SUPPLEMENTARY INFORMATION

Levels and risk assessment of hydrocarbons and organochlorines in aerosols from a North African coastal city (Bizerte, Tunisia)

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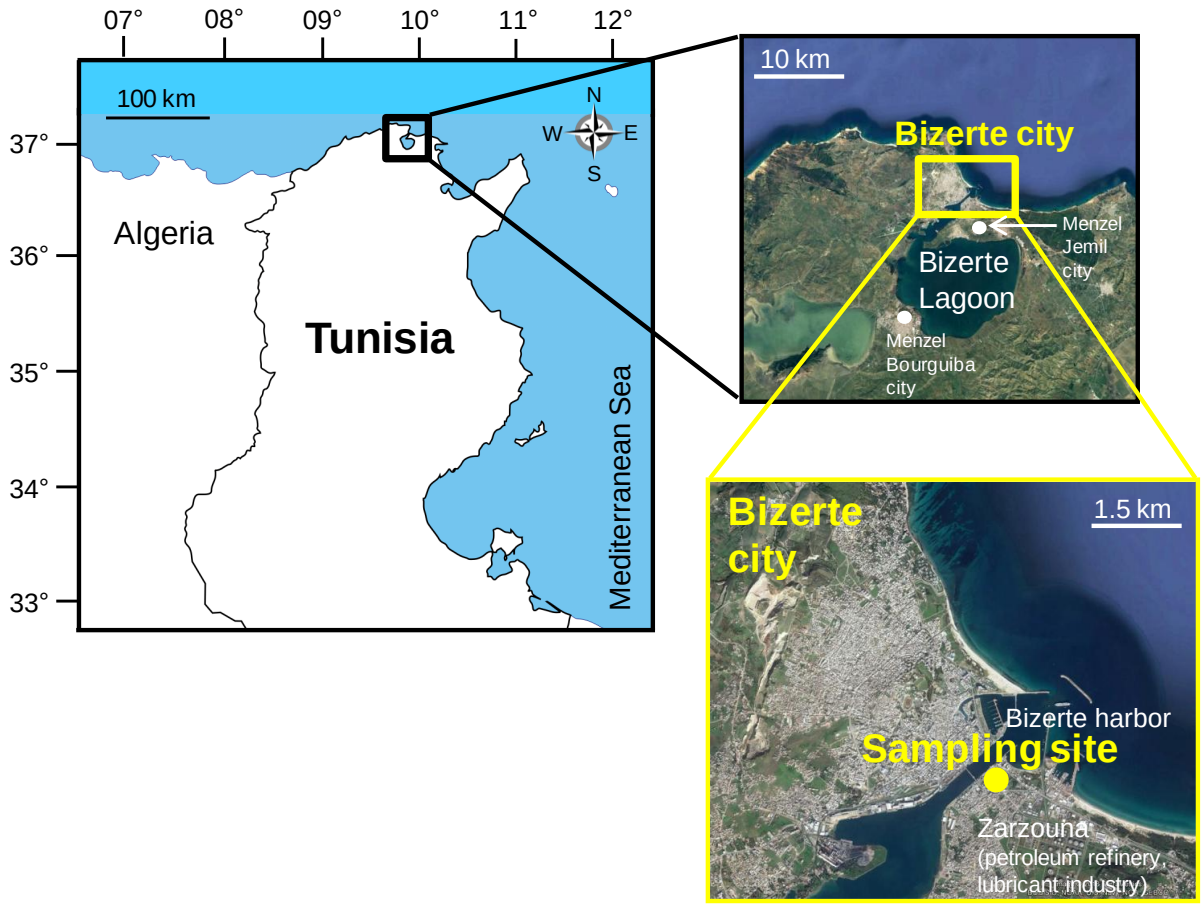

Table S1. Sampling details and meteorological data during the study period (March 2015-January 2016).

Month	Sampling period	Sampling duration (h)	Volume (m^3)	TSP ($\mu g\ m^{-3}$)	OC ($\mu g\ m^{-3}$)	ON ($\mu g\ m^{-3}$)	WS ($Km\ h^{-1}$)	RH (%)	AT (°C)	P (mm)
Mar-15	02-04/03/15	48	1888.67	50.78	13.57	1.77	10	76	18	0.0
Mar-15	04-06/03/15	48	1872.48	106.03	5.94	0.74	14	75	14	4.7
Mar-15	11-13/03/15	48	1868.54	99.74	10.27	1.50	12	77	11	0.0
Mar-15	13-15/03/15	48	1872.36	77.79	8.62	1.35	11	73	12	0.0
Mar-15	18-20/03/15	48	1879.90	44.36	5.81	1.02	22	81	14	0.0
Mar-15	25-27/03/15	48	1905.13	49.77	2.92	0.71	23	81	14	1.0
Mar-15	27-29/03/15	48	1888.65	71.34	2.12	0.56	37	75	14	0.0
Apr-15	07-09/04/15	48	1854.38	46.71	2.58	0.61	21	58	11	0.3
Apr-15	09-11/04/15	48	1883.44	40.71	5.36	1.34	14	68	14	0.0
Apr-15	17-19/04/15	48	1913.42	88.42	9.50	0.60	13	71	18	0.0
Apr-15	21-23/04/15	48	1894.08	45.70	5.87	0.73	13	71	17	0.0
Apr-15	23-25/04/15	48	1913.14	96.55	6.34	1.17	12	76	18	0.0
May-15	07-09/05/15	48	1927.44	111.52	9.20	0.45	20	67	21	0.0
May-15	12-14/05/15	48	1912.32	35.31	5.98	0.64	10	60	20	0.0
May-15	14-16/05/15	48	1943.55	81.70	7.11	0.26	13	59	21	0.3
May-15	19-21/05/15	48	1936.98	88.55	8.52	0.42	15	70	20	0.7
May-15	21-23/05/15	48	1898.44	82.75	8.28	0.64	27	63	17	1.3
May-15	26-28/05/15	48	1912.60	75.19	4.87	0.52	26	73	18	1.0
May-15	28-30/05/15	48	1926.39	37.64	5.34	0.64	16	61	20	0.0
Jun-15	02-04/06/15	48	1917.50	48.91	8.39	1.19	12	71	20	0.0
Jun-15	04-06/06/15	48	1926.31	22.34	4.59	0.65	10	68	22	0.0
Jun-15	09-11/06/15	48	1949.13	42.82	4.61	0.38	19	67	25	0.0
Jun-15	11-13/06/15	48	1967.08	92.43	6.55	0.58	24	58	27	0.0
Jun-15	18-20/06/15	48	1939.76	95.91	10.63	0.79	28	70	22	0.0
Jun-15	24-26/06/15	48	1944.57	16.32	3.29	0.36	13	68	24	0.3
Jun-15	26-28/06/15	48	1943.92	43.94	6.91	1.15	21	67	24	0.0
Jul-15	03-05/07/15	48	1942.73	9.63	4.27	0.84	11	71	25	0.0
Jul-15	05-07/07/15	48	1956.95	38.70	5.20	1.29	12	56	28	0.0
Jul-15	10-12/07/15	48	1954.09	41.23	4.16	0.23	14	71	25	0.0
Jul-15	12-14/07/15	48	1962.44	89.41	5.23	0.62	23	67	27	0.0
Jul-15	25-27/07/15	48	1973.14	50.80	4.79	0.25	13	63	27	0.0
Jul-15	29-31/07/15	48	1995.17	58.48	12.73	0.94	8	55	30	0.0
Aug-15	04-06/08/15	48	1973.60	47.29	13.49	0.82	7	64	28	0.0
Aug-15	07-09/08/15	48	1975.97	34.37	8.12	1.01	6	68	28	0.0
Aug-15	11-13/08/15	48	1964.42	25.85	5.90	0.50	21	67	27	0.3
Aug-15	13-15/08/15	48	1973.31	70.33	14.33	2.15	8	75	26	0.0
Aug-15	19-21/08/15	48	1951.85	60.67	11.88	1.86	16	63	26	0.0
Aug-15	21-23/08/15	48	1956.63	30.98	8.74	0.65	10	67	26	0.0
Aug-15	27-29/08/15	48	1965.70	68.17	8.28	0.92	21	68	29	0.0
Sep-15	02-04/09/15	48	1973.14	105.67	20.99	2.47	7	65	27	0.3
Sep-15	05-07/09/15	48	1996.96	44.93	9.95	0.91	14	63	25	0.3
Sep-15	11-13/09/15	48	1967.20	78.01	4.00	0.68	18	63	28	0.0
Sep-15	16-18/09/15	48	1989.74	119.06	11.50	1.72	17	56	29	0.0
Oct-15	06-08/10/15	48	1951.08	95.61	4.63	0.89	14	67	24	0.3
Oct-15	16-18/10/15	48	1935.82	57.69	5.75	0.97	7	67	21	0.0
Oct-15	22-24/10/15	48	1909.94	64.10	1.80	0.53	20	67	17	1.0
Oct-15	27-29/10/15	48	1921.69	41.94	3.76	0.73	10	79	20	0.0
Nov-15	04-06/11/15	48	1908.41	56.28	4.73	0.36	10	84	19	0.0
Nov-15	09-11/11/15	48	1893.29	37.83	3.95	0.32	13	78	19	0.0
Nov-15	11-13/11/15	48	1893.83	65.30	5.10	0.55	5	84	17	0.0
Nov-15	20-22/11/15	48	1914.44	99.46	6.03	0.74	27	66	18	0.0
Nov-15	27-29/11/15	48	1879.78	37.71	1.02	0.37	26	71	14	2.0
Dec-15	04-06/12/15	48	1857.30	83.53	11.06	1.78	4	78	15	0.0
Dec-15	11-13/12/15	48	1846.31	60.78	17.36	1.48	5	80	9	0.0
Dec-15	18-21/12/15	48	1853.05	64.44	24.49	2.88	3	85	12	0.0
Dec-15	23-25/12/15	48	1852.71	54.24	20.87	3.50	7	84	12	0.0
Jan-16	06-08/01/16	48	1882.64	40.03	9.11	0.82	25	79	13	2.0
Jan-16	13-15/01/16	48	1866.33	58.06	12.62	1.29	11	76	12	0.7
Jan-16	27-29/01/16	48	1840.11	50.24	8.42	1.00	14	82	14	0.0
Jan-16	29-31/01/16	48	1863.08	86.22	7.69	0.95	25	63	13	0.0

TSP total suspended particles, *OC* organic carbon, *ON* organic nitrogen, *WS* wind speed, *RH* relative humidity, *AT* ambient temperature, *P* precipitation.
Meteorological data from Bizerte city were provided by the National Institute of Meteorology.

Text S1. Details on analytical methodology

Reagents and standards

All the chemicals used were of analytical grade and mainly obtained from Supelco (USA), Sigma-Aldrich (France) or Cambridge Isotope Laboratories (USA). Several standard mixtures were used: AH standard mixture solution, containing *n*-alkanes (*n*-C_8–*n*-C_{40}) and isoprenoids (phytane (Phy) and pristane (Pr)), PAH standard mixture solution, containing 16 parent PAHs, as well as individual standard solutions of α-hexachlorocyclohexane (α-HCH), β-HCH, γ-HCH, δ-HCH, heptachlor (Hepchl) and hexachlorobenzene (HCB). Moreover, standard reference material SRM 1493 (chlorinated biphenyl congeners in 2,2,4-Trimethylpentane), SRM 2273 (DDTs and metabolites in isooctane) and SRM 1649b (Urban Dust) were provided by the National Institute of Standards and Technology (NIST, Gaithersburg, USA). The compounds used as internal standards for the quantification of PAHs were naphthalene-d_8 (Naph-d_8), fluorene-d_{10} (Flu-d_{10}), anthracene-d_{10} (Ant-d_{10}) and pyrene-d_{12} (Per-d_{12}), while acenaphtene-d_{10} (Ace-d_{10}), phenanthrene-d_{10} (Phe-d_{10}) and chrysene-d_{12} (Chrys-d_{12}) were used as syringe standards. Hexadecane-d_{34} (*n*-C_{16}-d_{34}), tetracosane-d_{50} (*n*-C_{24}-d_{50}) and hexatriacontane-d_{74} (*n*-C_{36}-d_{74}) were used as internal standards for the quantification of AHs, with nonadecane-d_{40} (*n*-C_{19}-d_{40}) and triacontane-d_{72} (*n*-C_{30}-d_{72}) used as syringe standards. For OCs, CB30, CB155 and CB198 were used as internal standards and BDE77 as syringe standard.

Silica, alumina (70-230 mesh ASTM, Merk, Germany) and granular anhydrous sodium sulfate (< 60 mesh, CHEM-LAB, Belgium) were activated in a furnace at 450°C for 6 h and then kept in sealed desiccators. All solvents used for sample processing and analyses (dichloromethane (DCM), hexane, acetone) were of organic trace analysis quality (Rathburn, Interchim). All glassware were intensively cleaned and baked at 450°C for 6 h.

Sample extraction and clean-up

The extraction and cleanup procedures for AHs, PAHs, PCBs and OCPs were carried out as follows. Each half of filter (blanks and samples) was spiked with a known amount of 10 internal standards, few grams of activated copper were added to remove sulfur and targeted compounds were extracted with DCM using an Accelerated Solvent Extraction system (ASE 350, DIONEX, 100°C, 110 bars, 3 cycles of 5 min, 100% of rinsing volume, 60 s purging time). Prior to use, each ASE cell was pre-cleaned by rinsing and extracting with DCM. The obtained extract was concentrated to approximately 10 mL under reduced pressure at 22°C water-bath using a rotary evaporator. Additional 10 mL *n*-hexane was added to the pear-shaped flask and evaporated down to few hundreds of µL. This concentrated extract was loaded onto the silica-alumina chromatograph column (10 mm i.d., made of glass), prepared by adding, from bottom to top, 3 g alumina deactivated with 3% w/w Milli-Q water, 3 g activated silica, and 1 g dehydrated sodium sulfate. Subsequently, the column was eluted after conditioning with 15 mL of *n*-hexane. The first fraction (F1) containing all AHs and PCBs, and some OCPs, was eluted from the column with 20 mL of *n*-hexane. PAHs and the remaining OCP compounds (fraction 2, F2) were eluted with a mixture of 40 mL *n*-hexane/DCM (v:v = 80:20). F1 and F2 were concentrated by rotary evaporation (35°C) and then reduced down to 0.2 mL under nitrogen stream and stored in a sample vial capped with a Teflon-lined septum. Before analysis each sample was spiked with a known amount of syringe standards.

Instrumental analysis

Both F1 and F2 were analyzed separately by gas chromatograph–mass spectrometer (GC–MS) (Trace ISQ, Thermo Electron) equipped with a HP-5 column (25 m × 0.32 mm × 0.52 µm, J&W Scientific, Agilent Technologies, USA) and operating in electron impact (EI) mode

(70 eV), using hydrogen as carrier gas at a flow rate of 1.2 mL min^{-1}. The injector (used in splitless mode) and detector temperatures were 250 and 320°C, respectively. The initial column temperature was held for 3 min at 70°C, then ramped at 15°C min^{-1} (ramp 1) to 150°C and then at 7°C min^{-1} (ramp 2) to a final temperature of 320°C, which was held for 10 min. PAHs and AHs were identified and quantified in full scan and selected ion monitoring (SIM) modes simultaneously, using two distinct methods (Guigue et al., 2011, 2014, 2017). The instrument quickly switches between the full scan mode over a mass range (m/z) of 50–600 amu, and the SIM mode, using the molecular ion of each compound over the m/z of 50–600 amu (PAHs), or using the fragment ions with m/z of 57, 71 and 85 amu (AHs) (Table S2). PAHs and AHs were identified using both the retention time from the GC chromatograph and comparisons of the MS spectra with standards. Quantification was carried out using internal standards. Sample concentrations are expressed in ng m^{-3}.

For AHs, we determined R, which corresponds to the sum of the concentrations of the resolved *n*-alkane series from *n*-C_{15} to *n*-C_{40} with two isoprenoids, Pr and Phy. We also determined the unresolved complex mixture (UCM) concentrations by integrating the hump using the mean response factor of the resolved compounds (relationship between the area of the peak and the mass of each AH). The UCM hump corresponds to a mixture of many structurally complex isomers and homologues of branched and cyclic hydrocarbons that cannot be resolved by capillary GC columns (Bouloubassi and Saliot, 1993; Guigue et al., 2014). For PAHs, we determined the concentrations of 19 parent PAHs (PAHs-P), namely naphthalene (Nap), acenaphthylene (Acy), acenaphtene (Ace), fluorene (Fl), dibenzothiophene (DBT), phenanthrene (Phe), anthracene (Ant), fluoranthene (Flu), pyrene (Pyr), benz[a]anthracene (BaA), chrysene (Chr), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), Benzo[e]pyrene (BeP), benzo[a]pyrene (BaP), Perylene (Per), dibenz[a,h]anthracene (DahA), benzo[g,h,i]perylene (BghiP), indeno[1,2,3-cd]pyrene (IcdP),

as well as the concentrations of alkylated homologues (methyl, dimethyl, trimethyl) of the five target compounds Naph, Flu, Phe, Pyr and Chr, which lead to a total of 34 PAHs. The 19 parent PAHs considered here are classified into 3 groups with regard to their aromatic ring number: low molecular weight (LMW)-PAHs, including 2 + 3 rings (Nap, Acy, Ace, Fl, DBT, Phe and Ant), medium molecular weight (MMW)-PAHs, including 4 rings (Flu, Pyr, BaA and Chr), and high molecular weight (HMW)-PAHs, with 5 + 6 rings (BbF, BkF, BeP, BaP, Per, DahA, BghiP and IcdP). The nine compounds (Flu, Pyr, BaA, Chr, BbF, BkF, BaP, IcdP, BghiP) mainly produced through combustion processes (Takada et al., 1990) are named combustion PAHs (CPAHs).

After analysis of AHs and PAHs, both F1 and F2 fractions were combined, reduced down to 0.1 mL and analyzed by gas chromatography (GC; Agilent 6890 Series gas chromatography system; Agilent Technologies, USA), equipped with a ^{63}Ni electron capture detector, and both HP-5MS (30 m × 0.25 mm i.d., 0.25 µm film thickness, J&W Scientific, Agilent Technologies, USA) and SPB-608 (30 m × 0.25 mm i.d., 0.25 µm film thickness, Supelco, USA) capillary columns. The optimized GC-ECD program for OCPs was as follows: injector temperature, 270°C, detector temperature, 300°C. The GC temperature program was 70°C (1 min hold) to 140°C at a rate of 25°C min^{-1}, 179°C at 2°C min^{-1}, 210°C at 1°C min^{-1} and then to 300°C (10 min hold) at 5°C min^{-1}. The carrier gas was helium, set at a flow rate of 1 mL min^{-1}. The detector make-up gas was nitrogen, set at a flow rate of 60 mL min^{-1}. Sample injection volume was 1 µL and injection mode was splitless (1 min). The data presented in this paper were obtained using the HP-5MS column, while the SPB-608 capillary column was used to confirm the identification of OCs and resolve the co-elution problem between CB8 and HCB. PCBs and OCPs were quantified by means of internal standards (CB30, CB155, CB198). Sample concentrations are expressed in pg m^{-3}. The targeted compounds include 20 PCB congeners (8, 18, 28, 52, 44, 66, 77, 101, 105, 118, 126, 128, 138, 153, 170, 180, 195,

206, 187 and 209), and 12 OCPs (HCB, HCHs (α-,β-, γ- and δ-HCH), Hepchl and DDTs (DDT: *p,p′*-DDT and *o,p′*-DDT; DDE: *p,p′*-DDE and *o,p′*-DDE; DDD: *p,p′*-DDD and *o,p′*-DDD)).

Table S2. Monitored ions, average field blanks, and method detection limits of the target compounds.

Compounds	Molecular/fragment ions monitored (m/z)[a]	Field blanks (pg)	Method detection limits (pg m^{-3})
PAHs			
Nap	128	n.d.	0.2
Acy	152	n.d.	0.4
Ace	154	n.d.	0.4
Fl	166	n.d.	0.4
DBT	184	n.d.	0.4
Phe	178	n.d.	0.4
Ant	178	n.d.	0.4
Flu	202	n.d.	0.4
Pyr	202	n.d.	0.4
BaA	228	n.d.	0.4
Chr	228	n.d.	0.4
BbF	252	n.d.	2
BkF	252	n.d.	2
BeP	252	n.d.	2
BaP	252	n.d.	2
Per	252	n.d.	2
DahA	278	n.d.	2
IcdP	276	n.d.	2
BghiP	276	n.d.	2
Methyl-Nap	142	n.d.	0.4
Dimethyl-Nap	156	n.d.	0.4
Trimethyl-Nap	170	n.d.	0.4
Methyl-Fl	180	n.d.	0.4
Dimethyl-Fl	194	n.d.	0.4
Trimethyl-Fl	208	n.d.	0.4
Methyl-Phe	192	n.d.	0.4
Dimethyl-Phe	206	n.d.	0.4
Trimethyl-Phe	220	n.d.	0.4
Methyl-Pyr	216	n.d.	0.4
Dimethyl-Pyr	230	n.d.	0.4
Trimethyl-Pyr	244	n.d.	2
Methyl-Chr	242	n.d.	2
Dimethyl-Chr	256	n.d.	2
Trimethyl-Chr	270	n.d.	2
AHs			
C15	57, 71, and 85	500.8	2.7
C16	57, 71, and 85	1061.4	4.3
C17	57, 71, and 85	1003.7	5.3
C18	57, 71, and 85	1492.2	7.9
C19	57, 71, and 85	460.3	2.4
C20	57, 71, and 85	588.0	3.1
C21	57, 71, and 85	425.8	2.2
C22	57, 71, and 85	484.4	2.4
C23	57, 71, and 85	320.3	1.5
C24	57, 71, and 85	411.7	1.8
C25	57, 71, and 85	678.6	3.3
C26	57, 71, and 85	275.7	1.2
C27	57, 71, and 85	619.6	3.0
C28	57, 71, and 85	198.2	0.9
C29	57, 71, and 85	607.1	3.0
C30	57, 71, and 85	190.1	0.9
C31	57, 71, and 85	368.5	1.8
C32	57, 71, and 85	120.6	0.6

C33	57, 71, and 85	175.7	0.9
C34	57, 71, and 85	159.4	0.9
C35	57, 71, and 85	162.1	0.9
C36	57, 71, and 85	106.3	0.6
C37	57, 71, and 85	111.6	0.6
C38	57, 71, and 85	n.d.	1.1
C39	57, 71, and 85	n.d.	1.6
C40	57, 71, and 85	n.d.	1.9
Pr	57, 71, and 85	1419.6	7.3
Phy	57, 71, and 85	1447.4	7.4
PCBs			
CB-8		n.d.	0.4
CB-18		250.3	0.4
CB-28		n.d.	0.4
CB-52		n.d.	0.4
CB-44		n.d.	0.2
CB-66		n.d.	0.2
CB-77		n.d.	0.2
CB-101		n.d.	0.4
CB-105		n.d.	0.4
CB-118		n.d.	0.3
CB-126		n.d.	0.3
CB-128		n.d.	0.3
CB-138		n.d.	0.07
CB-153		n.d.	0.07
CB-170		n.d.	0.05
CB-180		n.d.	0.05
CB-195		n.d.	0.02
CB-206		n.d.	0.03
CB-187		n.d.	0.03
CB-209		n.d.	0.03
OCPs			
HCB		10.7	0.01
α-HCH		n.d.	0.2
β-HCH		n.d.	0.6
γ-HCH		n.d.	0.2
δ-HCH		n.d.	0.2
Hepchl		n.d.	0.1
o,p′-DDE		n.d.	0.1
p,p′-DDE		n.d.	0.2
o,p′-DDD		n.d.	0.1
p,p′-DDD		n.d.	0.3
o,p′-DDT		n.d.	0.1
p,p′-DDT		n.d.	0.1

n.d.: not detectable.

[a]Mass-to-charge ratio of ions for identification and quantification.

Table S3. Comparison between concentrations of PAHs, PCBs and OCPs certified and those measured in SRM 1649b Urban Dust (mg kg^{-1}) (*n=3*).

Compound	Certified SRM 1649b	Measured value (this study)	Recovery (%)
PAHs			
Nap	0.95 ± 0.09	0.62 ± 0.08	65%
Acy	0.19 ± 0.02	0.12 ± 0.03	64%
Ace	0.20 ± 0.04	0.13 ± 0.02	66%
Fl	0.22 ± 0.04	0.17 ± 0.01	77%
DBT	0.19 ± 0.00	0.12 ± 0.07	63%
Phe	4.03 ± 0.06	3.76 ± 0.94	93%
Ant	0.41 ± 0.00	0.40 ± 0.03	96%
Flu	6.24 ± 0.08	5.93 ± 0.79	95%
Pyr	4.98 ± 0.14	4.61 ± 1.01	93%
BaA	2.11 ± 0.05	1.45 ± 0.71	69%
Chr	3.05 ± 0.03	2.47 ± 0.18	81%
BbF	6.18 ± 0.18	3.74 ± 2.62	60%
BkF	1.70 ± 0.05	1.82 ± 0.02	107%
BeP	2.97 ± 0.05	2.69 ± 0.59	90%
BaP	2.47 ± 0.24	2.17 ± 0.26	88%
Per	0.61 ± 0.01	0.60 ± 0.32	97%
IcdP	2.89 ± 0.16	2.18 ± 0.52	75%
DahA	0.29 ± 0.002	0.29 ± 0.11	97%
BghiP	3.97 ± 0.04	2.94 ± 2.16	74%
PCBs			
CB-8	10.80 ± 1.10	11.67 ± 2.34	108%
CB-18	15.80 ± 0.40	8.59 ± 1.18	54%
CB-28	17.80 ± 0.80	10.07 ± 0.37	57%
CB-52	24.30 ± 6.10	25.20 ± 6.63	104%
CB-44	14.50 ± 5.70	8.51 ± 1.05	59%
CB-66	21.00 ± 16.00	24.76 ± 2.31	118%
CB-101	56.40 ± 5.20	61.79 ± 5.52	110%
CB-118	24.00 ± 5	27.54 ± 4.82	115%
CB-153	76.6 ± 0.4.00	89.20 ± 15.22	116%
CB-105	10.00 ± 1.00	8.36 ± 1.06	84%
CB-138	61.00 ± 15.00	74.11 ± 6.71	121%
CB-187	39.30 ± 2.80	48.71 ± 1.39	124%
CB-128	9.70 ± 3.00	11.93 ± 0.59	123%
CB-180	74.20 ± 1.10	82.36 ± 9.43	111%
CB-170	36.30 ± 9.40	42.08 ± 7.18	116%
CB-195	9.00 ± 2.40	10.64 ± 0.02	118%
CB-206	17.00 ± 2.10	20.07 ± 3.12	118%
CB-209	5.70 ± 1.40	6.21 ± 1.34	109%
OCPs			
HCB	3.00 ± 1.20	2.69 ± 0.74	90%
α-HCH	13.70 ± 3.40	14.01 ± 0.97	102%
γ-HCH	3.10 ± 1.10	3.33 ± 0.78	108%
o,p′-DDE	4.71 ± 0.40	4.56 ± 0.08	97%
p,p′-DDE	50.70 ± 0.40	59.42 ± 3.48	117%
o,p′-DDD	13.60 ± 0.70	14.37 ± 2.20	106%
p,p′-DDD	37.70 ± 3.10	34.51 ± 7.07	92%
o,p′-DDT	39.00 ± 3.00	26.24 ± 9.42	67%
p,p′-DDT	235.00 ± 59.00	256.71 ± 59.53	109%

Table S4. Concentrations of PAHs and AHs (ng m^{-3}) in aerosol samples of Bizerte city from March 2015 to January 2016.

Compound	PAHs (ng m^{-3})					Compound	AHs (ng m^{-3})				
	Min	Max	Mean	SD	Median		Min	Max	Mean	SD	Median
Nap	<lod	0.05	0.01	0.02	0.01	*n*-C_{15}	0.02	0.23	0.06	0.04	0.04
Acy	<lod	0.03	0.01	0.01	<lod	*n*-C_{16}	<lod	0.35	0.10	0.09	0.09
Ace	<lod	0.01	0.01	0.01	<lod	*n*-C_{17}	<lod	0.59	0.13	0.12	0.16
Fl	<lod	0.02	0.01	0.01	0.01	*n*-C_{18}	<lod	0.37	0.11	0.10	0.15
DBT	<lod	0.02	0.01	0.01	<lod	*n*-C_{19}	0.01	0.49	0.10	0.09	0.10
Phe	0.01	0.17	0.05	0.04	0.03	*n*-C_{20}	0.02	0.90	0.22	0.17	0.22
Ant	<lod	0.04	0.01	0.01	<lod	*n*-C_{21}	0.04	1.35	0.30	0.29	0.22
Flu	0.02	1.63	0.13	0.29	0.06	*n*-C_{22}	0.07	2.17	0.61	0.44	0.54
Pyr	0.02	1.84	0.16	0.35	0.07	*n*-C_{23}	0.10	3.40	0.85	0.80	0.63
BaA	0.01	0.43	0.04	0.07	0.02	*n*-C_{24}	0.19	5.03	1.38	1.09	1.06
Chr	0.01	1.03	0.13	0.18	0.08	*n*-C_{25}	0.39	6.89	2.27	1.52	1.99
BbF	0.04	1.97	0.27	0.38	0.17	*n*-C_{26}	0.41	3.85	1.82	1.07	1.55
BkF	0.01	0.52	0.07	0.10	0.05	*n*-C_{27}	0.51	49.96	4.83	6.64	3.50
BeP	0.01	1.26	0.22	0.27	0.15	*n*-C_{28}	0.18	3.94	1.82	0.98	1.66
BaP	<lod	0.85	0.09	0.16	0.04	*n*-C_{29}	0.38	36.92	6.56	6.13	4.79
Per	<lod	0.14	0.02	0.03	0.01	*n*-C_{30}	0.11	4.53	1.56	0.96	1.36
DahA	0.01	0.19	0.04	0.04	0.02	*n*-C_{31}	0.41	40.66	3.61	6.66	2.16
IcdP	0.03	1.65	0.24	0.35	0.14	*n*-C_{32}	0.10	1.83	0.53	0.47	0.42
BghiP	0.04	2.07	0.34	0.44	0.22	*n*-C_{33}	0.22	6.68	1.16	1.22	0.80
Methyl-Nap	<lod	0.06	0.02	0.03	0.01	*n*-C_{34}	0.07	3.27	0.37	0.52	0.22
Dimethyl-Nap	<lod	0.12	0.03	0.05	0.01	*n*-C_{35}	0.10	1.24	0.38	0.37	0.26
Trimethyl-Nap	0.01	0.14	0.04	0.04	0.02	*n*-C_{36}	0.05	1.12	0.25	0.31	0.15
Methyl-Fl	0.01	0.04	0.01	0.01	0.01	*n*-C_{37}	0.02	2.33	0.24	0.34	0.13
Dimethyl-Fl	0.01	0.07	0.02	0.01	0.02	*n*-C_{38}	0.04	2.39	0.29	0.46	0.14
Trimethyl-Fl	<lod	<lod	<lod	<lod	<lod	*n*-C_{39}	0.03	1.64	0.20	0.25	0.13
Methyl-Phe	0.01	0.24	0.05	0.04	0.04	*n*-C_{40}	0.01	2.19	0.21	0.32	0.11
Dimethyl-Phe	0.02	0.32	0.07	0.06	0.04	Pr	<lod	0.81	0.12	0.15	0.20
Trimethyl-Phe	0.02	0.18	0.06	0.05	0.04	Phy	<lod	0.40	0.13	0.15	0.22
Methyl-Pyr	0.01	2.05	0.13	0.29	0.05						
Dimethyl-Pyr	<lod	1.18	0.11	0.17	0.05						
Trimethyl-Pyr	<lod	0.99	0.09	0.15	0.04						
Methyl-Chr	0.02	0.41	0.13	0.11	0.10						
Dimethyl-Chr	<lod	0.40	0.12	0.10	0.08						
Trimethyl-Chr	<lod	0.26	0.08	0.06	0.06						
Σ_{34}PAHs	0.48	17.77	2.77	3.43	1.74	Σ_{28}AHs	6.69	126.45	30.20	21.09	25.07

Table S5. Comparison of the concentrations of PAHs (ng m^{-3}), AHs (ng m^{-3}), PCBs (pg m^{-3}) and OCPs (pg m^{-3}) with other studies. ND indicates "not detected".

Location	Type	Atmospheric particle	Period	Congener numbers	Concentration	References
PAHs						
Agra, India	Urban	TSP	2006/11–2007/02	23	1212	Masih et al., 2010
Beijing, China	Megacity	$PM_{2.5}$	2008/06–2009/12	12	42.3 (3.5–217.4)	Wu et al., 2014
Konya, Turkey	Urban	TSP	2006/08–2007/05	16	80	Ozcan and Aydin, 2009
Thessaloniki, Greece	Urban	TSP	2007–2008	18	8.54–24.6	Chrysikou and Samara, 2009
Algiers City, Algeria	Urban	TSP	1998/05–1998/09	14	5.5–43.4	Yassaa et al., 2001
SE Mediterranean	Coastal	TSP	2006/06–2007/05	30	1.26 (0.5–3.0)	Castro-Jiménez et al., 2012
Elche, Southeastern Spain	Urban background	PM10	2008/10–2009/09	16	0.37–1.23	Chofre et al., 2016
Venice, Italy	Urban-industrial	TSP	2009–2012	15	0.82–0.34	Gregoris et al., 2014
Bizerte city, Tunisia	Urban	TSP	2015/03–2016/01	34	2.77 (0.48–17.77)	*This study*
AHs						
Qingdao, China	Urban	TSP	2001/06–2002/05	*n*-C_{14}–*n*-C_{36}	217.1 (19.1–502.4)	Guo et al., 2003b
Barcelona, Spain	Urban	TSP	2003/03–2003/10	*n*-C_{13}–*n*-C_{37}	198–314	Gogou et al., 1994
Athens, Greece	Urban	TSP	2001/06–2001/07	*n*-C_{18}–*n*-C_{35}	446	Karanasiou et al., 2007
Hong Kong, China	Megacity	$PM_{2.5}$	1996–1997	*n*-C_{14}–*n*-C_{36}	20±9–27±5	Zheng et al., 2000
Algiers City, Algeria	Urban	TSP	1998/05–1998/09	*n*-C_{18}–*n*-C_{33}	14.3–92.3	Yassaa et al., 2001
Prato, Italy	industrial	PM10	2000/05–2001/01	*n*-C_{13}–*n*-C_{34}	36.7–205	Cincinelli et al., 2003
Bizerte city, Tunisia	Urban	TSP	2015/03–2016/01	*n*-C_{15}–*n*-C_{40}	30.20 (6.96–126.45)	*This study*
PCBs						
WE Mediterranean	Coastal	TSP	2006/06–2007/05	41	92.9 (21.4–653.8)	Berrojalbiz et al., 2014
Seoul, Korea	Urban	TSP	1999/07–2000/01	41	130	Yeo et al., 2004
Athens, Greece	Urban	TSP	2000	38	181.1	Mandalakis et al., 2002
Venice, Italy	Urban-industrial	TSP	2009–2012	127	0.95–120	Gregoris et al., 2014
Konya, Turkey	Urban	TSP	2006/08–2007/05	6	29	Ozcan and Aydin, 2009
Finokalia, Greece	Urban	TSP	2000/04–2001/10	54	3.3–19	Mandalakis et al., 2005
Marseille, France	Urban-industrial	TSP	2015/03–2016/01	18	0.5–2.7	Castro-Jiménez et al., 2017
Sub-alpine, northern Italy	Semi-rural	TSP	2005/04–2006/04	7	3–10	Castro-Jiménez et al., 2009
Etang de Thau, France	Urban-industrial	TSP	2007/02–2008/02	18	0.6–10	Castro-Jiménez et al., 2011
Moravia, Czech Republic	Urban-industrial	PM_{10}	2007/08–2008/02	7	2–11	Landlová et al., 2014
Bizerte city, Tunisia	Urban	TSP	2015/03–2016/01	20	3.51 (0.35–10.97)	*This study*
OCPs						
Konya, Turkey	Urban	TSP	2006/08–2007/05	18	3270	Ozcan and Aydin, 2009
Bolu, Turkey	Urban	TSP	2007	11	240 (23–940)	Yenisoy-Karakaş et al., 2012
Jinan, China	Agricultural	TSP	2009/07–2010/06	18	92±82	Xu et al., 2011
Moravia, Czech Republic	Urban-industrial	PM_{10}	2007/08–2008/02	3	1.21–10.97	Landlová et al., 2014
Czech Republic	Background	TSP	2012/01–2013/12	10	ND–1.96	Degrendele et al., 2016
Bizerte city, Tunisia	Urban	TSP	2015/03–2016/01	12	1.09 (0.23–3.59)	*This study*

Figure S2. Average concentrations of individual PAHs (ng m^{-3}) (A), and relative contribution (%) of 2 + 3-, 4- and 5 + 6-ring groups (B) in 19 PAHs in aerosol samples of Bizerte city at the four seasons and for the whole year (March 2015-January 2016).

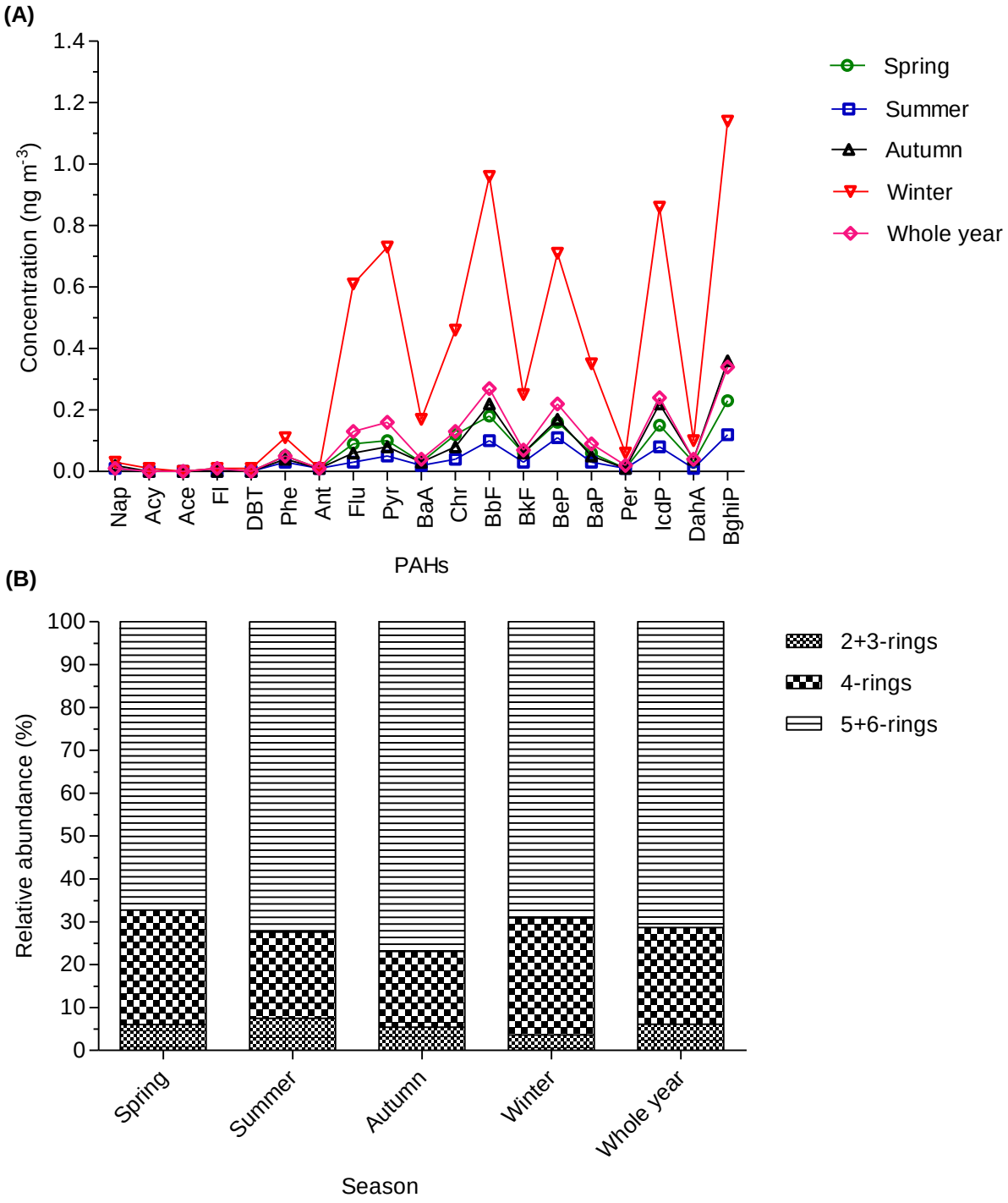

Figure S3. Relative abundances (%) of individual PAHs (A), AHs(B), PCBs (C) and OCPs (D) to $\sum_{34}$PAHs, $\sum_{28}$AHs, $\sum_{20}$PCBs and $\sum_{6}$OCPs, respectively, in aerosol samples of Bizerte city from March 2015 to January 2016.

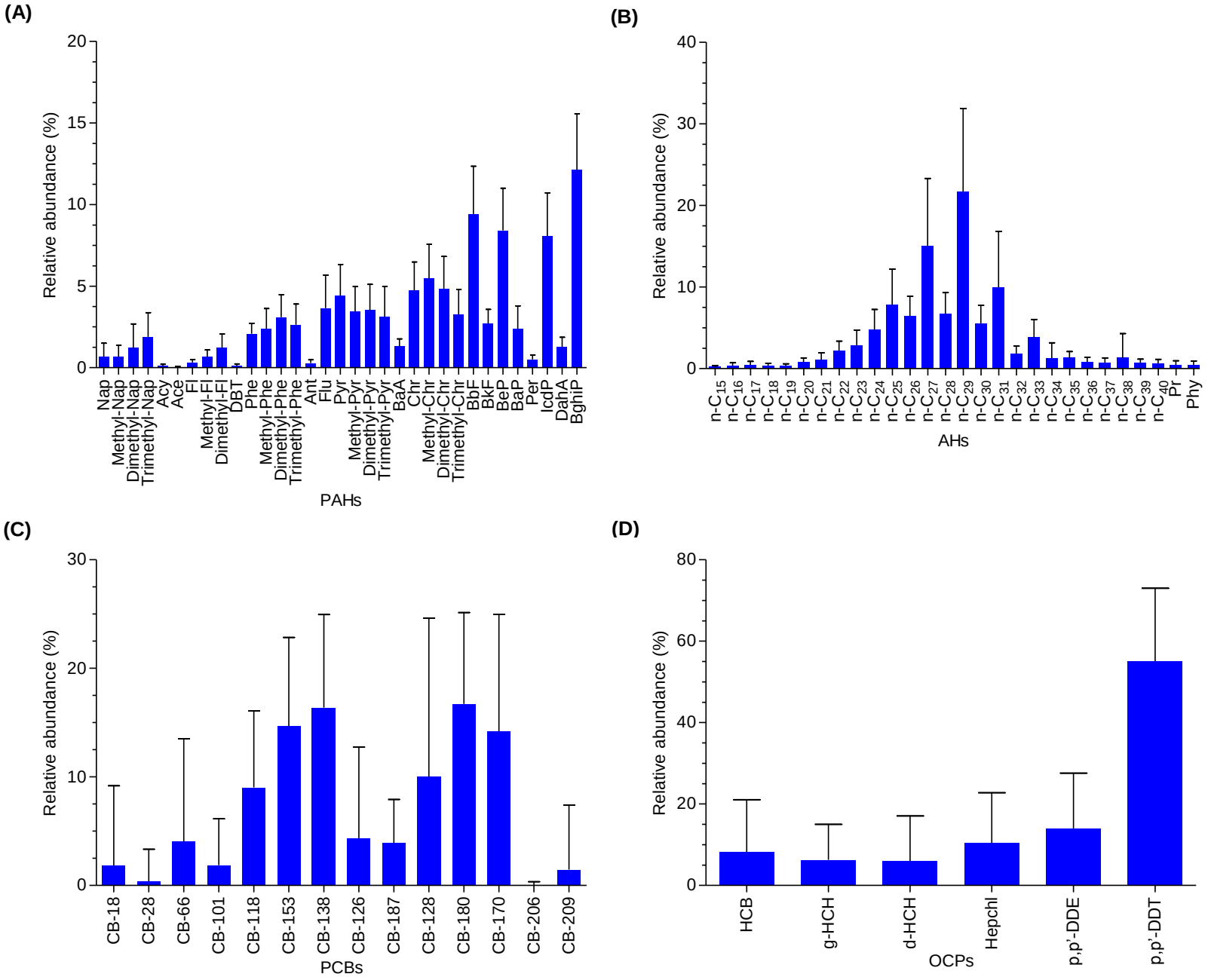

Figure S4. Box-and-whisker plots of total PAH (A), AH (B), PCB (C) and OCP (D) concentrations in aerosol samples of Bizerte city regarding the season. The bottom and top of the box are the 25th and 75th percentiles, respectively, whereas the central line is the 50th percentile (the median). The ends of the error bars correspond to the 10th percentile (bottom) and to 90th percentile (top). The dots are the minimal and maxima values. The red cross is the mean. Means values which are significantly different from each other (U-test, $p < 0.05$) have different (blue) letters: *a*, *b*, or *c*. According to H-test, only PAHs and OCPs display significantly different distributions ($p < 0.05$) throughout seasons.

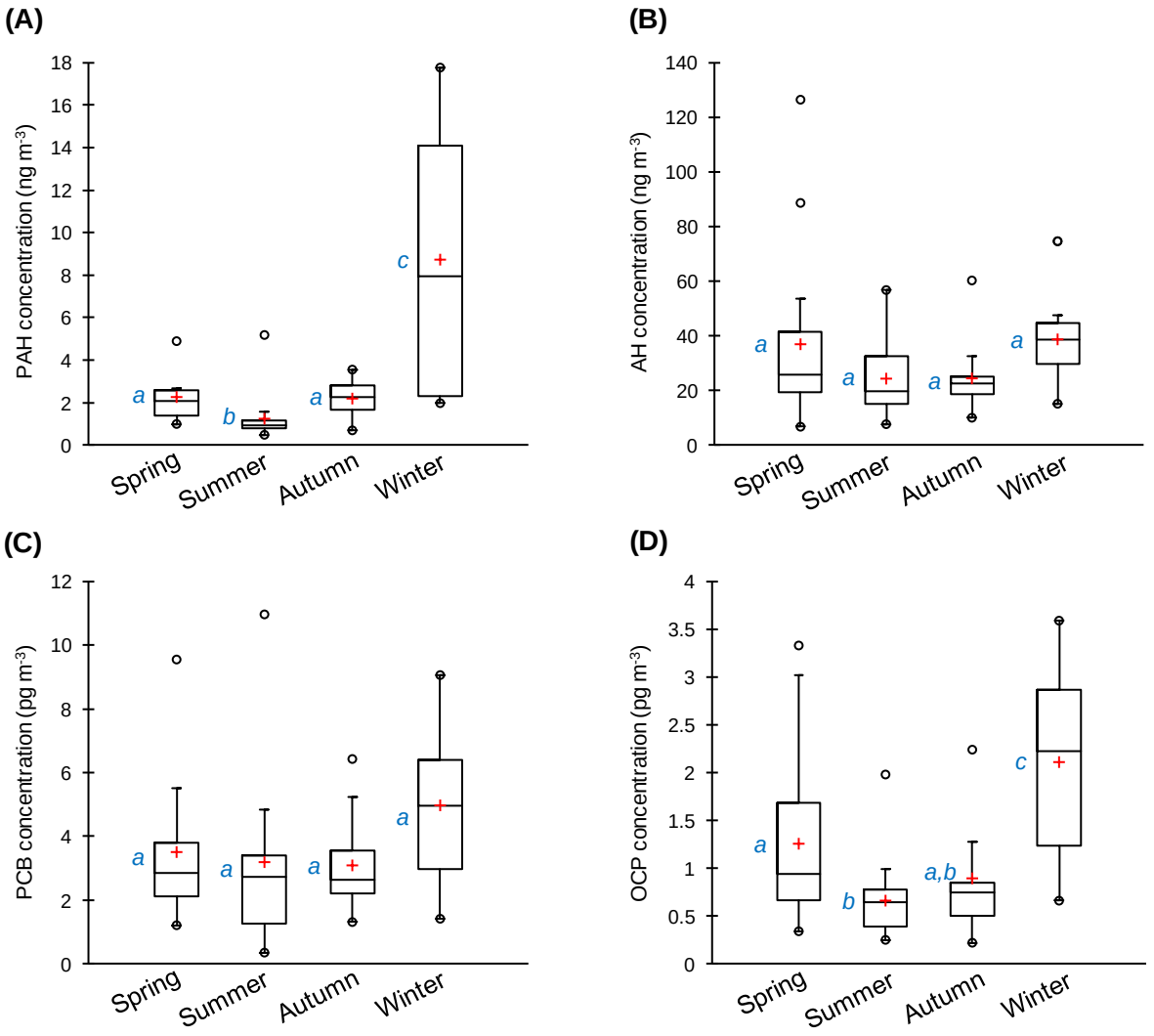

Figure S5. Average concentrations of individual AHs in aerosol samples of Bizerte city at the four seasons and for the whole year (March 2015-January 2016).

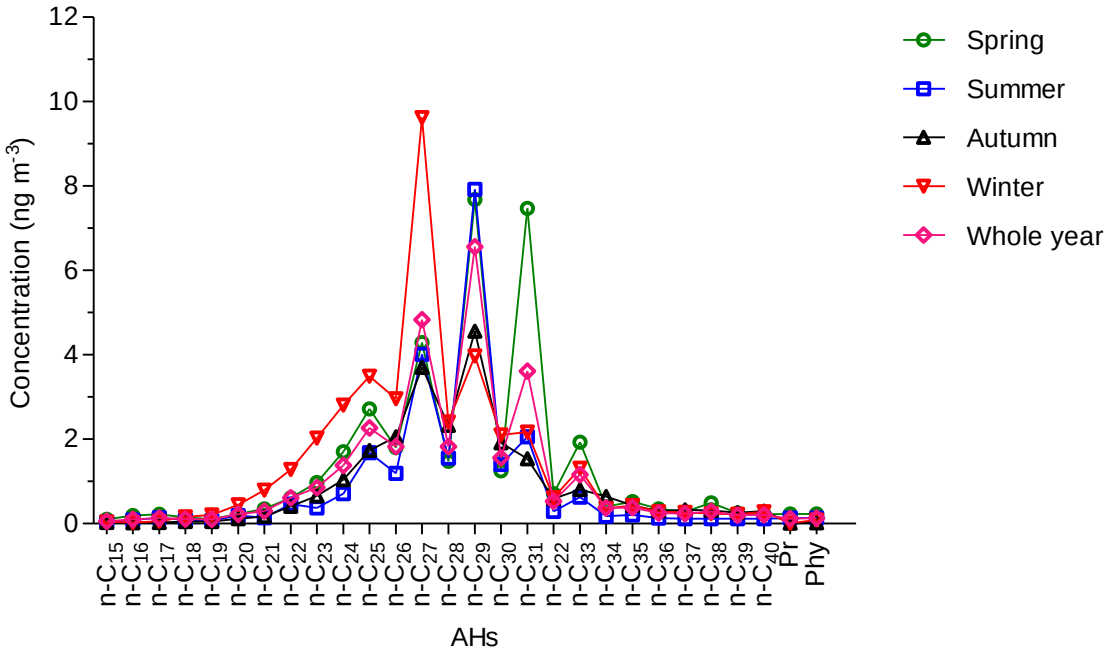

Table S6. Concentrations of organochlorines (pg m^{-3}) in aerosol samples of Bizerte city from March 2015 to January 2016.

Compound	Min	Max	Mean	SD	Median
PCBs					
Di-CBs					
CB-8	<lod	<lod	<lod	<lod	<lod
Tri-CBs					
CB-18	<lod	1.33	0.04	0.18	0.80
CB-28	<lod	1.82	0.03	0.23	<lod
Tetra-CBs					
CB-52	<lod	<lod	<lod	<lod	<lod
CB-44	<lod	<lod	<lod	<lod	<lod
CB-66	<lod	1.22	0.14	0.36	<lod
CB-77	<lod	<lod	<lod	<lod	<lod
Penta-CBs					
CB-101	<lod	1.38	0.11	0.29	<lod
CB-105	<lod	<lod	<lod	<lod	<lod
CB-118	<lod	2.34	0.32	0.36	0.22
CB-126	<lod	0.85	0.14	0.26	<lod
Hexa-CBs					
CB-128	<lod	2.31	0.35	0.56	<lod
CB-138	<lod	2.30	0.54	0.45	0.44
CB-153	<lod	2.55	0.51	0.47	0.36
Hepta-CBs					
CB-170	<lod	3.04	0.50	0.48	0.40
CB-180	<lod	3.91	0.61	0.65	0.41
Octa-CBs					
CB-195	<lod	<lod	<lod	<lod	<lod
Nona-CBs					
CB-206	<lod	0.19	0.01	0.02	<lod
Deca-CBs					
CB-187	<lod	0.72	0.14	0.16	0.10
CB-209	<lod	1.06	0.06	0.23	<lod
ΣPCBs	0.35	10.97	3.51	2.38	3.59
Pesticides					
HCB	0.01	2.27	0.10	0.29	0.03
α-HCH	<lod	<lod	<lod	<lod	<lod
β-HCH	<lod	<lod	<lod	<lod	<lod
γ-HCH	<lod	0.21	0.05	0.06	0.03
δ-HCH	<lod	0.54	0.08	0.14	<lod
Hepchl	<lod	0.91	0.12	0.20	0.05
o,p'-DDE	<lod	<lod	<lod	<lod	<lod
p,p'-DDE	<lod	0.78	0.18	0.24	0.09
o,p'-DDD	<lod	<lod	<lod	<lod	<lod
p,p'-DDD	<lod	<lod	<lod	<lod	<lod
o,p'-DDT	<lod	<lod	<lod	<lod	<lod
p,p'-DDT	0.07	2.10	0.55	0.42	0.43
ΣOCPs	0.23	3.59	1.09	0.84	0.76
*Σ*OCs	0.72	12.88	4.60	2.89	4.49

<lod: below the limit of detection.

Figure S6. Average concentrations of individual PCBs (pg m^{-3}) (A), and relative contribution (%) of homologue PCBs (B) in 14 PCBs in aerosol samples of Bizerte city at the four seasons and for the whole year (March 2015-January 2016).

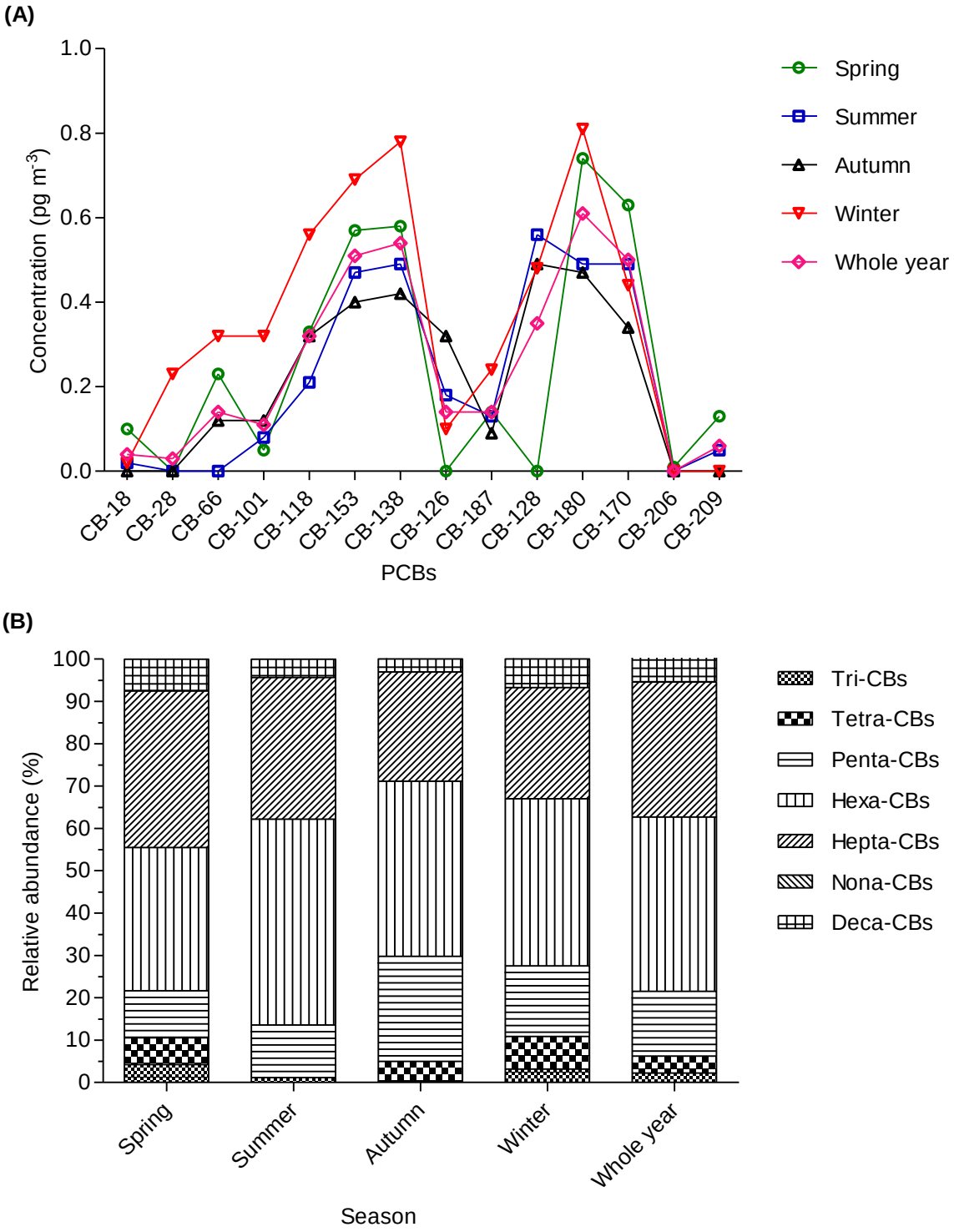

Figure S7. Average concentration of individual OCPs (pg m^{-3}) in aerosol samples of Bizerte city at the four seasons and for the whole year (March 2015-January 2016).

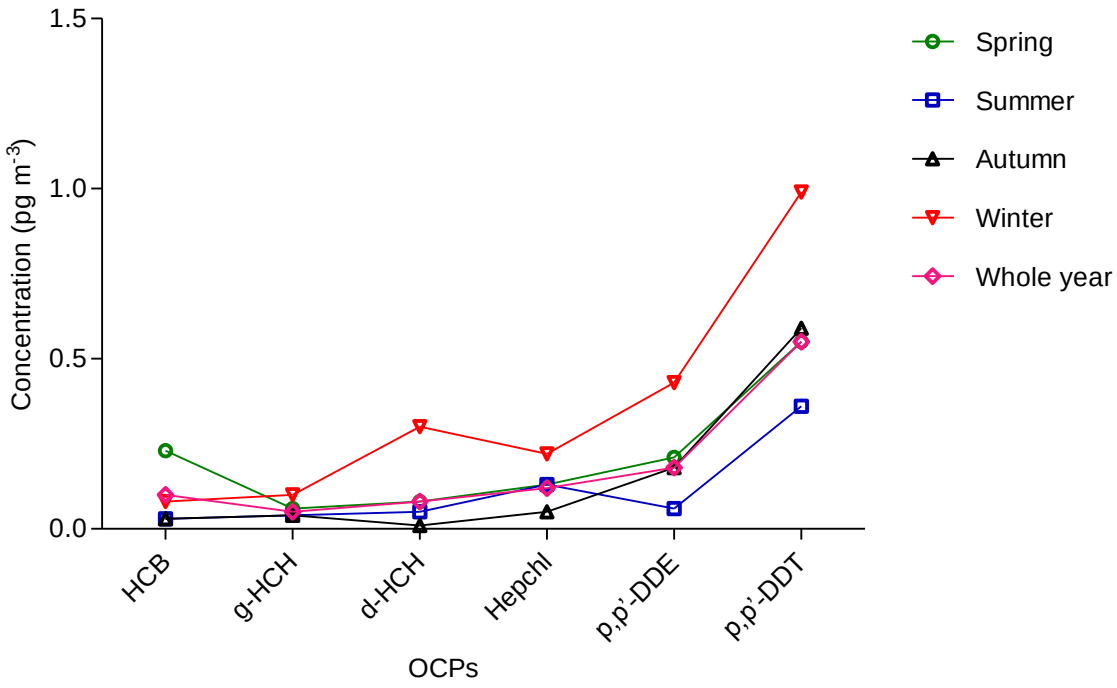

Table S7. Pearson correlation matrix between chemical variables, total suspended particles (TSP), organic carbon (OC), organic nitrogen (ON) and meteorological parameters.

	$\sum_{34}$PAHs	$\sum_{20}$PCBs	$\sum_{6}$OCPs	TSP (μg m^{-3})	OC (μg m^{-3})	ON (μg m^{-3})	Wind speed (Km h^{-1})	Relative humidity (%)	Ambient temperature (°C)	Precipitation (mm)
$\sum_{34}$PAHs (ng m^{-3})	1	**0.373****	**0.591*****	0.042	**0.662*****	**0.650*****	**-0.434****	**0.466*****	**-0.482*****	-0.073
$\sum_{28}$AHs (ng m^{-3})	0.186	0.234	**0.325***	**0.310***	0.206	0.058	-0.050	0.018	-0.055	-0.115
$\sum_{20}$PCBs (pg m^{-3})		1	**0.489*****	0.207	**0.282***	0.186	0.031	0.134	-0.206	-0.079
$\sum_{6}$OCPs (pg m^{-3})			1	0.227	**0.472*****	**0.509*****	**-0.268***	**0.387****	**-0.479*****	-0.024

Correlation coefficients in bold are significant. *p < 0.05, **p < 0.01, ***p < 0.001.

Text S2. Dry deposition flux estimation and cancer risk assessment

The dry deposition fluxes (F ng m^{-2} d^{-1}) were calculated as follows:

$$F = V_d \times C_{TSP} \quad (1)$$

Where V_d is the deposition velocity and C_{TSP} is the volumetric concentration of pollutants in the TSP (ng m^{-3}). V_d is known to be variable, depending on particle size and meteorological conditions. In the Mediterranean Sea, Castro-Jiménez et al. (2012) used a V_d value of 0.2 cm s^{-1} for an average wind speed of 5 m s^{-1}. Here, since our wind speed values were very close (4 m s^{-1} on average), we chose a V_d value of 0.2 cm s^{-1}. The results from this calculation only provide indicative estimation of fluxes that should be eventually validated by experimental measurements.

In this study, the Eq. (2) was used to evaluate the total daily carcinogenic potential of PAH mixture (Valotto et al., 2017):

$$\sum \mathrm{BaP_{TEQ}} = \sum_i \mathrm{PAHi} \times \mathrm{TEFi} \quad (2)$$

Where, PAHi (ng m^{-3}) is the atmospheric concentration of congener i and TEFi is the respective Toxic Equivalent Factor. Here, we used the TEF proposed by Malcolm et al. (1994) and Nisbet and LaGoy (1992) for the non-volatile PAH (MW ≥ 228), BaA (0.1), Chr (0.01), BbF (0.1), BkF (0.1), BaP (1), BghiP (0.01), IcdP (0.1) and DahA (1).

To calculate the lifetime excess cancer risk (ECR) due to the inhalation of the mixture of PAHs, we used the Eq. (3), where UR_{BaP} is the inhalation cancer unit risk factor of BaP (Jia et al., 2011; OEHHA, 2003; Ramírez et al., 2011).

$$\mathrm{ECR} = \sum \mathrm{BaP_{TEQ}} \times \mathrm{UR_{BaP}} \quad (3)$$

The UR_{BaP} (= 1.1 × 10^{-6} (ng m^{-3})$^{-1}$) is defined as the number of people presenting a risk of contracting cancer from inhalation at a BaP equivalent concentration of 1 ng m^{-3} within their

lifetime of 70 years (OEHHA, 2003). ECR > 10^{-6} and > 10^{-4} are considered as a potential health risk and a high potential health risk, respectively (US. EPA, 2005).

Table S8. Average dry deposition fluxes of $\sum_{34}$PAHs (ng m^{-2} day^{-1}), $\sum_{28}$AHs (ng m^{-2} day^{-1}), $\sum_{20}$PCBs (pg m^{-2} day^{-1}) and $\sum_{6}$OCPs (pg m^{-2} day^{-1}) at the four seasons and for the whole year from March 2015 to January 2016.

	Spring		Summer		Autumn		Winter		Annual	
	Mean	Range	Mean	Range	Mean	Range	Mean	Range	Mean	Range
$\sum_{34}$PAHs	390	171–845	214	83–895	378	119–614	1508	340–3070	478	83–3070
$\sum_{28}$AHs	6376	1157–21850	4199	1318–9806	4218	1726–10412	6683	2610–12894	5219	1157–21850
$\sum_{20}$PCBs	606	209–1650	551	61–1895	534	226–1111	859	244–1566	606	61–1895
$\sum_{6}$OCPs	217	58–575	114	43–342	155	38–388	365	115–620	189	38–620

Table S9. Dry deposition fluxes of particulate PAHs, PCBs and OCPs reported from different regions.

Location	Type	Period	Congener numbers	Flux (ng m^{-2} day^{-1})	References
PAHs					
Butal, Turkey	Urban-industrial	2008/09–2009/06	16	21000	Birgül et al., 2011
North China	Urban	2005–2006	15	5200–18500	Wang et al., 2014
Shanghai, China	Megacity	2010/11–2011/10	18	4060	Cheng et al., 2016
North China	Background	2005–2006	15	800–1700	Wang et al., 2014
São Paulo State, Brazil	Megacity	2003/04–2004/05	13	536–2803	Vasconcellos et al., 2011
Kocaeli, Turkey	Urban-industrial	2006/03–2007/03	10	280	Binici et al., 2014
Kozani, Greece	Urban	2001/01–2001/10	12	260	Terzi and Samara., 2005
Bizerte city, Tunisia	Urban	2015/03–2016/01	34	478 (83–3070)	*This study*
PCBs					
Tainan City, Taiwan	Urban	1994/01–1994/05	106	4730	Lee et al., 1996
Paris, France	Urban	1989	12	79	Granier et al., 1997
Hudson River Bay, USA	Urban-industrial	1997/09–2001/05	93	2.1–53	Totten et al., 2004
Bursa, Turkey	Suburban	2004/07–2005/05	41	41±41	Cindoruk and Tasdemir, 2007
Mundanya, Turkey	Urban	2008/06–2009/07	82	21	Cindoruk and Tasdemir, 2007
Thau Lagoon, France	Urban	2007/02–2008/02	18	0.4	Castro-Jiménez et al., 2011
Eastern Atlantic Ocean	Rural	1999/05–2000/07	19	1.7	Van Drooge et al., 2001
Bizerte city, Tunisia	Urban	2015/03–2016/01	20	0.61 (0.06–1.89)	*This study*
OCPs					
Izmir, Turkey	Industrial	2004/08–2005/04	12	187–776	Bozlaker et al., 2009
Mid-Atlantic, USA	Urban-industrial	2000/01–2001/05	22	7.57	Gioia et al., 2005
Mid-Atlantic, USA	Suburban	2000/01–2001/05	22	4.64	Gioia et al., 2005
Bizerte city, Tunisia	Urban	2015/03–2016/01	12	0.19 (0.04–0.62)	*This study*

Figure S8. Total integrated dry deposition flux of $\sum_{34}$PAHs +$\sum_{28}$AHs +$\sum_{20}$PCBs +$\sum_{6}$OCPs in Bizerte city from March 2015 to January 2016.

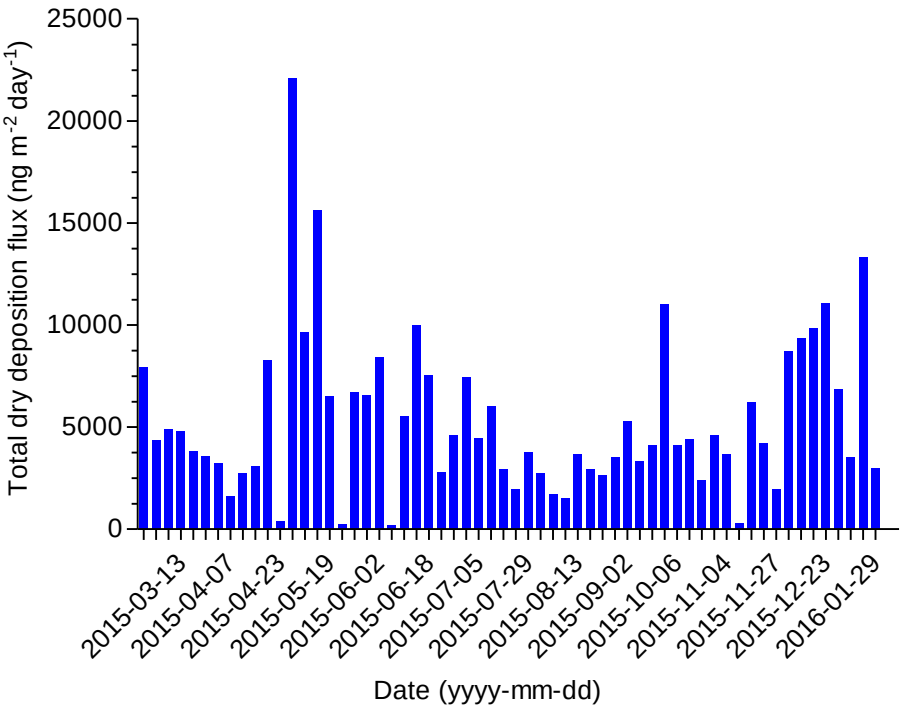

Table S10. Carcinogenic potential of PAHs mixture (∑BaPTEQ) in the Bizerte city and in other regions.

City	PM	TEQ	Period	References
Bizerte city, Tunisia	TSP	0.01–1.49	2015–2016	*This study*
Elche, Spain	PM_{10}	0.1	2008–2009	Chofre et al., 2016
Zaragoza, Spain	TSP	0.08–4.42	1999–2001	Mastral et al., 2003
Algiers, Algeria	TSP	1.9–7.7	1998	Yassaa et al., 2001
Venice, Italy	TSP	23–130	2009 and 2012	Gregoris et al., 2014

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