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Supplementary data

Evaluation of processing factors for selected organic contaminants during virgin olive oil production: distribution of BTEXS during olives processing

Rafael López-Blanco, Bienvenida Gilbert-López*, Rubén Rojas-Jiménez, José Robles-Molina, Natividad Ramos-Martos, Juan F. García-Reyes and Antonio Molina-Díaz

Analytical Chemistry Research Group, Department of Physical and Analytical Chemistry, University of Jaén, Campus Las Lagunillas, Edif. B-3, 23071 Jaén, Spain.

* *Corresponding author:* Dr. Bienvenida Gilbert-López. Analytical Chemistry Research Group, Department of Physical and Analytical Chemistry, University of Jaén, Campus Las Lagunillas, Edif. B-3, 23071 Jaén, Spain. Tel: +34 953 212 147; Fax: +34 953 212 940. E-mail: bgilbert@ujaen.es

Abstract. Details on the statistical analysis conducted, main physicochemical properties of the analytes under study (**Table S4**), analytical figures of merit of the olives and olive oil HS-GC-MS method used (**Tables S1 and S2**), features of the samples analyzed within the olive grove and their nomenclature (**Table S3 and Figure S1**), Detailed information about BTEXS concentrations found in different samples of Andalusia (μg·kg^{-1}) (**Table S7**), Concentrations of BTEXS in gasoil and gasoline (**Table S5**), Olives mass spiked (50 μg·kg^{-1}), oil mass extracted and efficiencies obtained (**Table S6**), and detailed values of statistical study of BTEXS concentration obtained from different locations (**Table S8)**.

Table S1. Analytical figures of merit for the determination of BTEXS in olives using HS-GCMS. For details see: Gilbert-López et al., 2010.

Compound	LODs (μg·kg^{-1})	LOQs (μg·kg^{-1})	RSD (%) 50 μg·kg^{-1}	R^2
Benzene	1.15	3.84	5.97	0.9987
Toluene	0.11	0.36	5.41	0.9989
Ethylbenzene	0.014	0.049	4.67	0.9983
m-, p-Xylene	0.08	0.29	4.83	0.9991
o-Xylene	0.16	0.50	4.96	0.9992
Styrene	0.37	1.25	4.59	0.9969

Table S2. Analytical figures of merit for the determination of BTEXS in olive oil using HS-GCMS. For details see: Gilbert-López et al., 2010.

Compound	LODs (μg·kg^{-1})	LOQs (μg·kg^{-1})	RSD (%) 50 μg·kg^{-1}	R^2
Benzene	0.5	2.20	18.11	0.9991
Toluene	0.4	1.25	14.49	0.9993
Ethylbenzene	3.7	14.40	11.30	0.9987
m-, p-Xylene	4.4	15.00	10.74	0.9992
o-Xylene	2.6	10.00	11.25	0.9983
Styrene	3.5	13.30	9.58	0.9980

Table S3. Features of each sample analyzed within the olive grove.

	Number	Mass Sample (g)	Observations
Samples near the road A-401	2x4	973.6	Secondary road
	2x6	1051.6	Secondary road
	5x1	944.0	-
	9x1	1040.4	Secondary road
	12x1	955.4	-
	13x2	1076.0	Second line
	15x1	946.5	-
	16x1	1171.2	-
	17x1	1113.7	-
	18x1	1095.3	-
	19x1	853.2	-
	19x7	1092.3	Secondary road
	20x1	1153.8	-
	21x1	1080.4	-
	27x1	1125.1	-
Samples inside the grove	4x6	1126.1	-
	5x7	1077.7	-
	6x7	1015.0	-
	10x7	964.0	-
	10x8	1076.6	-
	11x5	875.3	-
	12x6	935.0	-
	12x8	919.5	-
	14x4	749.2	-
	15x6	1052.4	-
	15x7	1220.0	-
	16x5	1165.5	-
	16x6	886.0	-
	17x4	1008.9	-
	17x6	904.6	-

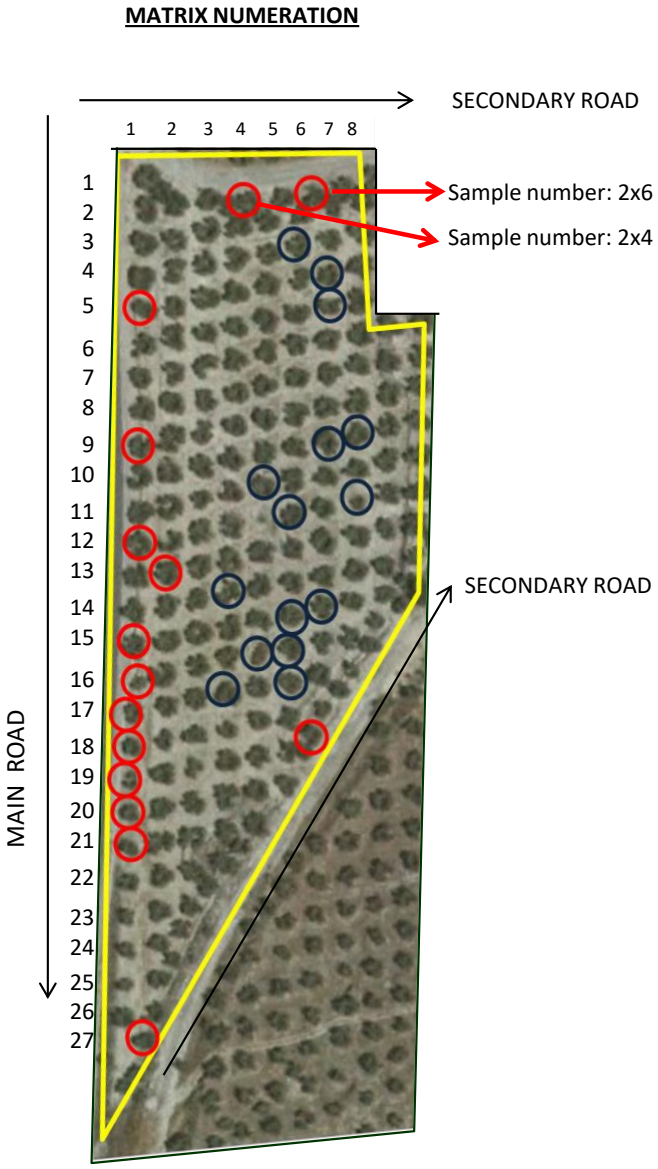

Figure S1. Plot of studied olive grove and nomenclature used for sample ID.

Table S4. Main physicochemical properties of BTEXS.

Compound	Molecular formula	Vapor pressure (mmHg; 25°C)	Water solubility (mg/L; 25°C)	Log Kow
Benzene	C_6H_6	94.80	1790	2.13
Toluene	C_7H_8	28.40	526	2.73
Ethylbenzene	C_8H_{10}	9.60	169	3.15
m-Xylene	C_8H_{10}	8.29	161	3.20
p-Xylene	C_8H_{10}	8.84	162	3.15
o-Xylene	C_8H_{10}	6.61	178	3.12
Styrene	C_8H_8	6.40	300	2.95

Table S5. Concentrations of BTEXS in gasoil and gasoline.

Compound	Benzene	Toluene	Ethylbenzene	m-, p-Xylene	o-Xylene	Styrene
C gasoil ($mg \cdot L^{-1}$)	4576	56998	956	18298	13531	1841
C gasoline ($mg \cdot L^{-1}$)	172	2387	575	1004	1068	66

Table S6. Olives mass spiked (50 $\mu g \cdot kg^{-1}$), oil mass extracted and efficiencies obtained.

Samples	Olives mass milled (g)	Oil mass (g)	Yield (%)
Sample 1	250.4	26.0	10.38
Sample 2	249.8	20.2	8.09
Sample 3	250.5	31.8	12.69
Sample 4	250.0	27.7	11.08
Sample 5	250.0	27.4	10.96
Average	250.14	26.62	10.64
SD	0.30	4.19	1.66
RSD(%)	0.12	15.74	15.63

Table S7. Detailed information about BTEXS concentrations found in different samples of Andalusia ($\mu g \cdot kg^{-1}$).

	Sample	Benzene	Toluene	Ethylbenzene	m-+p-Xylene	o-Xylene	Styrene	ΣBTEXS
Far	Azuaga Badajoz	-	-	1.1	1.7	4.9	-	7.7
	Alhaurín de la Torre Málaga	-	-	1.2	6.1	7.0	2.0	16.6
	Churriana Málaga	-	-	1.0	-	-	4.2	5.2
	Torrequebradilla	-	-	0.6	2.6	-	3.1	6.2
Close to a contamination source	[Infanta-Bailén] (road)	-	-	0.5	2.8	5.1	7.3	15.7
	[Jaén-Mancha Real] (highway)	-	-	-	-	-	-	-
	Alhaurín de la Torre (A-404)	-	-	0.4	4.9	5.7	-	11.0
	Alhaurín de la Torre (C/Giacomo Puccini)	-	-	0.8	3.3	6.1	-	10.2
	Azuaga	-	-	0.7	3.4	5.1	-	9.3
	Baena	-	-	1.4	19.9	16.1	5.9	43.3
	Baena	-	-	1.2	25.7	13.5	4.4	44.8
	Bailén (road)	-	-	0.5	2.0	-	-	2.6
	Churriana (road de Álora)	-	-	0.7	5.4	6.4	8.9	21.4
	Corcoya	-	-	0.9	5.7	7.0	-	13.6
	Ibros	-	-	0.4	4.5	6.8	2.9	14.6
	Jaén (Bulevar)	-	-	0.8	10.4	9.7	6.3	27.2
	Jaén (road A-6050)	-	-	1.3	2.5	5.2	-	9.0
	Jaén (EDAR)	-	-	0.7	4.2	5.5	-	10.3
	Jaén (Gas station)	-	-	0.3	2.7	5.9	-	8.9
	Jaén (Gran Eje)	-	-	0.6	3.0	6.1	-	9.6
	Jaén (La Magdalena)	-	-	0.8	5.0	7.7	2.7	16.2
	Jaén (parque Bulevar)	-	-	1.0	12.4	12.5	5.7	31.6
	Jaén (Parque Industrial)	-	-	0.6	6.9	6.9	-	14.4
	Jaén (Políg. Los Olivares 1)	-	9.9	1.2	13.3	11.0	4.3	39.8
	Jaén (Políg. Los Olivares 2)	-	-	0.9	8.9	8.7	-	18.5
	Jaén (Ronda los olivares)	-	-	1.0	6.3	7.4	2.8	17.5
	Jamilena	-	-	0.8	6.2	6.7	-	13.7
	Las Infantas	-	-	-	4.4	7.7	9.4	21.4
	Linares (Road de Baños)	-	-	0.8	-	-	2.2	3.0
	Los Villares	-	-	0.4	4.2	6.3	2.3	13.2
	Mancha Real	-	-	0.4	3.2	4.9	2.2	10.7
	Mancha Real (entrada)	-	-	0.4	5.0	6.9	11.0	23.3
	Mengibar (IFAPA)	-	-	0.5	4.0	4.9	2.2	11.6
	Montillana	-	-	1.2	6.1	7.0	2.0	16.3
	Puente de Génave	-	-	1.2	6.3	7.2	4.3	19.0
	Torredelcampo	-	-	0.5	1.7	-	-	2.2

	Torredonjimeno	-	-	-	2.1	-	-	2.1
	Torremolinos (Pinos)	-	-	-	-	-	-	-
	Torrequebradilla (highway)	-	-	0.4	3.3	6.1	5.8	15.6
	Villargordo	-	-	0.4	4.2	5.6	3.8	14.0

1. Statistical analysis

With the aim to find out if significant differences exist among the BTEXS concentrations of the samples closer to the contamination sources compared with those far from them, a statistical study using the Student's t-test (Miller & Miller, 1988) has been accomplished to compare the concentration averages from the two groups. The F-test of equality of variances (Miller et al., 1988) showed a significant difference between the variances of the two groups of olives samples in the case of all compounds, except for styrene. Equations I and II were used for benzene, toluene, ethylbenzene, the three isomers of xylenes (ortho, meta and para), while equations III and IV were used for styrene.

Equation I: $\textit{Degrees of freedom} = \frac{\left(\frac{s_1^2}{n_1}+\frac{s_2^2}{n_2}\right)^2}{\left(\frac{s_1^4}{n_1^2(n_1-1)}+\frac{s_2^4}{n_2^2(n_2-1)}\right)}$,

Equation II: $t_{experimental} = \frac{(x_1-x_2)}{\sqrt{\frac{s_1^2}{n_1}+\frac{s_2^2}{n_2}}}$,

Equation III: $\textit{Degrees of freedom} = n_1 + n_2 - 2$

Equation IV: $t_{experimental} = \frac{(x_1-x_2)}{S\sqrt{\frac{1}{n_1}+\frac{1}{n_2}}}$, where $S=\sqrt{\frac{(n_1-1)s_1^2+(n_2-1)s_2^2}{(n_1+n_2-2)}}$

where s_1 and s_2 are the variances (standard deviations) of the concentration of BTEXS in the two olives samples groups, and n_1 and n_2 are the number of samples in each group.

Table S8. Detailed values of statistical study of BTEXS concentration obtained from different locations.

	Benzene	Toluene	Ethylbenzene	m-, p-Xylene	o-Xylene	Styrene
Average road samples (x_1)	0.0	6.0	2.4	3.0	1.9	0.3
Average inside samples (x_2)	0.0	1.0	0.2	0.1	0.0	0.5
Standard deviation road (s_1)	0.0	6.7	2.9	3.4	2.9	0.3
Standard deviation inside (s_2)	0.0	0.5	0.2	0.3	0.0	0.3
Degrees of freedom	-	14	14	14	14	28
$t_{experimental}$	-	2.85	2.87	3.32	2.52	-1.83
$t_{tabulated}$	-	2.14	2.14	2.14	2.14	2.05
Δt	-	0.71	0.73	1.18	0.38	- 3.88