Improving Chemical Transport Model Predictions of Organic Aerosol: Measurement and Simulation of Semivolatile Organic Emissions from Mobile and Non-Mobile Sources

Allen L. Robinson

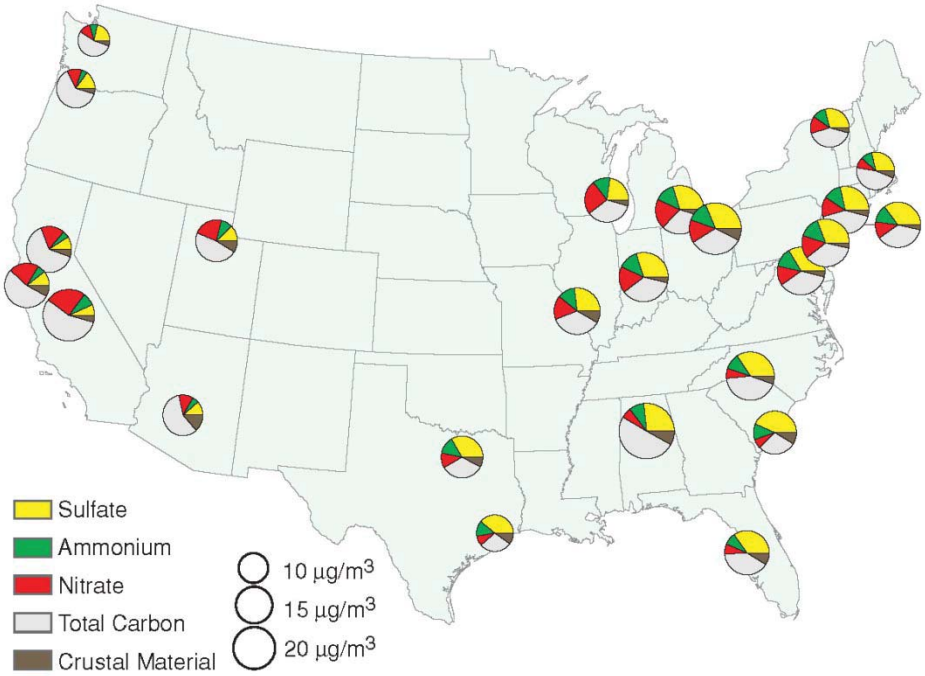
Center for Atmospheric Particle Studies

Carnegie Mellon University

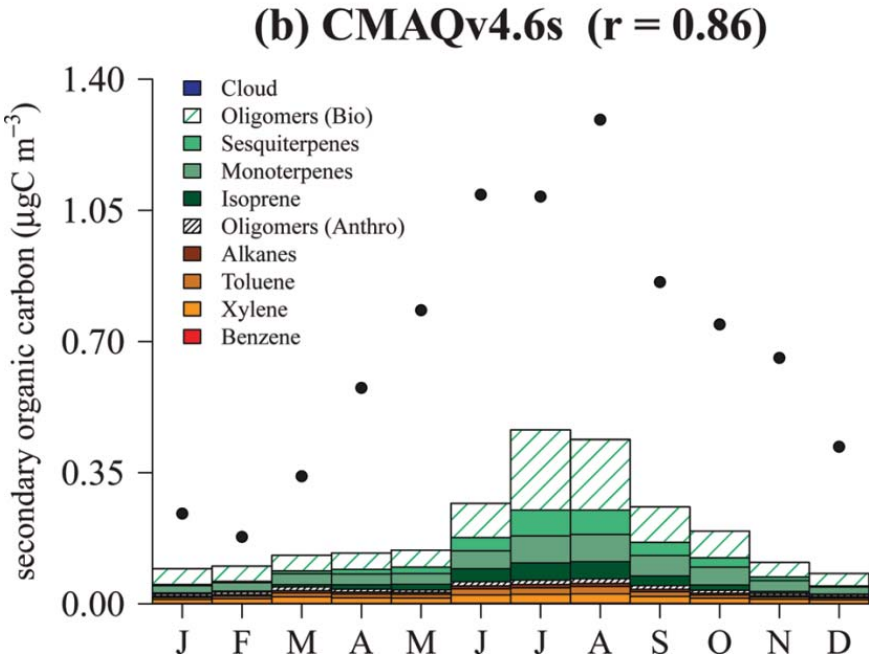
Presented at EPA STAR-OTAQ Transportation Emissions Research Forum,
U.S. Environmental Protection Agency, RTP, NC, March 4-5, 2014.

Carnegie Mellon

Organic aerosols are ubiquitous, but often underpredicted by CTMs



(NARSTO PM Assessment 2004)



(Carlton et al. EST 2010)

Carnegie Mellon

How do PM emissions evolve after leaving a source?

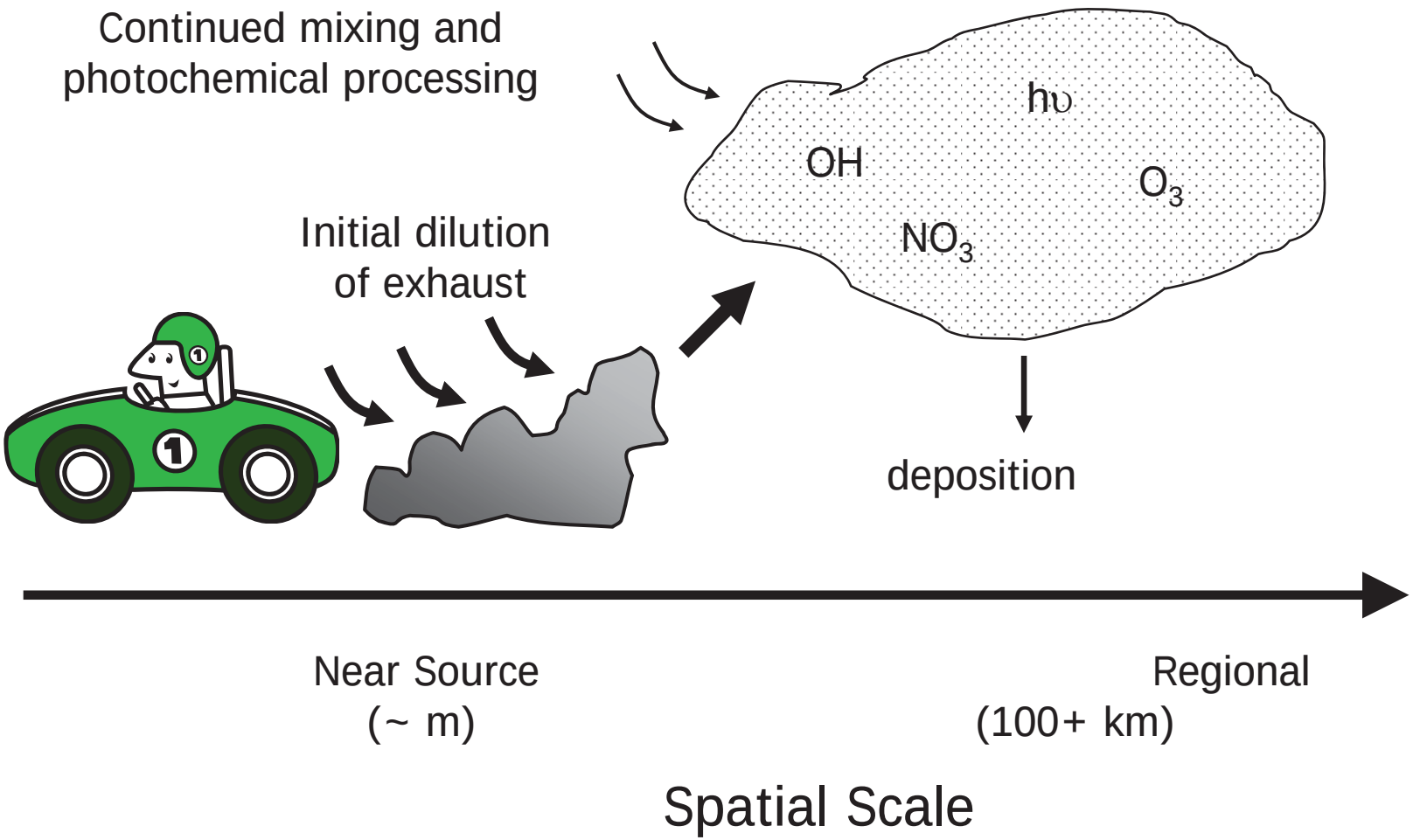
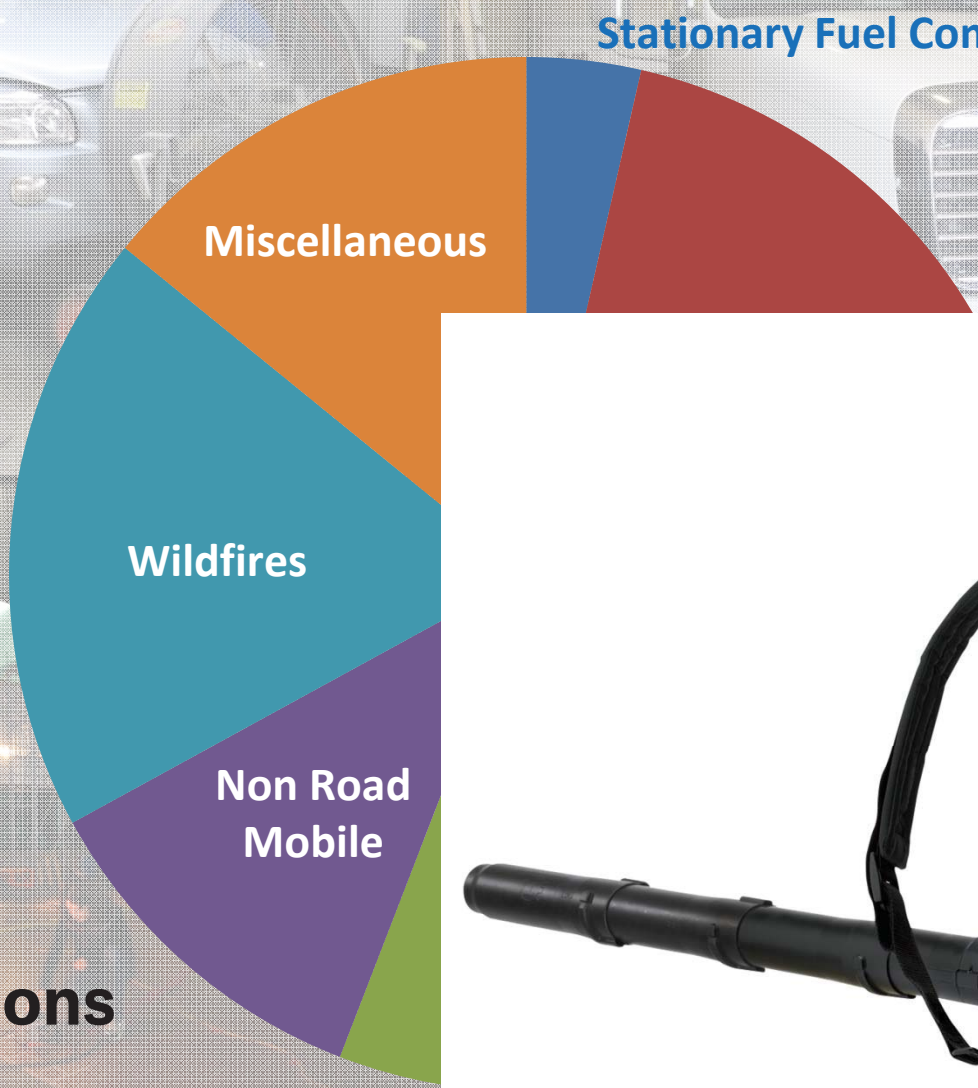


Photo: Daniel Welsh-Bon

US Anthropogenic VOC Emissions



2013
17,624 short tons

http://www.epa.gov/ttnchie1/trends/

Experimental Set-up

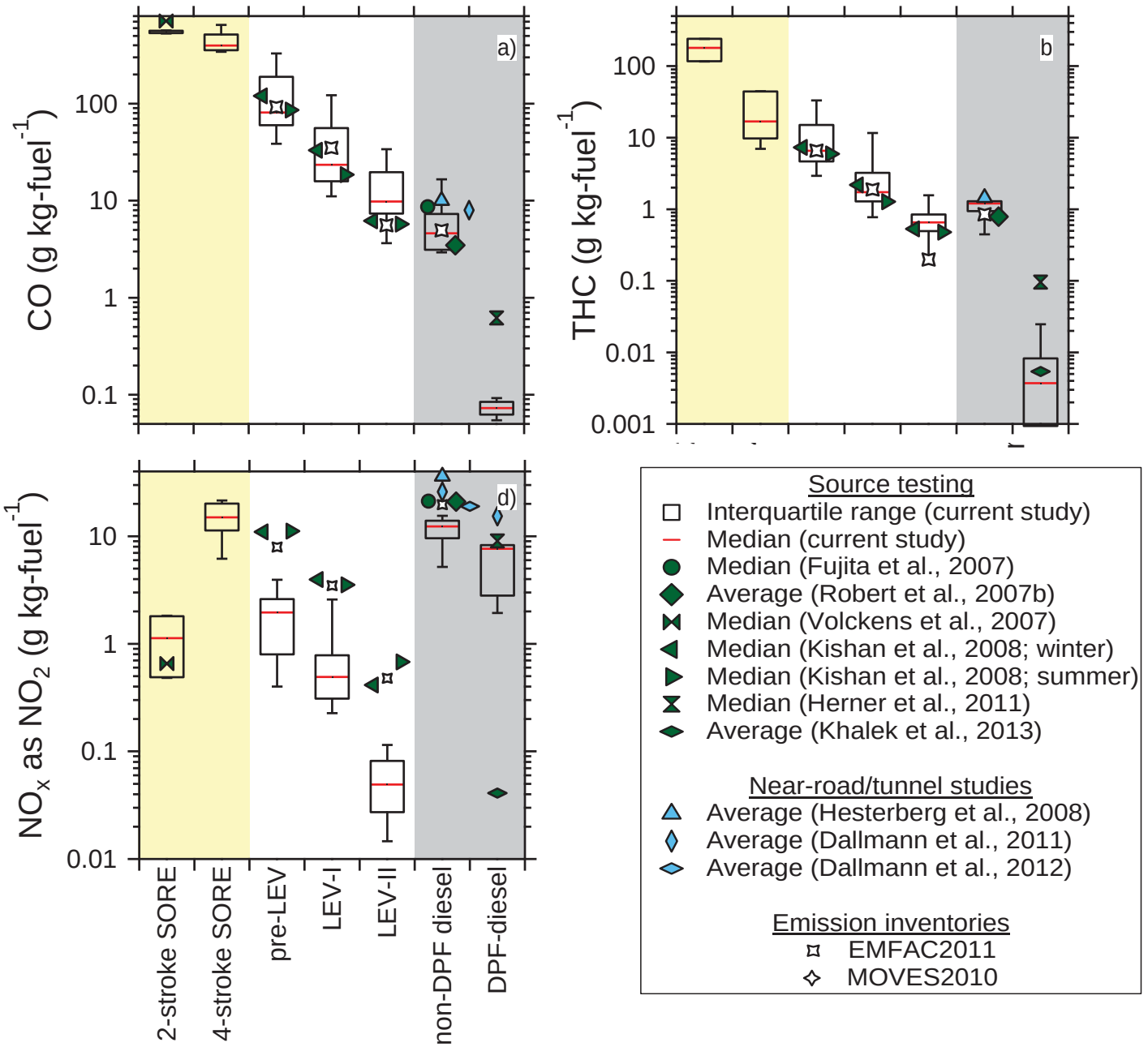
Light-duty vehicles tested using cold start Unified Cycle

Primary emissions characterized with CVS

Secondary aerosol production investigated with portable smog

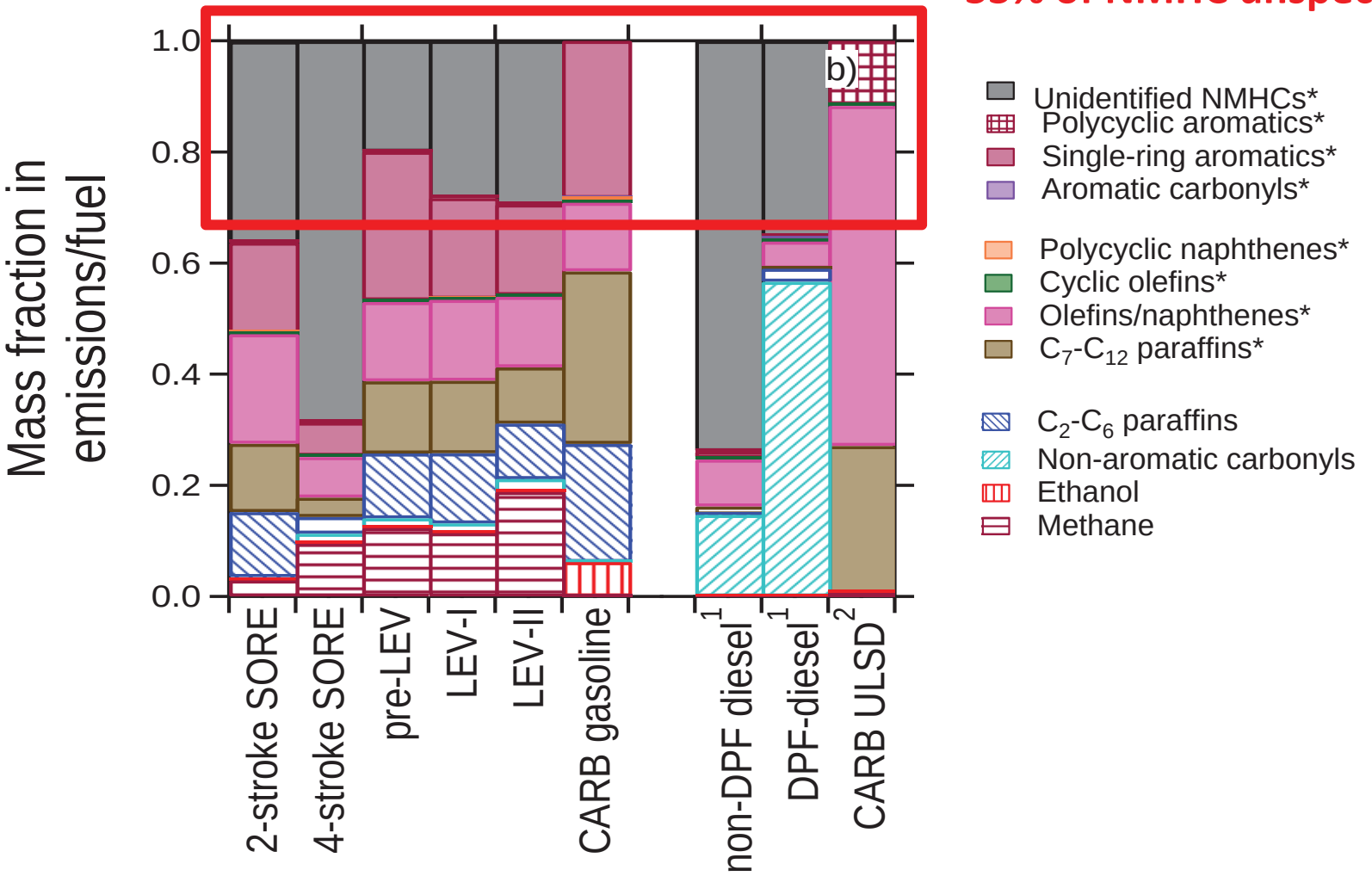
Carnegie Mellon

Gaseous Emissions



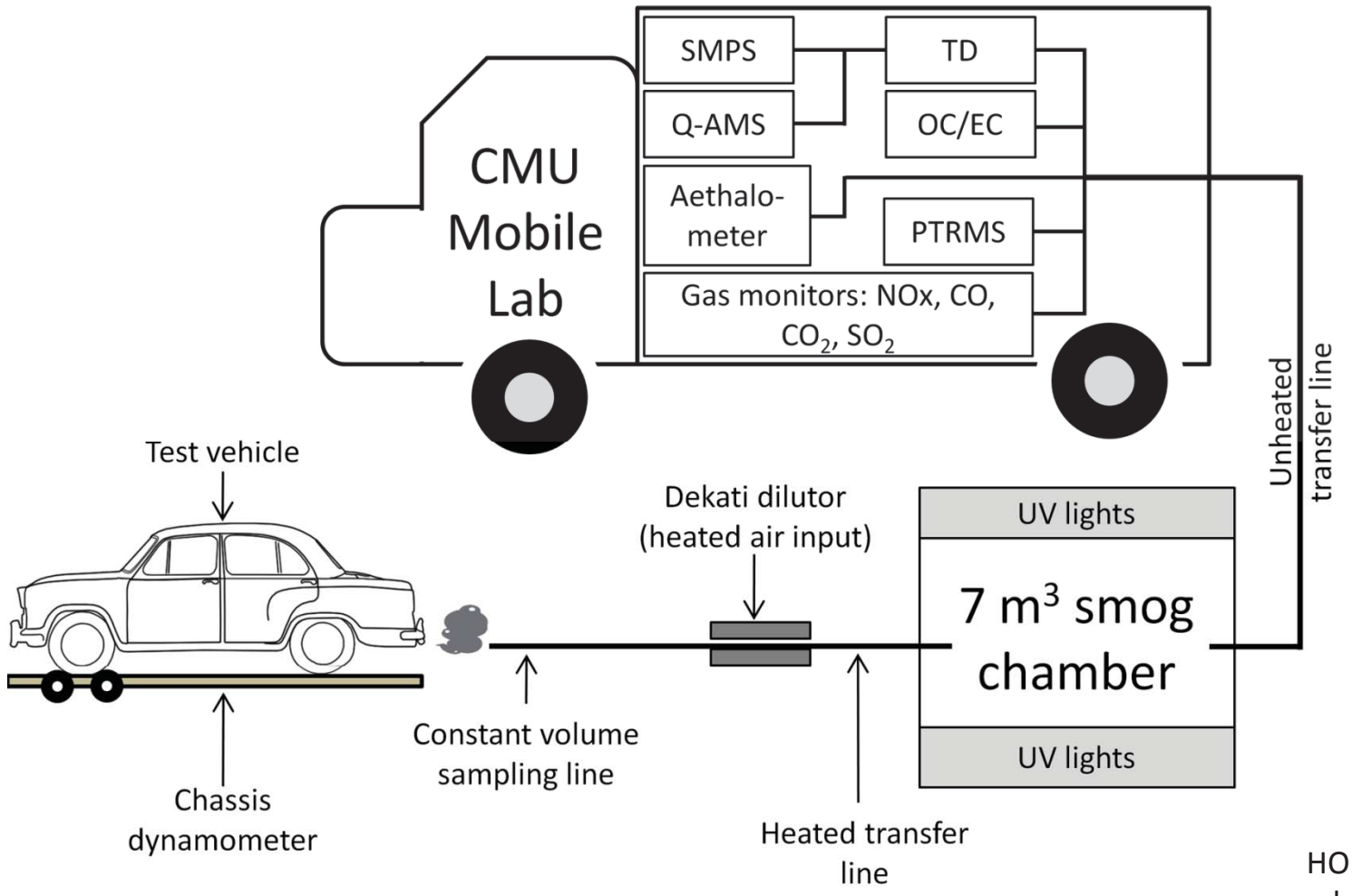
May et al. AE in press

VOC speciation



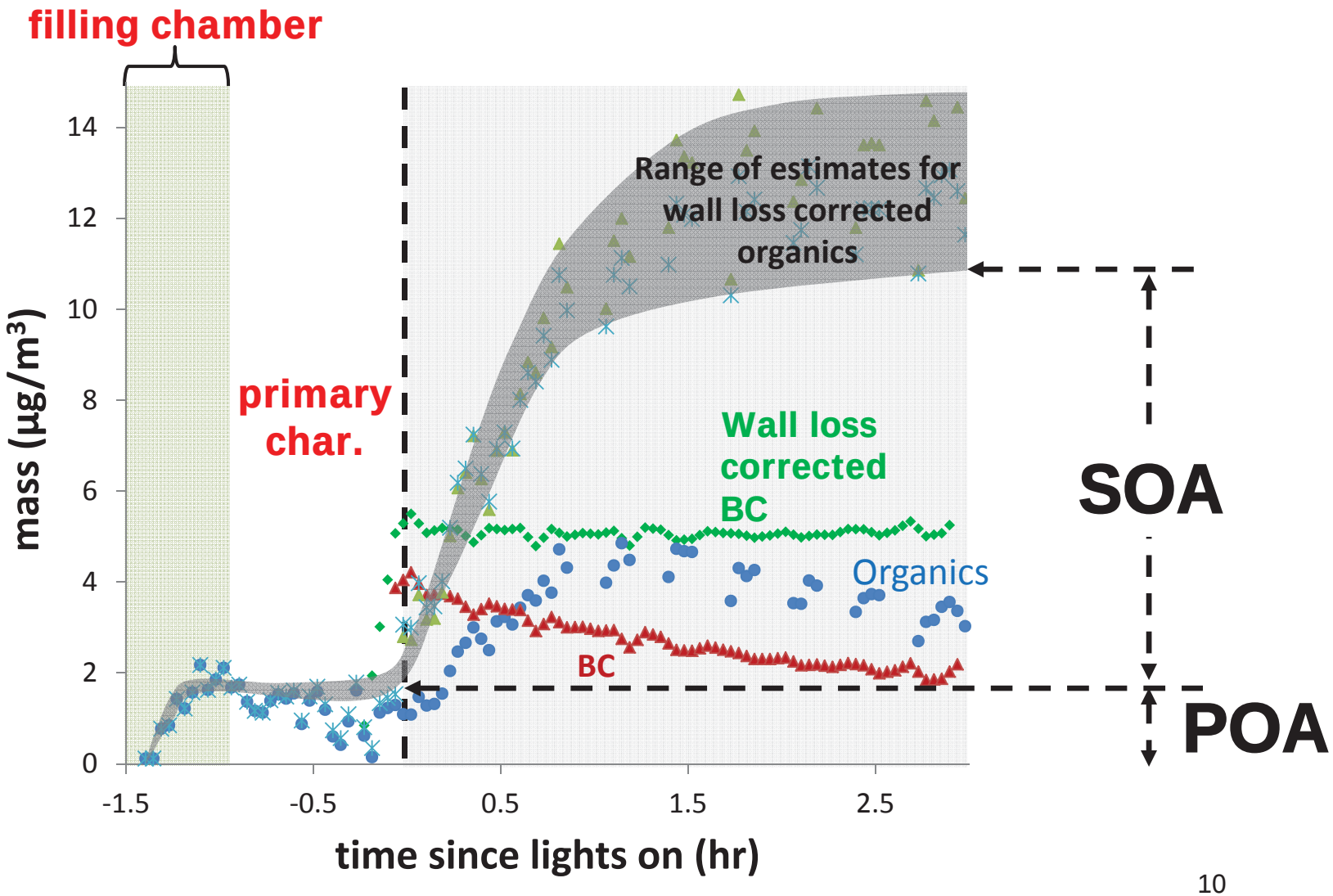
May et al. AE in press

Smog chamber aging experiments

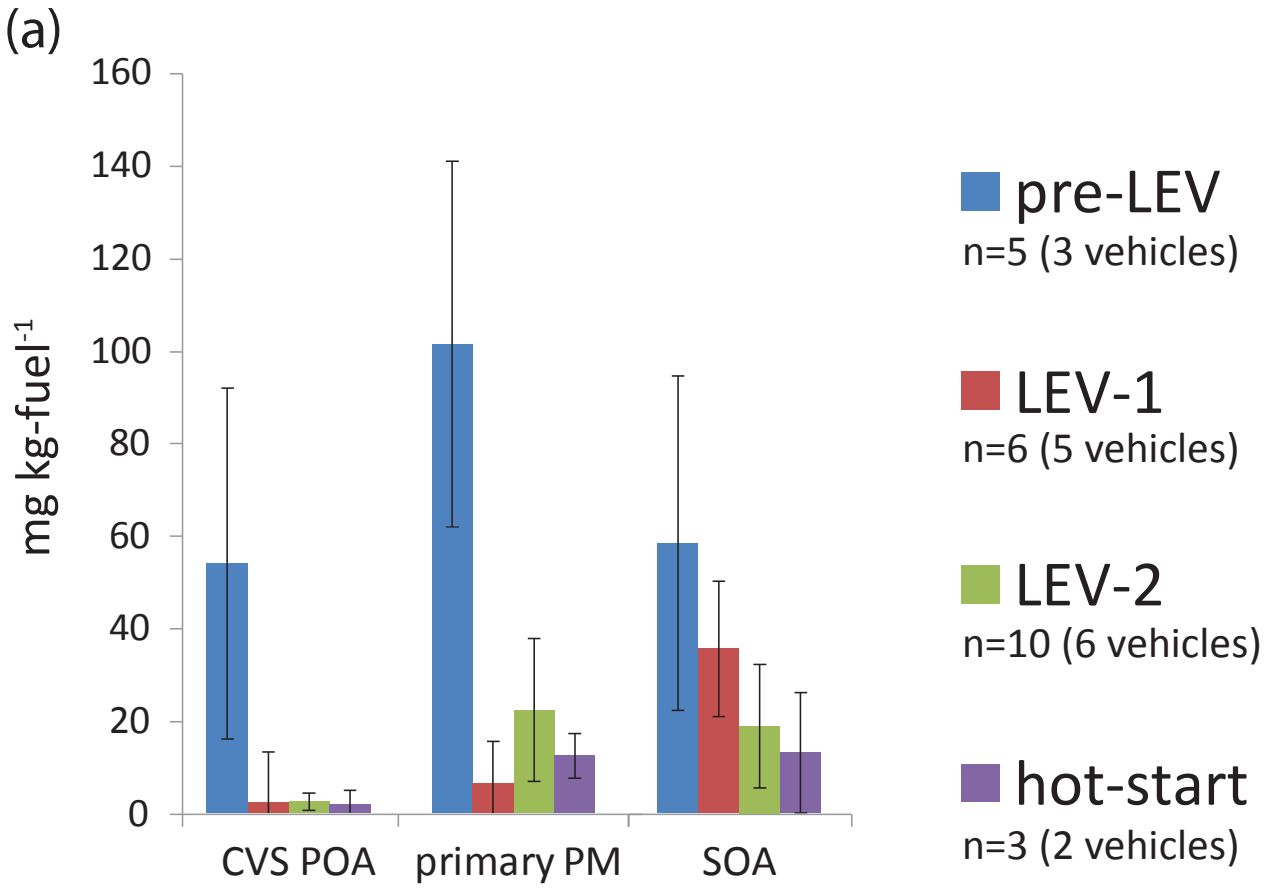


HONO & propene addition

Production of Secondary Organic Aerosol

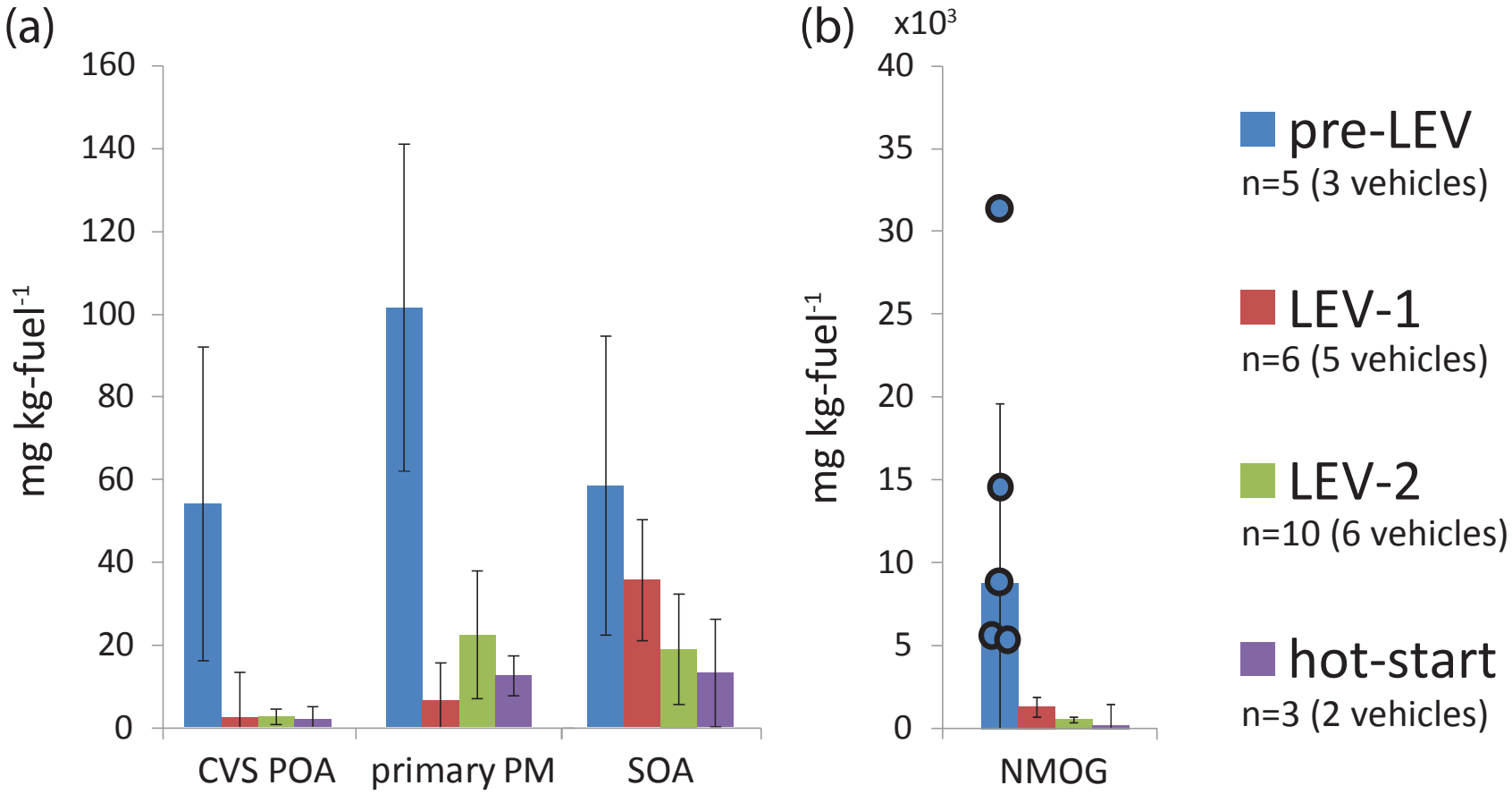


Summary of LDGV Results



Gordon et al. ACP 2014

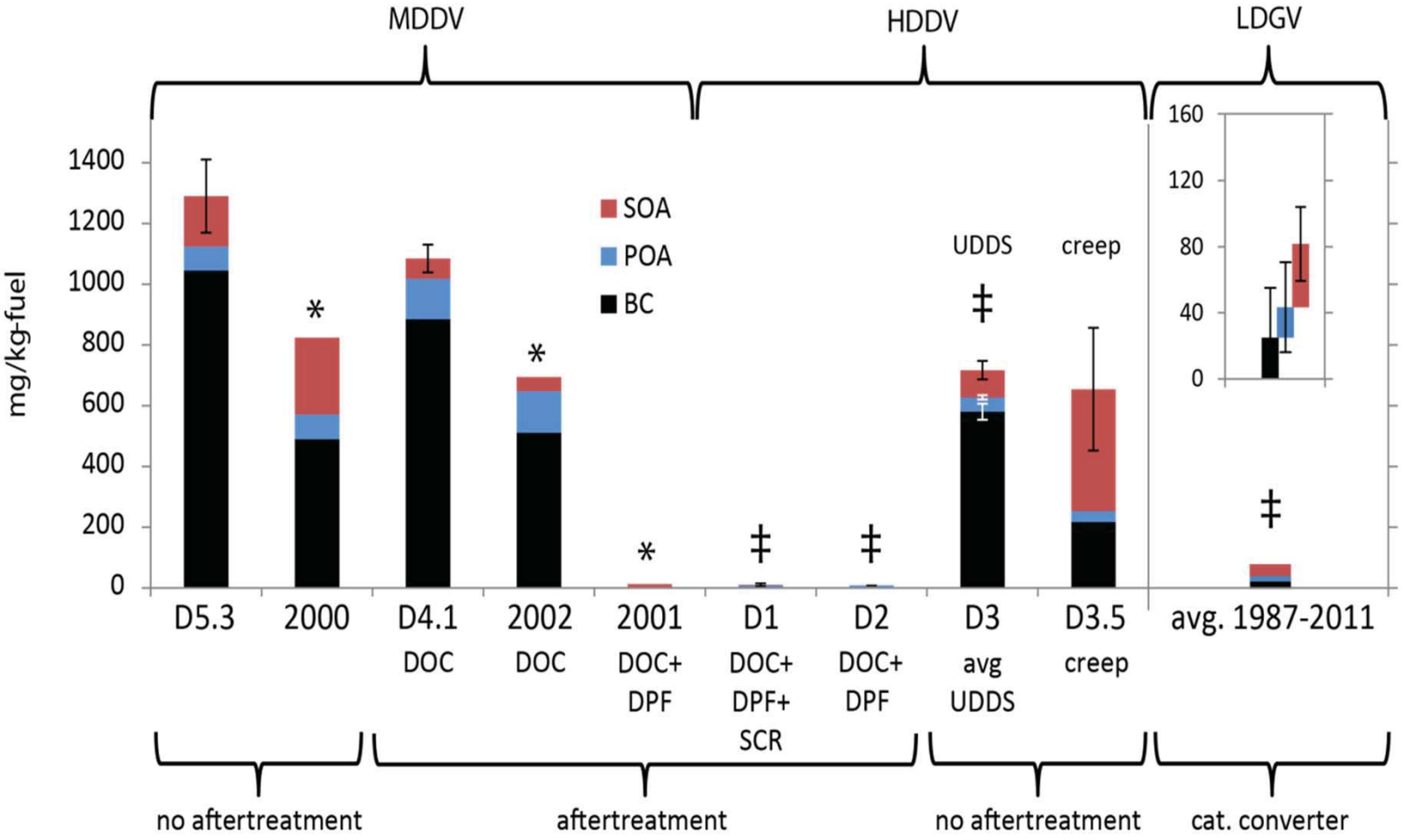
Summary of LDGV Results



Reduction in SOA < Reduction in NMOG → LEV2 emissions more potent SOA precursors ?

Gordon et al. ACP 2014

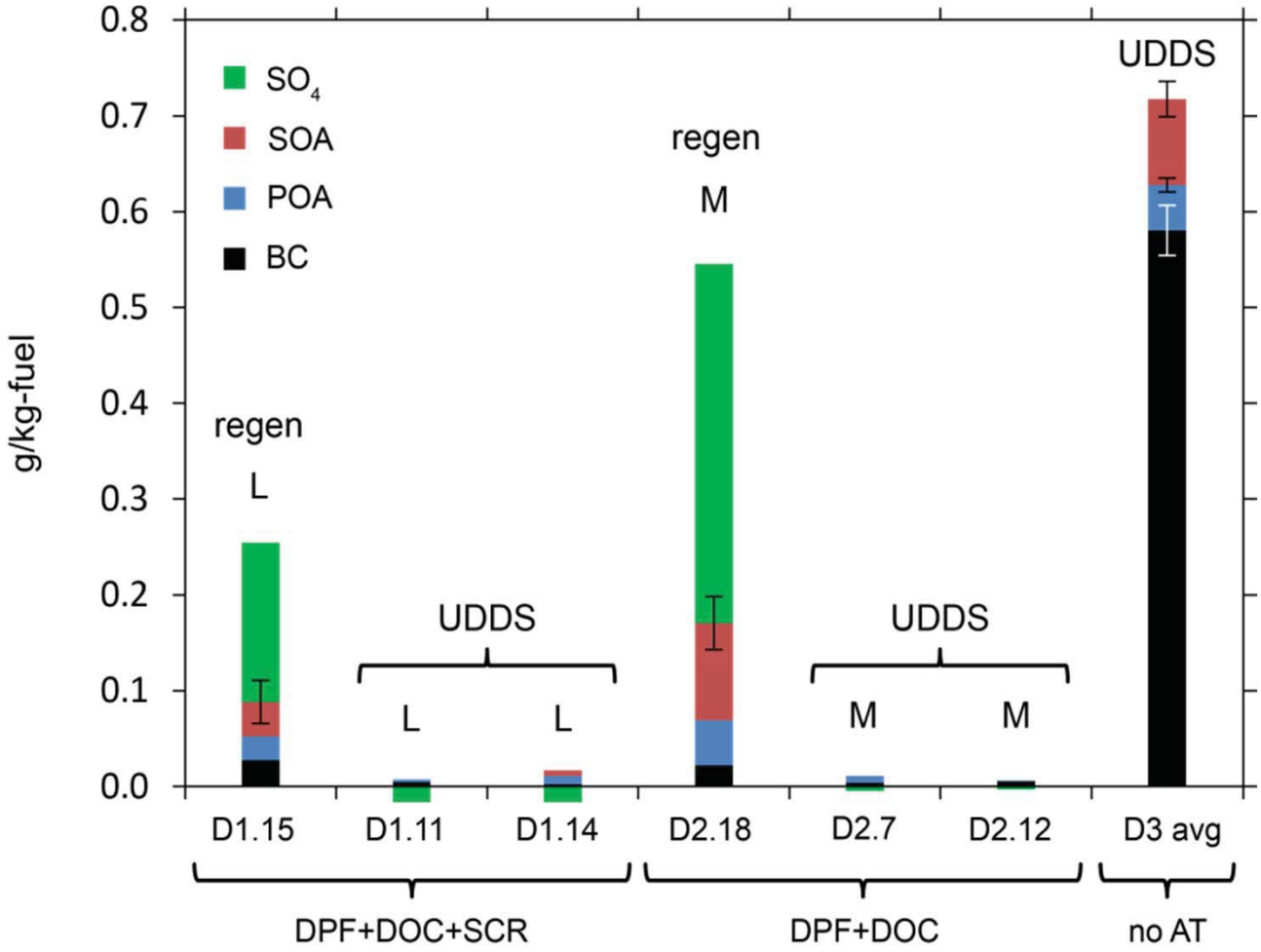
Diesel SOA



Gordon et al. ACP 2014

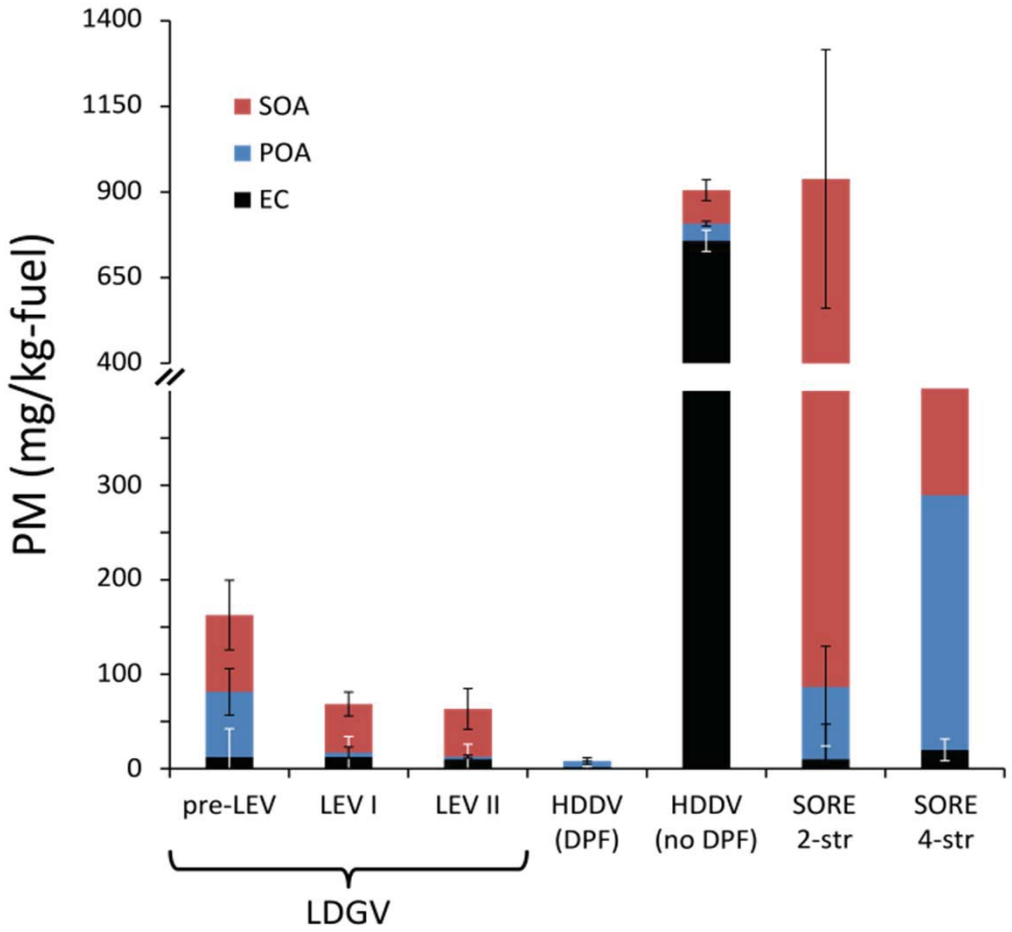
DPF – very effective. Non-DPF – primary PM (BC) very significant

Active Forced Regeneration



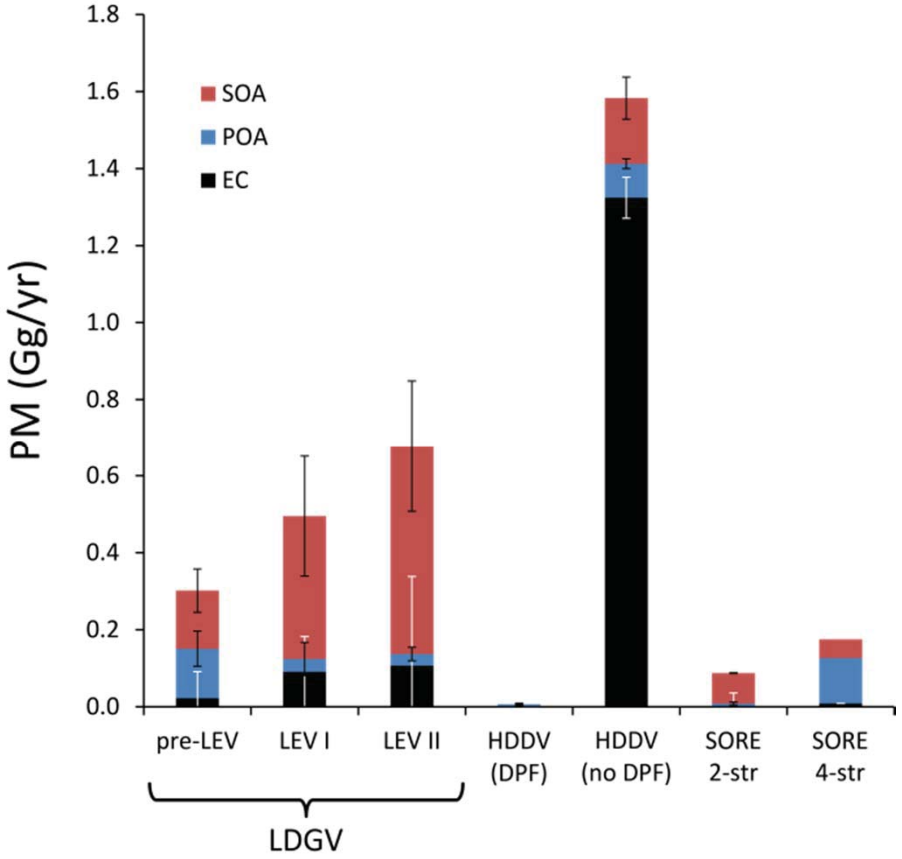
Gordon et al. ACP 2014

Summary of Mobile Source Data



Gordon et al. EST 2013

Scaling Data by SoCAB Fuel Consumption



Gordon et al. EST 2013

Conclusions

- LDGV
  - Lots of SOA relative to POA or primary PM
  - LEV2 emissions appear to have more potent SOA precursor mix than emissions from older vehicles
- Diesel
  - DPF– Very clean (except for forced regeneration)
  - Non-DPF – some SOA formation but primary PM dominated
- SORE
  - Lots of SOA formation (and primary PM)
- SoCAB budget:
  - After three hours of oxidation: diesel PM (mainly BC) ~ gasoline PM (mainly SOA)
  - **What happens at longer time scales?**
- Unspeciated organics appear to be important SOA precursors
- SOA yields parameterized with volatility basis set

Papers

Primary Emissions data

- "Gas- and particle-phase primary emissions from in-use, on-road gasoline and diesel vehicles" (A.A. May et al.) Atmospheric Environment, in press.
- "Primary Gas- and Particle-Phase Emissions and Secondary Organic Aerosol Production from Gasoline and Diesel Off-Road Engines" (T.D. Gordon et al.), Environmental Science & Technology, *47* (24), pp 14137–14146, 2013.

Secondary Organic Aerosol Formation

- "Secondary Organic Aerosol Formation Exceeds Primary Particulate Matter Emissions for Light-Duty Gasoline Vehicles" (T.D. Gordon et al.), Atmospheric Chemistry and Physics, in press (discussion paper is online http://www.atmos-chem-phys-discuss.net/13/23173/2013/acpd-13-23173-2013.html).
- "Secondary Organic Aerosol Production from Diesel Vehicle Exhaust: Impact of Aftertreatment, Fuel Chemistry and Driving Cycle" (T.D. Gordon et al.), Atmospheric Chemistry and Physics, in press (discussion paper is online http://www.atmos-chem-phys-discuss.net/13/24223/2013/acpd-13-24223-2013.html).
- "Primary Gas- and Particle-Phase Emissions and Secondary Organic Aerosol Production from Gasoline and Diesel Off-Road Engines" (T.D. Gordon et al.), Environmental Science & Technology, *47* (24), pp 14137–14146, 2013.
- "Primary to secondary organic aerosol: evolution of organic emissions from mobile combustion sources" (A. A. Presto et al.) Atmospheric Chemistry and Physics, submitted (discussion paper is online http://www.atmos-chem-phys-discuss.net/13/24263/2013/acpd-13-24263-2013.html).

Gas-particle partitioning of primary organic aerosol

- "Gas-particle partitioning of primary organic aerosol emissions: (2) diesel vehicles" (A. A. May et al.) Environmental Science & Technology, 47 (15), 8288–8296, 2013.
- "Gas-particle partitioning of primary organic aerosol emissions: (1) gasoline vehicle exhaust" (A. A. May et al.) Atmospheric Environment, 77, 128-139, 2013.
- "Gas-particle partitioning of primary organic aerosol emissions 3. Biomass burning" (A.A. May et al.) Journal of Geophysical Research, 118(19), 2013JD020286, 2013.

Acknowledgments

- **CMU:** Albert Presto, Tim Gordon, Ngoc Nguyen, Chris Hennigan, Andy May, Mrunmayi Karve, Shantanu Jathar, Yunliang Zhao, Eric Lipsky, Neil Donahue, Peter Adams
- **ARB:** Alváro Gutierrez, William H. Robertson, Mang Zhang, Oliver Chang, Shiyan Chen, Pablo Cicero-Fernandez, Mark Fuentes, Shiou-Mei Huang, Richard Ling, Jeff Long, Christine Maddox, John Massetti, Eileen McCauley, Kwangsam Na, Yanbo Pang, Paul Rieger, Todd Sax, Tin Truong, Thu Vo, Christopher Brandow, Lyman Dinkins, Richard Ong Antonio Miguel, Sulekha Chattopadhyay, Hector Maldonado, Bob Torres, Eileen McCauley
- **Ford:** Matti Maricq
- **Financial Support:**
  - EPA STAR – CMU primary emissions measurements
  - ARB – Vehicle procurement, testing, and emissions characterization
  - CRC A74/E96 – CMU smog chamber experiments
  - NSF – graduate student fellowships
- This presentation reflects the views of the authors and not any of the funding agencies. No official endorsement should be inferred.