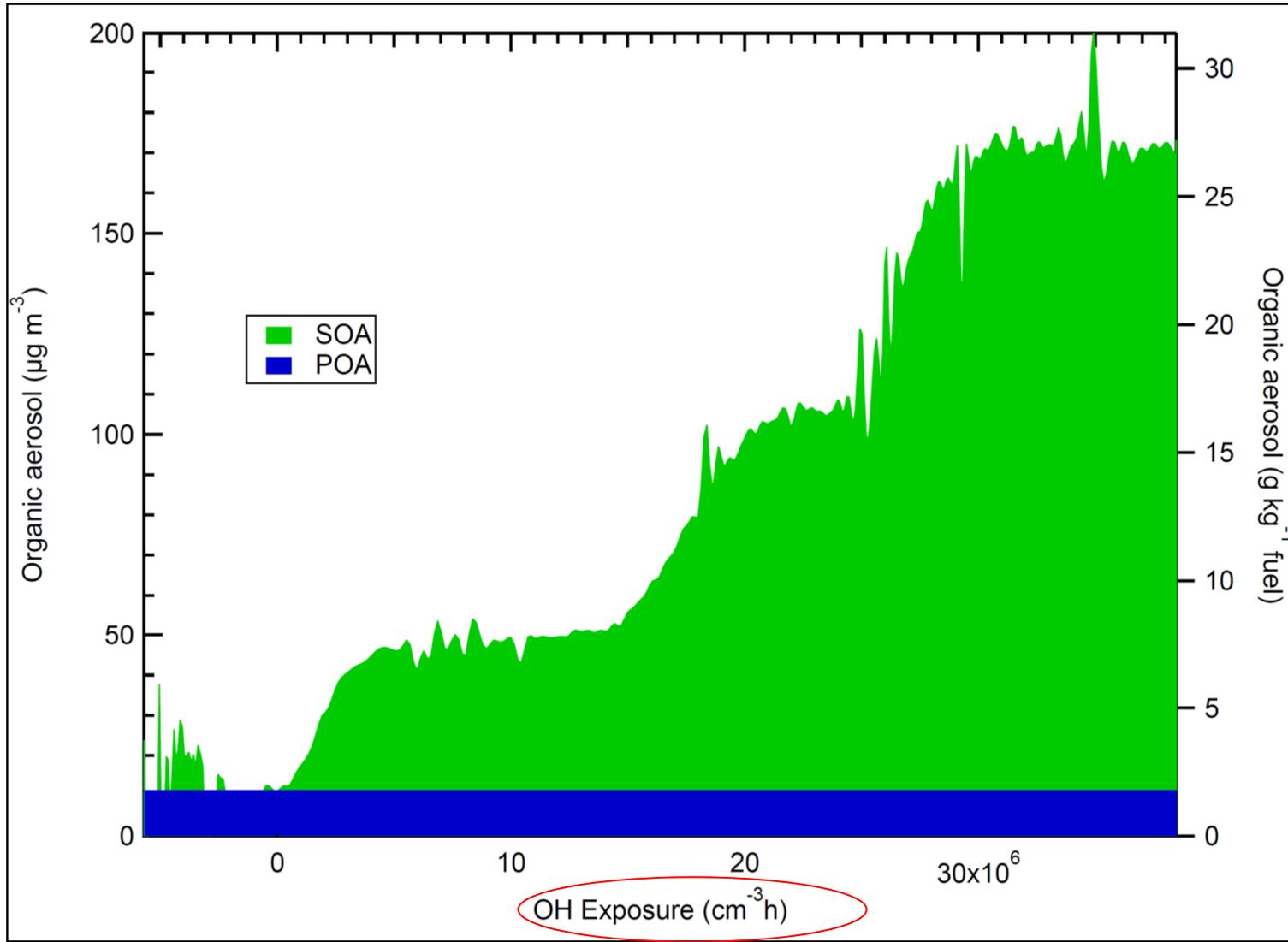


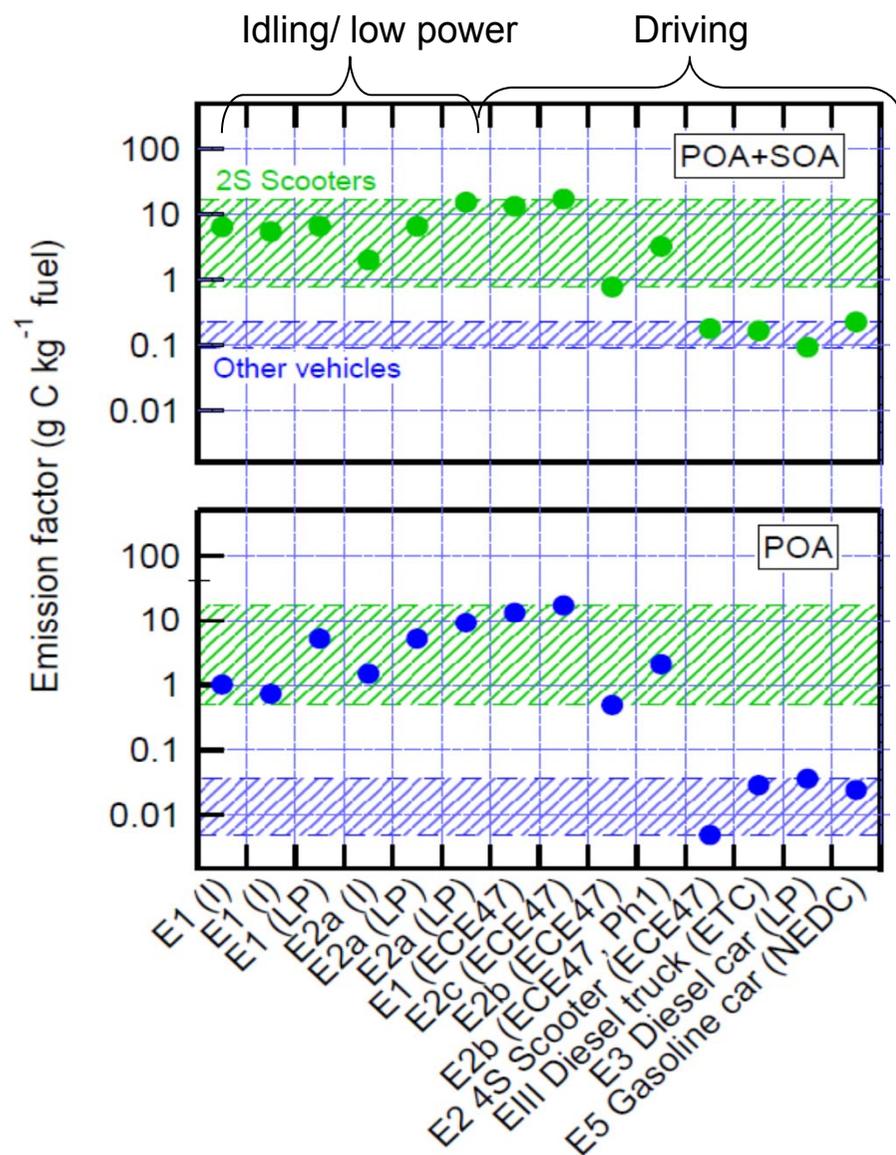
ECE 47 (Scooters):



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 ₂ O CO, CO ₂)			
	PTR-ToF-MS			
	T, RH Sensors			

AMS organic aerosol (Euro 2 idling scooter emissions)



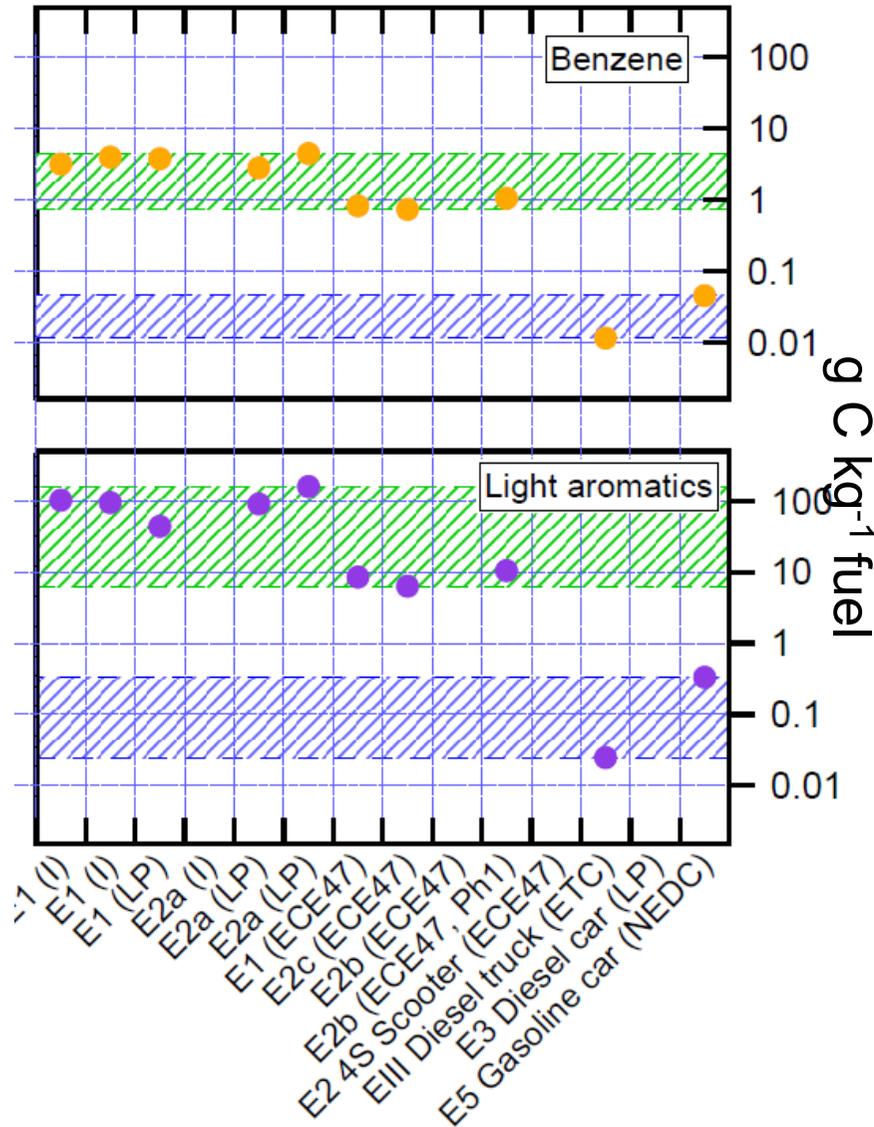


Generally higher for 2S scooters at least 10 up to 1000s (15 hours in atmosphere and OA around 100 $\mu\text{g m}^{-3}$)

Small number needed to have large influence

Consequence of engine: 2S is 'leaky' and needs lube oil, and scooters run on rich combustion

E1=Euro 1, E2=Euro 2..etc, I=Idle, LP=simulated low power, ECE47/NEDC/ETC= driving cycle Ph1=phase 1 only

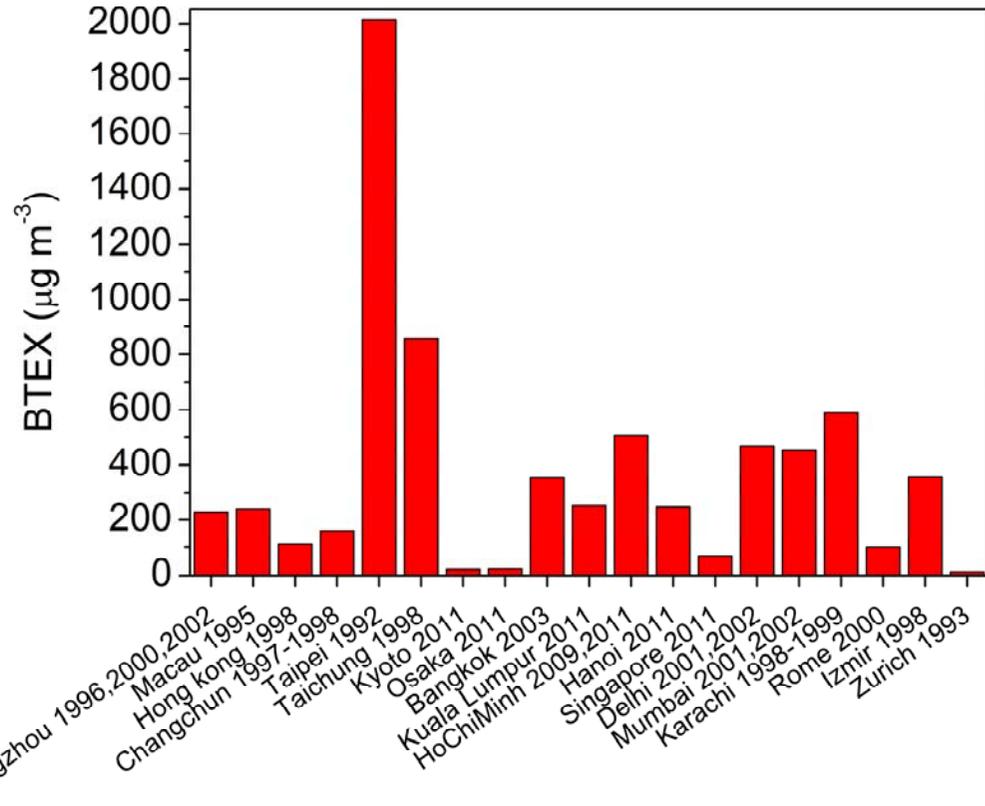
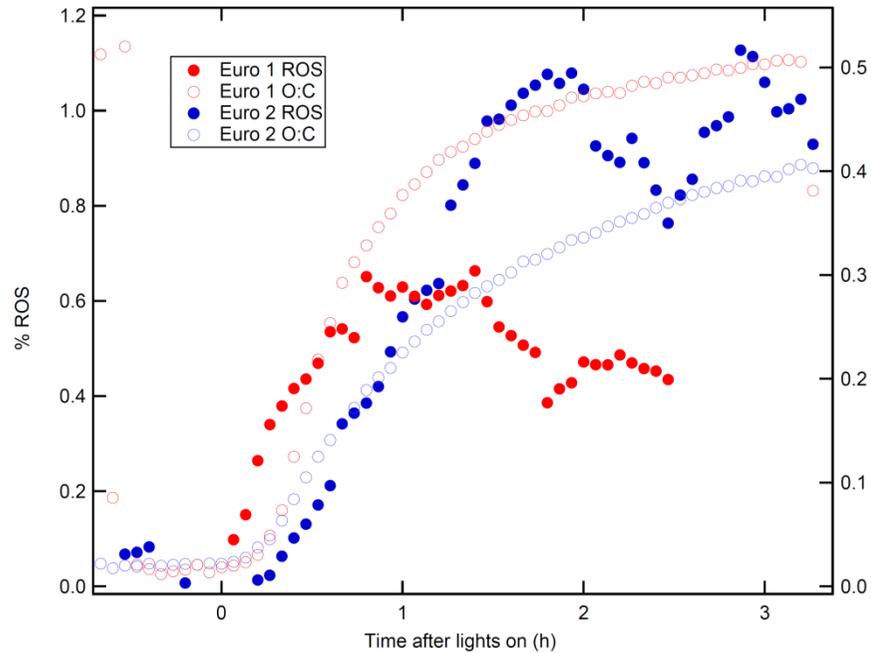


PTR-MS
measurements:
300000 $\mu\text{g m}^{-3}$ in
raw idling scooter
exhaust, EU limit
for safety is 5 $\mu\text{g m}^{-3}$
(annual
mean)



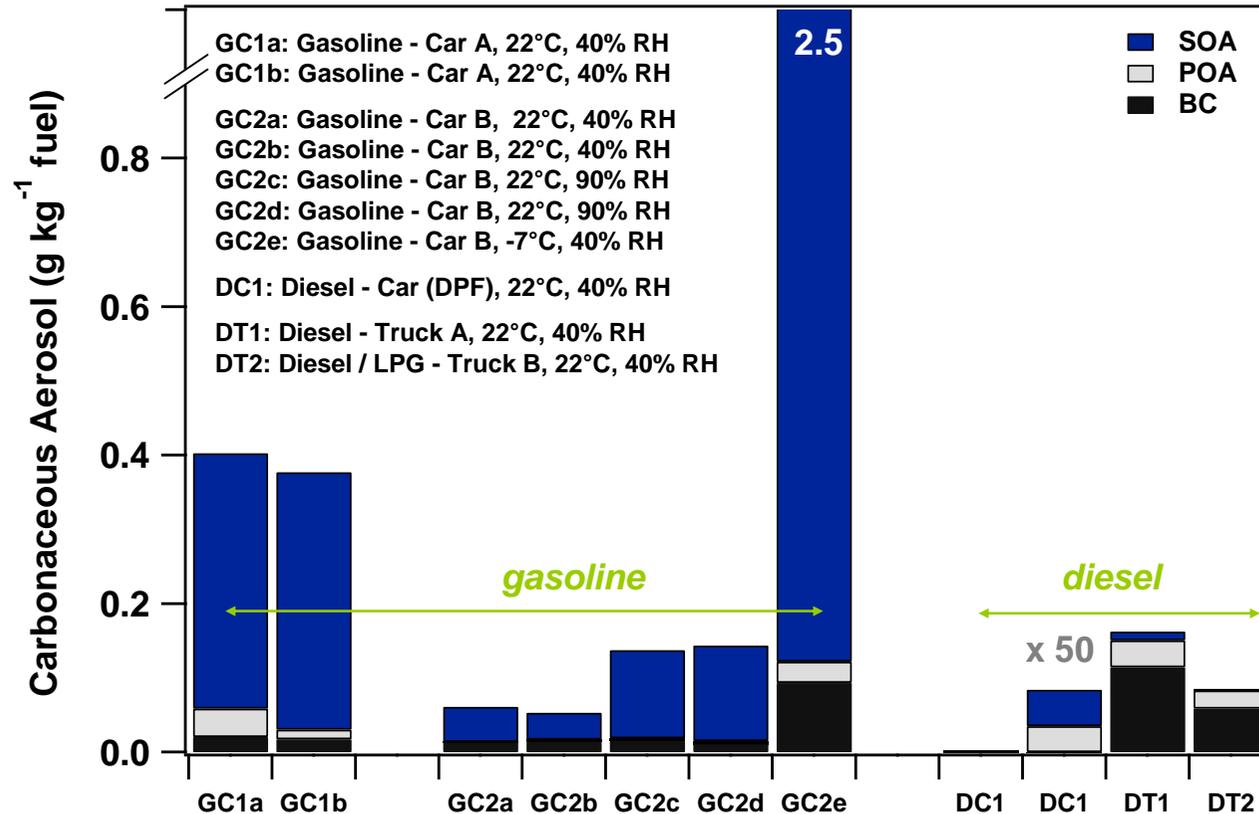
Again generally much higher as
a result of engine type

Aromatics are known SOA
precursors



Scooter SOA increases in toxicity with age and O:C

Taiwan: 7,000,000 2S scooters in Taipei in 1997, and incredibly high aromatic (roadside measured), such correlations generally strong

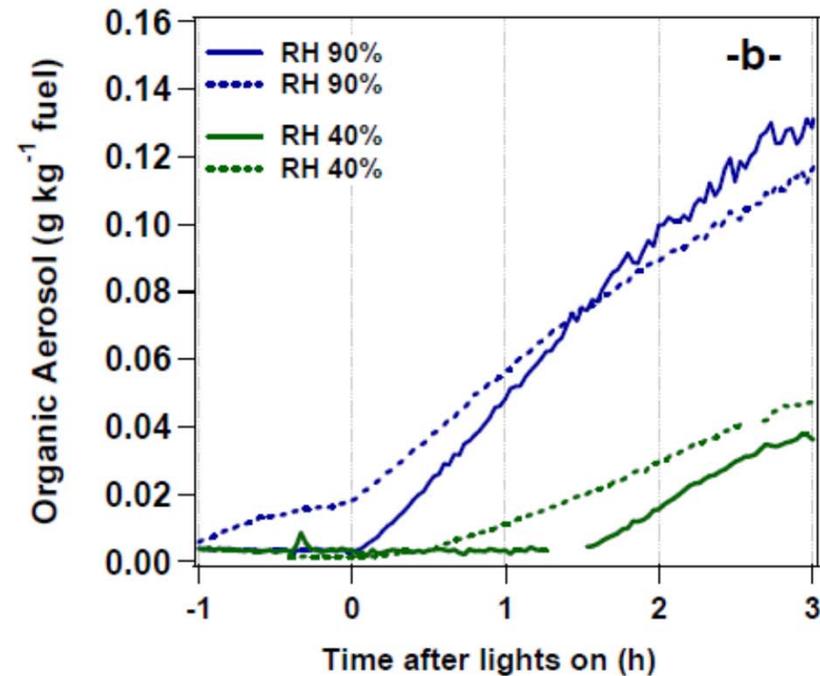
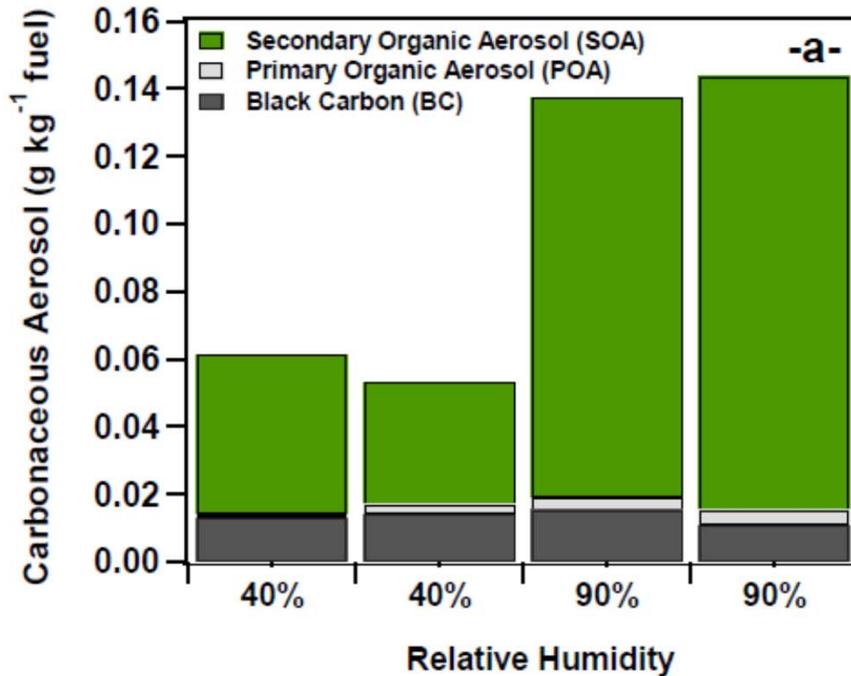


Older diesel or diesel without DPF has high primary (BC+POA)

Gasoline cars high SOA, diesel low SOA

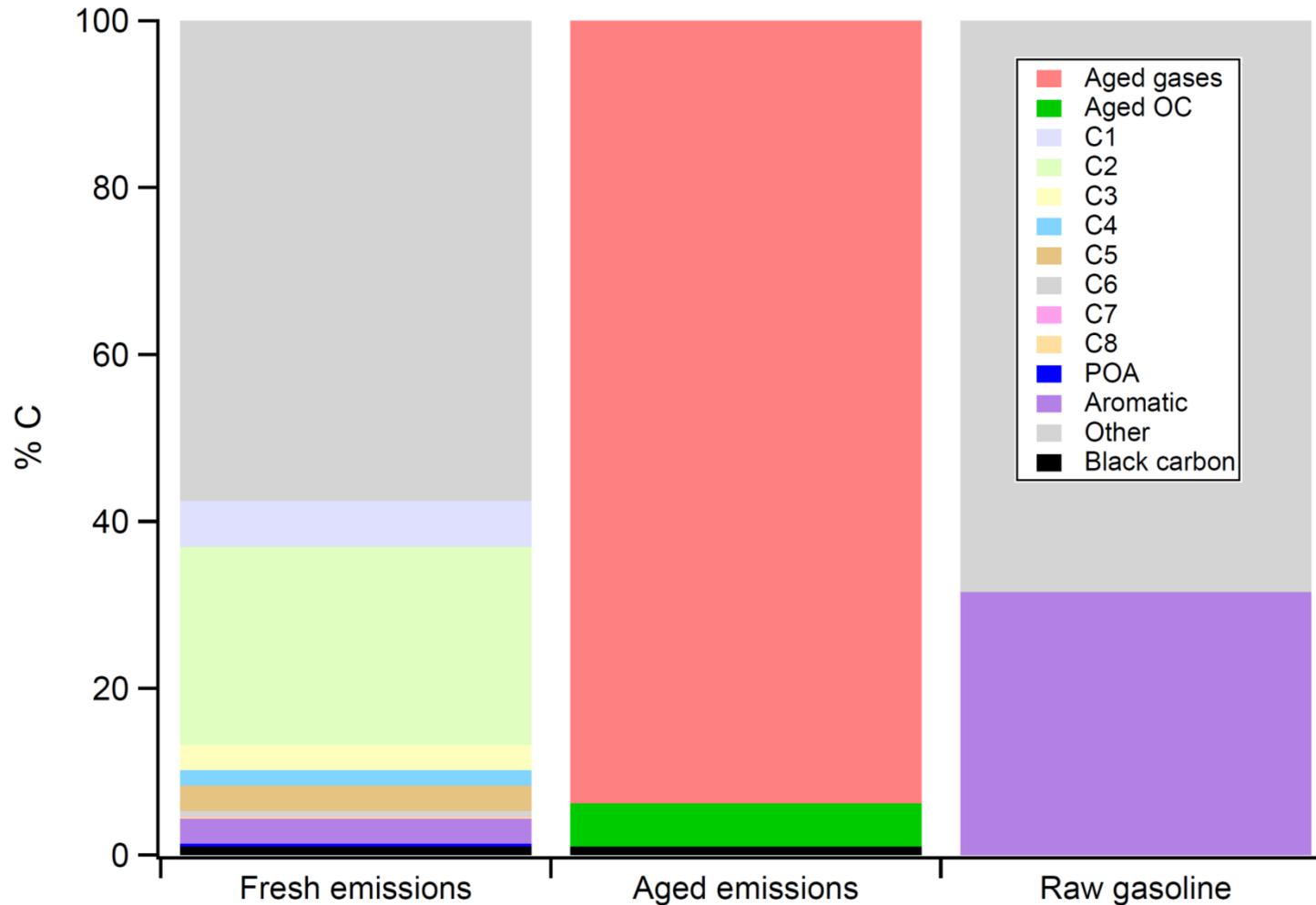
DPF incredibly effective (as expected), but filters expected to be ineffective for SOA

Gasoline car (DI):



Considerable increase in SOA formation (hydrocarbon is constant)

SOA 'formation factors' will always depend on conditions and be highly variable dependant on VOC/ NOX, humidity, light, temperature, acidity, NH₃



Emitted aromatic carbon is less than total, SOA: must be non traditional

Raw gasoline is not a good proxy for real exhaust, leads to false conclusions

Conclusions

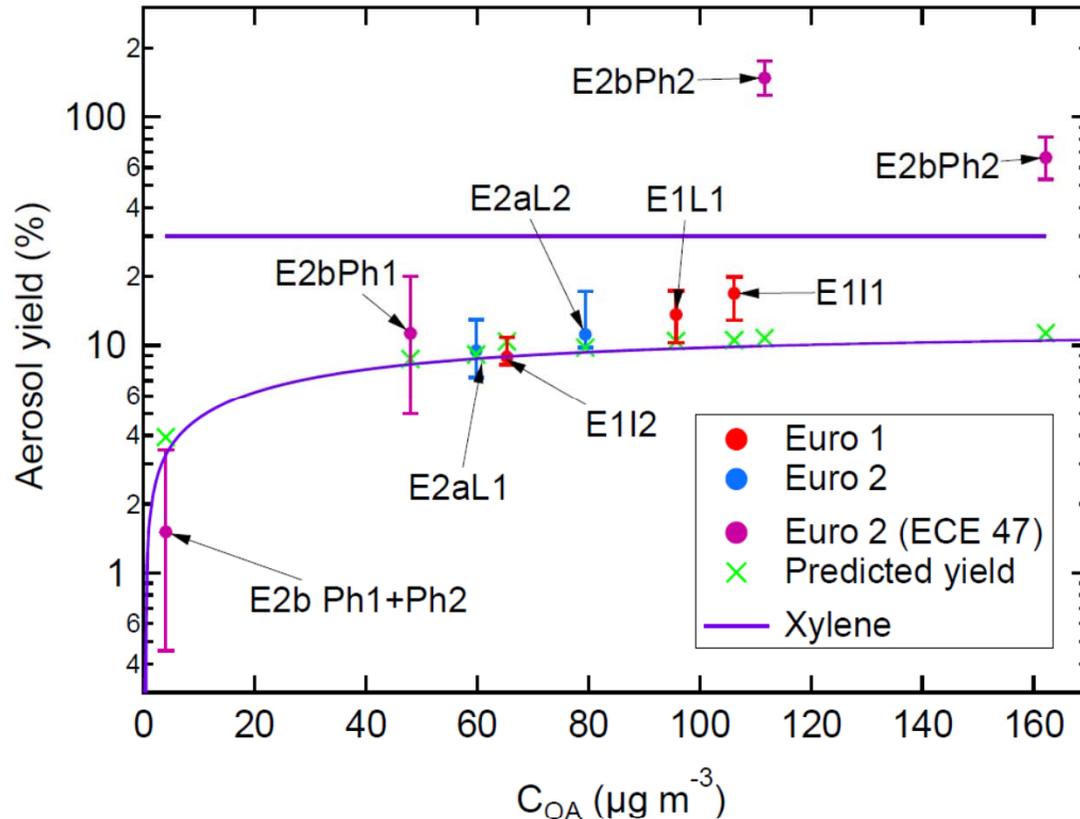
2S Scooters can dominate urban pollution aerosol and aromatic VOC even at a low proportion of the total fleet

No one size fits all SOA formation, but diesel generally produces less SOA. Modern diesel produces less PM pollution when SOA is accounted for

Demonstrable effect of relative humidity on SOA formation (increase 'yield' under same conditions)

Since SOA can be the largest part of vehicular PM it should be considered when thinking about how to reduce or regulate ambient nanoparticles

Back up slides



$$Y = \Delta M_o \left[\frac{\alpha_1 K_{om,1}}{1 + K_{om,1} M_o} + \frac{\alpha_2 K_{om,2}}{1 + K_{om,2} M_o} \right]$$

C_o =organic mass, α are fitting parameters,
 K_{OM} = partitioning coefficient (Ng et al., 2007)

Apparent yield: assume all SOA is from reacted aromatics
($Y = M_{SOA} / \Delta Aromatics$)

Theoretical, predicted yields: xylene low
 $NO_x = 30\%$ (upper limit), high NO_x (more realistic)

So far, so traditional (except for the phase 2 only)

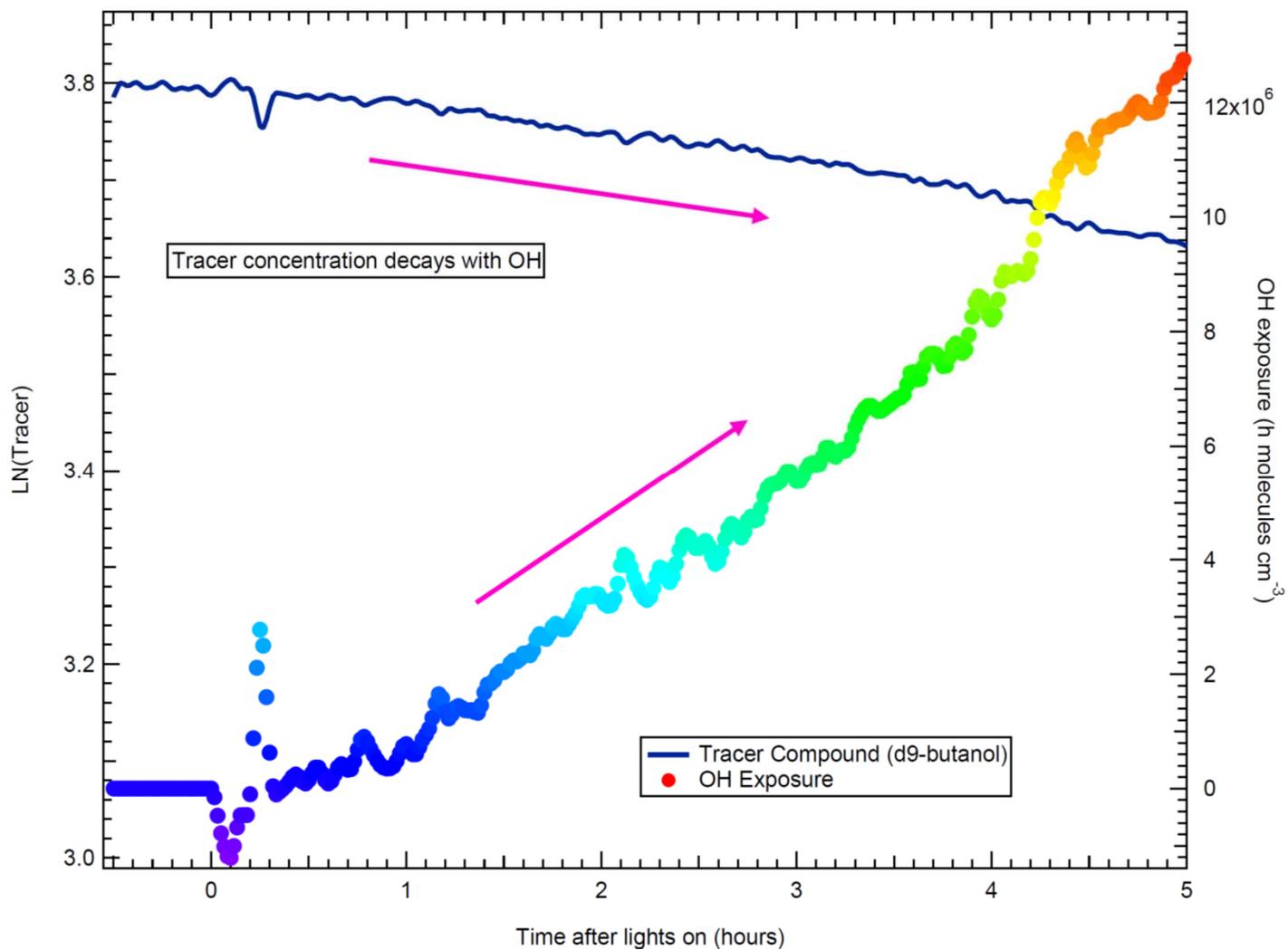
Cars

Tier	Date	<u>CO</u>	<u>THC</u>	<u>NMHC</u>	<u>NO_x</u>	<u>HC+NO_x</u>	<u>PM</u>	<u>P^{***}</u>
Diesel								
Euro 1†	July 1992	2.72 (3.16)	-	-	-	0.97 (1.13)	0.14 (0.18)	-
Euro 2	January 1996	1.0	-	-	-	0.7	0.08	-
Euro 3	January 2000	0.64	-	-	0.50	0.56	0.05	-
Euro 4	January 2005	0.50	-	-	0.25	0.30	0.025	-
Euro 5	September 2009	0.50	-	-	0.180	0.230	0.005	-
Euro 6 (future)	September 2014	0.50	-	-	0.080	0.170	0.005	-
Petrol (Gasoline)								
Euro 1†	July 1992	2.72 (3.16)	-	-	-	0.97 (1.13)	-	-
Euro 2	January 1996	2.2	-	-	-	0.5	-	-
Euro 3	January 2000	2.3	0.20	-	0.15	-	-	-
Euro 4	January 2005	1.0	0.10	-	0.08	-	-	-
Euro 5	September 2009	1.0	0.10	0.068	0.060	-	0.005**	-
Euro 6 (future)	September 2014	1.0	0.10	0.068	0.060	-	0.005**	-
<p>* Before Euro 5, passenger vehicles > 2500 kg were type approved as light commercial vehicles N₁-I</p> <p>** Applies only to vehicles with direct injection engines</p> <p>*** A number standard is to be defined as soon as possible and at the latest upon entry into force of Euro 6</p> <p>† Values in brackets are conformity of production (COP) limits</p>								

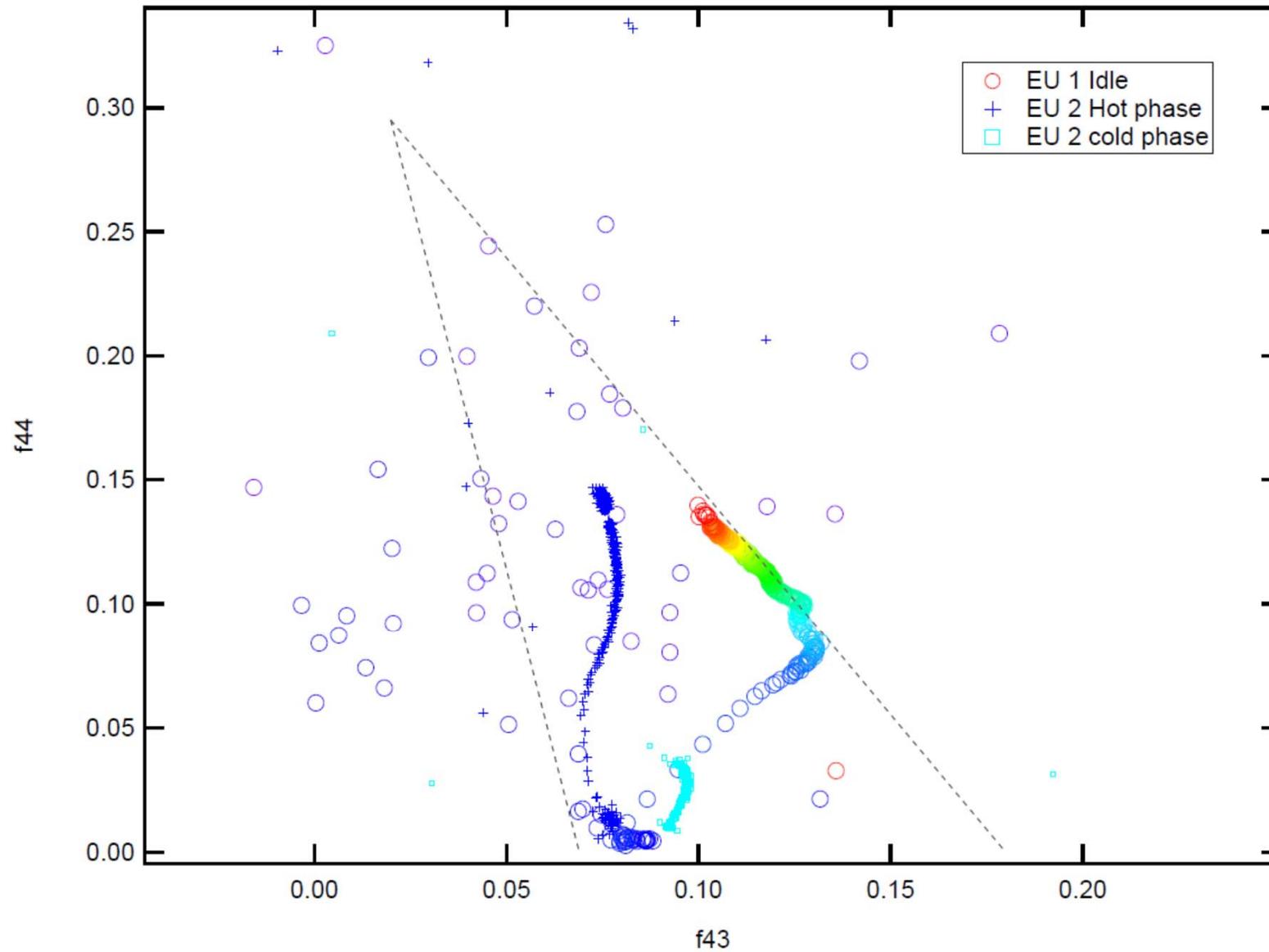
Euro Limits

	CO	HCNOx
E1	6000	3000
E2	1000	1200

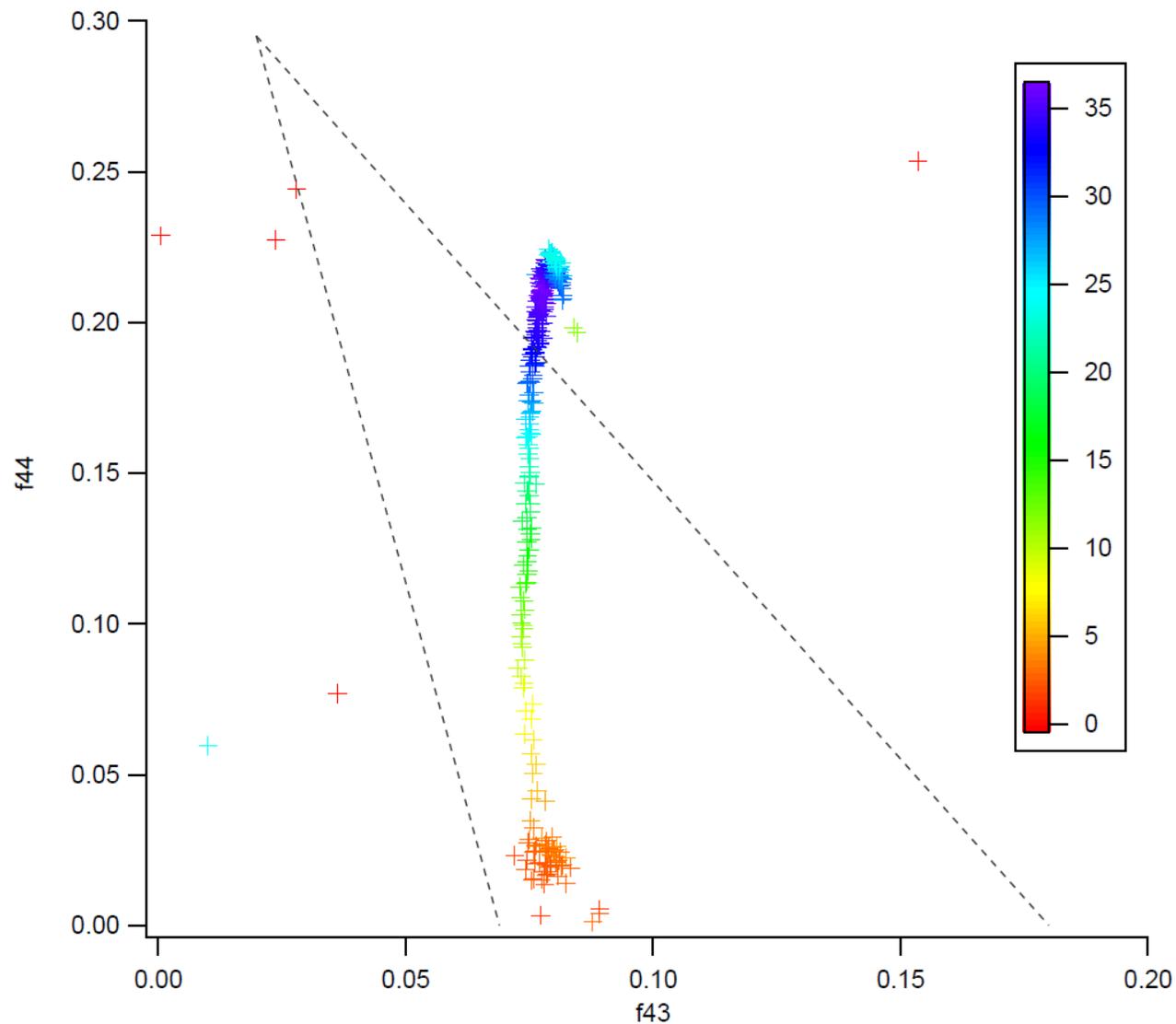
OH Clock



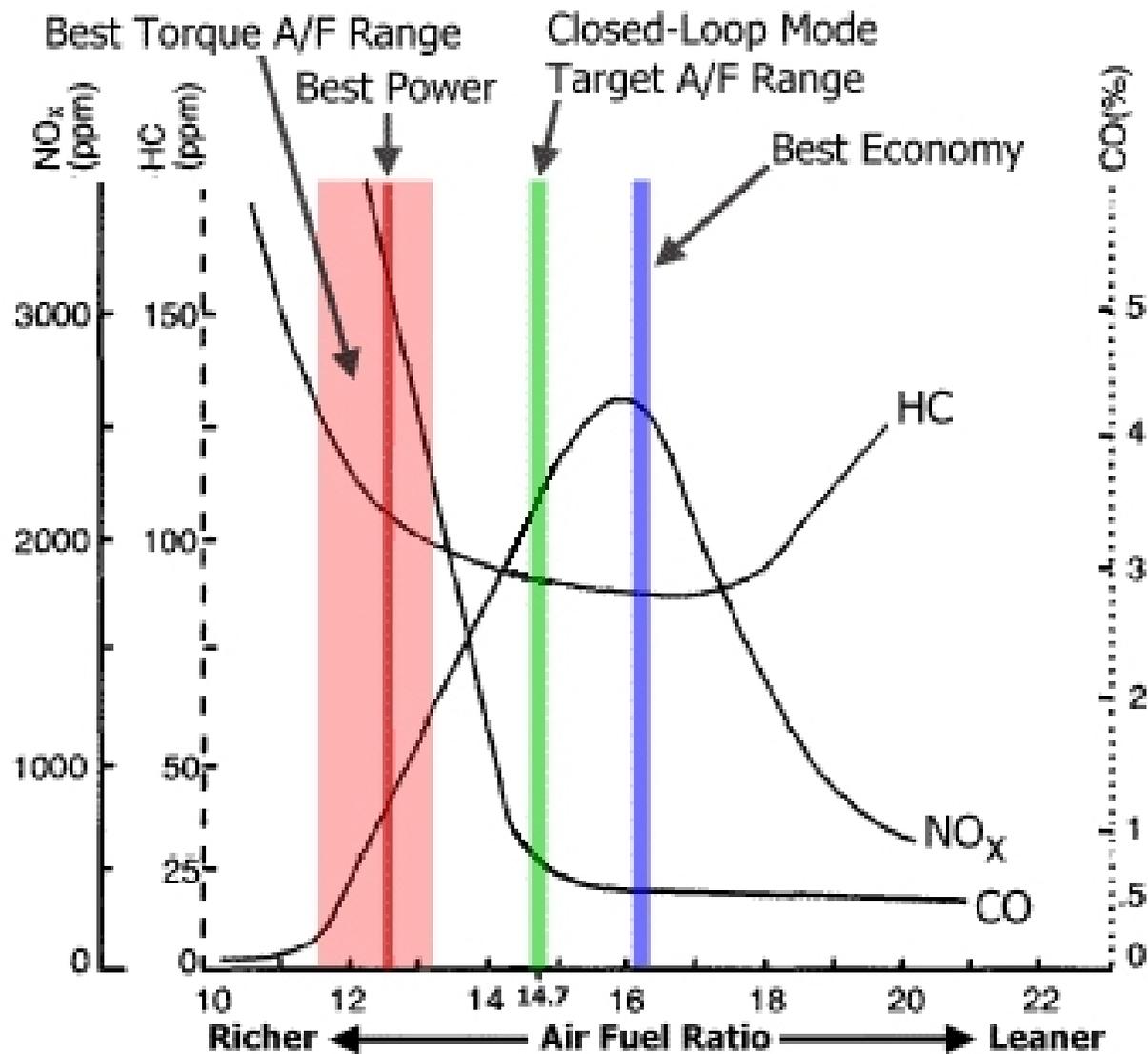
Scooters in Sally triangle



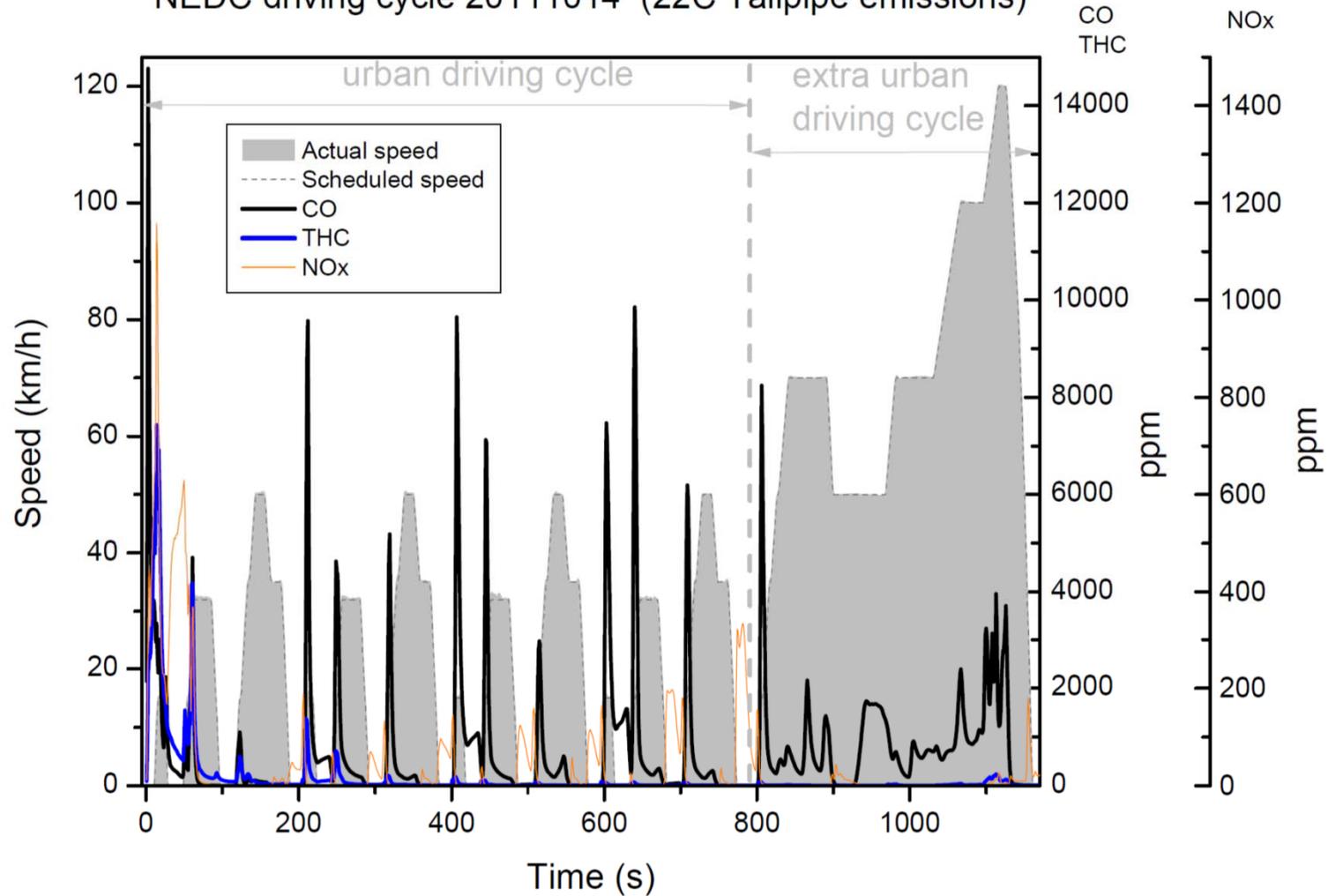
Car in Sally triangle



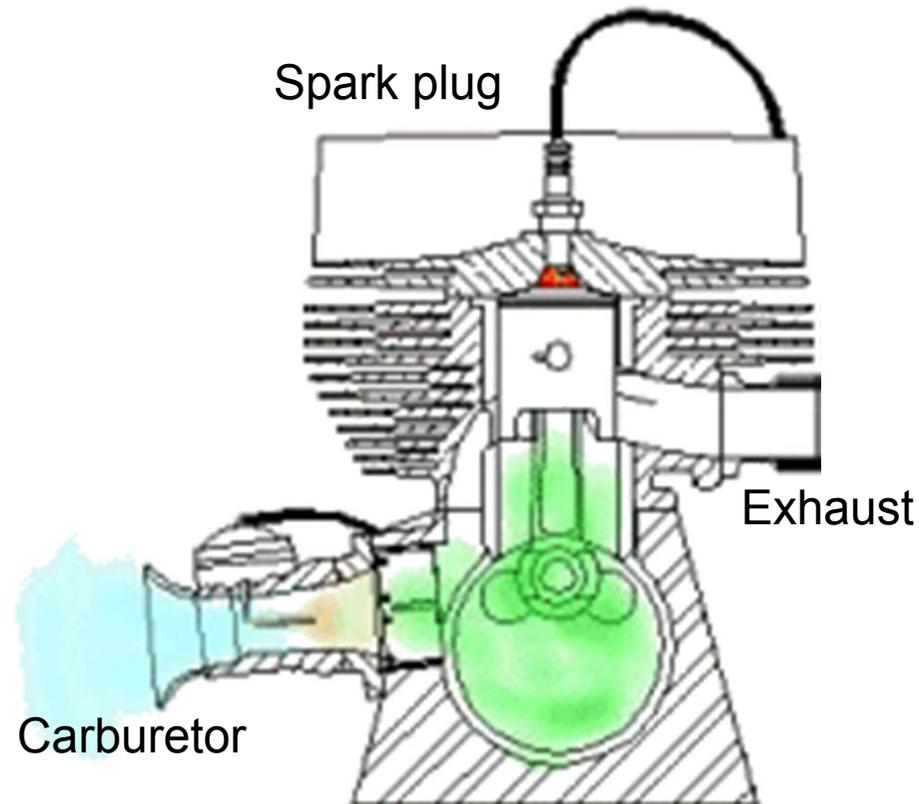
Combustion stoichiometry



NEDC driving cycle 20111014 (22C Tailpipe emissions)



Why 2-strokes suck



(Goes much faster than this...)

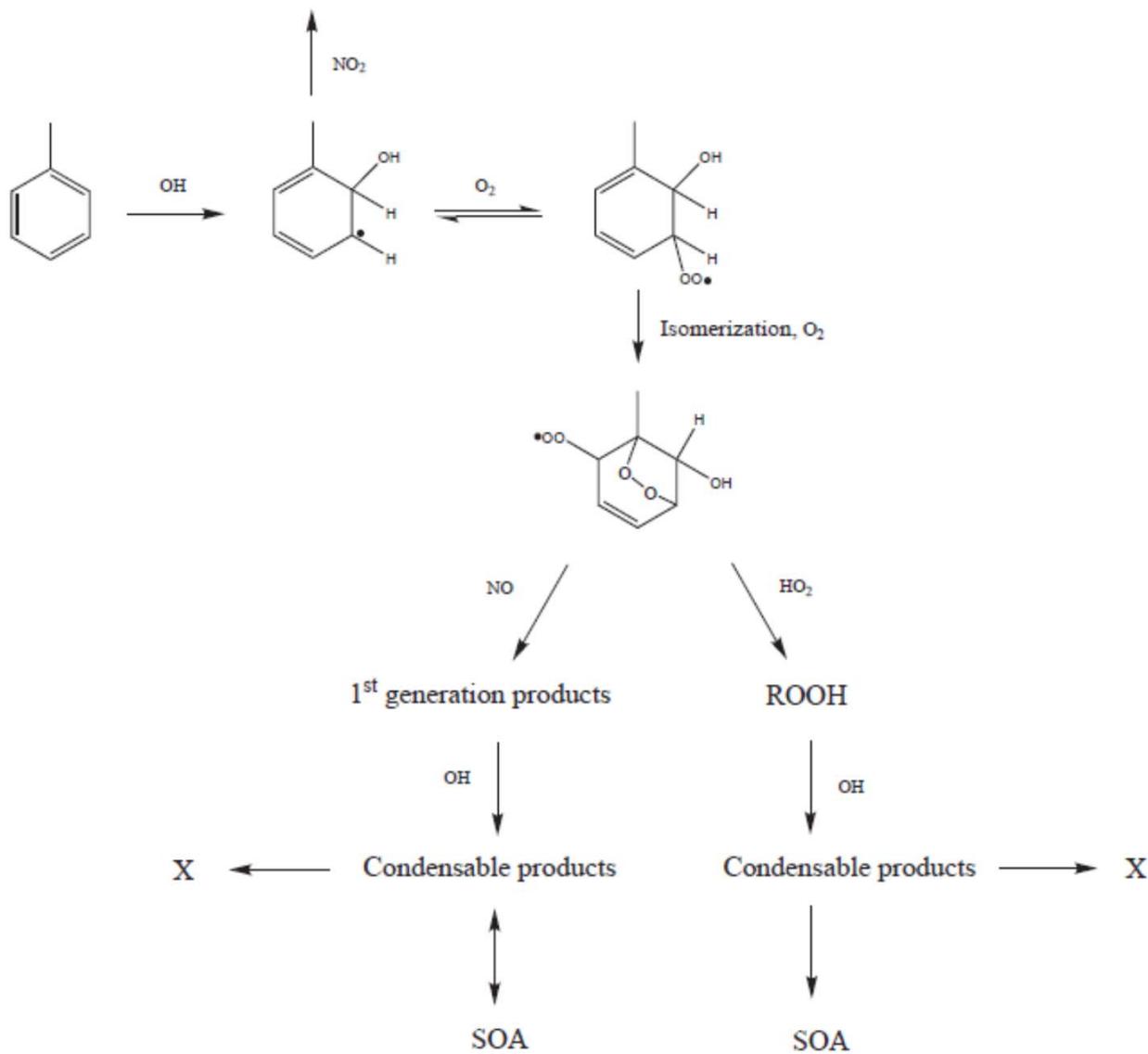
Lubricant oil

High hydrocarbon when open
to atmosphere

High primary aerosol emission

Secondary aerosol from
hydrocarbons?

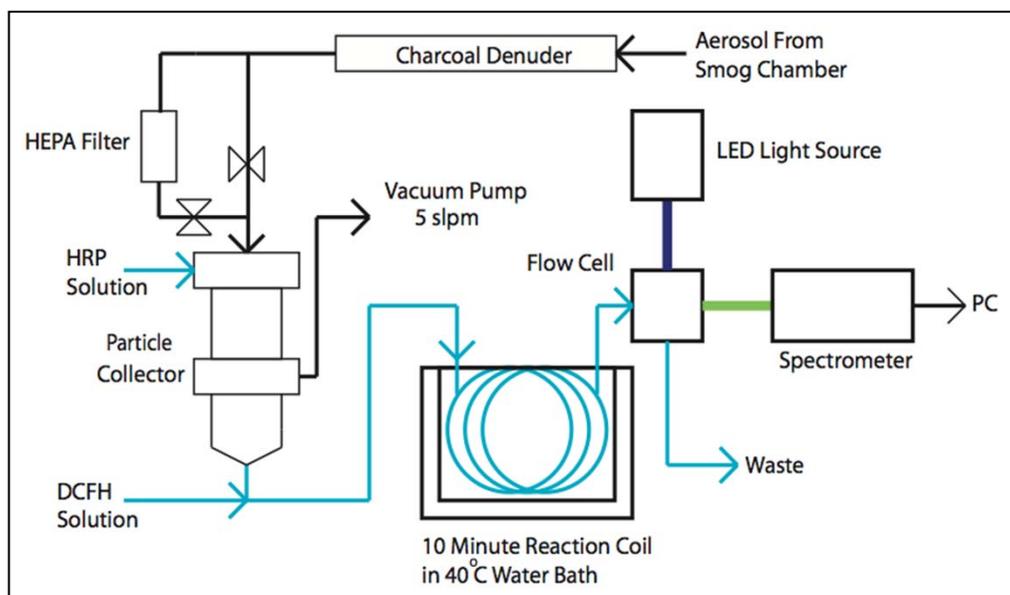
2 product toluene



Indicative of health effects: high particle-bound ROS, more cell damage from oxidative stress

Reaction in solution: Horseradish peroxidase (HRP) with ROS, HRP also with fluorescent dye dichlorofluorescein (DCFH), more ROS leads to higher fluorescence

Fluorescence calibrated to hydrogen peroxide



-Platt et al., in prep.

