

Measurement of secondary organic aerosol emissions from a gasoline engine and small-scale wood combustion with a new photochemical flow tube reactor

O. Sippula¹, M. Ihalainen¹, P. Tiitta¹, P. Yli-Pirilä^{1,2}, M. Kortelainen¹, M. Sklorz³, B. Stengel⁴, L. Müller³, S. Stanglmaier³, V. Kohlmeier³, G. Jakobi³, A. Hartikainen¹, J. Orasche³, H. Lamberg¹, J. Tissari¹, I. Methuen¹, H. Suhonen¹, A. Leskinen², T. Streibel³, H. Harndorf⁴, R. Zimmermann³, J. Jokiniemi¹

¹Department of Environmental Sciences, Univ. of Eastern Finland, P.O. Box 1627, FI-70211 Kuopio, Finland

²Department of Applied Physics, Univ. of Eastern Finland, Kuopio, Finland

³Joint Mass Spectrometry Centre, Cooperation Group Comprehensive Molecular Analytics, University of Rostock, Institut für Chemie, Rostock / Helmholtz Zentrum München, München Germany.

⁴Chair of Piston Machines and Internal Combustion Engines, University of Rostock, Rostock, Germany

Background

Secondary organic aerosol (SOA) is an important emission component for many combustion processes. These emissions are currently neglected in emission legislation and their emission factors as well as environmental and health effects are still poorly known. In this work combustion emissions were studied with the new photochemical emission aging reactor (PEAR), which is based on the PAM-tube concept (Kang et al., 2007)

Objectives

- Evaluation of the novel flow reactor setup for determining SOA emissions from combustion processes
- Quantify SOA-emission factors for a gasoline engine and log wood combustion
- Investigate the effects of aging on physico-chemical properties of emissions

Flow reactor setup

- Computer-controlled sampling & dilution system based on porous tube – ejector dilution system (Fig. 1)
- Stainless steel tube (Ø 34 cm) with four 254 nm UV lamps (Fig. 1)
- Flow rate 50-200 lpm
→ 30-180 s residence time
- O₃ and H₂O supply into the reactor to generate OH radicals
- Design aided with 3D CFD simulations (Ansys 15.0 Fluent) and trace gas experiments (Fig. 2)
- The reactor inlet consists of flow diffuser, which is designed to achieve a nearly optimal laminar flow profile in the reactor.
- Outlet flow is divided into a “ring flow” (taken from the perimeter of the tube) and center flow
- Online monitoring of photochemical age via D9-butanol according to Barnet et al. (2012)

SOA emissions from a gasoline engine

- EURO5, 2 l turbo charged flexible fuel gasoline engine
- Engine operation according to NEDC cycle
- For gasoline 4-times higher SOA emission than primary organic aerosol (POA) emission (Fig. 3)
- E85 ethanol fuel generates lower POA and SOA.

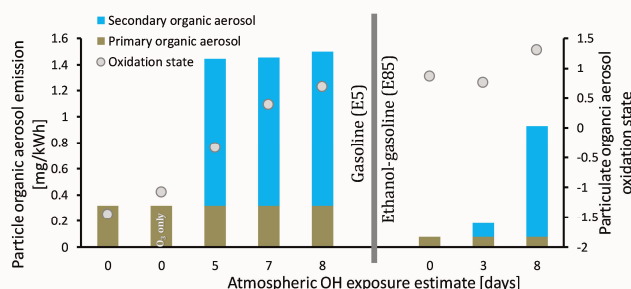


Fig. 3. Primary and secondary organic aerosol emissions and the carbon oxidation state of the organic aerosols in gasoline engine emissions of two different fuel blends. Measured with AMS-ToF.

Evaluation of the photochemical tube reactor

- Range of OH exposures ~ 10^9 – 10^{12} molec cm⁻³ s
→ up to 2 weeks of atmospheric age
- Particle losses: 1-10% for 50 nm particles
- Laminar flow profile (Fig. 2)
- Toluene SOA yield without seed particles 0.44 to 0.5 at OH exposures 1.2-3.4¹¹ molec cm⁻³ s

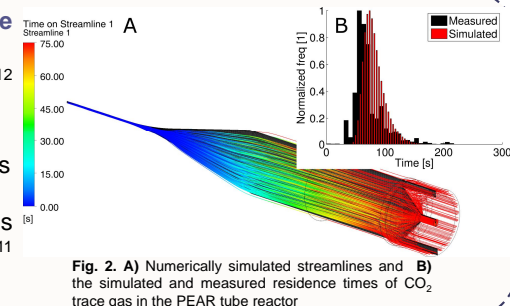


Fig. 2. A) Numerically simulated streamlines and B) the simulated and measured residence times of CO₂ trace gas in the PEAR tube reactor

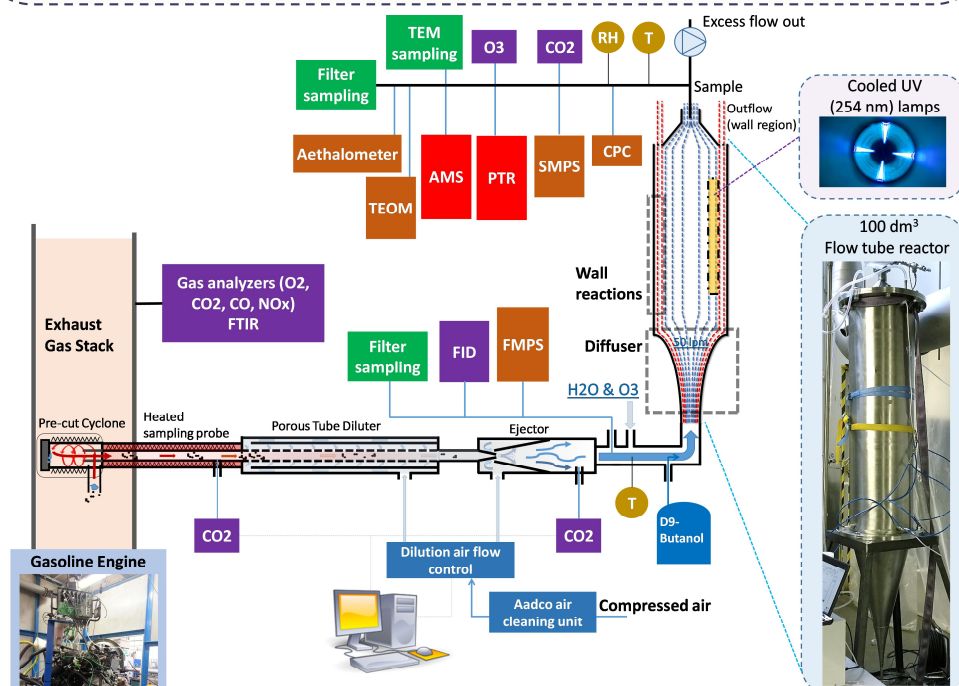


Fig. 1. PEAR experimental setup in the gasoline engine experiments

SOA emissions from log-wood combustion

- Northern modern masonry heater
- Fired with spruce logs with three different moisture contents
- Each measurement includes combustion of 1-3 batches
- OH-exposure: 1.5-1.6¹¹ molec cm⁻³ s → ~ 2 days in atmosphere
- thermal-optical carbon analysis of PM1 filters (NIOSH 5040)
- Organic carbon increases by 20-250 % upon aging (Fig. 4)
- Fuel moisture content has enormous effects on POA and SOA emissions



Fig. 4. Primary and secondary particulate emissions from combustion of spruce wood logs with different moisture contents.

Support by the Academy of Finland (Grant: 258315) and the Helmholtz Virtual Institute of Complex Molecular Systems in Environmental Health (HICE) is gratefully acknowledged.

REFERENCES:

- Kang, E., Toohey, D. W., and Brune, W. H. (2007) *Atmos. Chem. Phys.* 11, 1837-1852.
- Barnet, P., Dommen, J., DeCarlo, P.F., Tritscher, T., Praplan, A.P., Platt, S.M., Prevot, A.S.H., Donahue, N.M., Baltensperger, U. (2012) *Atmos. Meas. Tech.*, 5, 647-656.



UNIVERSITY OF
EASTERN FINLAND



HICE Aerosols and Health
Helmholtz Virtual Institute of Complex
Molecular Systems in Environmental Health