

## Supplementary Material

### Methodology for the source apportionment of carbons

In brief, the separation of fossil and non-fossil carbon species was calculated using mass concentration and corresponding  $f_c$  values:

For non-fossil-derived OC:

$$OC_{nf} = WSOC_{nf} + WIOC_{nf} \quad (1)$$

$$WIOC_{nf} = WSOC \times f_{c(WIOC)} \quad (2)$$

$$WSOC_{nf} = WSOC \times f_{c(WSOC)} \quad (3)$$

For fossil-derived OC:

$$OC_f = WSOC_f + WIOC_f \quad (4)$$

$$WIOC_f = WSOC \times (1 - f_{c(WIOC)}) \quad (5)$$

$$WSOC_f = WSOC \times (1 - f_{c(WSOC)}) \quad (6)$$

EC is formed directly by incomplete combustion processes of fossil ( $EC_f$ ) and biomass fuels ( $EC_{nf} = EC_{bb}$ ):

$$EC = EC_{nf} + EC_f \quad (7)$$

$$EC_{nf} = EC \times f_{c(EC)} \quad (8)$$

$$EC_f = EC \times (1 - f_{c(EC)}) \quad (9)$$

$f_c$  values were converted from  $f_M$  as follows:  $f_c = f_M / f_{M,ref}$ ;

$f_{M,ref}$  is a reference value representing  $f_M$  for non-fossil carbon sources, which include biogenic and biomass burning emissions. In this study,  $f_c$  values for EC, WSOC and WIOC were  $1.10 \pm 0.05$ ,  $1.08 \pm 0.05$ ,  $1.08 \pm 0.05$  respectively (Zhang *et al.*, 2015; Szidat *et al.*, 2016).

### References

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