A Preliminary Assessment of the Impacts of Multiple Temporal-scale Variations in Particulate Matter on its Source Apportionment

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Evaluating the SOC

Secondary organic carbon (SOC) concentrations were estimated using the OC/EC ratio, as follows:

$$OC_{pri} = EC \times (OC / EC)_{pri} \tag{S1}$$

$$SOC = OC_t - EC \times (OC / EC)_{min} \tag{S2}$$

where OC_{pri} is primary OC, OC_t is total OC, and $(OC / EC)_{pri}$ is the OC/EC ratio of primary aerosol. Because $(OC / EC)_{pri}$ is difficult to estimate, it was replaced by the minimum ratio of OC/EC ($(OC / EC)_{min}$), according to the suggestion proposed by Castro et al. (1999).

Figures

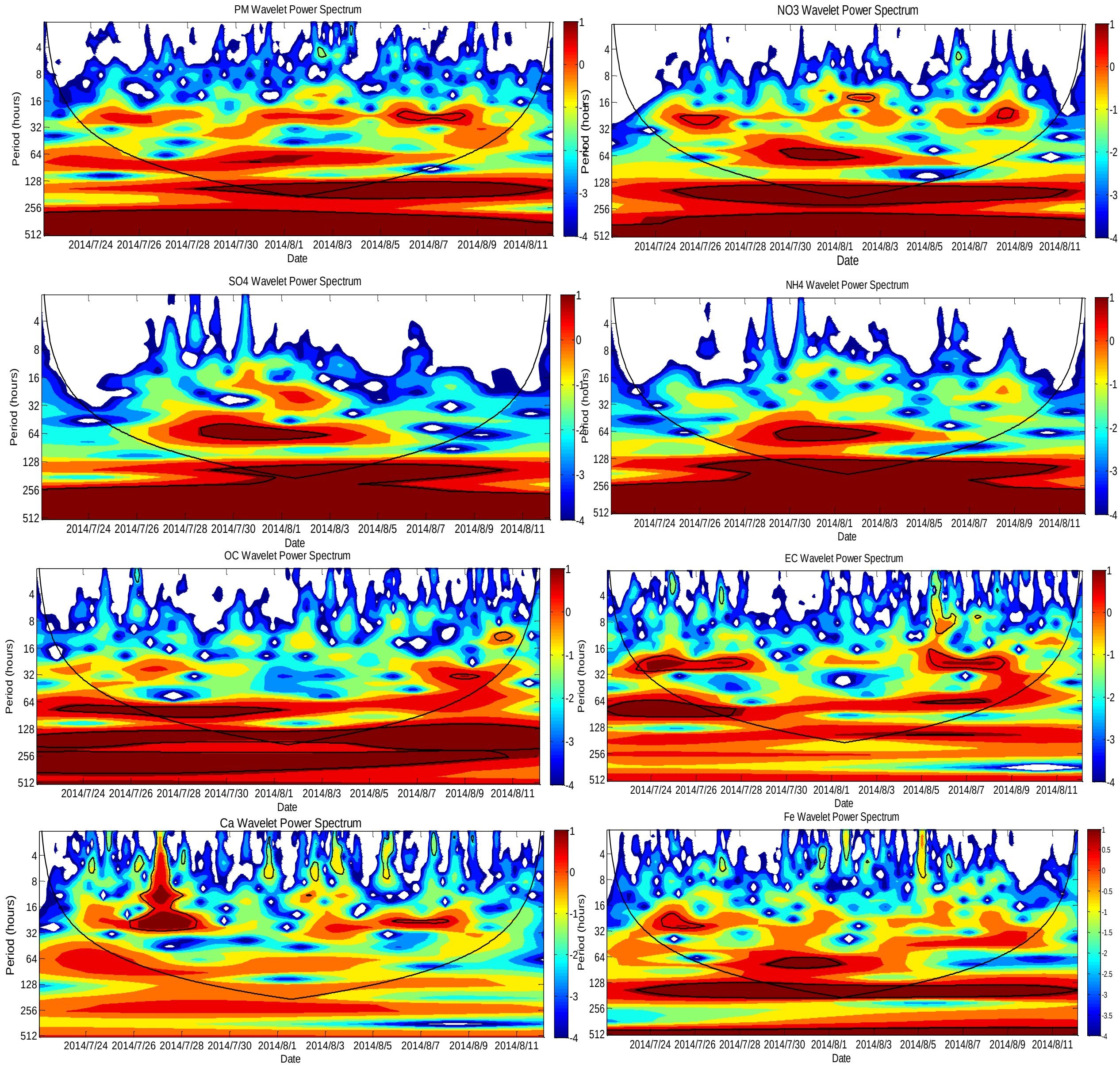

Figure S1. The wavelet variance diagrams of $PM_{2.5}$ and chemical species from original data.

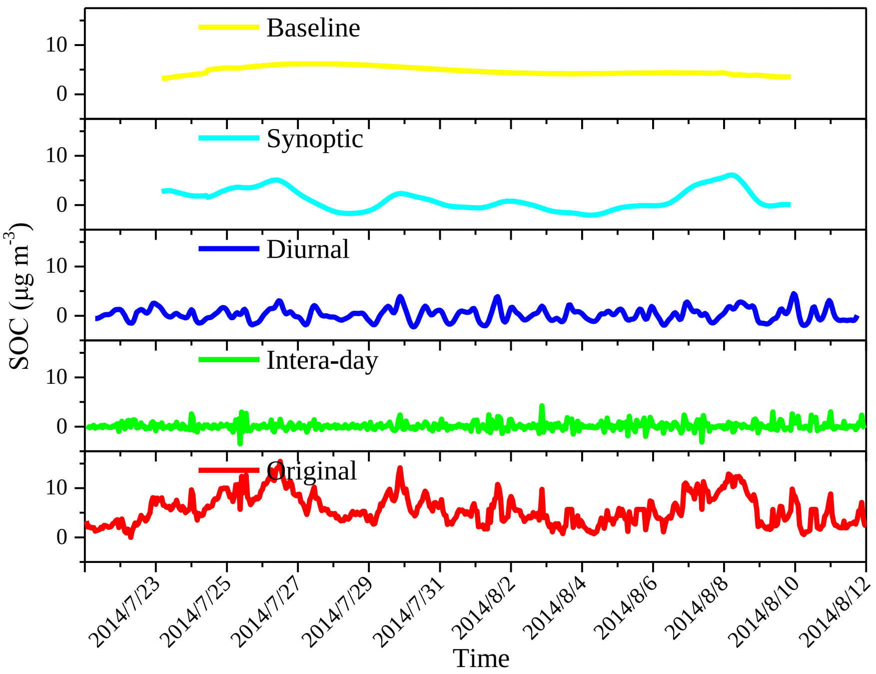

Figure S2. Variance and level of SOC concentrations influenced by intra-day (time period less than 12 h), diurnal (12-24 h), synoptic (2-21 days), and baseline (greater than 21 days) temporal-scale (TS) components, for the period of 22 July 2014 to 13 Aug 2014 at Beijing, China.

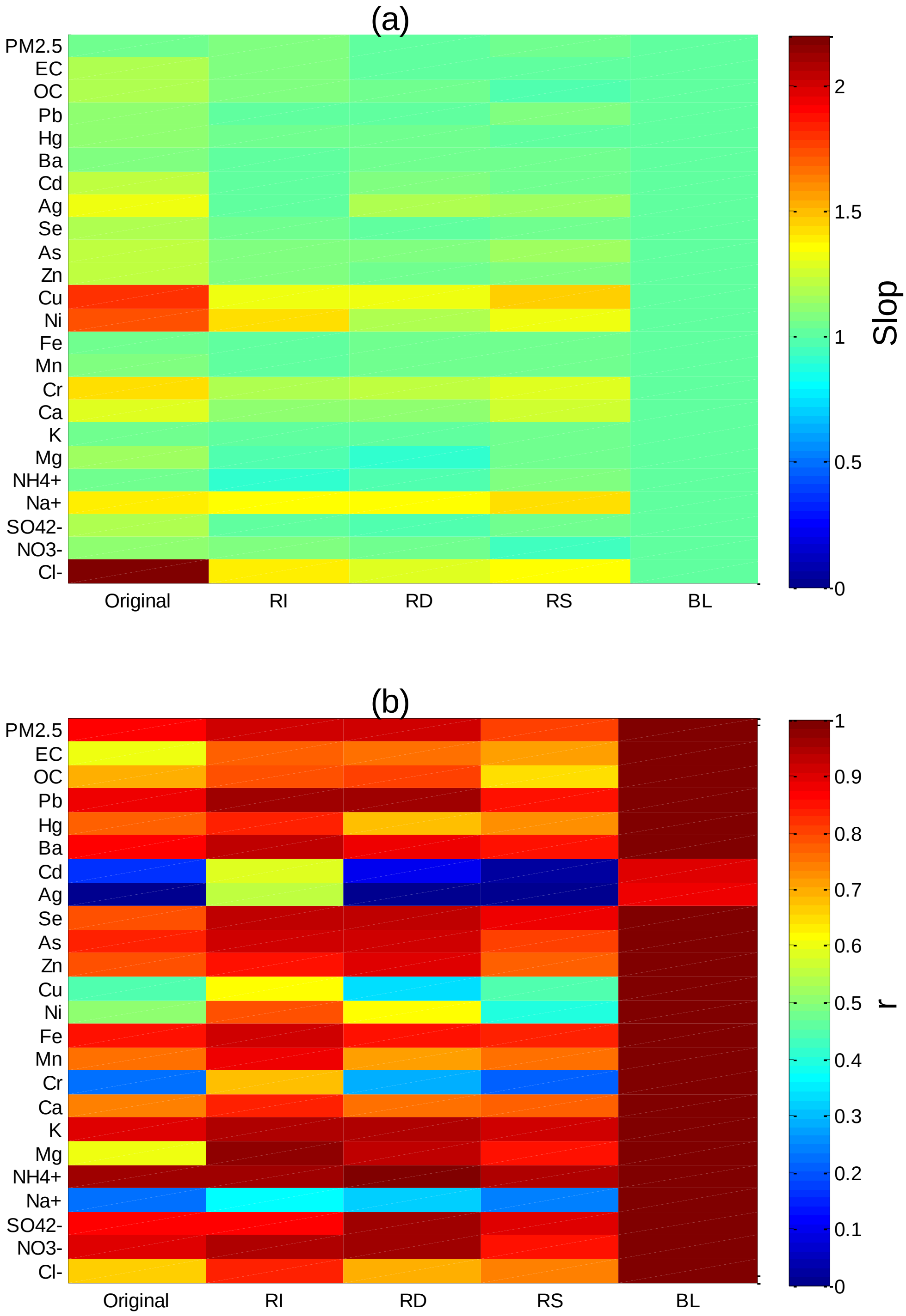

Figure S3. The performance of ME-2 from five datasets. (a): the slops between model and measured concentrations of chemical species and $PM_{2.5}$. (RI: intra-day removed dataset, RD: diurnal removed dataset, RS: synoptic removed dataset, RBL: baseline removed dataset) (b)The correlation coefficients between modeled and measured concentrations of chemical species and $PM_{2.5}$.

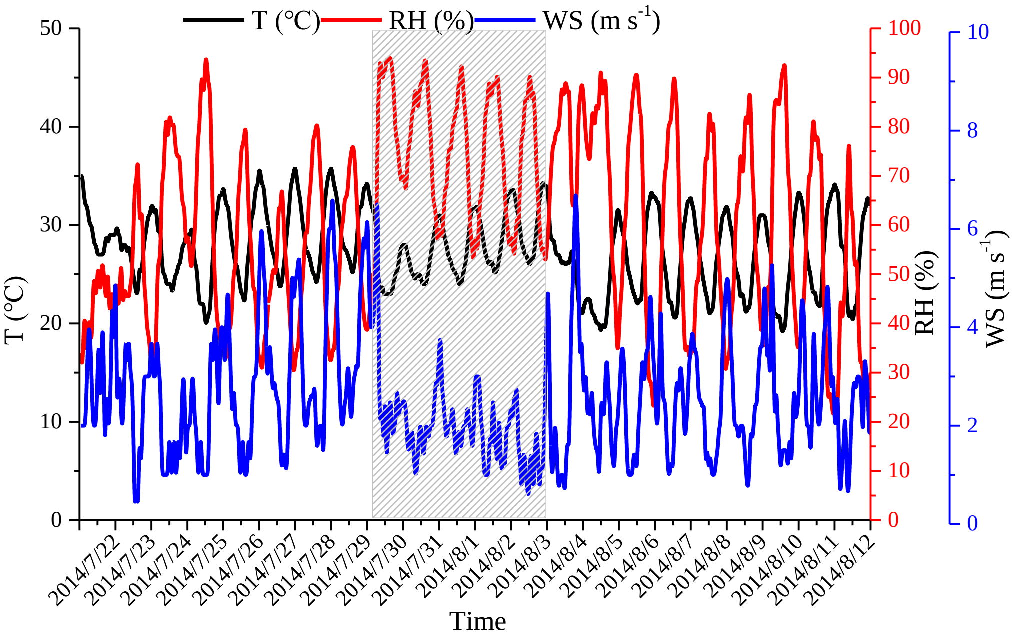

Figure S4. Time series of the temperature, RH and wind speed. The vertical gray lines demarcate the heavy pollution period.

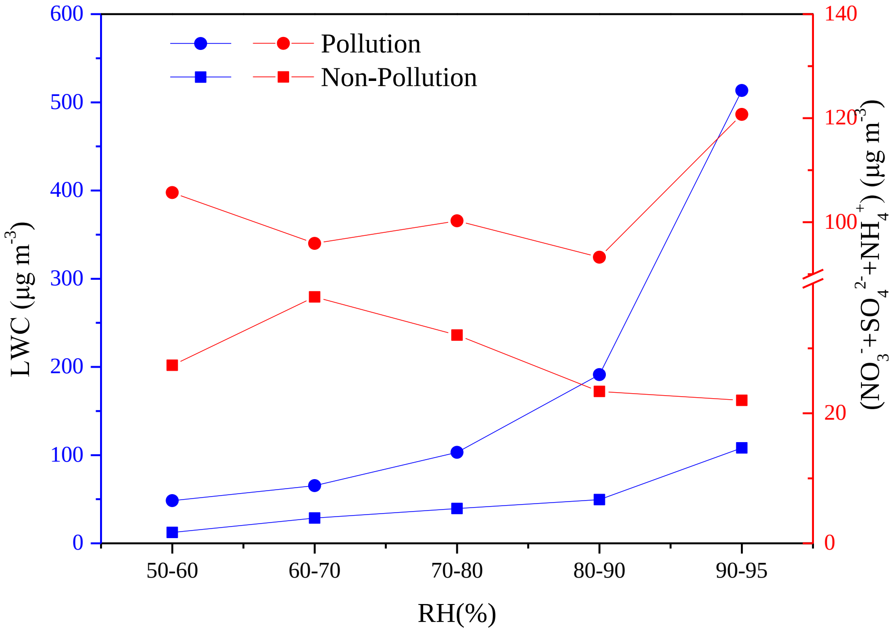

Figure S5. The total ions (NO_3^-+SO_4^{2-}+NH_4^+) and LWC concentrations depend on RH during the pollution and non-pollution periods.

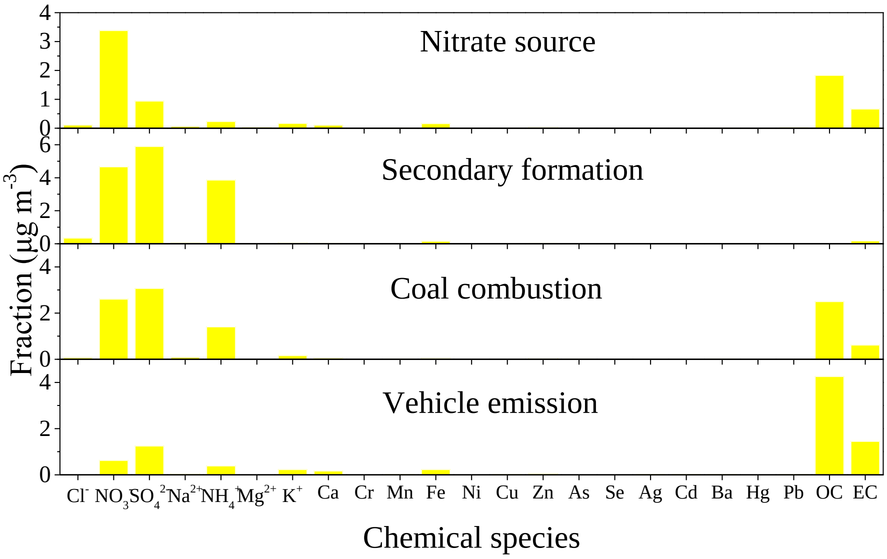

Figure S6. The factor profiles obtained from ME-2 from baseline dataset.

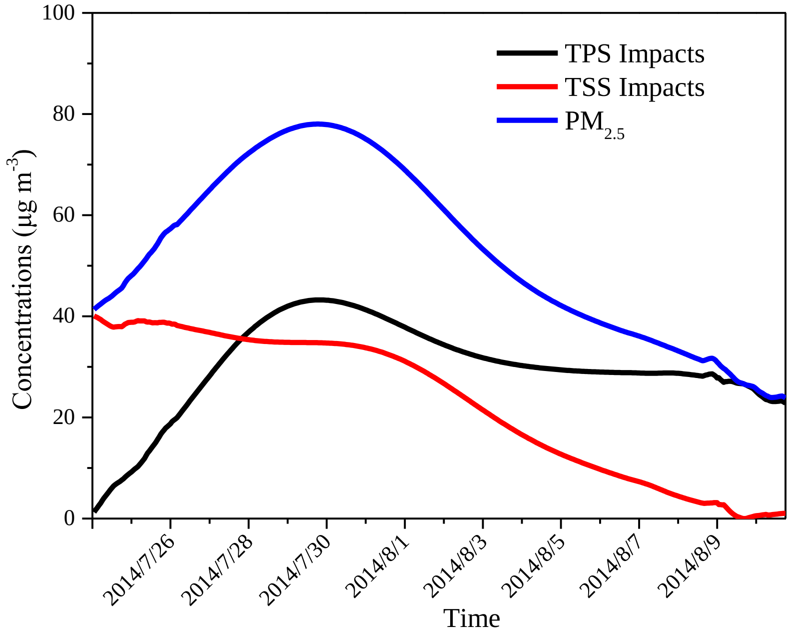

Figure S7. TPS, TSS impacts and $PM_{2.5}$ concentration from baseline dataset. (TPS: the total contributions of vehicle emission and coal combustion. TSS: the total contributions of secondary formation and nitrate source).

Tables

Table S1. Average absolute error (AAE) between original and RI, RD, RS, and RBL for $PM_{2.5}$, seven species and SOC.

	RI[a]	RD[b]	RS[c]	RBL[d]
$PM_{2.5}$	18	40	63	149
OC	8	15	23	100
EC	10	20	20	106
NO_3^-	12	57	77	149
SO_4^+	7	30	79	127
NH_4^+	9	41	108	148
Ca	20	38	24	124
Fe	12	30	31	116
SOC	11	21	31	102

[a]RI: intra-day removed dataset. [b]RD: diurnal removed dataset. [c]RS: synoptic removed dataset. [d]RBL: baseline removed dataset.

Table S2. Correlation coefficients between original and RI, RD, RS, and RBL for $PM_{2.5}$, seven species and SOC.

	RI	RD	RS	RBL
$PM_{2.5}$	0.96**	0.85**	0.88**	0.49**
NO_3^-	0.98**	0.86**	0.88**	0.51**
SO_4^+	0.99**	0.95**	0.88**	0.73**
NH_4^+	0.99**	0.95**	0.85**	0.69**
OC	0.97**	0.92**	0.77**	0.31**
EC	0.94**	0.84**	0.90**	0.19**
Ca	0.88**	0.88**	0.97**	0.26**
Fe	0.92**	0.87**	0.86**	0.34**
SOC	**0.96****	**0.92****	**0.72****	**0.30****

** Significant correlation at 0.01 level.

Table S3. The Q values of each solution obtained by ME-2.

	Q	Q_{the}
Original	12593	9940
RI	10732	9353
RD	9873	9712
RS	10847	8002

Table S4. The results obtained from PCA from RBL dataset.

Components	Factor 1	Factor 2	Factor 3	Factor 4	Factor 5	Factor 6	Factor 7
Cl^-	0.09	0.12	0.41	**0.67**	0.30	0.02	-0.04
NO_3^-	0.13	0.23	**0.80**	0.33	0.17	0.04	-0.08
SO_4^{2-}	0.22	0.03	**0.86**	-0.01	-0.03	0.04	0.14
Na^+	-0.01	0.00	0.07	-0.01	0.01	0.02	**0.93**
NH_4^+	0.18	0.12	**0.92**	0.22	0.12	0.05	0.05
Mg^{2+}	0.22	0.30	0.14	0.52	0.02	-0.06	0.33
K	**0.87**	0.38	0.06	0.00	-0.09	-0.05	0.00
Ca	0.18	**0.83**	-0.05	0.17	-0.11	0.00	0.11
Cr	-0.03	0.08	0.02	0.04	**0.92**	-0.04	0.03
Mn	0.57	0.58	0.19	0.05	0.16	-0.09	-0.10
Fe	0.52	0.58	0.31	0.14	0.40	-0.05	-0.09
Ni	-0.15	-0.01	0.16	0.13	**0.91**	0.04	-0.01
Cu	0.17	0.12	0.15	**0.76**	0.12	0.03	0.01
Zn	0.67	0.16	0.16	0.39	-0.18	-0.02	0.18
As	**0.79**	0.12	0.02	0.37	-0.11	0.00	0.00
Se	**0.85**	0.11	0.38	-0.05	-0.06	-0.07	-0.02
Ag	-0.06	-0.03	0.03	0.00	0.02	**0.97**	-0.01
Cd	-0.02	-0.01	0.07	0.04	-0.02	**0.97**	0.02
Ba	0.19	**0.87**	0.22	0.21	0.15	0.00	0.03
Hg	0.37	0.54	0.28	0.33	0.08	-0.11	-0.03
Pb	**0.86**	0.15	0.18	0.31	0.06	0.02	0.01
OC	0.44	0.35	-0.03	0.56	-0.09	0.04	-0.14
EC	0.15	0.58	0.05	0.59	-0.03	0.10	-0.09
Variance contribution (%)	19.60	14.25	12.70	11.91	9.18	8.45	4.82

Table S5. Correlation coefficients (426 samples) between original and intra-day removed dataset, diurnal removed dataset, synoptic removed dataset for source contributions.

	RI	RD	RS
Crustal dust[a]	**0.68****	-	-
Vehicle emission	**0.45****	0.51**	0.25**
Coal combustion	0.82**	**0.77****	0.74**
Secondary formation	0.96**	0.91**	**0.53****
Nitrate source[b]	-	0.63**	0.32**

[a]Only ME-2 from original and RI datasets identified the crustal dust. [b]ME-2 from original dataset did not identify the correlation coefficients between RI and RD, and RS for the nitrate sources. **Significant correlation at 0.01 level.

References

Castro, L. M., Pio, C. A., Harrison, R. M., and Smith, D. J. T.: Carbonaceous aerosol in urban and rural European atmospheres: estimation of secondary organic carbon concentrations. Atmos. Environ., 33, 2771-2781, doi: 10.1016/S1352-2310(98)00331-8, 1999.