# Supplementary Materials:

# Carbonaceous Aerosols in Contrasting Atmospheric Environments in Greek Cities: Evaluation of the EC-tracer Methods for Secondary Organic Carbon Estimation

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**Table 1.** Measurement periods and number of samples at each site in the current study.

| Site/Campaign | Period | # samples |
|---|---|---|
| **Thissio, Athens** | 1/11/2013–2/19/2013 | 60 ($PM_{2.5}$) (day/night) |
| **Ioannina** | 1/12/2013–2/14/2013 | 61 (TSP) (day/night) |
| **Ioannina** | 3/3/2013–10/25/2013 | 20 (TSP) (daily) |
| **Ioannina** | 12/9/2013–2/12/2014 | 51 (TSP) (day/night) |
| **Heraklion** | 1/10/2013–2/7/2013 | 55 (TSP) (day/night) |

**Table 2.** Pearson correlation coefficients (r) from the linear regressions of TSP, OC and EC concentrations with meteorological variables in Ioannina.

| | Temp. | Temp.$_{min}$ | Wind Speed | Rainfall | Rainfall (>0.2 mm) |
|---|---|---|---|---|---|
| **TSP** | −0.29 | −0.63 | −0.30 | −0.44 | −0.57 |
| **OC** | −0.59 | −0.66 | −0.54 | −0.35 | −0.40 |
| **EC** | −0.39 | −0.58 | −0.35 | −0.31 | −0.42 |

**Table 3.** Pearson correlation coefficient (r) matrix between carbonaceous aerosols and ionic species in Ioannina. Values in boldface correspond to statistically significant correlations at the 95% confidence level. .

|  | EC | Cl$^-$ | NO$_3^-$ | SO$_4^{-2}$ | C$_2$O$_4^{-2}$ | Na$^+$ | NH$_4^+$ | K$^+$ | Mg$^{+2}$ | Ca$^{+2}$ |
|---|---|---|---|---|---|---|---|---|---|---|
| OC | **0.83** | −0.01 | **0.64** | **0.57** | **0.34** | −0.19 | **0.50** | **0.87** | −0.03 | **0.30** |
| EC |  | −0.07 | **0.66** | **0.59** | **0.34** | **−0.22** | **0.43** | **0.74** | −0.05 | **0.49** |
| Cl$^-$ |  |  | 0.01 | **0.23** | **0.22** | **0.75** | −0.01 | 0.08 | **0.65** | 0.12 |
| NO$_3^-$ |  |  |  | **0.68** | **0.69** | −0.02 | **0.63** | **0.57** | −0.04 | **0.52** |
| SO$_4^{-2}$ |  |  |  |  | **0.57** | 0.16 | **0.64** | **0.59** | 0.13 | **0.53** |
| C$_2$O$_4^{-2}$ |  |  |  |  |  | 0.08 | **0.41** | 0.28 | **0.22** | **0.37** |
| Na$^+$ |  |  |  |  |  |  | −0.08 | −0.06 | **0.52** | 0.12 |
| NH$_4^+$ |  |  |  |  |  |  |  | **0.56** | −0.18 | 0.14 |
| K$^+$ |  |  |  |  |  |  |  |  | −0.07 | 0.25 |
| Mg$^{+2}$ |  |  |  |  |  |  |  |  |  | **0.37** |

**Table 4.** Same as in Suppl. Table 3, but for Heraklion.

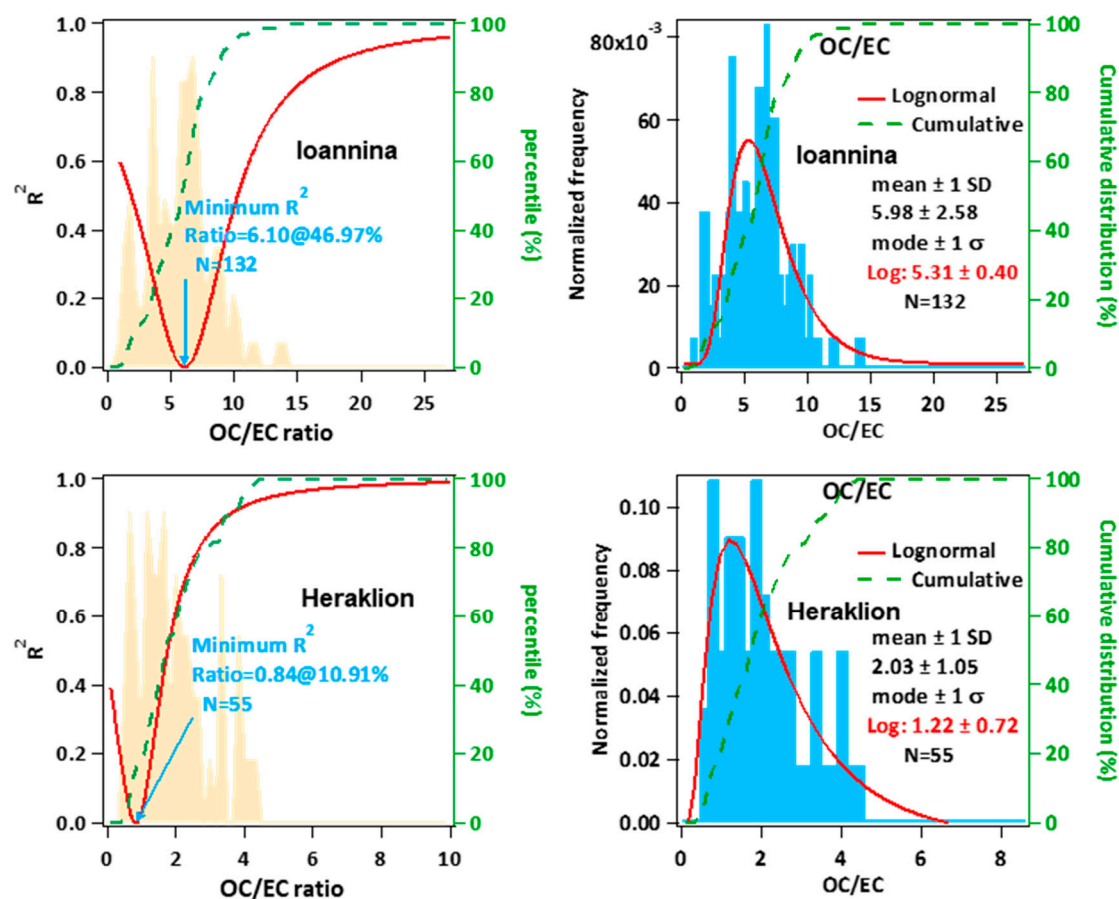|  | EC | Cl$^-$ | NO$_3^-$ | SO$_4^{2-}$ | C$_2$O$_4^{2-}$ | Na$^+$ | NH$_4^+$ | K$^+$ | Mg$^{2+}$ | Ca$^{2+}$ |
|---|---|---|---|---|---|---|---|---|---|---|
| OC | **0.67** | −0.14 | −0.06 | 0.18 | **0.44** | −0.23 | **0.36** | **0.50** | −0.17 | 0.07 |
| EC |  | 0.11 | −0.02 | 0.21 | **0.72** | −0.03 | 0.12 | **0.28** | 0.23 | **0.63** |
| Cl$^-$ |  |  | 0.11 | 0.26 | **0.37** | **0.96** | −0.18 | 0.02 | **0.92** | **0.43** |
| NO$_3^-$ |  |  |  | **0.76** | 0.17 | 0.16 | **0.48** | 0.07 | 0.16 | 0.07 |
| SO$_4^{2-}$ |  |  |  |  | **0.43** | 0.25 | **0.66** | 0.16 | 0.25 | 0.14 |
| C$_2$O$_4^{2-}$ |  |  |  |  |  | 0.26 | 0.16 | **0.31** | **0.45** | **0.58** |
| Na$^+$ |  |  |  |  |  |  | −0.21 | 0.02 | **0.87** | **0.30** |
| NH$_4^+$ |  |  |  |  |  |  |  | 0.18 | −0.25 | **−0.29** |
| K$^+$ |  |  |  |  |  |  |  |  | −0.08 | −0.01 |
| Mg$^{2+}$ |  |  |  |  |  |  |  |  |  | **0.61** |

**Figure 1.** Application of the MRS method for Ioannina and Heraklion. At Ioannina, $(OC/EC)_{pri}$=6.10; @46.97% means that ~53% of data would have positive SOC, while in Heraklion 89% of the data have positive SOC values. Lognormal curves are fitted in the OC/EC frequency distributions.
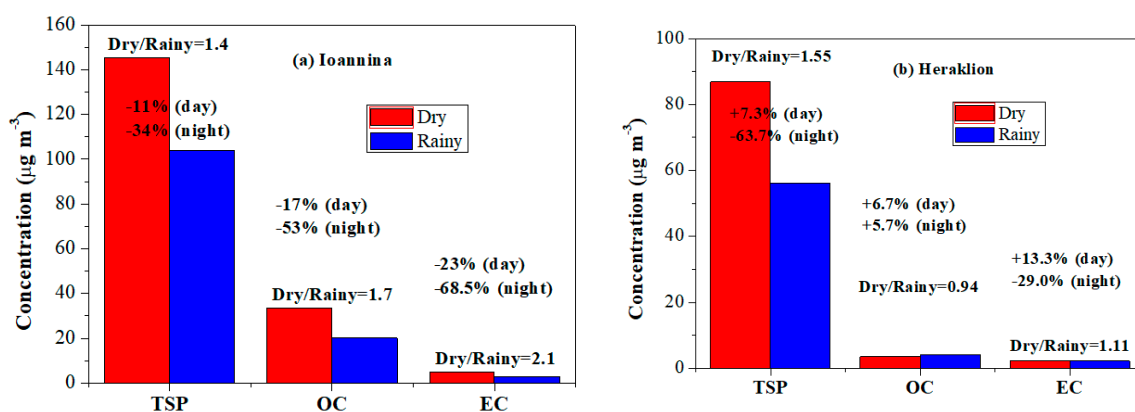


**Figure 2.** TSP, OC, EC concentrations and their ratios during dry and rainy days in Ioannina (**a**) and Heraklion (**b**). The percentage changes due to rainfall are also given for day and night samples.
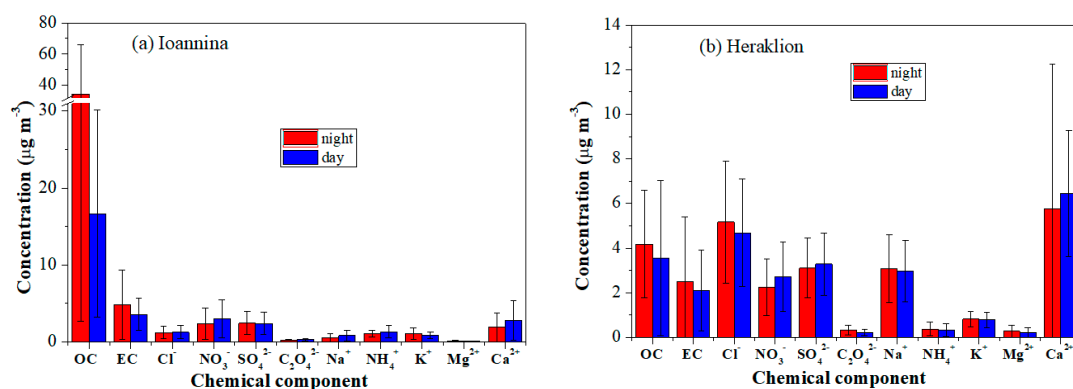
**Figure 3.** Day vs night concentrations of the carbonaceous-aerosol and inorganic species (in TSP) in Ioannina (**a**) and Heraklion (**b**).
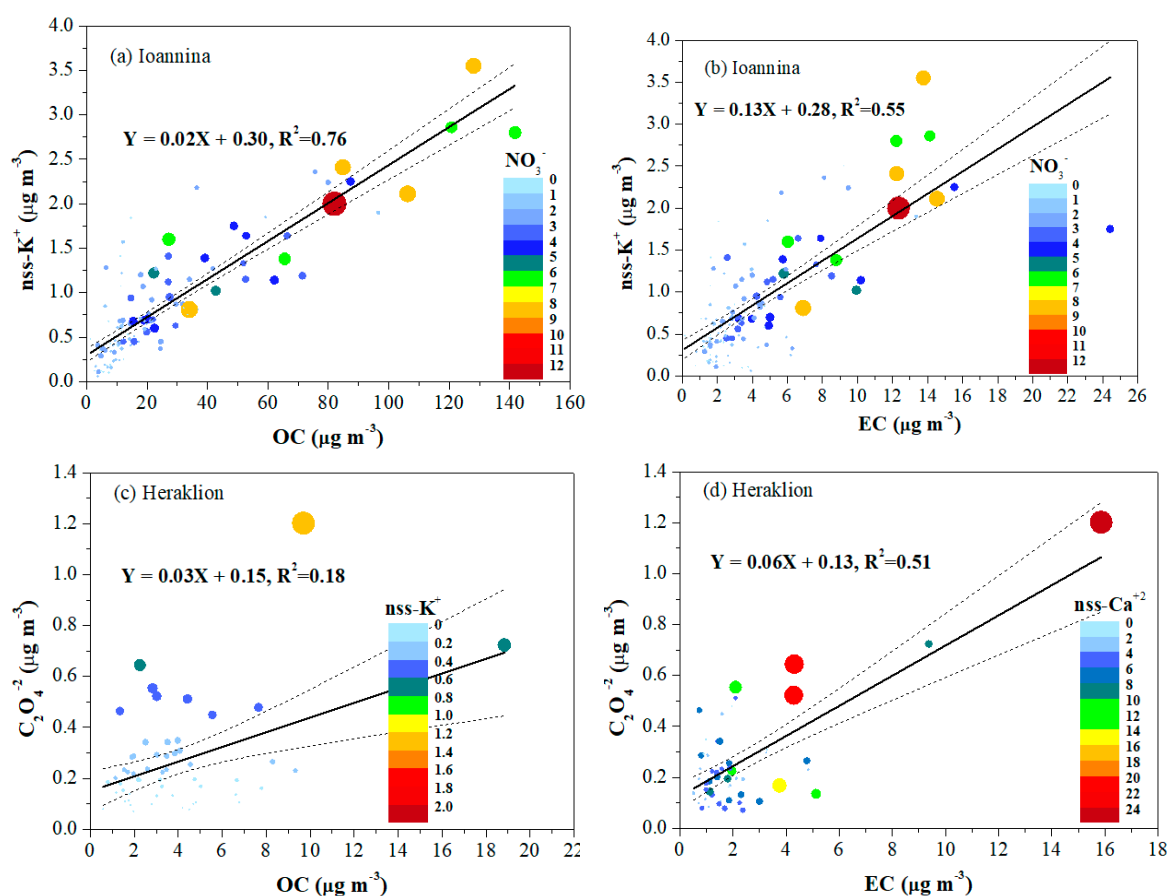


**Figure 4.** Correlations between OC, EC concentrations and nss-K$^+$, C$_2$O$_4^{2-}$ in Ioannina (**a,b**) and Heraklion (**c,d**).
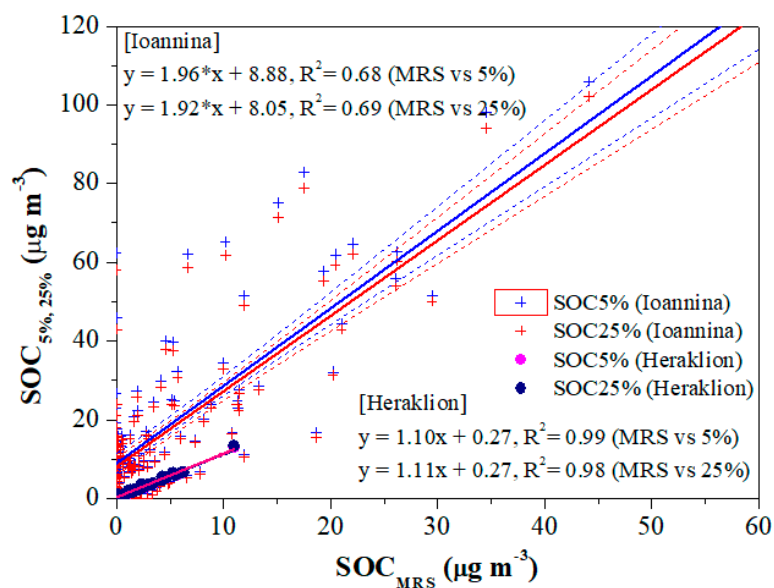
**Figure 5.** Correlations between the estimated SOC concentrations obtained from the MRS and percentile (5%, 25%) methods in Ioannina and Heraklion.
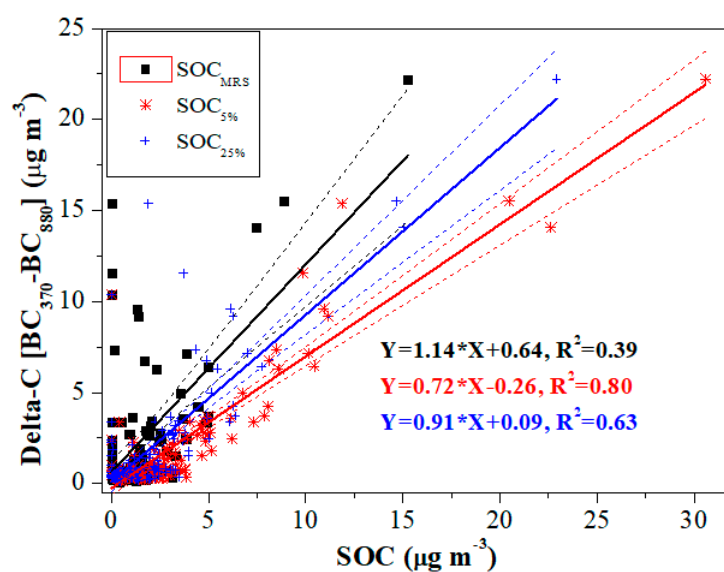


**Figure 6.** Correlation between the SOC estimates from the various SOC estimation methods and the Delta-C values derived from Aethalometer (AE-33) measurements in Athens during December 2016–January 2017. .