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Time-Resolved Characterisation of Carbonaceous Aerosols from Real-World Cookstoves

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Outline

- **Motivation**
- **Experiments and Instrumentation**
- **Results**
- **Conclusion**

Motivation

- 3 billion people in the developing world rely on solid fuels for domestic energy.
- 4 million annual premature deaths attributed to the emissions from residential solid fuel combustion.
- No detailed measurements of chemical and microphysical properties of emissions from developing world stoves.
- Large uncertainty in models estimating radiative forcing by BC particles from solid fuel combustion.

Cookstoves and solid fuels

Heating stove

- Willow logs
- Pine
- Coal

CarbonZero

- Oak (dry)
- Oak (wet)
- Willow stick (dry)

Gyapa

- Oak (dry)
- Oak (wet)
- Charcoal

Lucia stove

- White wood pellets

Experiments and Instrumentation

cToF-AMS: Inorg and Org aerosols in PM_1.

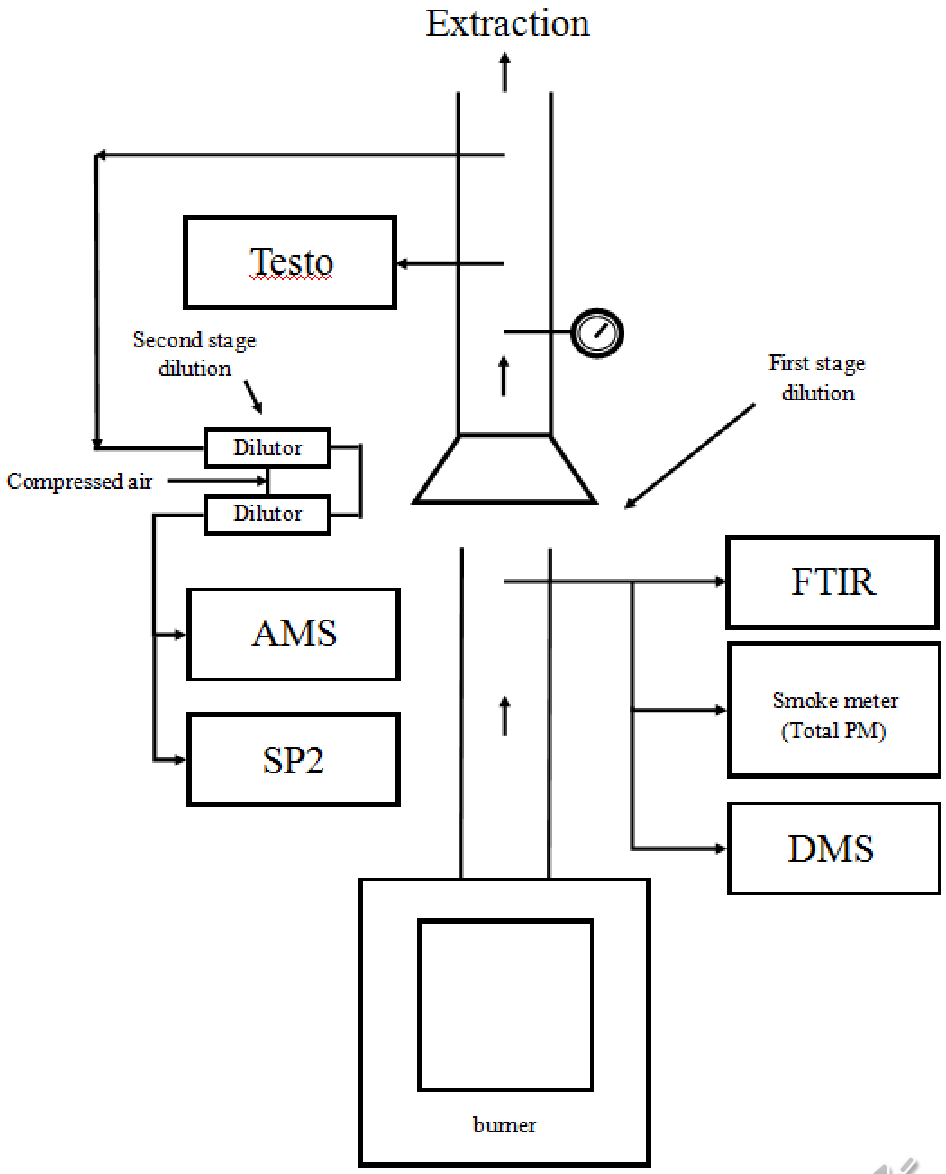
SP2: Physical and optical properties of refractory BC (rBC).

DMS500: Number concentrations and size distribution of particles.

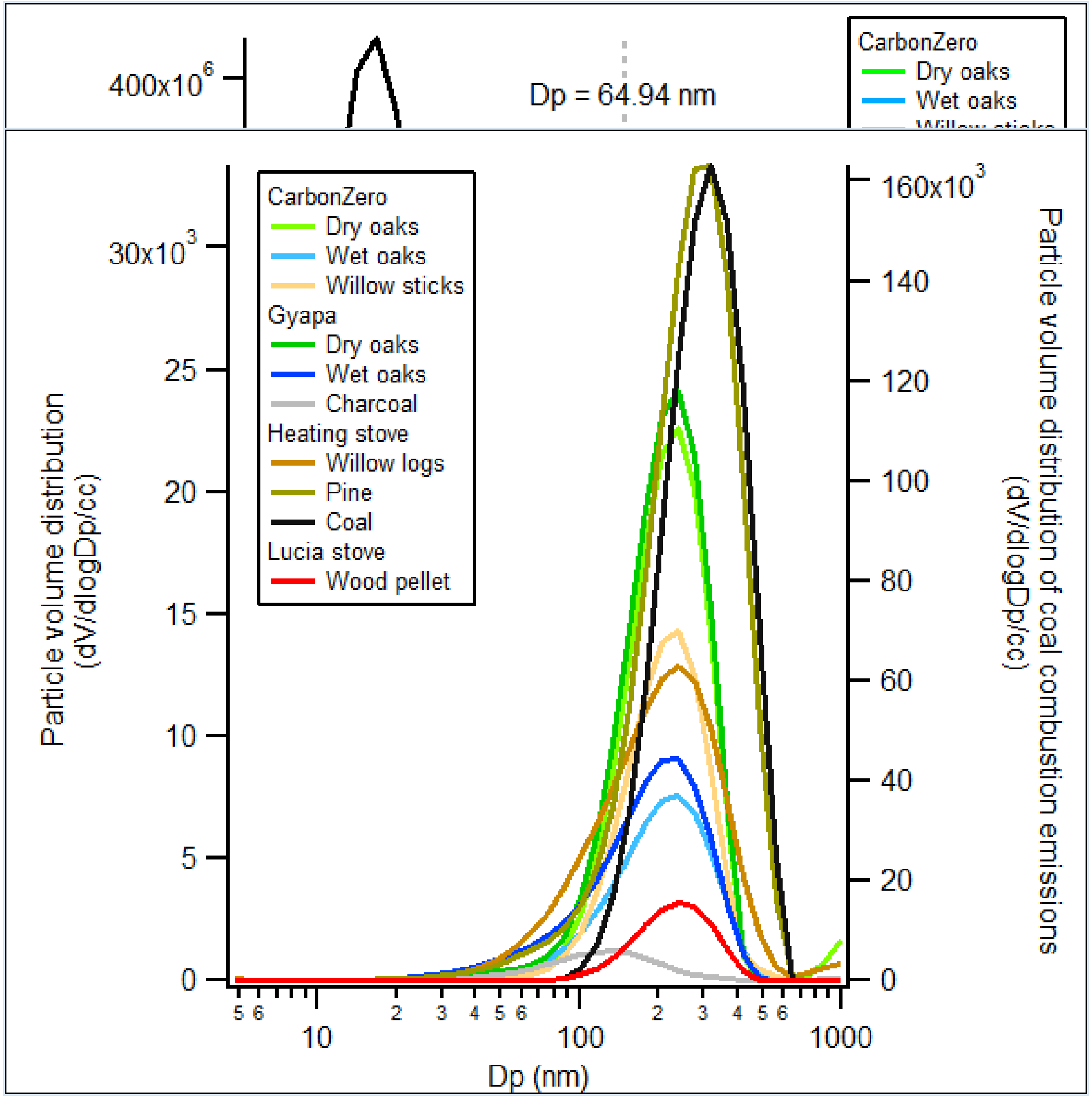
FTIR: H_2O, CO_2, O_2, CO, NO, NO_2, N_2O, NH_3, SO_2, HCL, HF, CH_4, C_2H_6, C_2H_4, C_3H_8, C_6H_{14}, HCN, CHOH, C_6H_6, C_2H_2, furfural and acetic acid.

Testo 340: O_2, CO, NO, NO_x, CO_2 and temperature.

Two Dekati dilutors before the inlet of the AMS and SP2; dilution factors of dilutors were 150 ~ 2000, depending on the burning conditions and one of the dilutor was found to be blocked in the final experiment (coal).



Result- Size distribution



Number size distribution:

Dry oak: 20 and 150 nm; Wet oak: 30 and 110 nm.

Willow stick: 23 and 150 nm; Willow log: 56 nm.

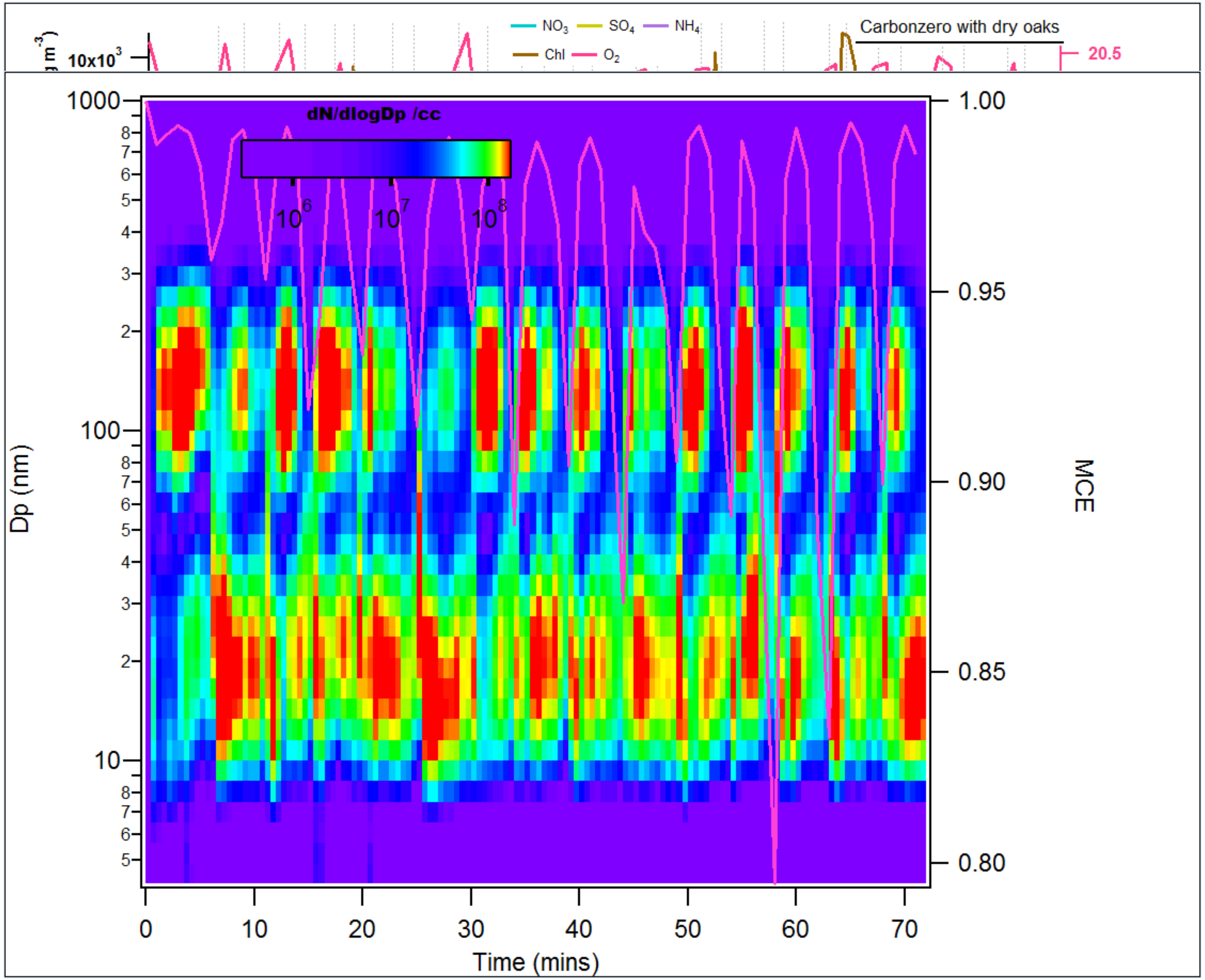
Charcoal: 20-100 nm; Coal: 12 and 200 nm.

Pellet: 12 and 150nm.

Volume size distribution:

Most particle volumes peak at the range of 80 and 700 nm within the detection limits of the AMS and SP2 (80-800 nm).

Result-Combustion process



- **Modified combustion efficiency (MCE):**

  $\Delta CO_2 / (\Delta CO + \Delta CO_2)$

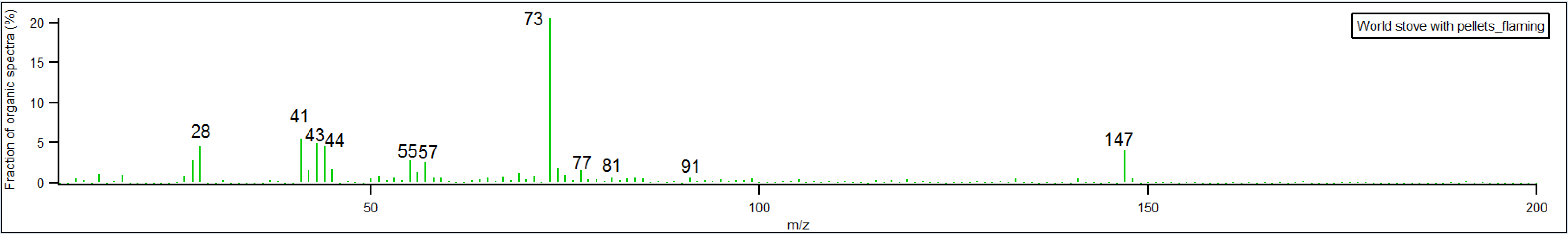
- OM loadings spiked after reloading fuels (pyrolysis) with lower MCE, followed by flaming or poor-burning.

- BC and salt particles (Chl and SO_4, ~ 5%) increased with MCE and temperature.

- Bimodal mode mainly occurred at high MCE.

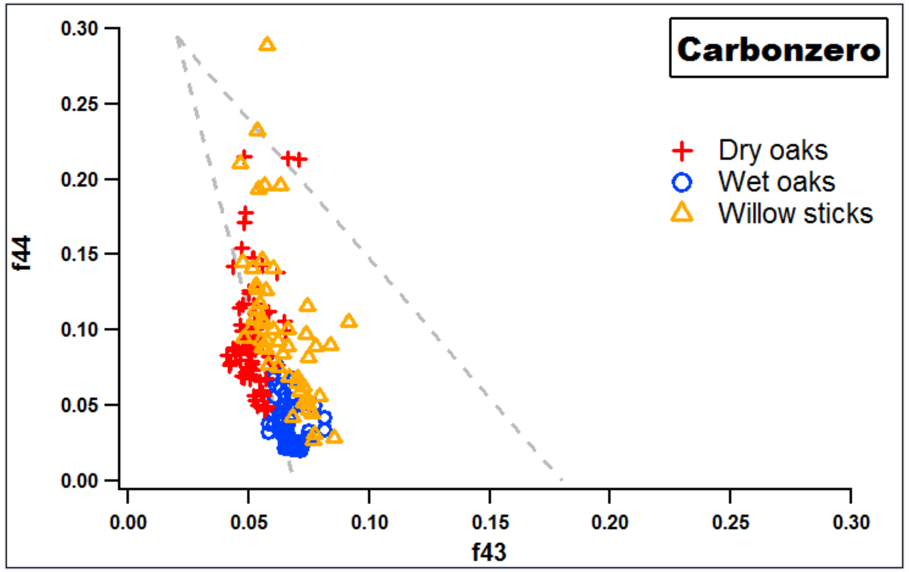
Result- Correction for the Silicone tubing effect

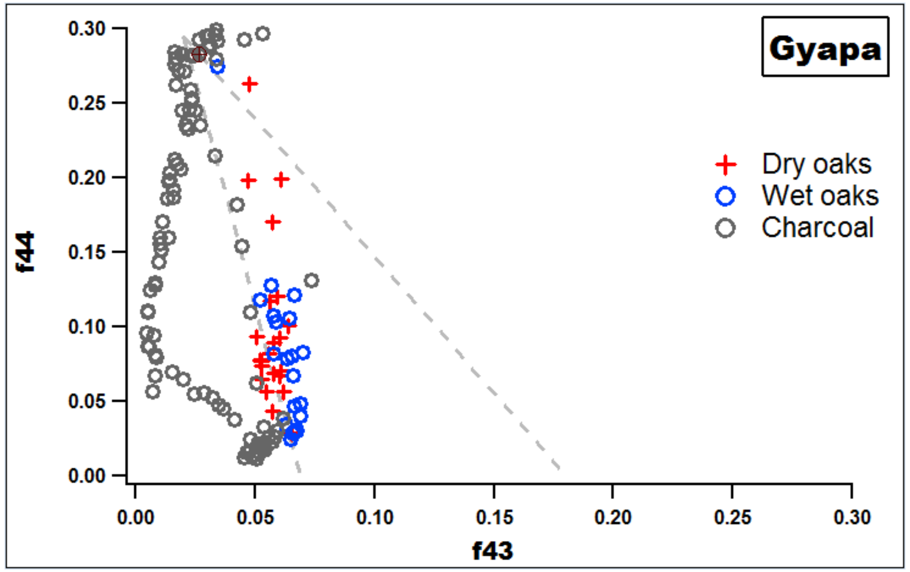
- Signals at m/z 60 and 73 have been considered as marker fragments of Levoglucosan produced from biomass burning (Schneider et al. 2006).
- Signals at m/z 73, 147, 207, 221 and 281 are signatures for siloxane.

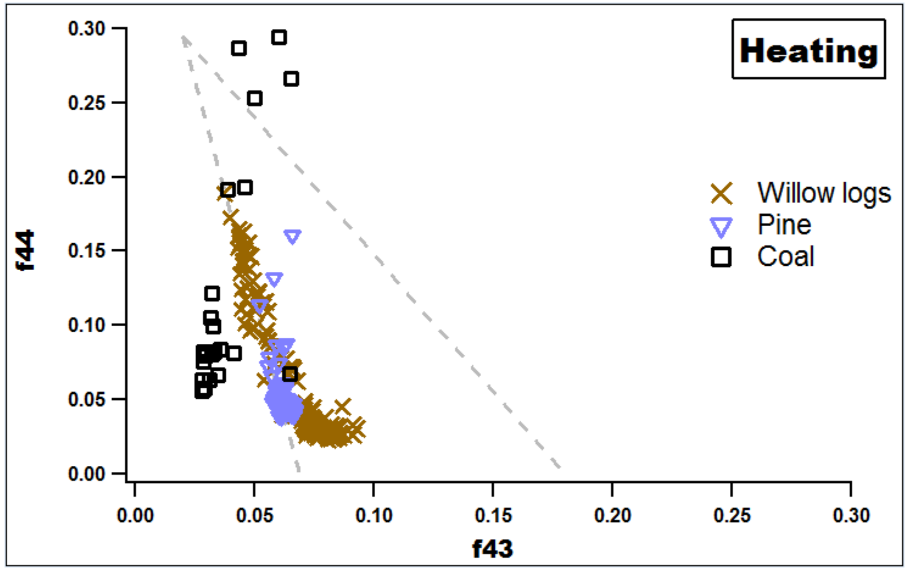


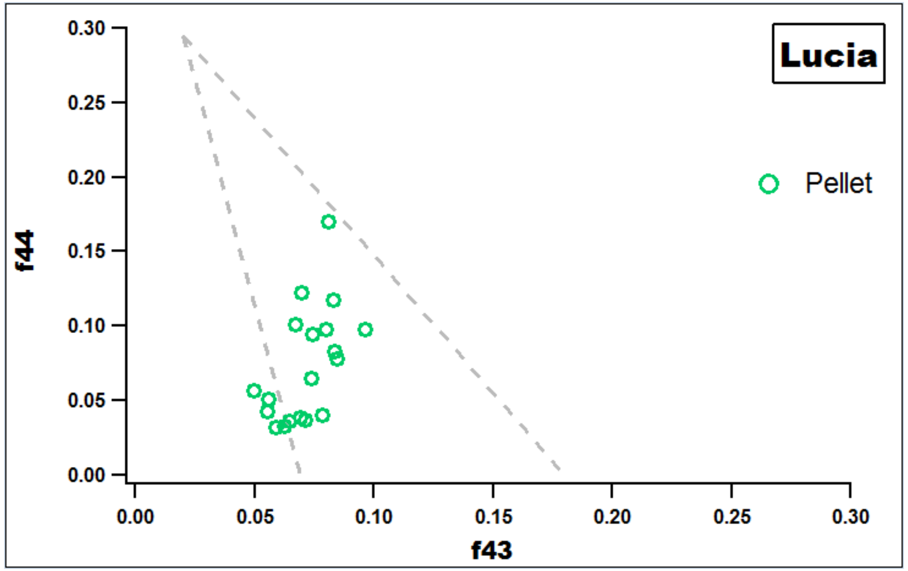
- Ratio of signals at m/z 73 : 147 for pellet in flaming phase = 5.07 : 1.
- m/z 60 : 73 for levoglucosan = 1 : 0.28 (Schneider et al. 2006).

Result- Organic mass spectra (f44 vs f43)

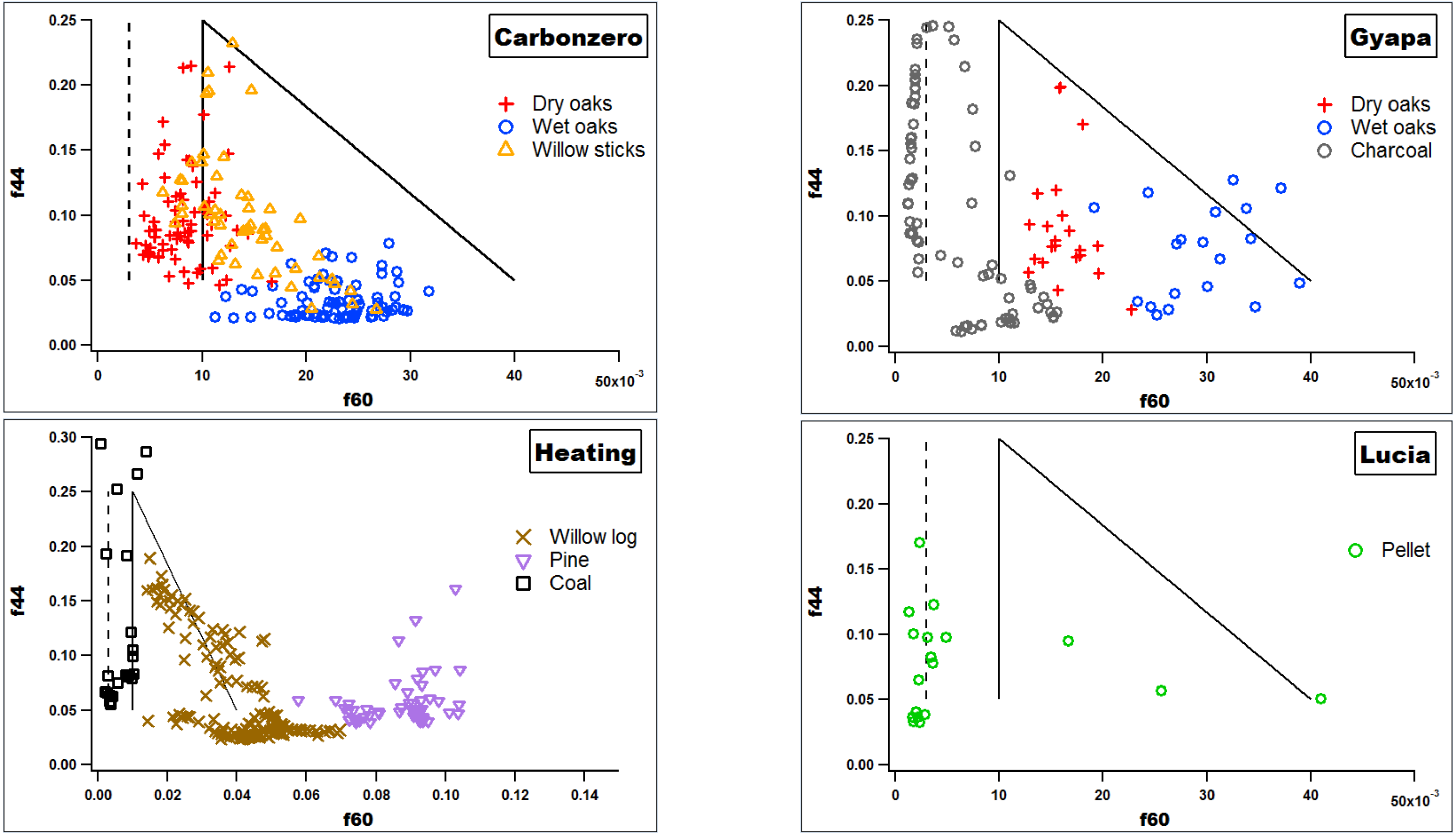




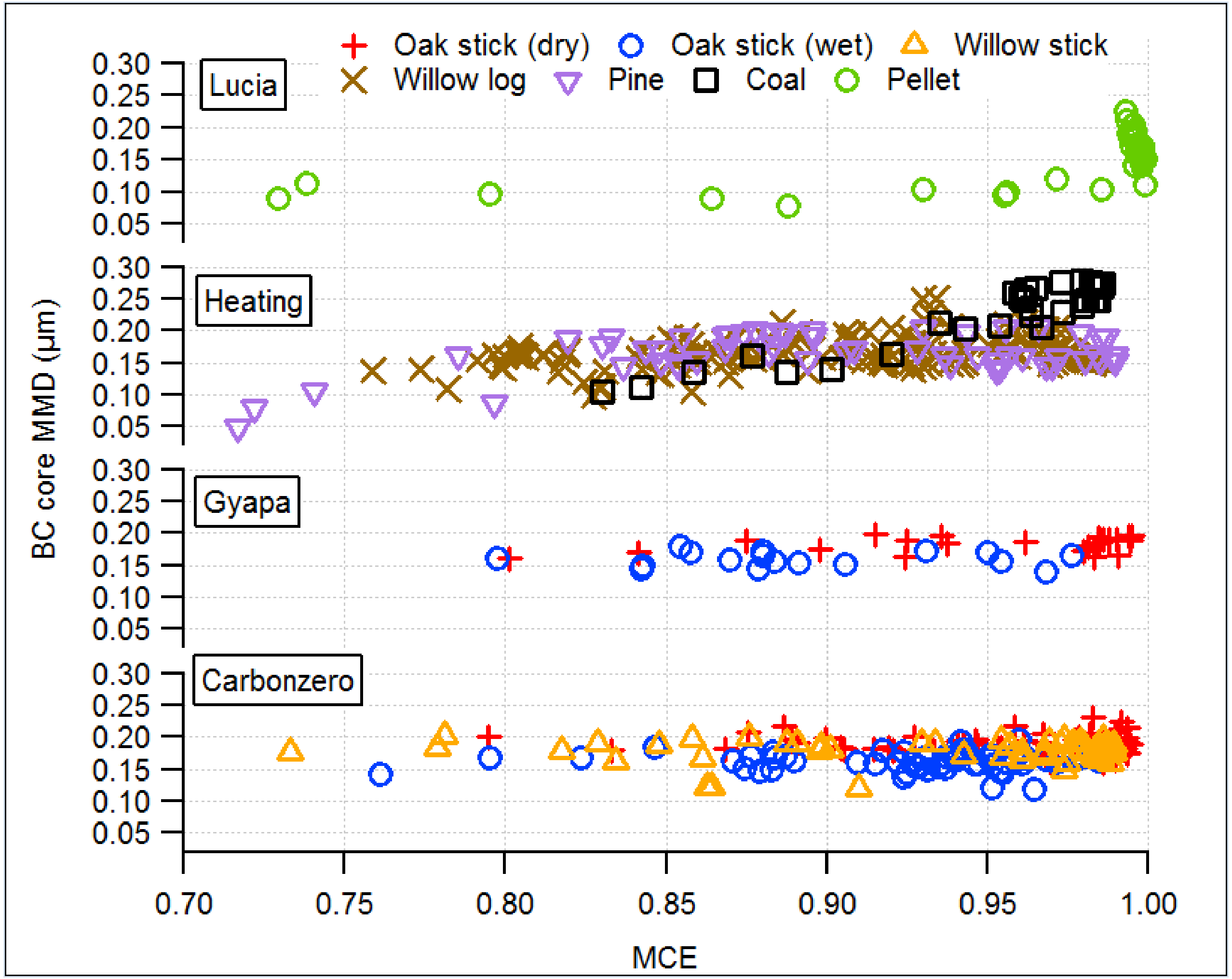




Result- Organic mass spectra (f44 vs f60)



Result- Mixing state of OM and BC



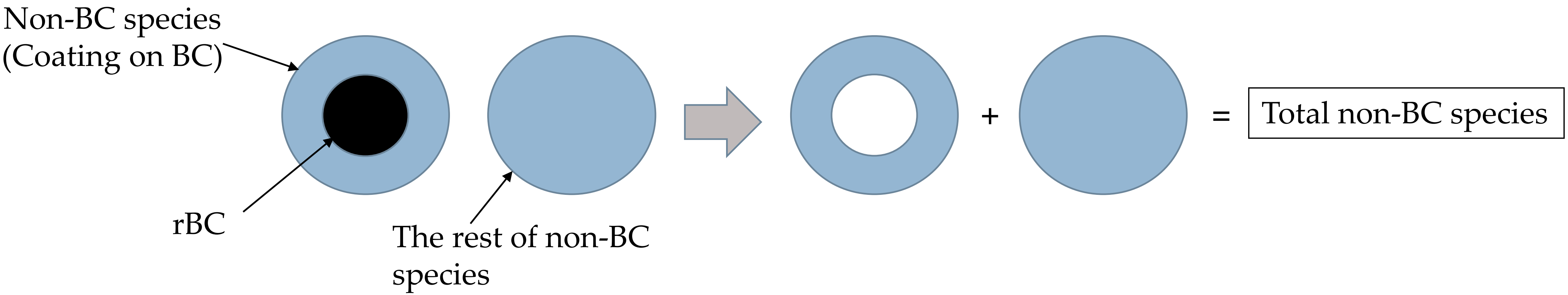
- **Mass median diameter (MMD)** of BC cores has been used as the dominant size of BC (Liu et al. 2014).
- BC core MMD of dry oak and willow stick emissions was of ~ 0.20 µm; wet oak emissions of ~ 0.15 µm (No prominent relationship with MCE).
- BC core MMD from combustion emissions using heating stove correlated with MCE; the largest MMD from the coal emissions was up to ~ 0.25 µm.

Result- Mixing state of OM and BC

- Mass ratios **(MR)** of non-black carbon species to black carbon have an impact on light absorption enhancement (Liu et al. 2017) .

$$M_R = \frac{M_{non-BC}}{M_{rBC}}$$

- $M_{non\text{-}BC}$: The mass of non-BC material in a BC-containing particle (coating).
- M_{rBC}: The mass of refractory BC (rBC).



Result- Mixing state of OM and BC

$$M_{non-BC} = \frac{\left(\left(\frac{D_p}{D_c}\right)^3 - 1\right) * \rho_{OA}}{\rho_{BC}} * BC$$

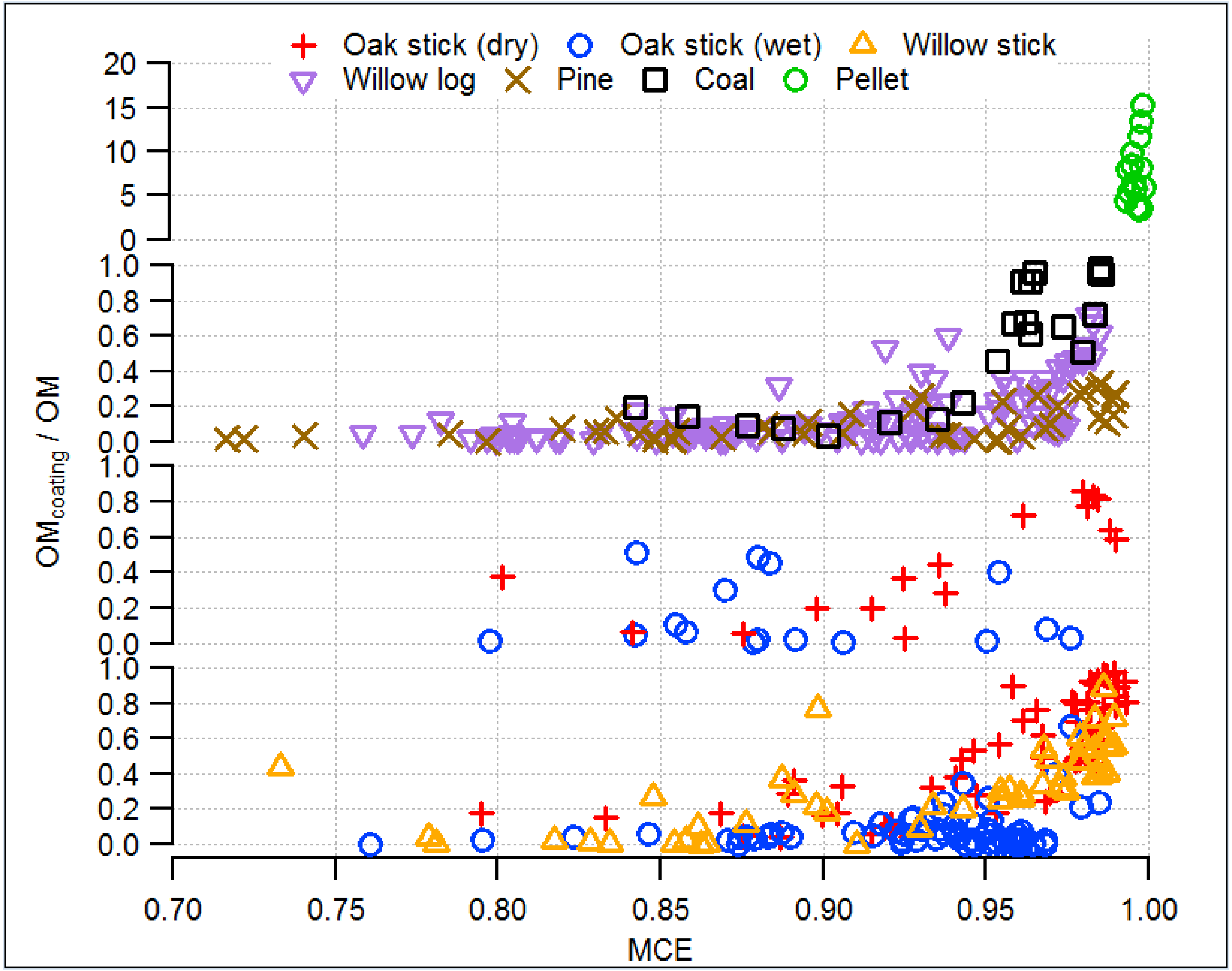
Dp/Dc: Relative coating thickness.

Dp: Coated BC particle diameter; Dc: BC core diameter.

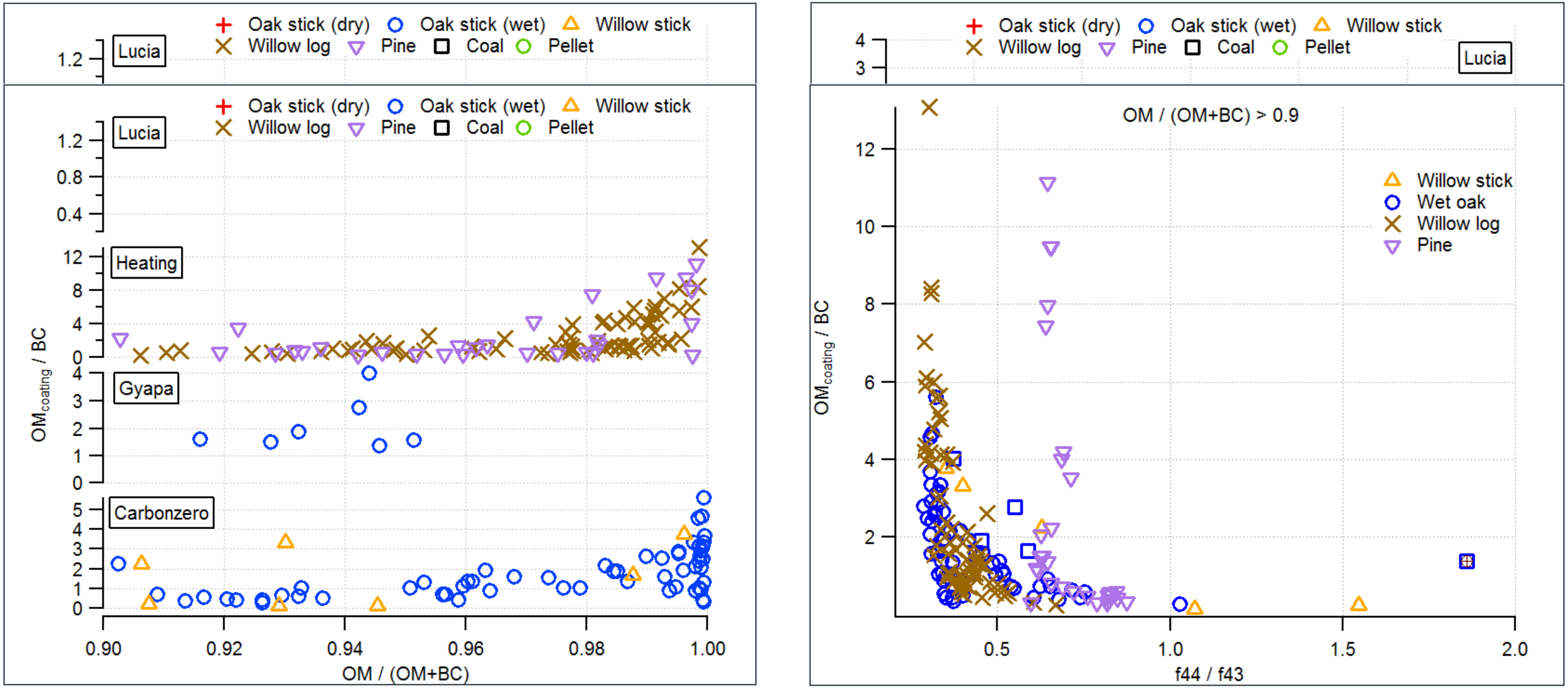
p_{OA} and p_{BC} are the densities of OA and BC assumed to be 1.2 (Turpin et al. 2001) and 1.8 g cm^{-3}

- The technique of leading edge only (LEO) fitting; Refractive index of the particles; Mie theory. Details described in Liu et al. (2014).
- Coatings on BC mainly come from condensation of OOA and SO_4^{2-}.
- $M_{non\text{-}BC}$ are assumed to be totally contributed by OM ($OM_{coating}$) as SO_4^{2-} less than 5% of emissions.

Result- Mixing state of OM and BC



Result- Mixing state of OM and BC



Conclusion

- Most of OM loadings were produced during pyrolysis phase, where as the BC particles were mainly produced during flaming phase after pyrolysis of solid fuels.
- The BC sizes have a wide range between ~ 0.10 and 0.25 µm, depending on the fuel types and MCE.
- MCE<0.9: ~20% of OM coating on BC; MCE>0.9: the OM coatings increase with MCE.
- $OM_{coating}$-to-BC ratios (MR) increased with a large diversity only when OM fractions were above 0.9 (up to 12 from heating stove); $OM_{coating}$-to-BC ratios are less than 1 when OM fractions were less than 0.9.
- Lower oxidation level of OOA (higher f43) facilitates the $OM_{coating}$-to-BC ratios.

Thank you for your attention!!