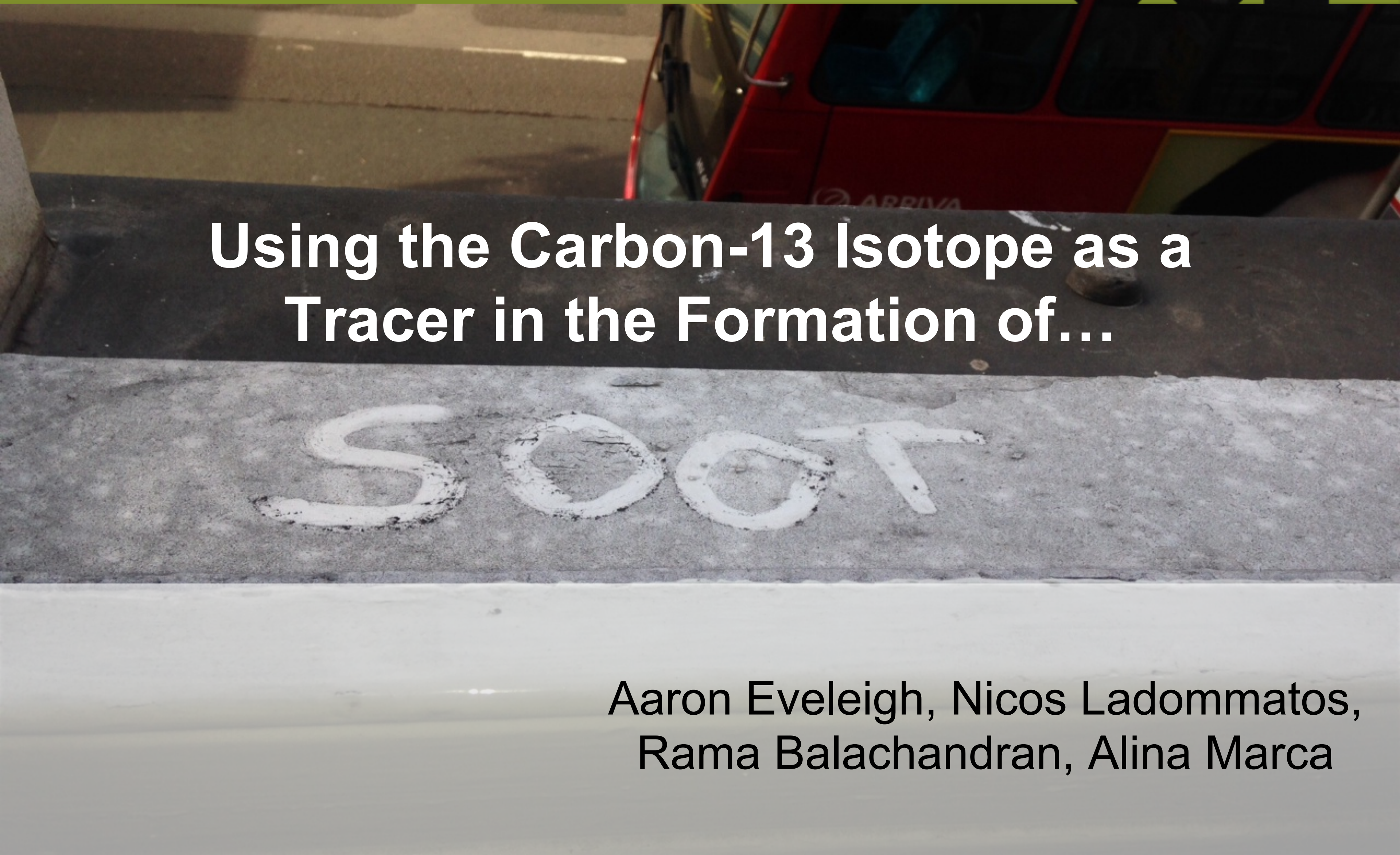
Using the Carbon-13 Isotope as a Tracer in the Formation of…



Aaron Eveleigh, Nicos Ladommatos,
Rama Balachandran, Alina Marca

1. Motivation
2. History & Background
3. Experimental Method
4. Results
5. Potential

Motivation

- Detrimental health and environmental effects of particulate emissions; and
- Increasingly stringent legislation limits these.

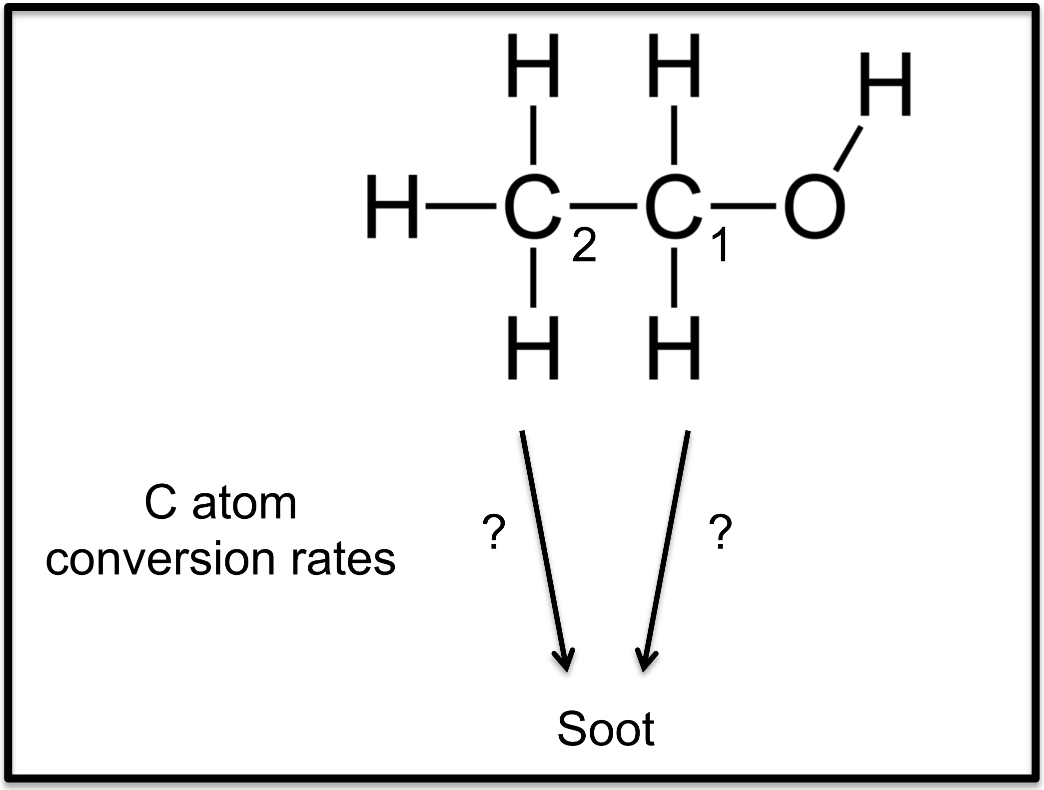
Overall Approach

- Understand how single-component and multicomponent fuels form PM, and other emissions;
- With the aim of improving fuel characteristics by post-processing or bioengineering.

Aims

- To show how individual atoms within a molecule convert to soot, in:

- Ethanol
- Propanol(s)
- Pentanol(s)
- Ethyl Acetate
- Toluene



Use of Isotopes in Combustion Studies

- Isotopes have not been widely used for combustion research, and only few examples exist;

- Most examples of tracer studies in combustion use radiocarbon, ^{14}C, to track atoms from fuel to soot.

- Some more recent examples of isotopes being used with laser techniques in flames.

Isotope measurement by EA-IRMS

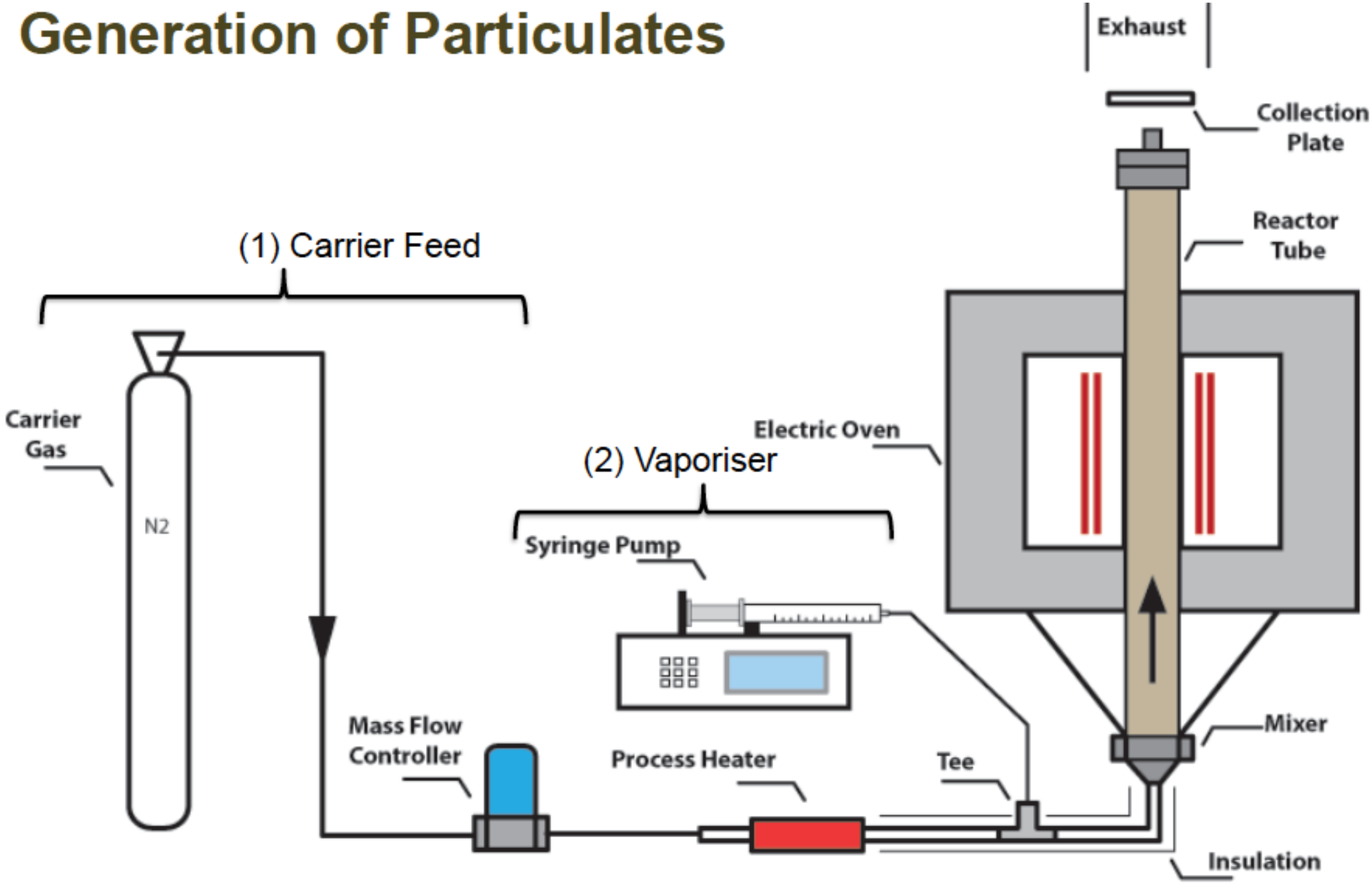
- Isotope ratio measurement by IRMS (Isotope ratio Mass spectrometry) are routine in Earth sciences, for isotopes of: Carbon, Oxygen, Hydrogen, and Nitrogen.
- Elemental Analysis (EA), is required to convert the sample to CO_2 which is required for IRMS.
- The CO_2 is passed to an isotope ratio mass spectrometer where beams of $^{12}CO_2$ and $^{13}CO_2$ are split and detected.
- Measurements represent ^{13}C abundance relative to an international standard (PDB).

Bloomsbury Environmental Isotope Facility

Labelling Fuels

- Typically less than 200ul of enriched fuel required per 100ml of unenriched fuel.
- Tests have been conducted with single-component fuels.
- In principle, also possible to apply the method to trace a particular component in multi-component fuels.

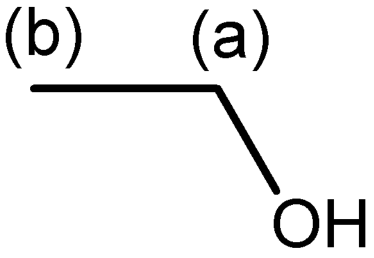
Generation of Particulates

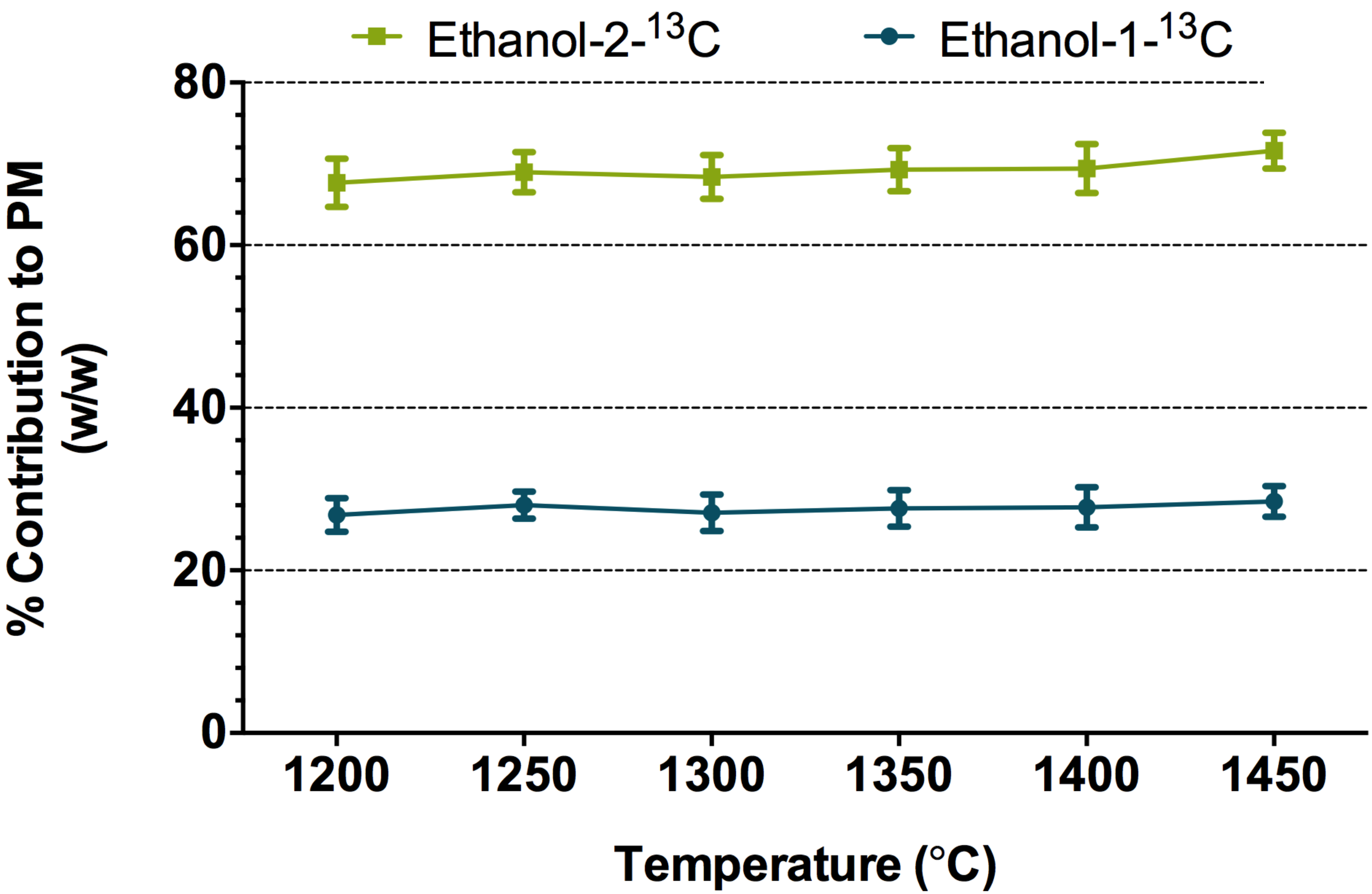


Generation of Particulates (2)

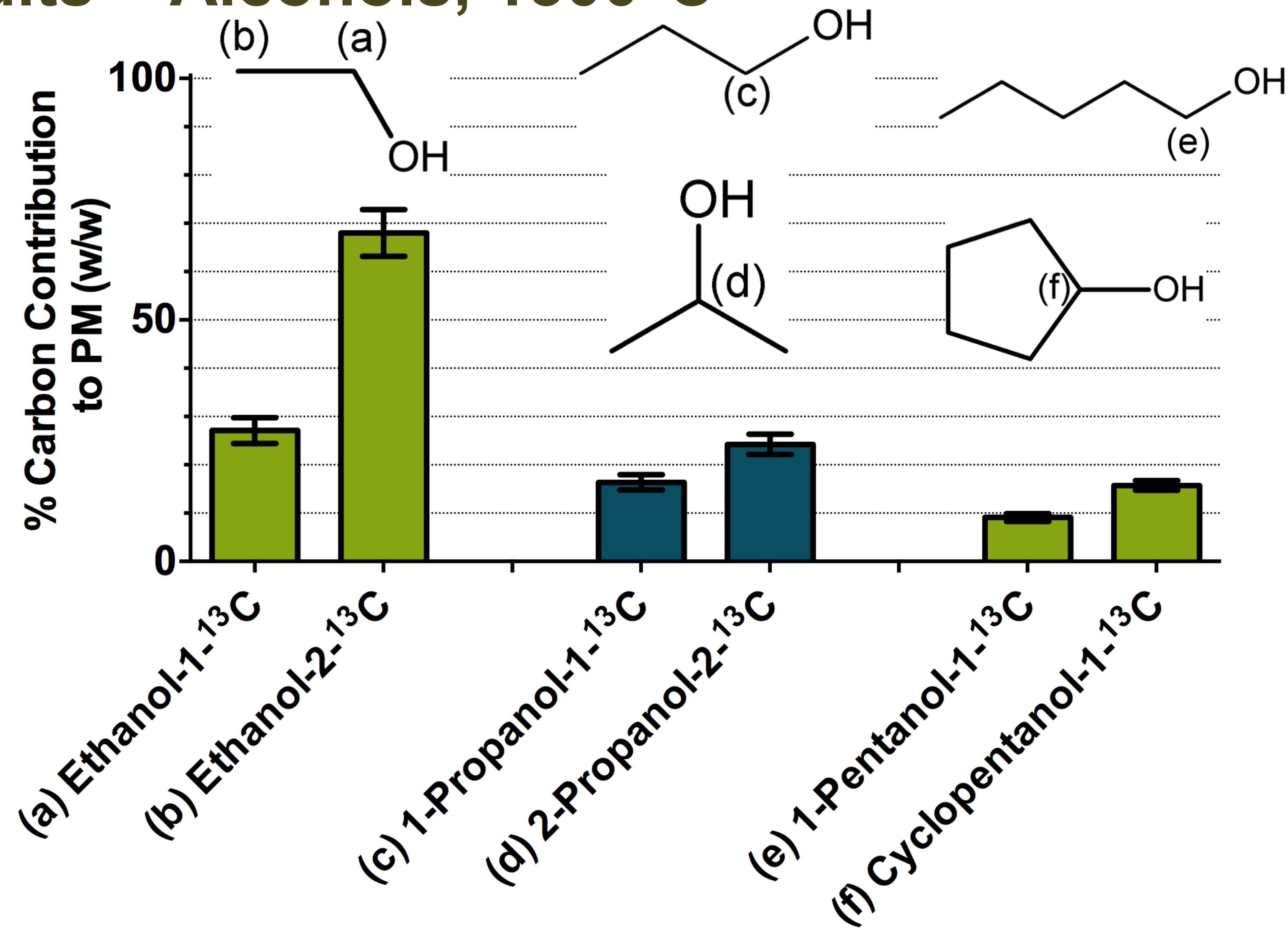
- Temperature range: 1000-1450°C
- Reactor length: 600 mm
- Reactor Diameter: 104 mm
- Carrier flow rate 20 lpm
- Residence time ~1s

Ethanol - Pyrolysis

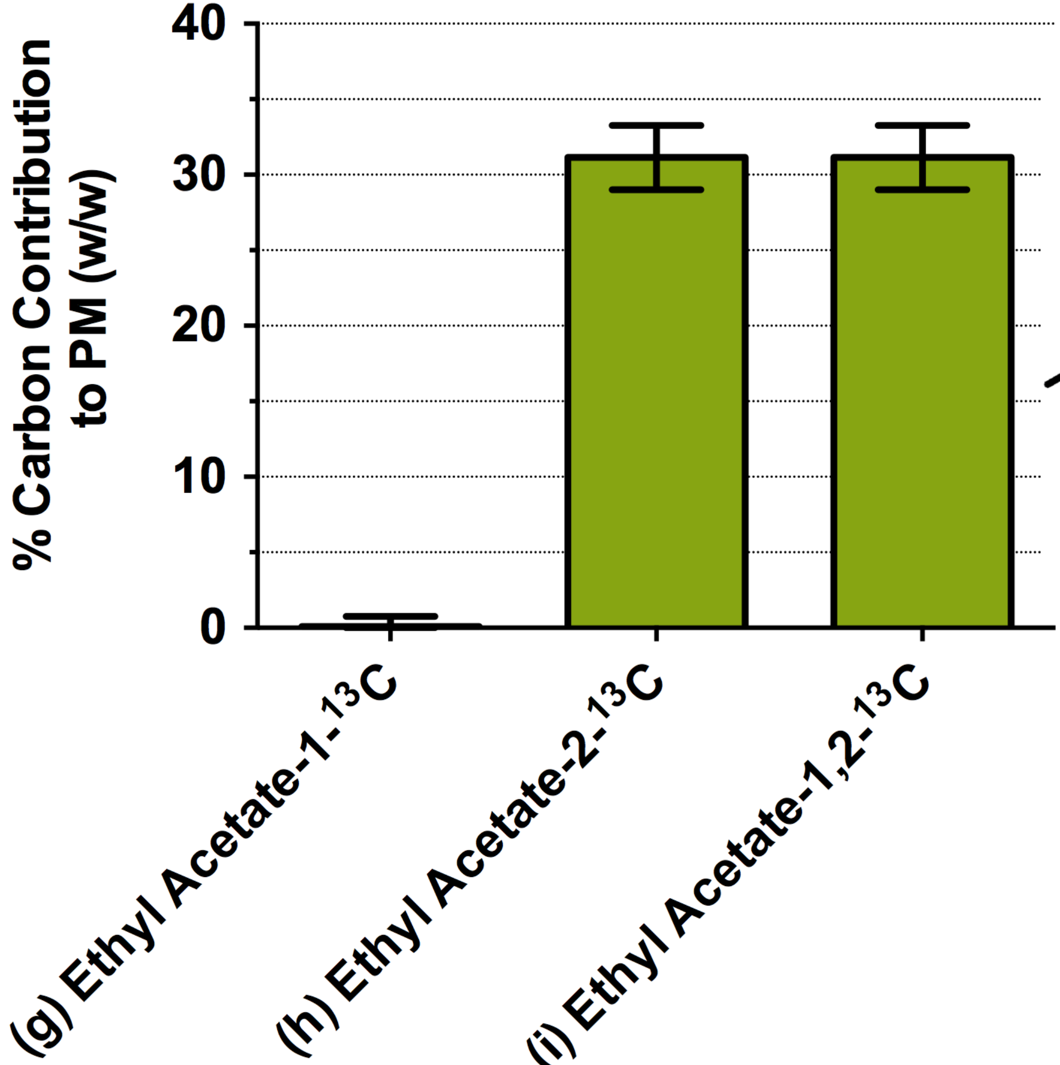


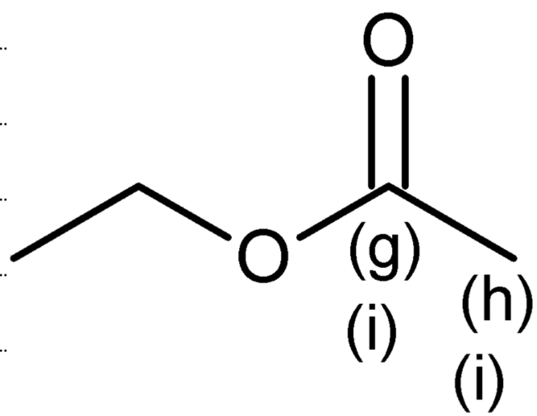


Results – Alcohols, 1300°C

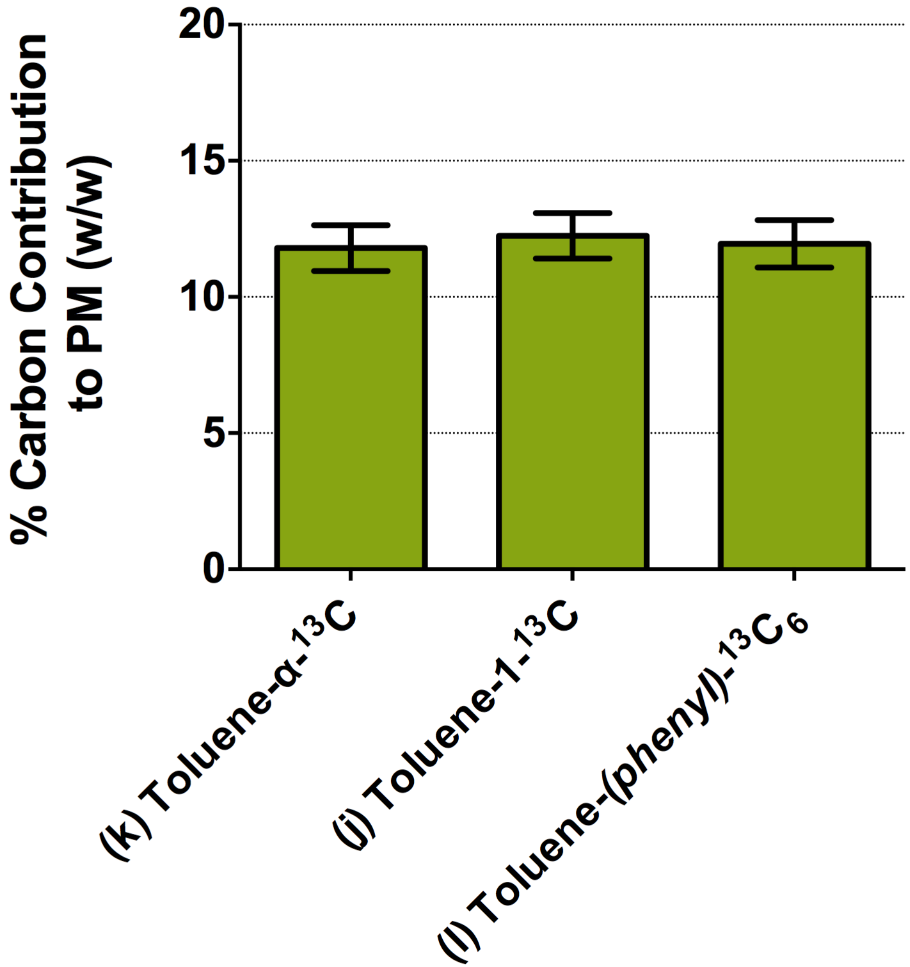


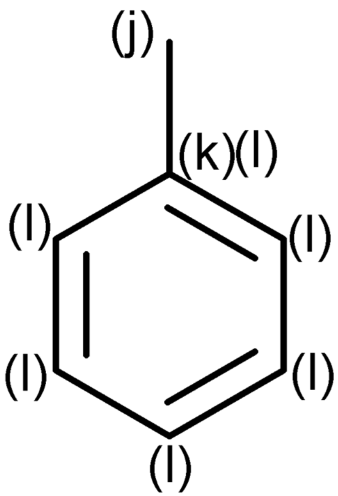
Ethyl Acetate, 1300°C



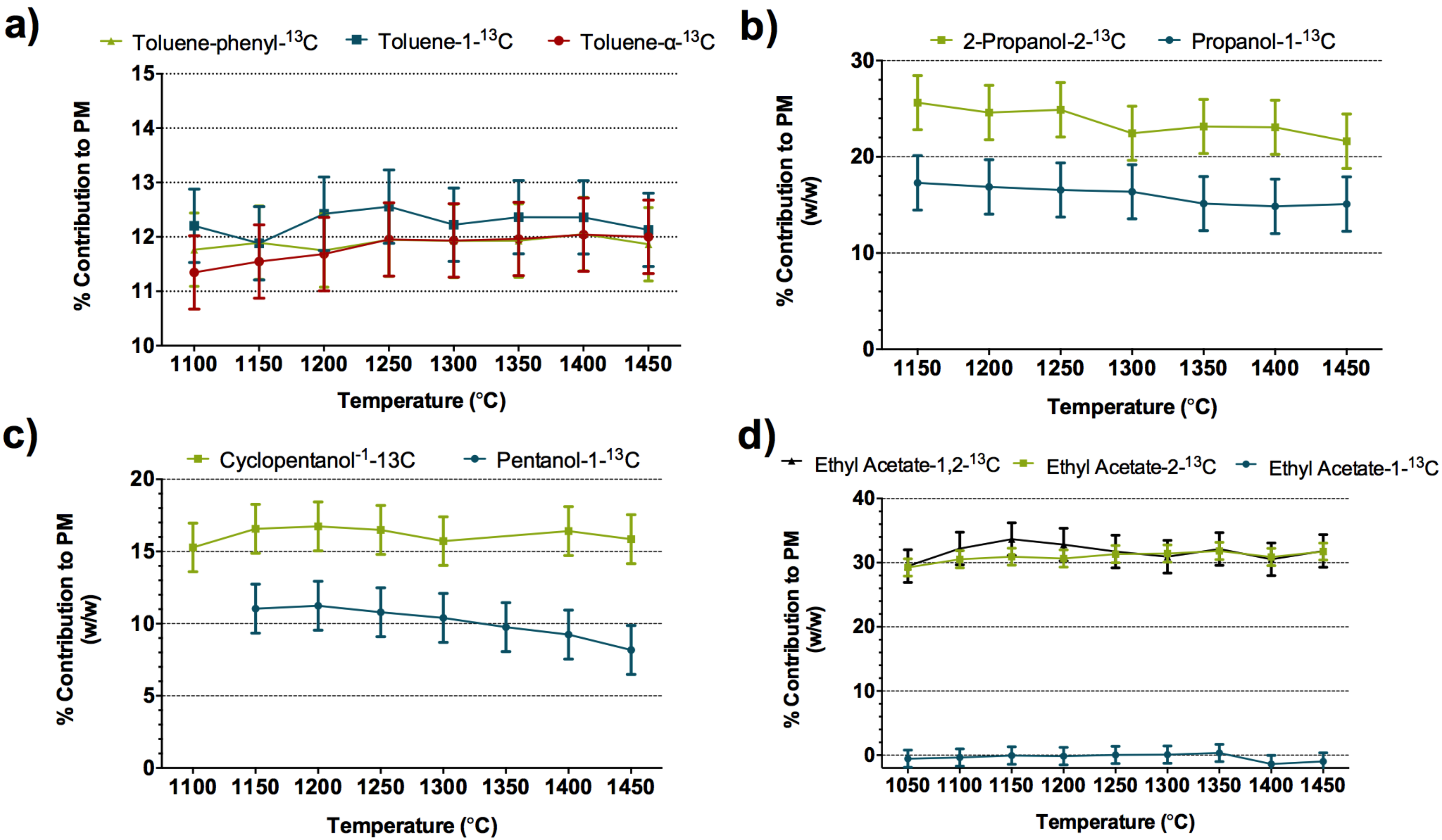


Toluene, 1300°C

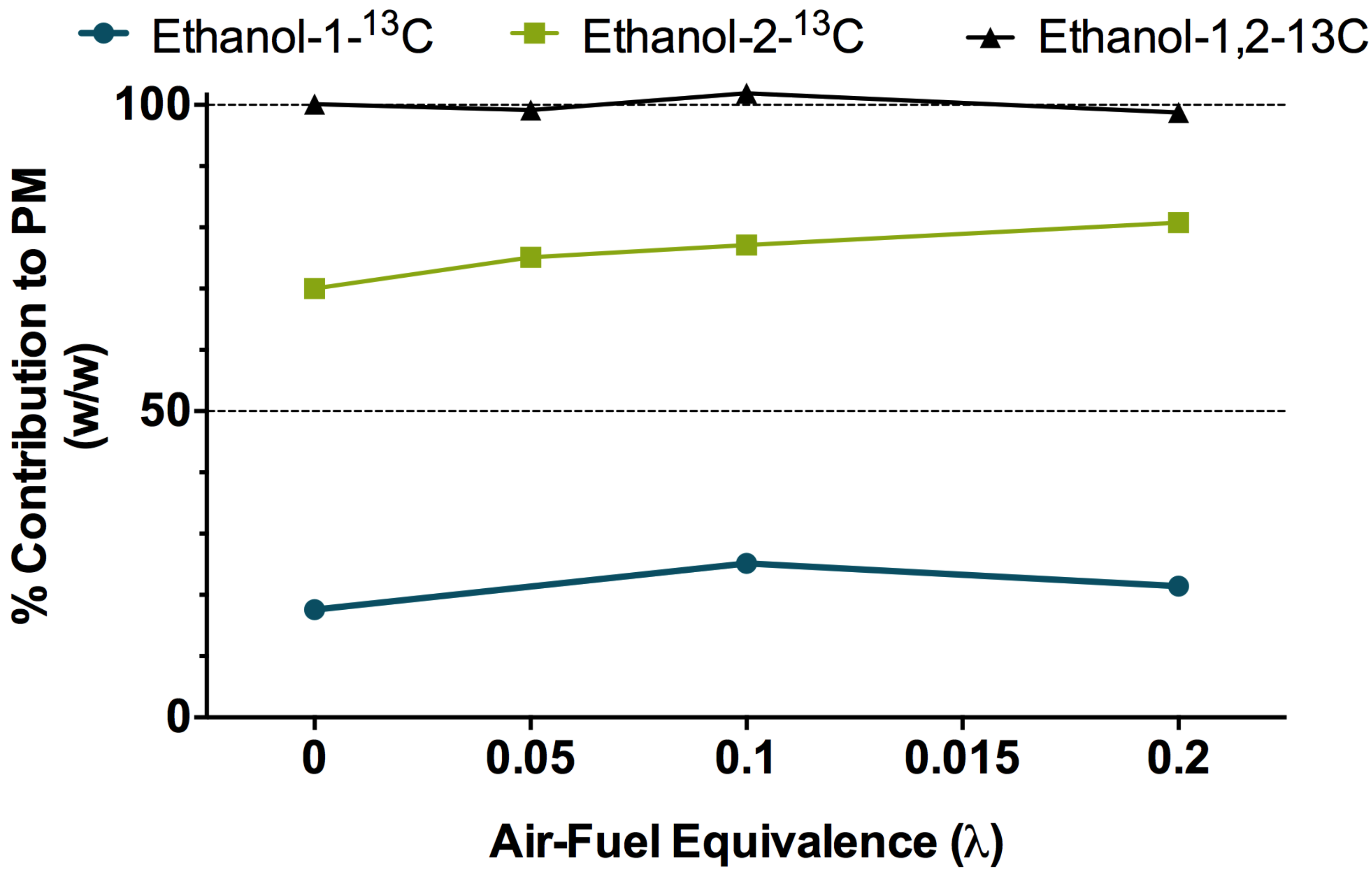




All fuels- With Temperature

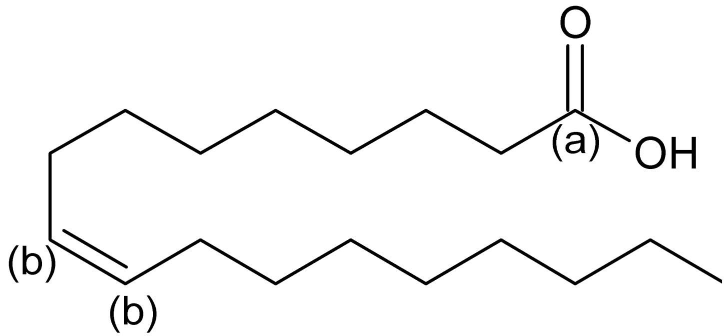


Ethanol and Air- 1300°C

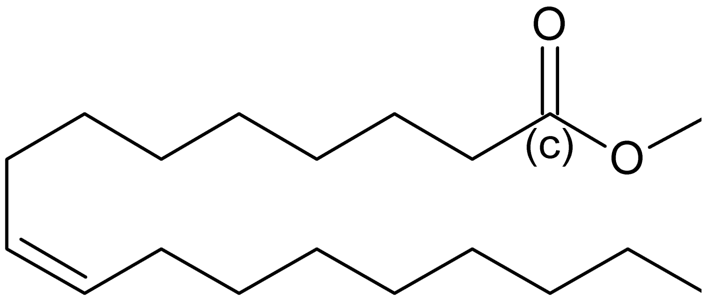


Next Phase of Testing

- Relate reactor results to practical combustion systems.
- Diesel engine and reactor tests with ‘biofuels’: oleic acid, and methyl oleate.



Oleic Acid



Methyl Oleate

Advantages ^{13}C labeling

- IRMS is about 50-100 times cheaper than AMS.
- ^{13}C labeled compounds are significantly cheaper than ^{14}C analogues.
- Off-the-shelf availability of 13C labeled compounds.
- Eliminates procedural precautions required for handling radioactive ^{14}C.

Potential

- Can be used as a diagnostic tool to shed light on the kinetic effects on fuels during combustion.
- Improve estimates of sooting tendency of untested fuels.
- Identify individual atom contributions to PM or components in a mixture.
- In principle, can be applied to CO_2, CO.

Conclusions

- Large differences in contributions to PM depending on the origin of carbon within a molecule;
- Particularly in the case of oxygenated molecules, where conversion of the adjacent carbon is reduced.
- Measurement by IRMS is precise enough to resolve the very small enrichments– considering large background of ^{13}C, and small amount of enriched fuel added.

Thank you

Research supported by:

EPSRC

Engineering and Physical Sciences Research Council

Corresponding open-access article:

A. Eveleigh et al., Combust. Flame. (2014), http://dx.doi.org/10.1016/j.combustflame.2014.05.008

Contact: aaron.eveleigh.10@ucl.ac.uk