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Characteristics of brown carbon in the urban Po Valley atmosphere

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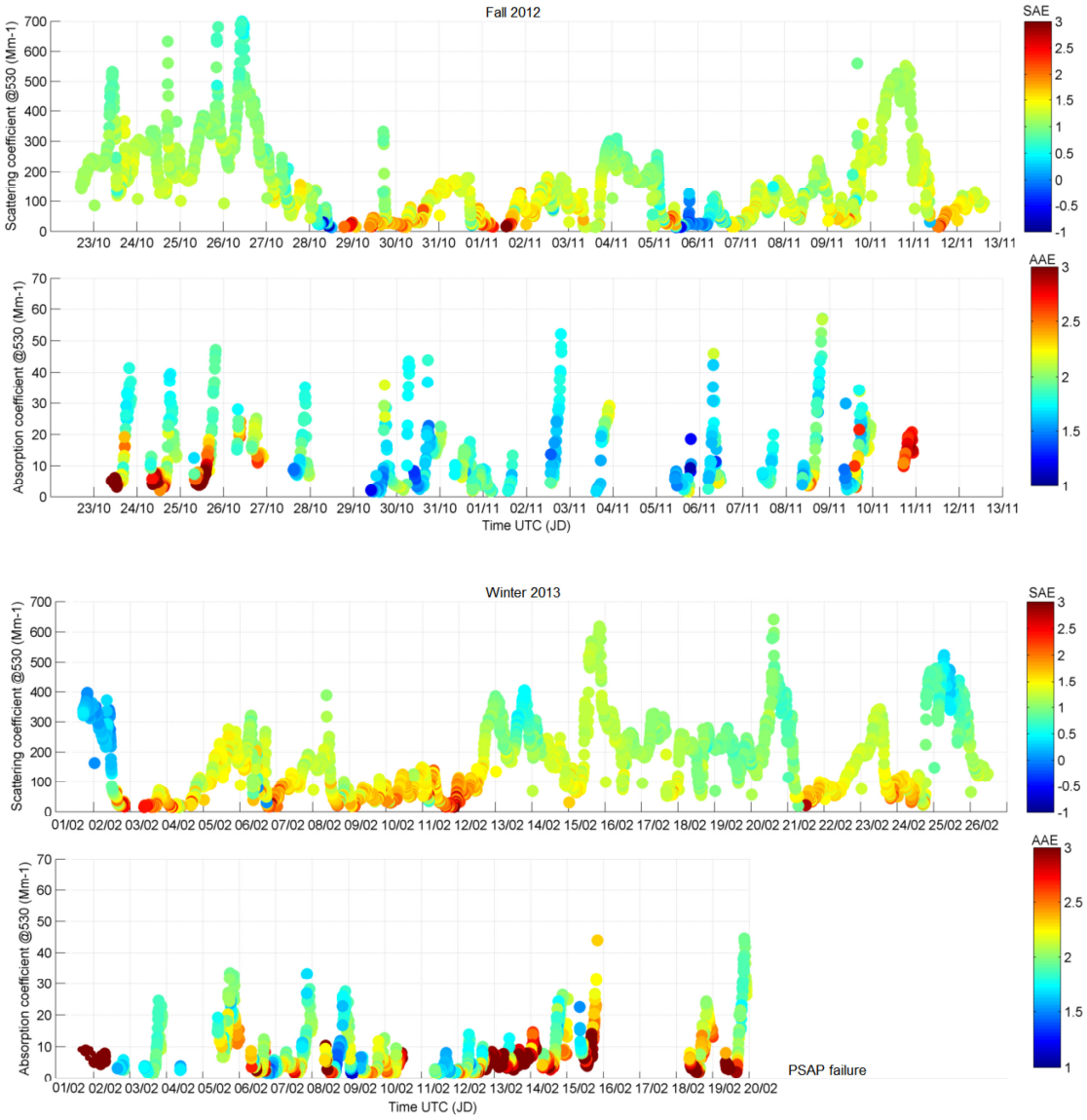
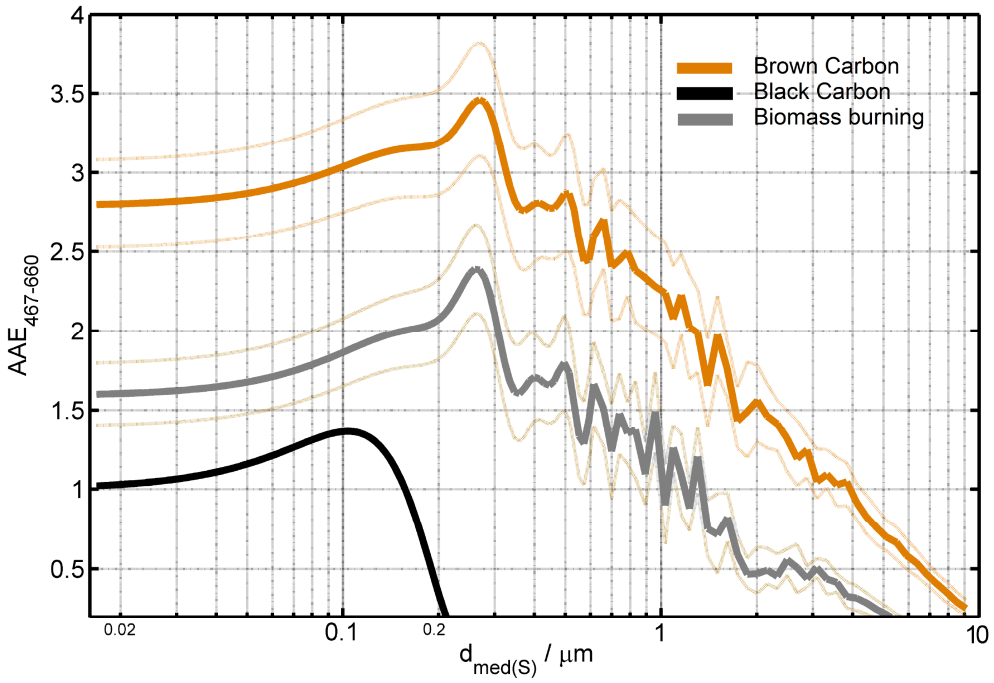


Figure 1. Spectral optical properties measured during the field campaigns: y-axis shows data of scattering and absorption coefficients, colored by the Scattering Ångström Exponent (SAE), and Absorption Ångström Exponent (AAE), respectively. Note that the case study is February 1^{st}, the first day of the Winter field campaign (indicated as 01/02).



	$m_{(\lambda)} = n_{(\lambda)} - i \cdot k_{(\lambda)}$		λ (nm)	Reference
	$n_{(\lambda)}$	$k_{(\lambda)}$		
	1.470 ± 0.01	0.026 ± 0.001	467	Flowers et al. (2010) Moise et al. (2015)
	1.426 ± 0.01	0.017 ± 0.001	530	
	1.470 ± 0.01	0.014 ± 0.001	660	
	1.95 ± 0.01	0.79 ± 0.001	467,530,660	Alexander et al. (2008)
	1.512 ± 0.01	0.027 ± 0.001	467	Costabile et al. (2013)
	1.510 ± 0.01	0.021 ± 0.001	530	
	1.511 ± 0.01	0.022 ± 0.001	660	

Figure 2. Numerical simulations (Mie theory) of the Absorption Ångström Exponent ($AAE_{467-660}$) in the visible range (λ=467-660 nm) resolved by particle diameter d_p (x-axis) and refractive index n (relevant $n(\lambda)$ are showed). Black and brown lines are intended to represent black carbon (BC) and brown carbon (BrC), respectively. Values of the complex refractive index ($n(\lambda)$) are indicated. Brown and black lines indicate patterns theoretically expected (Mie theory) for BrC and BC, respectively.

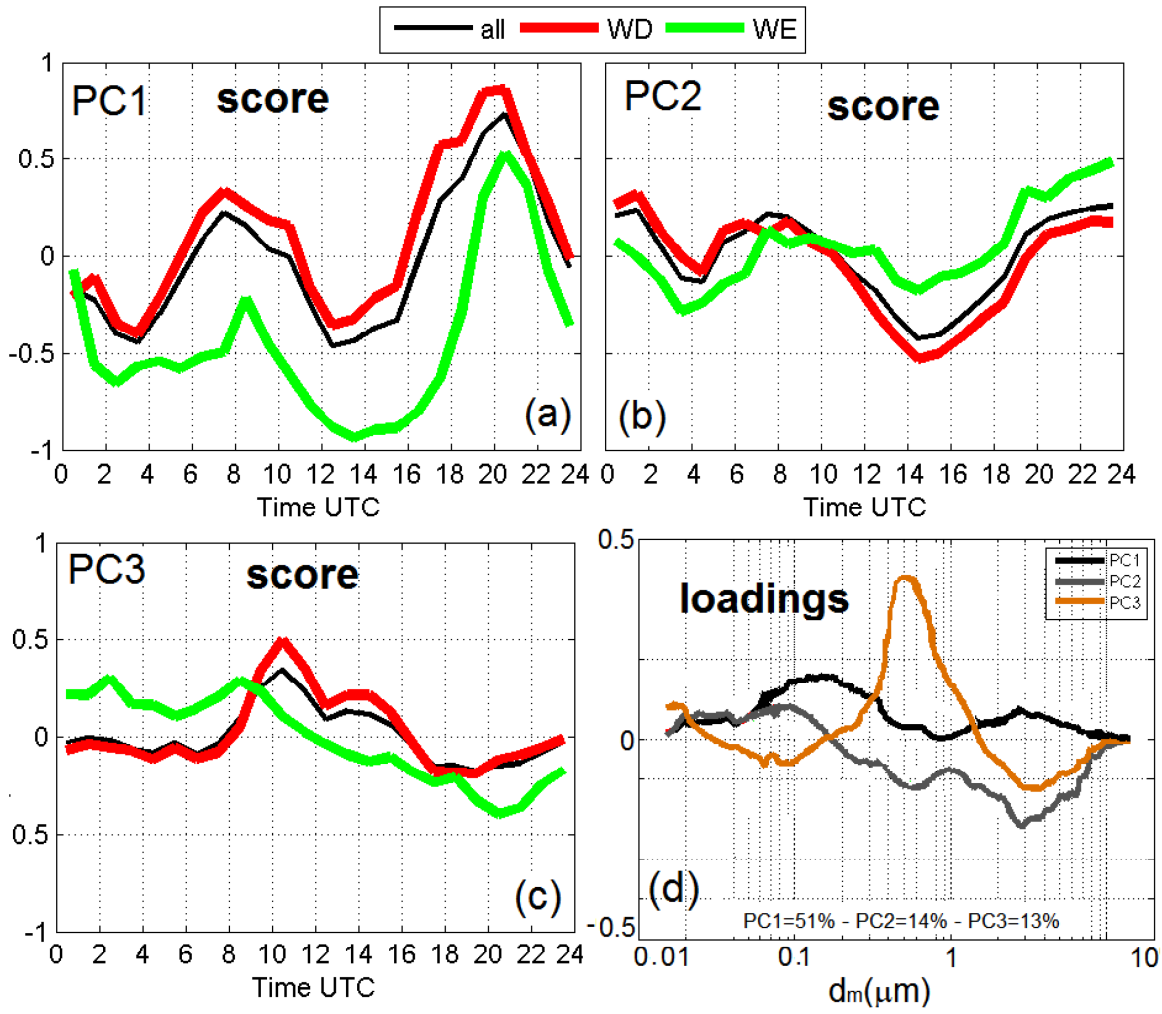


Figure 3. Principal components (PC1-PC3) of Particle Number Size Distributions extracted by principal Component Analysis: (a-c) weekly diurnal cycles of scores (red, Week Day; green, Week End; black, all data), and (d) loadings.

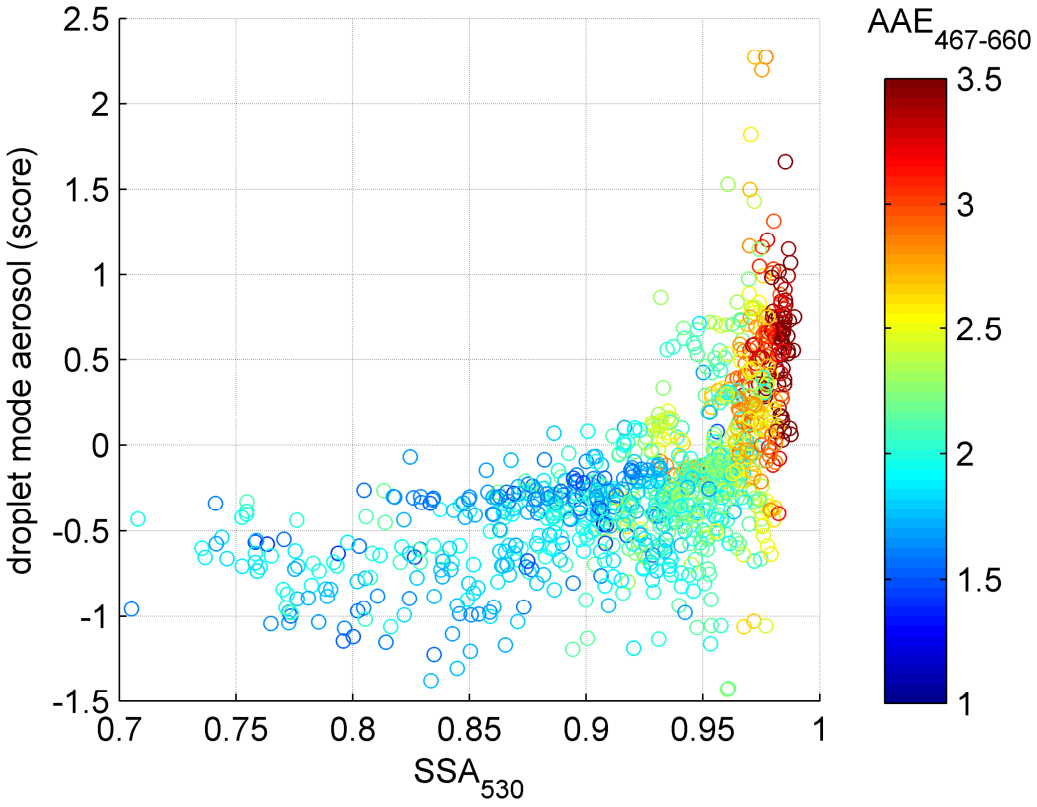


Figure 4. Single Scattering Albedo at 530 nm (SSA) against droplet mode aerosol score. Markers are colored by the Absorption Ångström Exponent ($AAE_{467-660}$).

Table 1. Pearson's correlation coefficients (r) during both field campaigns (first row), in the winter (second row), and the Fall (third row) between: Absorption Ångström Exponent (AAE) at 467-660 nm; scores of major aerosol types identified by PCA, being PC1 the traffic emissions related organic aerosol, PC2 the heating emissions related nocturnal aerosol, and PC3 the droplet mode aerosol; mass concentration and mass fractions of Black Carbon (BC), organics (OA), nitrate (NO_3^-), sulfate (SO_4^{2-}), and ammonium (NH_4^+); optically relevant aerosol size represented by the median mobility diameter of the particle surface size distribution ($d_{med(S)}$); OA-to-BC ratios (OA-to-BC). Relevant matrix of Bonferroni Probabilities (p) associated to these r are shown in Table 2.

r_{all} r_{winter} r_{fall}	AAE	$d_{med(S)}$	BC	f_{BC}	OA	f_{OA}	OA-to-BC	NO_3	f_{NO3}	SO_4	f_{SO4}	NH_4	f_{NH4}
$d_{med(S)}$			-0.24	-0.38	0.04	-0.68	0.37	0.48	0.50	0.30	0.25	0.49	0.69
			-0.34	-0.64	0.06	-0.71	0.47	0.49	0.63	0.32	0.24	0.51	.72
			-0.58	-0.55	0.01	-0.67	0.39	0.56	0.65	0.40	0.26	0.60	0.79
AAE		0.60	-0.26	-0.52	0.40	-0.34	0.78	0.60	0.40	0.67	0.18	0.65	0.44
		0.60	-0.24	-0.55	0.42	-0.36	0.82	0.61	0.27	0.71	0.31	0.65	0.39
		0.71	-0.18	-0.52	0.18	-0.33	0.55	0.51	0.41	0.48	0.20	0.58	0.50
PC1	0.15	-0.02	0.56	0.14	0.83	0.15	0.07	0.52	-0.09	0.40	-0.19	0.52	-0.17
	0.27	-0.05	0.61	0.06	0.90	0.07	0.12	0.58	-0.02	0.43	-0.17	0.57	-0.07
	-0.06	-0.15	0.57	0.22	0.50	0.44	0.01	0.11	-0.32	0.27	-0.30	0.14	-0.44
PC2	-0.17	-0.52	-0.04	-0.06	0.10	0.31	-0.02	-0.19	-0.17	-0.11	-0.18	-0.20	-0.27
	-0.22	-0.53	0.31	0.40	0.12	0.40	-0.24	-0.23	-0.33	-0.16	-0.17	-0.23	-0.41
	-0.41	-0.45	-0.01	0.10	0.01	0.40	-0.06	-0.29	-0.34	-0.40	-0.07	-0.33	-0.38
PC3	0.66	0.60	-0.38	-0.53	0.12	-0.52	0.54	0.46	0.50	0.45	0.17	0.49	0.58
	0.67	0.63	-0.22	-0.52	0.13	-0.50	0.53	0.43	0.40	0.42	0.25	0.48	0.50
	0.63	0.73	-0.65	-0.53	0.04	-0.64	0.34	0.61	0.72	0.53	0.18	0.65	0.73

Table 2. Pearson's correlation coefficients between all the measured variables with relevant matrix of Bonferroni Probabilities (p) associated. These p are probabilities adjusted by the Bonferroni method that provides protection for multiple tests. When the p value (or Bonferroni adjusted probability) is less than the critical value set (p<0.001), there is a significant correlation (significant correlations are shown in bold).

Pearson s correlation matrix

	AAE	DmedS	BC	fBC	OA	fOA	OAtoBC	NO3	fNO3	SO4	fSO4	NH4	fNH4	S1	S2	S3	BCtoOA
AAE	**1,00**																
DmedS	**0,60**	**1,00**															
BC	**-0,26**	**-0,24**	**1,00**														
fBC	**-0,52**	**-0,38**	**0,70**	**1,00**													
OA	**0,40**	0,04	**0,39**	-0,12	**1,00**												
fOA	**-0,34**	**-0,68**	**0,45**	**0,42**	**0,32**	**1,00**											
OAtoBC	**0,78**	**0,37**	**-0,49**	**-0,59**	**0,33**	**-0,25**	**1,00**										
NO3	**0,60**	**0,48**	-0,10	**-0,45**	**0,60**	**-0,45**	**0,53**	**1,00**									
fNO3	**0,40**	**0,50**	**-0,53**	**-0,62**	-0,16	**-0,87**	**0,33**	**0,54**	**1,00**								
SO4	**0,67**	**0,30**	-0,06	**-0,30**	**0,54**	**-0,22**	**0,58**	**0,61**	0,05	**1,00**							
fSO4	**0,18**	**0,25**	-0,16	0,00	**-0,18**	-0,11	0,11	-0,17	**-0,30**	**0,49**	**1,00**						
NH4	**0,65**	**0,49**	-0,09	**-0,45**	**0,63**	**-0,44**	**0,56**	**0,99**	**0,48**	**0,71**	-0,06	**1,00**					
fNH4	**0,44**	**0,69**	**-0,62**	**-0,64**	**-0,25**	**-0,96**	**0,37**	**0,49**	**0,88**	**0,26**	0,14	**0,48**	**1,00**				
S1	0,15	0,02	**0,56**	0,14	**0,83**	0,15	0,07	**0,52**	-0,09	**0,40**	**-0,19**	**0,52**	-0,17	**1,00**			
S2	-0,17	**-0,52**	-0,04	-0,06	0,10	**0,31**	-0,02	**-0,19**	-0,17	-0,11	-0,18	**-0,20**	**-0,27**	-0,12	**1,00**		
S3	**0,66**	**0,60**	**-0,38**	**-0,53**	**0,12**	**-0,52**	**0,54**	**0,46**	**0,50**	**0,45**	0,17	**0,49**	**0,58**	**0,20**	0,06	**1,00**	
BCtoOA	**-0,49**	**-0,21**	**0,69**	**0,95**	**-0,23**	0,17	**-0,60**	**-0,40**	**-0,43**	**-0,30**	0,05	**-0,40**	**-0,42**	0,08	**-0,19**	**-0,45**	**1,00**

Matrix of Bonferroni Probabilities

	AAE	DmedS	BC	fBC	OA	fOA	OAtoBC	NO3	fNO3	SO4	fSO4	NH4	fNH4	S1	S2	S3	BCtoOA
AAE	**0.000**																
DmedS	**0.000**	**0.000**															
BC	**0.000**	**0.000**	**0.000**														
fBC	**0.000**	**0.000**	**0.000**	**0.000**													
OA	**0.000**	1.000	**0.000**	0.287	**0.000**												
fOA	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**											
OAtoBC	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**										
NO3	**0.000**	**0.000**	1.000	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**									
fNO3	**0.000**	**0.000**	**0.000**	**0.000**	0.003	**0.000**	**0.000**	**0.000**	**0.000**								
SO4	**0.000**	**0.000**	1.000	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	1.000	**0.000**							
fSO4	**0.000**	**0.000**	0.002	1.000	**0.000**	0.552	0.738	0.002	**0.000**	**0.000**	**0.000**						
NH4	**0.000**	**0.000**	1.000	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	1.000	**0.000**					
fNH4	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	0.025	**0.000**	**0.000**				
S1	0.020	1.000	**0.000**	0.043	**0.000**	0.017	1.000	**0.000**	1.000	**0.000**	**0.000**	**0.000**	0.001	**0.000**			
S2	0.020	**0.000**	1.000	1.000	1.000	**0.000**	1.000	**0.000**	0.001	0.662	0.001	**0.000**	**0.000**	0.188	**0.000**		
S3	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	0.001	**0.000**	**0.000**	**0.000**	1.000	**0.000**	
BCtoOA	**0.000**	**0.000**	**0.000**	**0.000**	**0.000**	0.001	**0.000**	**0.000**	**0.000**	**0.000**	1.000	**0.000**	**0.000**	1.000	**0.000**	**0.000**	**0.000**