

## SUPPLEMENTARY MATERIAL

### Contaminants of Emerging Concern in the Basque coast (N Spain): occurrence and risk assessment for a better monitoring and management decisions

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**Table S1.** Description of the sampling stations in the Ibaizabal, Oka and Oiartzun estuaries (Nerbioi interior, Oka interior and Oiartzun transitional water bodies) and in the Mompas-Pasaia coastal water body.

Water body	Station code	UTMX ETRS89	UTMY ETRS89	Description	Sample type
Nerbioi interior (Ibaizabal estuary)	GALINDO	500067	4794579	Galindo -WWTP (Consorcio de Aguas Bilbao Bizkaia)	Wastewater
	E-N15	502111	4793583	Barakaldo (Rontegi bridge)	Receiving water
	E-N17	500185	4795862	Leioa (Lamiako)	Receiving water
Oka	GERNIKA	526407	4796855	Gernika -WWTP (Consorcio de Aguas de Busturialdea)	Wastewater
	E-OK5	527059	4798683	Close to Gernika -WWTP discharge point	Receiving water
Oiartzun	E-OI15	586667	4797168	Pasaia – San Pedro (Herrera basin)	Receiving water
Mompas-Pasaia	ULIA	584628	4798540	Ulia station of the Loiola WWTP (Mancomunidad de Aguas del Añarbe)	Wastewater
	L-UR20	584725	4798981	Mompas-Pasaia litoral	Receiving water

**Table S2.** GC-EI-MS/MS experimental conditions used for analysis of EHMC and BTH.

	Precursor ion	Product ion	CE (V)
HCB-C13*	292	257	25
EHMC_1	290	178	10
EHMC_2	290	161	10
GAMMA-HCH-D6*	224	187	10
BTH_1	220	205	20
BTH_2	220	177	20
Naphthalene-D8**	136	108	25

Column: HP5-MS 5% phenyl methyl siloxane (30m x 250 $\mu$ m x 0.25 $\mu$ m, from Agilent).

Injection volume: 2  $\mu$ L (splitless mode).

Gradient temperature: initial T = 50 $^{\circ}$ C, hold for 5min, increase to 200 $^{\circ}$ C at 40 $^{\circ}$ C/min, hold for 1 min, increase to 300  $^{\circ}$ C at 50  $^{\circ}$ C/min, hold for 5 min.

MS source T: 250  $^{\circ}$ C; Dwell time: 100 ms.

\*Used as surrogate standards: HCB-C13, hexachlorobenzene-C13; GAMMA-HCH-D6, gamma-hexachlorocyclohexane-D6.

\*\*Used for quality control.

**Table S3.** Number of samples (n), and minimum (min.), maximum (max.) and mean temperature (TEMP), salinity (SAL), concentration of dissolved oxygen (DO), oxygen saturation (%O2), pH, and concentration of suspended solids (SS) in receiving waters.

		TEMP °C	SAL	DO mg·L <sup>-1</sup>	%O2 %	pH	SS mg·L <sup>-1</sup>
E-N15	n	7	8	7	7	7	8
	Min.	8.1	0.47	4.90	64.9	7.86	4.33
	Max.	22.4	30.43	12.64	107.3	8.33	32.22
	Mean	14.8	11.79	9.07	92.1	8.09	13.74
E-N17	n	8	8	8	8	8	8
	Min.	8.4	0.09	5.61	74.6	7.82	4.88
	Max.	22.3	31.09	12.19	106.9	8.08	28.74
	Mean	14.8	14.18	8.87	92.6	7.96	14.60
E-OK5	n	8	8	8	8	8	8
	Min.	10.8	0.17	4.52	54.4	7.65	9.28
	Max.	24.1	13.54	12.00	111.6	9.70	60.57
	Mean	15.6	3.91	8.26	82.7	8.54	24.29
E-O115	n	8	8	8	8	8	8
	Min.	12.1	27.13	4.73	59.0	7.77	3.68
	Max.	23.7	34.29	9.62	119.1	8.15	24.72
	Mean	16.3	32.04	7.01	87.0	8.01	8.04
L-UR20	n	8	8	8	8	8	8
	Min.	12.0	33.86	7.10	98.6	8.08	1.60
	Max.	23.0	35.12	8.95	107.6	8.27	9.96
	Mean	16.1	34.60	8.20	102.3	8.20	5.51

**Table S4.** Mean (and range) concentrations (ng·L<sup>-1</sup>) of Watch List substances determined in the Basque coast, by sampling station.

	Wastewater			Receiving water				
	Galindo	Gernika	Ulía	E-N15	E-N17	E-OK5	E-OI15	L-UR20
Diclofenac	103.5 (78-139)	51.3 (<3.3-134.1)	43.3 (<3.3-94.2)	5.7 (3.4-7.4)	18.1 (<3.3-40.4)	15.7 (6.1-26.2)	29.3 (<3.3-103)	5.1 (<3.3-15.3)
E2	<1 (<0.116-<1)	<1 (<0.116-<1)	<1 (<0.116-<1)	<1 (<0.116-<1)	<1 (<0.116-<1)	<1 (<0.116-<1)	<1 (<0.116-<1)	<1 (<0.116-1.15)
EE2	<1 (<0.116-<1)	<1 (<0.116-<1)	<1 (<0.116-<1)	<1 (<0.116-<1)	<1 (<0.116-<1)	<1 (<0.116-<1)	<1 (<0.116-<1)	<1 (<0.116-<1)
E1	<0.116 (<0.116)	<0.116 (<0.116)	<0.116 (<0.116)	<0.116 (<0.116)	<0.116 (<0.116-0.242)	<0.116 (<0.116)	<0.116 (<0.116)	<0.116 (<0.116)
BTH	223 (<180-620)	350 (<180-650)	818 (<180-3000)	225 (<180-630)	<180 (<180)	660 (<180-2370)	708 (<180-2160)	<180 (<180)
EHMC	<270 (<270-330)	<270 (<270)	<270 (<270-350)	<270 (<270)	306 (<270-820)	<270 (<270-330)	<270 (<270)	481 (<270-1520)
Erythromycin	25.61 (6-50)	25.89 (<0.08-62.2)	15.14 (<0.08-42)	<5 (0.2-<5)	<5 (0.3-5.4)	<5 (0.2-15.9)	5.20 (0.71-32.7)	<5 (<0.08-<5)
Clarithromycin	174.3 (58.7-535)	98.0 (<2.5-475)	33.7 (<2.5-59)	<2.5 (0.33-6.5)	29.2 (1.1-176)	15.9 (2.4-53.3)	24.1 (0.24-166)	<2.5 (<0.1-7.1)
Azithromycin	375.6 (198-549)	206.3 (19.4-470.2)	177.9 (34.3-672)	19.3 (0.8-66.2)	41.3 (0.9-190)	30.5 (1.5-68.7)	88.7 (1.7-649)	<15 (0.5-39)
Ciprofloxacin	382.13 (58.5-802)	57.59 (<1.67-187)	6.64 (<1.67-24.4)	16.13 (<1.67-62)	27.14 (<1.67-104)	63.82 (<1.67-236)	36.38 (<1.67-143)	54.17 (<1.67-165)
Methiocarb	<16.5 (<2-<16.5)	<16.5 (<2-<16.5)	<16.5 (<2-<16.5)	<16.5 (<2-<16.5)	<16.5 (<2-<16.5)	<16.5 (<2-<16.5)	<16.5 (<2-<16.5)	<16.5 (<2-<16.5)
Imidacloprid	47.1 (6.4-172)	13.3 (<0.17-65)	24.3 (0.55-58)	2.0 (<0.17-5.6)	5.7 (0.2-18.2)	9.4 (<0.17-46.9)	4.6 (<0.17-33)	0.3 (<0.17-1.7)
Thiacloprid	<0.17 (<0.17)	<0.17 (<0.17-0.6)	0.50 (<0.17-2)	<0.17 (<0.17)	0.37 (<0.17-2.4)	0.30 (<0.17-1.8)	<0.17 (<0.17-0.67)	0.25 (<0.17-1.4)
Thiamethoxam	1.193 (<0.116-5.4)	0.268 (<0.116-0.8)	1.042 (<0.116-5.2)	<0.116 (<0.116-0.15)	<0.116 (<0.116-0.45)	0.277 (<0.116-1.6)	0.322 (<0.116-1.3)	0.140 (<0.116-0.71)
Clothianidin	<5 (<0.17-<5)	<5 (<0.17-<5)	<5 (<0.17-<5)	<5 (<0.17-<5)	<5 (<0.17-<5)	<5 (<0.17-<5)	<5 (<0.17-<5)	<5 (<0.17-<5)
Acetamiprid	47.79 (<0.1-314)	5.76 (<0.1-29.1)	2.20 (<0.1-5.3)	1.35 (<0.05-9.1)	0.30 (<0.05-0.8)	0.80 (<0.1-1.9)	0.31 (<0.05-2.2)	<0.1 (<0.05-0.4)
Oxadiazon	0.21 (<0.17-0.60)	0.26 (<0.17-0.79)	0.24 (<0.17-0.41)	<0.17 (<0.17-0.22)	<0.17 (<0.17)	0.98 (<0.17-3.10)	0.22 (<0.17-0.51)	0.28 (<0.17-0.87)
Triallate	1.34 (<0.17-4.7)	1.39 (<0.17-5.3)	4.20 (<0.17-16.3)	0.20 (<0.17-0.4)	<0.17 (<0.17-0.4)	5.27 (<0.17-11.1)	0.99 (<0.17-2.8)	0.87 (<0.17-3)
Metaflumizone	<3.3 (<3.3)	<3.3 (<3.3)	<3.3 (<3.3)	<3.3 (<3.3)	<3.3 (<3.3)	<3.3 (<3.3)	<3.3 (<3.3)	<3.3 (<3.3)

**Table S5.** Concentrations (ng·L<sup>-1</sup>) of Watch List substances reported in estuarine and coastal water samples from different regions.

Substance	Range	Mean	Location	Reference
Diclofenac	<1.4 - 16.3		Saronic Gulf (Greece)	Alygizakis et al., 2016
	<1 - 9.2		Honfleur/Authie estuaries (France)	Amiard et al., 2009
		7.6 - 14	Garonne River Estuary (France)	Aminot et al., 2016
	<1.5 - 11.6		Singapore (Indian Ocean)	Bayen et al., 2013
	nd - 2.5		Gulf of Cadiz (Spain)	Biel- Maeso et al., 2018
	nd - 31.9		Cadiz Bay (Spain)	
	<0.02		Open sea waters of the Western Mediterranean Sea	Brumovský et al., 2017
	7.17		Qinzhou Bay, China	Cui et al., 2019
	<2.5 - 53.6		Danshui Estuary, Bali (Taiwan)	Fang et al., 2012
	1.2 - 486.5		Rostock coastline, Baltic Sea (Germany)	Fisch et al., 2017
	4 - 31		Arade Estuary (Portugal)	Gonzalez-Rey et al., 2015
	4		Gola de Ter beach, Girona (Spain)	Gros et al., 2012
	<0.33 - 84.5	13.2	Superficial waters (sites 11-14), Ebro Delta (Spain)	Gusmaroli et al., 2019
	250.8	51.44	Humber Estuary (United Kingdom)	
		35.35	Cromarty Estuary (United Kingdom)	Letsinger et al., 2019
		34.06	Thames Estuary (United Kingdom)	
		30.6	North Portuguese coast	Lolić et al., 2015
	<22 - 550		Marine surface waters (Ireland)	McEneff et al., 2014
	1 - 22		Estuary of Plentzia (Spain)	Mijangos et al., 2018
	2 - 35		Estuary of Urdaibai (Spain)	
1 - 650		Bilbao estuary (Spain)		
<0,12		North Scotland (United Kingdom)	Nebot et al., 2007	
<7.4 - 19.4		Santos Bay (Brasil)	Pereira et al., 2016	
1.6 - 51.8	14.84	Tejo Estuary (Portugal)	Reis-Santos et al., 2018	
0.819 - 11		Jiulong Estuary (China)	Sun et al., 2016	
<8 - 195		United Kingdom estuaries (Belfast Lough, Tyne, Tees, Mersey, Thames)	Thomas and Hilton, 2004	
~1500		Cortiou rocky inlet, Marseilles (France)	Togola and Budzinski, 2008	
6.2		Elbe estuary (Germany)	Weigel et al., 2002	
nd		Marine waters (North Sea)		
17β-estradiol (E2)	<LOD - 75.8	21.1	Jiulong Estuary (China)	Ashfaq et al., 2019
	<0.15 - 1.36	0.2	Superficial waters (sites 11-14), Ebro Delta (Spain)	Gusmaroli et al., 2019
	3.1 - 21.4		Medway Estuary (Kent, UK)	Hibberd et al., 2009
	<6.09		Puget Sound, Washington (USA)	Meador et al., 2016
	<0.25		Scheldt Estuary (Belgium-The	Noppe et al., 2007

Substance	Range	Mean	Location	Reference
			Netherlands)	
	<7		Duoro Estuary (Portugal)	Ribeiro et al., 2009
	nd		Yangtze River Estuary and East China Sea (China)	Shi et al., 2014
	<1 - 8.7		Manko tidal flat (East China Sea)	Tashiro et al., 2003
	nd		Rio de la Plata Estuary (Argentina)	Valdés et al., 2015
	<0.1 - 3.7		South China Sea (China)	Zhang et al., 2014
	5.6 - 63.8		Honfleur/Authie estuaries (France)	Amiard et al., 2009
	<LOD - 9.22	0.523	Jiulong Estuary (China)	Ashfaq et al., 2019
	<0.025 - 0.18		Little River Estuary (Australia)	Ferguson et al., 2013
	<0.34 - 2.88	0.5	Superficial waters (sites 11-14), Ebro Delta (Spain)	Gusmaroli et al., 2019
	<0.27 - 1.5		Medway Estuary (Kent, UK)	Hibberd et al., 2009
17 $\alpha$ -ethinylestradiol (EE2)	<6.05		Puget Sound, Washington (USA)	Meador et al., 2016
	<0.25		Scheldt estuary (Belgium-The Netherlands)	Noppe et al., 2007
	<18 - 101.9		Duoro Estuary (Portugal)	Ribeiro et al., 2009
	nd - 0.11		Yangtze River Estuary and East China Sea (China)	Shi et al., 2014
	nd		Rio de la Plata Estuary (Argentina)	Valdés et al., 2015
	<0.3 - 3.99		South China Sea (China)	Zhang et al., 2014
	<0.8 - 11		Singapore (Indian Ocean)	Bayen et al., 2013
	<0.08 - 6.9	1.8	Superficial waters (sites 11-14), Ebro Delta (Spain)	Gusmaroli et al., 2019
	<0.2 - 2.3		Chesapeake Bay (USA)	He et al. 2019
	0.6 - 14.3		Medway Estuary (Kent, UK)	Hibberd et al., 2009
	<4.33		Puget Sound, Washington (USA)	Meador et al., 2016
	<0.25 - 10		Scheldt estuary (Belgium-The Netherlands)	Noope et al., 2007
Estrone (E1)	<15 - 112.9		Duoro Estuary (Portugal)	Ribeiro et al., 2009
	nd - 1.43		Yangtze River Estuary and East China Sea (China)	Shi et al., 2014
	2.2 - 53		Manko tidal flat (East China Sea)	Tashiro et al., 2003
	nd		Rio de la Plata Estuary (Argentina)	Valdés et al., 2015
	<0.05 - 11.38		South China Sea (China)	Zhang et al., 2014
2,6-Ditert-butyl-4-methylphenol (BTH)	<9.73 - 28.7	19.2	Superficial waters (sites 11-14), Ebro Delta (Spain)	Gusmaroli et al., 2019
	<1.6 - 756.4		Water samples in beaches, Gran Canaria (Spain)	Sánchez Rodríguez et al. 2015
2-Ethylhexyl 4-methoxycinnamate (EHMC)	nd		Rostock coastline, Baltic Sea (Germany)	Fisch et al., 2017
	<0.16		Superficial waters (sites 11-14), Ebro Delta (Spain)	Gusmaroli et al., 2019
	nd - 115		Chesapeake Bay (USA)	He et al. 2019

Substance	Range	Mean	Location	Reference	
	2.11 - 70.5	16.8	Water samples in beaches (South Korea)	Kim et al., 2017	
	nd - 143		Okinawa Island (Japan)	Tashiro and Kameda, 2013	
	25 - 66		Arctic Ocean and Chukchi Sea		
	<0.41 - 79		Chaozhou (China)		
	89 - 4043		Hong Kong (China)		
	91 - 138		Los Angeles (USA)	Tsui et al., 2014b	
	89 - 150		New York (USA)		
	52 - 78		Shantou (China)		
	46 - 95		Tokyo Bay (Japan)		
	3.84 - 55.6	21.5	South China Sea coastal region (China)	Tsui et al., 2019	
	<LOD - 0.3		Gulf of Cadiz (Spain)	Biel- Maeso et al., 2018	
	nd - 2.3		Cadiz Bay (Spain)		
	<0.05 - 16.6		Hailing Island (China)	Chen et al. 2015	
	nd - 1.7	0.5	Yellow Sea (China)	Du et al. 2017	
nd		Gola de Ter beach, Girona (Spain)	Gros et al., 2012		
9.5 - 486		Victoria Harbour and Hong Kong coast (China)	Gulkowska et al., 2007		
<0.18 - 3.21	0.7	Superficial waters (sites 11-14), Ebro Delta (Spain)	Gusmaroli et al., 2019		
<0.259 - 1.02		Coastal and fish farm areas, South Sea (Korea)	Kim et al., 2017		
nd - 57.07		East China Sea (China)	Li et al., 2020		
<3.18 - 122		Pearl River Estuary (China)	Liang et al., 2013		
<2.49 - 3.3		Puget Sound, Washington (USA)	Meador et al., 2016		
Erythromycin	4.7 - 1900		Victoria Harbour and Hong Kong coast (China)	Minh et al., 2009	
	<0.3 - 78.4		Mar Menor (Spain)	Moreno-González et a., 2015	
	<0,07		North Scotland (United Kingdom)	Nebot et al., 2007	
	nd		Tejo Estuary (Portugal)	Reis-Santos et al., 2018	
	nd		United Kingdom estuaries (Tyne, Tees, Mersey, Thames)	Thomas and Hilton, 2004	
	<2 - 5.2		Victoria Harbour (China)	Xu et al., 2007a	
	<0.17 - 45.4		Yangzte Estuary (China)	Yan et al., 2013	
	0.9 - 8.5	2.6	Laizhou Bay (China)	Zhang et al., 2012	
	0.13 - 6.7	0.69	Bohai Sea and the Yellow Sea (China)	Zhang et al., 2013a	
	<0.23 - 25.2		Yellow Sea (China)	Zhang et al., 2013b	
	1.1 - 50.9		Beibu Gukf (China)	Zheng et al., 2012	
	<LOQ - 150		Bohai Sea (China)	Zou et al. 2011	
	Clarithromycin	<1 - 1.5		Saronic Gulf (Greece)	Alygizakis et al., 2016
		0.2 - 9.4		Cadiz Bay (Spain)	Biel- Maeso et al., 2018
nd			Gulf of Cadiz (Spain)		
<0.08			Hailing Island (China)	Chen et al. 2015	
nd - 0.89		3	Yellow Sea (China)	Du et al. 2017	
17			Gola de Ter beach, Girona (Spain)	Gros et al., 2012	

Substance	Range	Mean	Location	Reference	
	<0.06 - 13.5	4.3	Superficial waters (sites 11-14), Ebro Delta (Spain)	Gusmaroli et al., 2019	
	nd - 9.7		Chesapeake Bay (USA)	He et al. 2019	
	nd - 6.38		East China Sea (China)	Li et al., 2020	
	<1.62		Puget Sound, Washington (USA)	Meador et al., 2016	
	<2 - 9.6		Mar Menor (Spain)	Moreno-González et a., 2015	
	nd - 0.82	0.19	Laizhou Bay (China)	Zhang et al., 2012	
	nd - 0.51	0.07	Bohai Sea and the Yellow Sea (China)	Zhang et al., 2013a	
	nd - 2.6		Yellow Sea (China)	Zhang et al., 2013b	
	nd - 0.72		Beibu Gulf (China)	Zheng et al., 2012	
	nd - 17.8		Cadiz Bay (Spain)	Biel- Maeso et al., 2018	
	nd - 1.2		Gulf of Cadiz (Spain)		
	nd - 138.9	4.8	Yellow Sea (China)	Du et al. 2017	
	<0.6		Gola de Ter beach, Girona (Spain)	Gros et al., 2012	
	Azithromycin	<0.24 - 22.6	7.6	Superficial waters (sites 11-14), Ebro Delta (Spain)	Gusmaroli et al., 2019
nd - 2.7			Chesapeake Bay (USA)	He et al. 2019	
nd - 1.52			East China Sea (China)	Li et al., 2020	
<2.04 - 2.2			Puget Sound, Washington (USA)	Meador et al., 2016	
<11.2 - 163.8			Mar Menor (Spain)	Moreno-González et a., 2015	
<LOQ			Tejo Estuary (Portugal)	Reis-Santos et al., 2018	
nd - 1.2		0.14	Laizhou Bay (China)	Zhang et al., 2012	
nd - 0.39		0.06	Bohai Sea and the Yellow Sea (China)	Zhang et al., 2013a	
nd - 2.5			Yellow Sea (China)	Zhang et al., 2013b	
nd - 0.64			Beibu Gukf (China)	Zheng et al., 2012	
nd - 211.7			Cadiz Bay (Spain)	Biel- Maeso et al., 2018	
nd			Gulf of Cadiz (Spain)		
<0.04			Open sea waters of the Western Mediterranean Sea	Brumovský et al., 2017	
<1.05 - 186			Hailing Island (China)	Chen et al. 2015	
nd		Yellow Sea (China)	Du et al. 2017		
nd		Gola de Ter beach, Girona (Spain)	Gros et al., 2012		
Ciprofloxacin	<0.082 - 1.52		Coastal and fish farm areas, South Sea (Korea)	Kim et al., 2017	
	243 - 1230		Coastline of Bohai Sea, China Sea, Yellow Sea (China)	Lu et al., 2018	
	<6.49 - 7.3		Puget Sound, Washington (USA)	Meador et al., 2016	
	nd - 17		Estuary of Urdaibai (Spain)	Mijangos et al., 2018	
	nd - 540		Bilbao estuary (Spain)		
	nd		Estuary of Plentzia (Spain)		
	1.56 - 7.14	3.91	Tejo Estuary (Portugal)	Reis-Santos et al., 2018	
	<0.41 - 2.27		Yangzte Estuary (China)	Yan et al., 2013	
	<3.3 - 66.1	30.8	Laizhou Bay (China)	Zhang et al., 2012	
	<LOQ - 390		Bohai Sea (China)	Zou et al. 2011	
	Methiocarb	nd		Arade Estuary (Portugal)	Gonzalez-Rey et al., 2015
		<0.23 - 1.49	0.3	Superficial waters (sites 11-14),	Gusmaroli et al., 2019



Substance	Range	Mean	Location	Reference
			Ebro Delta (Spain)	
Imidacloprid	1 - 8		Arade Estuary (Portugal)	Gonzalez-Rey et al., 2015
	<0.17 - 182	24.1	Superficial waters (sites 11-14), Ebro Delta (Spain)	Gusmaroli et al., 2019
	0.3 - 10		Belgian part of the North Sea	Vanryckeghem, et al. 2019
Thiacloprid	<0.06 - 3.54	1.0	Superficial waters (sites 11-14), Ebro Delta (Spain)	Gusmaroli et al., 2019
	0.34 - 65		Belgian part of the North Sea	Vanryckeghem, et al. 2019
Thiamethoxam	nd		Arade Estuary (Portugal)	Gonzalez-Rey et al., 2015
	<0.18 - 1.62	0.5	Superficial waters (sites 11-14), Ebro Delta (Spain)	Gusmaroli et al., 2019
	<1 - 54		Belgian part of the North Sea	Vanryckeghem, et al. 2019
Clothianidin	<0.16 - 9.89	2.3	Superficial waters (sites 11-14), Ebro Delta (Spain)	Gusmaroli et al., 2019
	0.17 - 3.1		Belgian part of the North Sea	Vanryckeghem, et al. 2019
Acetamiprid	<0.35 - 57.8	5.1		
Oxadiazon	15.4 - 592	222.5	Superficial waters (sites 11-14), Ebro Delta (Spain)	Gusmaroli et al., 2019
Triallate	<0.12 - 1.85	0.4		

**Table S6.** Concentrations (ng·L<sup>-1</sup>) of Watch List substances reported in WWTP effluents from different regions.

Substance	Range	Mean	Location	Reference
Diclofenac		927	Psittalia WWTP (Greece)	Alygizakis et al., 2016
	5.68 - 31.6	26	Xiamen WWTP (China)	Ashfaq et al., 2017
	<20 - 2349	599	Sewage treatment works in UK	Ashton et al., 2004
	172 - 277		Jerez de la Frontera WWTP (Spain)	Biel- Maeso et al., 2018
	761 - 987	874	Psittalia WWTP (Greece)	Dasenaki & Thomaidis, 2015
	100 - 131		Bali WWTP (Taiwan)	Fang et al., 2012
	15087		Wastewater discharge in Hope Bay (Antartica Peninsula Region)	González-Alonso et al., 2017
	184 - 376		WWTP in Girona (Spain)	Gros et al., 2012
	200 - 614	440	WWTPs in Ebro Delta region (Spain)	Gusmaroli et al., 2019
	8.8 - 127	40	WWTPs in South Korea	Kim et al., 2007
			Stony Brook WWTP, New York (USA)	Lara-Martin et al., 2014
	<62 - 748	194	WWTPs in Ontario (Canada)	Lishman et al., 2006
	310 - 2630		WWTPs in Ireland	McEneff et al., 2014
	8 - 683		Gorliz WWTP (Spain)	
	127 - 1911		Galindo WWTP (Spain)	Mijangos et al., 2018
	10 - 1932		Gernika WWTP (Spain)	
	97.35		WWTP in North Scotland (United Kingdom)	Nebot et al., 2007
	58-599	WTTTPs in UK	Petrie et al., 2015	
6 - 431	220	Alcalá de Henares WWTP (Spain)	Rosal et al., 2010	
210.7 - 486.4		WWTPs in Hérault watershed (France)	Togola and Budzinski, 2008	
17β-estradiol (E2)	0.6 - 5.8		WWTPs in China	Ben et al., 2018
	<0.46		WWTPs in Ebro Delta region (Spain)	Gusmaroli et al., 2019
	0.3 - 2.5		Tokio and Ibaraki WWTPs (Japan)	Isobe et al., 2003
	<1		WWTPs in South Korea	Kim et al., 2007
	<5		South Queensland (Australia) and Canterbury (New Zealand WWTPs)	Leusch et al., 2006
	<6.09		Bremerton West and Tacoma Central WWTPs, Washington (USA)	Meador et al., 2016
		0.4 - 1.3	WTTTPs in UK	Petrie et al., 2015
	122 - 631	307.7	WWTPs in Pampas region (Argentina)	Valdés et al., 2015
	nd - 8.4	1.7	Gaobeidian WWTP, Beijing (China)	Zhou et al., 2012
17α-ethinylestradiol (EE2)	nd		WWTPs in China	Ben et al., 2018
	0.27 - 0.35		Western WWTP, Melborune (Australia)	Ferguson et al., 2013
	<2.28		WWTPs in Ebro Delta region (Spain)	Gusmaroli et al., 2019
	<0.2		Tokio and Ibaraki WWTPs (Japan)	Isobe et al., 2003
	<1 - 1.3		WWTPs in South Korea	Kim et al., 2007
	<6.05		Bremerton West and Tacoma Central WWTPs, Washington (USA)	Meador et al., 2016

Substance	Range	Mean	Location	Reference	
Estrone (E1)		0.2 - 0.47	WWTPs in UK	Petrie et al., 2015	
	65 - 187	110.7	WWTPs in Pampas region (Argentina)	Valdés et al., 2015	
	nd - 5.1	2	Gaobeidian WWTP, Beijing (China)	Zhou et al., 2012	
	0.1 - 15.3		WWTPs in China	Ben et al., 2018	
	8.8 - 10.1	9	WWTPs in Ebro Delta region (Spain)	Gusmaroli et al., 2019	
	2.5 - 34		Tokio and Ibaraki WWTPs (Japan)	Isobe et al., 2003	
	<1 - 36	14	WWTPs in South Korea	Kim et al., 2007	
	17.1 - 71		Eysines WWTP (near Bordeaux, France)	Labadie and Budzinski, 2005	
	<1 - 27		South Queensland (Australia) and Canterbury (New Zealand WWTPs)	Leusch et al., 2006	
	4.5 - 58		Bremerton West and Tacoma Central WWTPs, Washington (USA)	Meador et al., 2016	
		4.3 - 12	WWTPs in UK	Petrie et al., 2015	
	nd		WWTPs in Pampas region (Argentina)	Valdés et al., 2015	
	0.2 - 74.2	12.7	Gaobeidian WWTP, Beijing (China)	Zhou et al., 2012	
	2,6-Ditert-butyl-4-methylphenol (BTH)	610		Källby WWTP (Sweden)	Bendz et al., 2005
		22 - 258	132	WWTPs in Germany	Fries and Püttman, 2004
<12.3			WWTPs in Ebro Delta region (Spain)	Gusmaroli et al., 2019	
245 - 290		262	WWTPs in China	Liu et al., 2015	
<268 - 520			WWTPs in Canada	Lu et al., 2019	
<10 - 440			WWTPs, county of Jönköping (Sweden)	Remberger et al., 2004	
<200 - 251			WWTP in Santiago de Compostela (Spain)	Rodil et al., 2010	
<1 - 1480			WWTPs in Albany area, New York (USA)	Wang and Kannan, 2018	
2-Ethylhexyl 4-methoxycinnamate (EHMC)		143 - 168	151	WWTPs in Ebro Delta region (Spain)	Gusmaroli et al., 2019
		2 - 13	6	WWTPs in South Korea	Kim et al., 2017
	<0.85 - 505.2		WWTPs in Hong Kong (China)	Tsui et al., 2014a	
	<10 - 1842	109	Sewage treatment works in UK	Ashton et al., 2004	
Erythromycin	0.1 - 473.6		WWTPs in China	Ben et al., 2018	
	33 - 36		Jerez de la Frontera WWTP (Spain)	Biel- Maeso et al., 2018	
	<155		Psittalia WWTP (Greece)	Dasenaki & Thomaidis, 2015	
	14 - 17		WWTP in Girona (Spain)	Gros et al., 2012	
	29.7 - 53.7	42	WWTPs in Ebro Delta region (Spain)	Gusmaroli et al., 2019	
	<1 - 294	130	WWTPs in South Korea	Kim et al., 2007	
	87 - 138		Bremerton West and Tacoma Central WWTPs, Washington (USA)	Meador et al., 2016	
	246 - 4330		WWTPs in Victoria Harbour (China)	Minh et al., 2009	
	186		WWTP in North Scotland (UK)	Nebot et al., 2007	
		109-1385	WWTPs in UK	Petrie et al., 2015	
	<99 - 760	331	Alcalá de Henares WWTP (Spain)	Rosal et al., 2010	
	62-2054	WWTPs in Pearl River Delta (China)	Xu et al., 2007b		

Substance	Range	Mean	Location	Reference
Clarithromycin	1.3 - 214.6		WWTPs in China	Ben et al., 2018
	4 - 5		Jerez de la Frontera WWTP (Spain)	Biel- Maeso et al., 2018
	900 - 1476	1153	Psittalia WWTP (Greece)	Dasenaki & Thomaidis, 2015
	138		Wastewater discharge in Hope Bay (Antartica Peninsula Region)	González-Alonso et al., 2017
	19 - 192		WWTP in Girona (Spain)	Gros et al., 2012
	24 - 478	138	WWTPs in Ebro Delta region (Spain)	Gusmaroli et al., 2019
			Stony Brook WWTP, New York (USA)	Lara-Martin et al., 2014
			Bremerton West and Tacoma Central WWTPs, Washington (USA)	Meador et al., 2016
Azithromycin	1.4 - 795.1		WWTPs in China	Ben et al., 2018
	nd		Jerez de la Frontera WWTP (Spain)	Biel- Maeso et al., 2018
	123 - 245	171	Psittalia WWTP (Greece)	Dasenaki & Thomaidis, 2015
	31 - 170		WWTP in Girona (Spain)	Gros et al., 2012
	367 - 3474	1714	WWTP in Ebro Delta region (Spain)	Gusmaroli et al., 2019
	261 - 629		Bremerton West and Tacoma Central WWTPs, Washington (USA)	Meador et al., 2016
Ciprofloxacin	6.78 - 11.5	8.71	Xiamen WWTP China)	Ashfaq et al., 2017
	0.6 - 116		WWTPs in China	Ben et al., 2018
	553 - 564		Jerez de la Frontera WWTP (Spain)	Biel- Maeso et al., 2018
	523 - 1437	1116	Psittalia WWTP (Greece)	Dasenaki & Thomaidis, 2015
	104 - 245		WWTP in Girona (Spain)	Gros et al., 2012
			Stony Brook WWTP, New York (USA)	Lara-Martin et al., 2014
	158 - 192		Bremerton West and Tacoma Central WWTPs, Washington (USA)	Meador et al., 2016
	58 - 3194		Gorliz WWTP	
	155 - 3803		Galindo WWTP	Mijangos et al., 2018
	nd - 4719		Gernika WWTP	
<10 - 5692	2378	Alcalá de Henares WWTP (Spain)	Rosal et al., 2010	
Methiocarb	<0.55			
Imidacloprid	17 - 221	92		
Thiacloprid	<0.61			
Thiamethoxam	<0.70		WWTPs in Ebro Delta region (Spain)	Gusmaroli et al., 2019
Clothianidin	<0.77 - 25.4			
Acetamiprid	<0.59			
Oxadiazon	<0.17 - 8.7			
Triallate	<0.28			

**Table S7.** Mean concentration ( $\text{ng}\cdot\text{L}^{-1}$ ) of selected Watch List substances in wastewater samples collected from May 2017 to March 2019, and estimated total ( $\text{m}^3\cdot\text{year}^{-1}$ ) and normalized by the population equivalents ( $\text{mg}\cdot\text{kg}^{-1}\cdot\text{p.e.}^{-1}$ ) mass loads.

	WWTP	Diclofenac	Clarithromycin	Azithromycin	Ciprofloxacin	Imidacloprid
Mean ( $\text{ng}\cdot\text{L}^{-1}$ )	Galindo	103.5	150.4	371.7	258.3	50.5
	Gernika	51.3	82.5	185.9	58.6	14.3
	Loiola (Ulia)	43.3	34.3	190	4.5	28.7
Total mass load ( $\text{kg}\cdot\text{year}^{-1}$ )	Galindo	8.65	12.56	31.05	21.58	4.22
	Gernika	0.11	0.18	0.40	0.13	0.03
	Loiola (Ulia)	1.73	1.37	7.60	0.18	1.15
Normalised mass load ( $\text{mg}\cdot\text{year}^{-1}\cdot\text{p.e.}^{-1}$ )	Galindo	7.14	10.37	25.63	17.81	3.48
	Gernika	6.00	9.65	21.74	6.85	1.67
	Loiola (Ulia)	3.13	2.48	13.74	0.33	2.08

Notes:

Annual flow rate in 2019 (provided by the WWTPs):

- (i)  $83.54 \text{ Hm}^3\cdot\text{year}^{-1}$  in Galindo;
- (ii)  $2.17 \text{ Hm}^3\cdot\text{year}^{-1}$  in Gernika (raw water flow that will remain once the WWTP is out of service and connected to the new WWTP); and
- (iii)  $39.99 \text{ (Hm}^3\cdot\text{year}^{-1})$  in Loiola (Ulia).

Population equivalent served by the WWTPs:

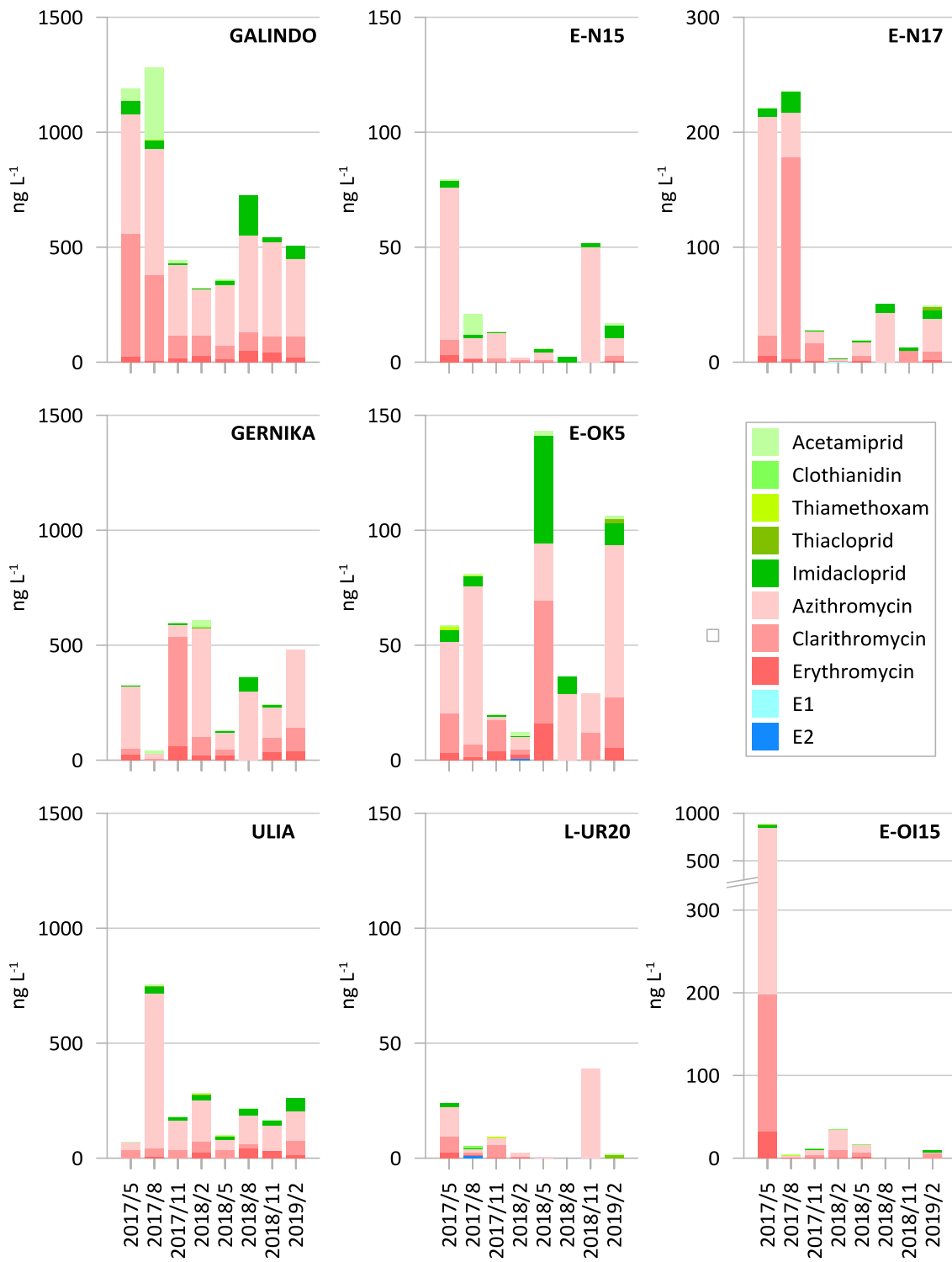
- (i) 1,211,500 p.e. in Galindo;
- (ii) 18,600 p.e. in Gernika; and
- (iii) 553,000 p.e. in Loiola (Ulia).

Total mass load was calculated using Eq (1):

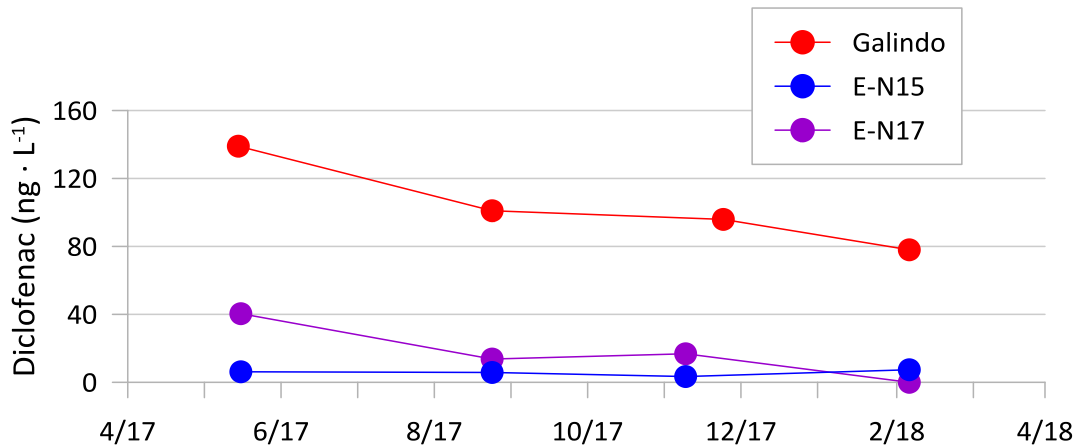
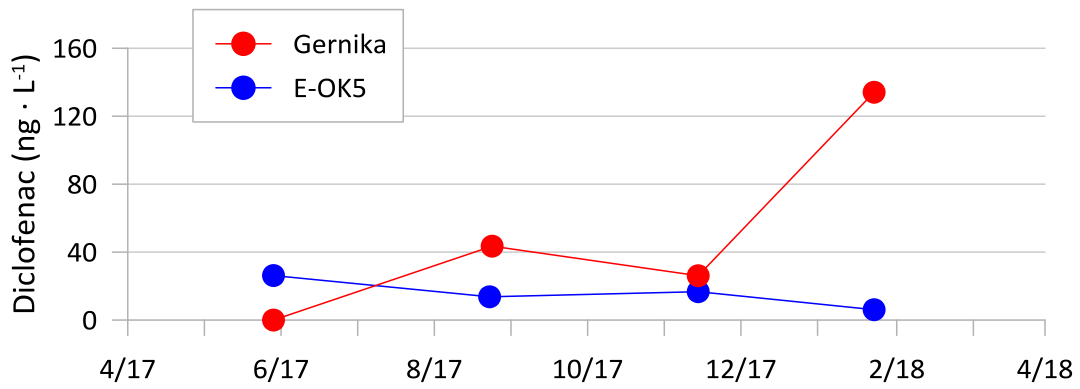
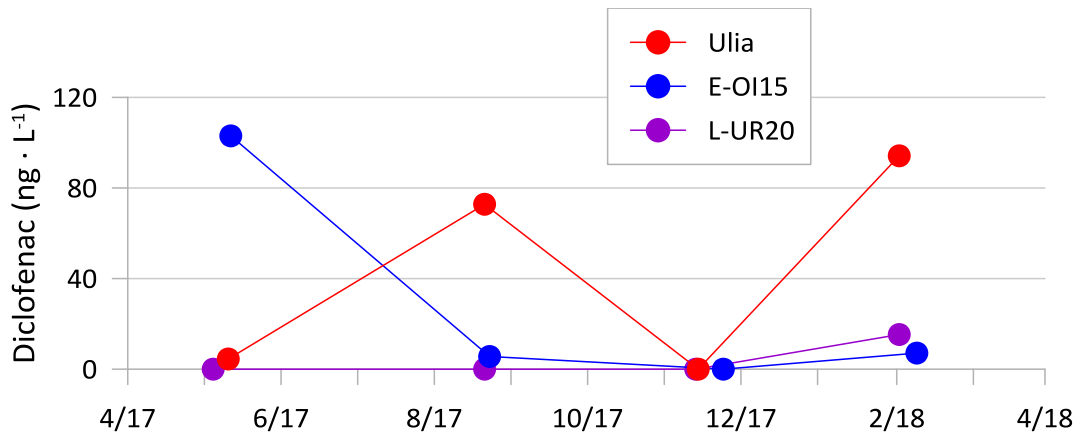
$$\text{Total mass load (kg} \cdot \text{year}^{-1}) = \frac{\text{Concentration (ng}\cdot\text{L}^{-1}) \times \text{Flow rate (Hm}^3\cdot\text{year}^{-1})}{10^3} \quad (1)$$

Normalised mass load was calculated using Eq (2):

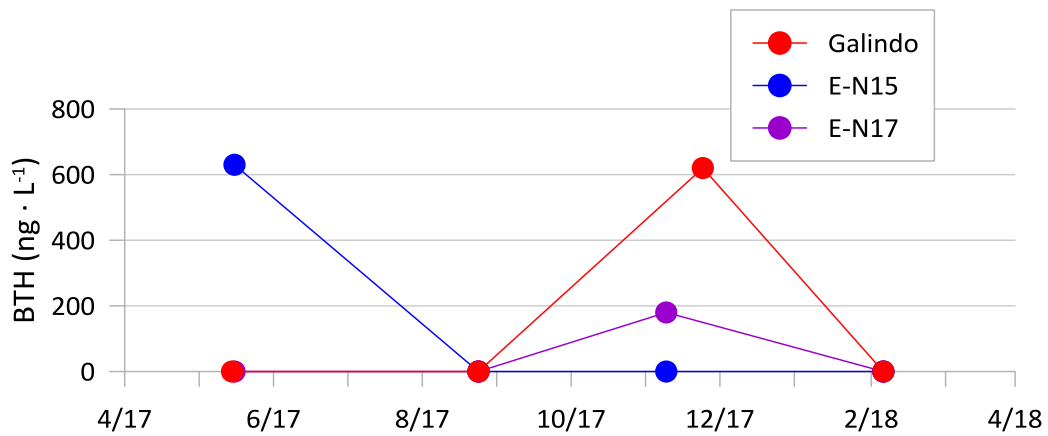
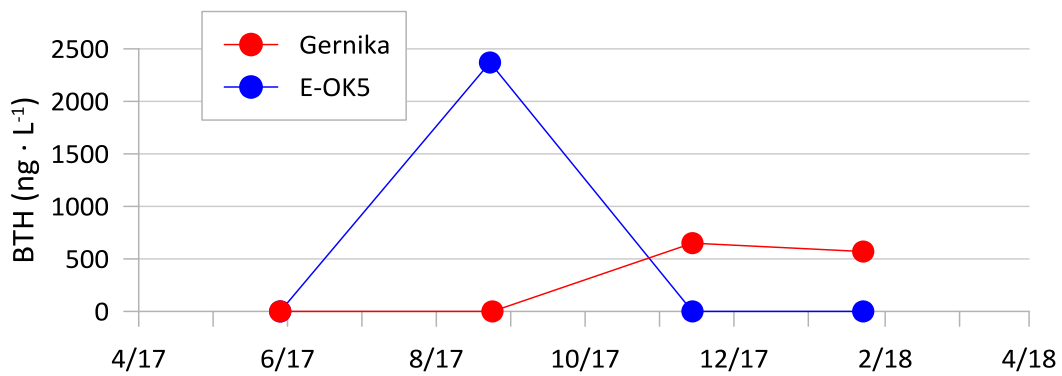
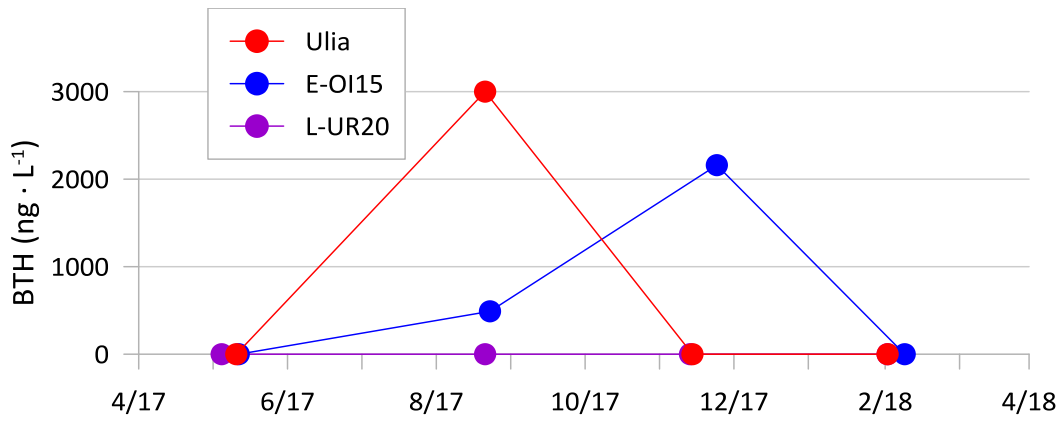
$$\text{Normalised mass load (mg} \cdot \text{year}^{-1} \cdot \text{p.e.}^{-1}) = \frac{\text{Mass load (mg}\cdot\text{year}^{-1})}{\text{Population equivalents (p.e.)}} \times 10^6 \quad (2)$$



**Figure S1.** Concentration of the substances included in both Watch Lists, in the Basque coast, from May 2017 to March 2019. EE2 and methiocarb are not represented since their concentrations were in all cases  $< \text{MQL}$ .

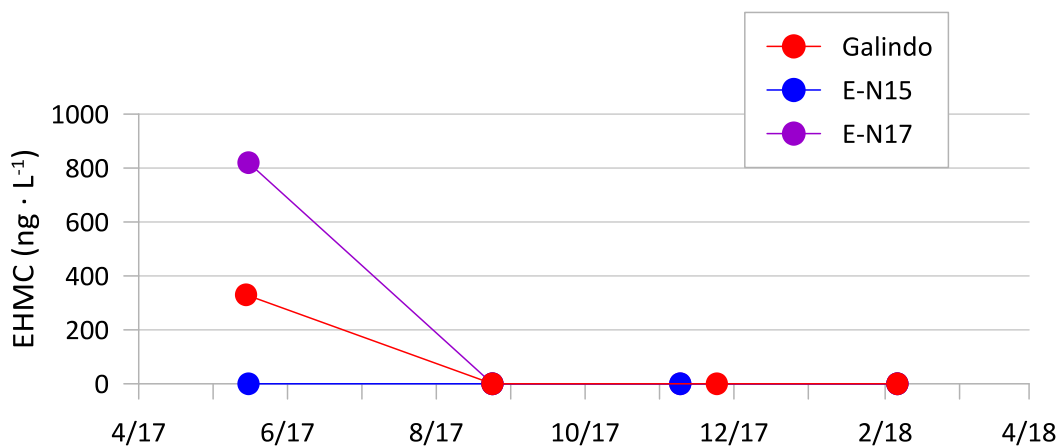
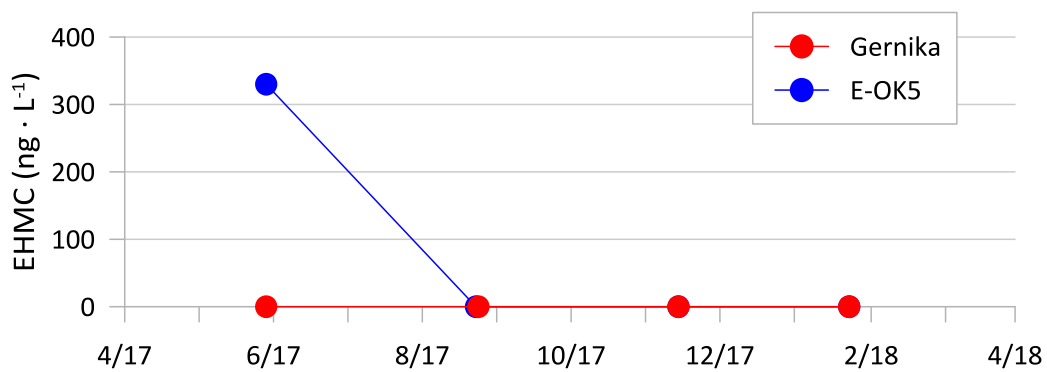
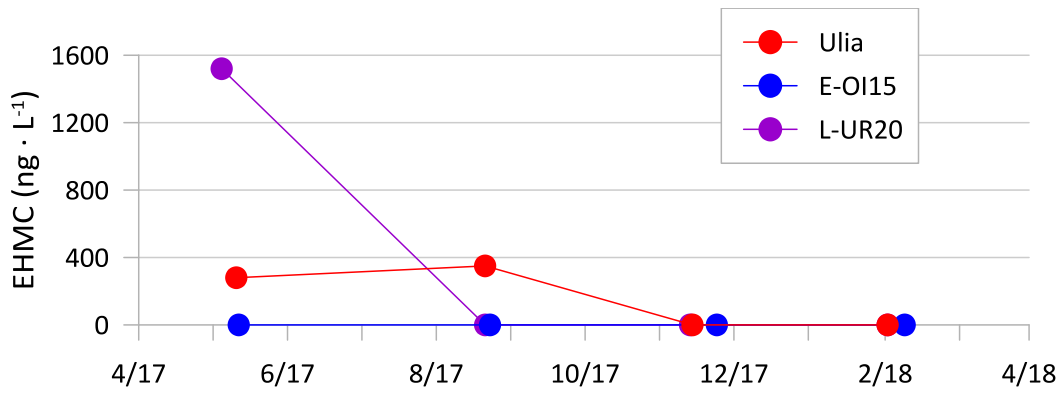


**Figure S2.** Temporal variability of the concentration of diclofenac in the Basque coast, from May 2017 to March 2019. Note: the values below the limit of quantification have been considered as zero.

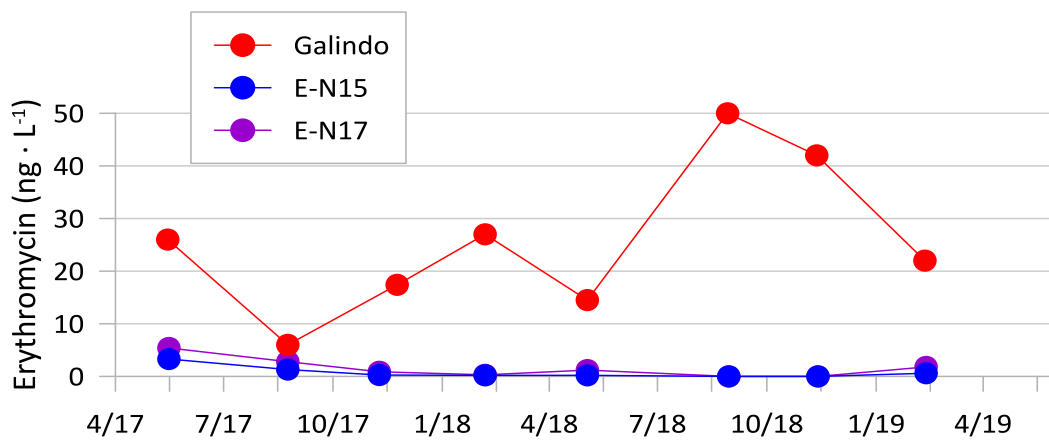
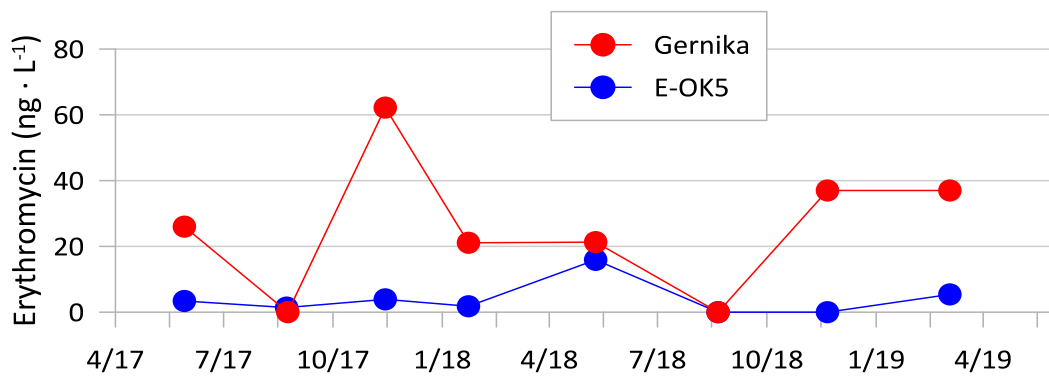
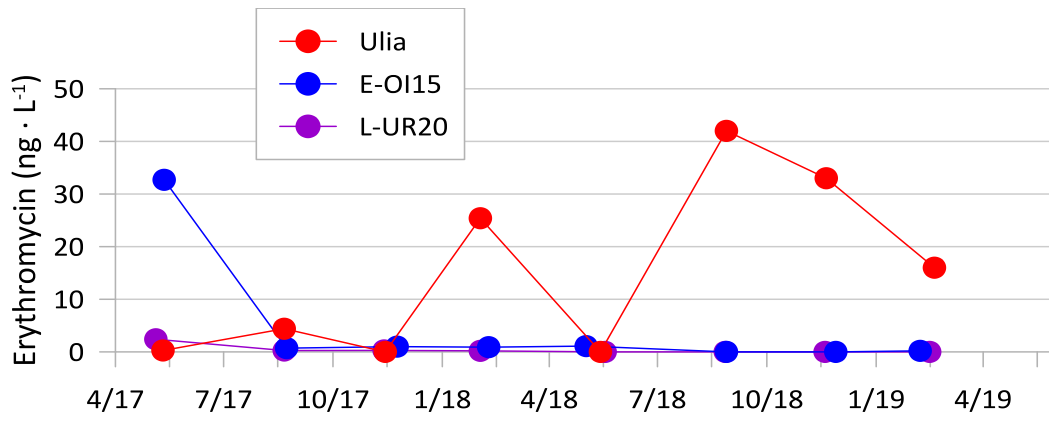


**Figure S3.** Temporal variability of the concentration of BTH in the Basque coast, from May 2017 to March 2019. Note: the values below the limit of quantification have been considered as zero.

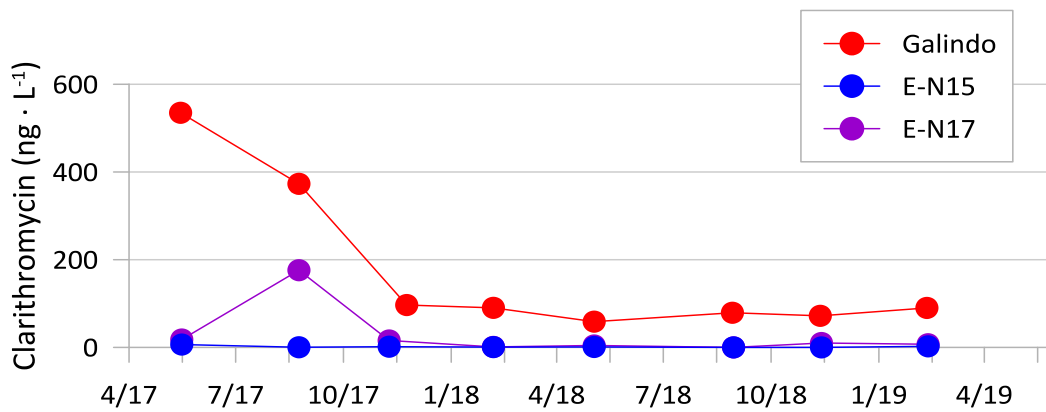
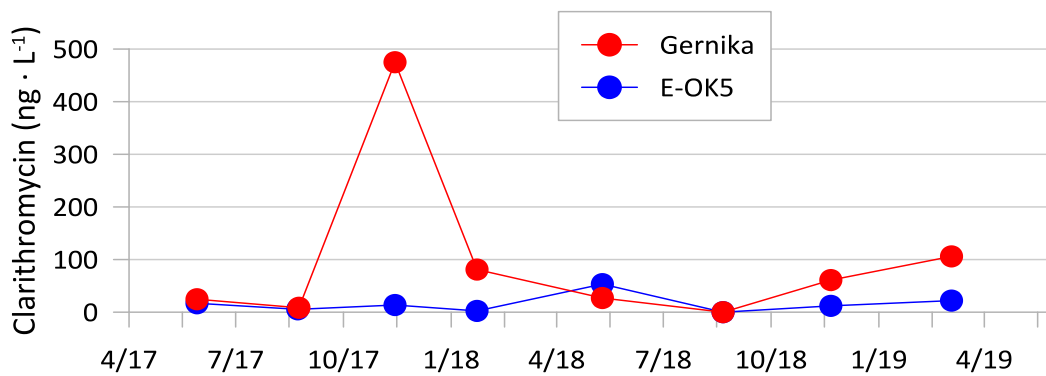
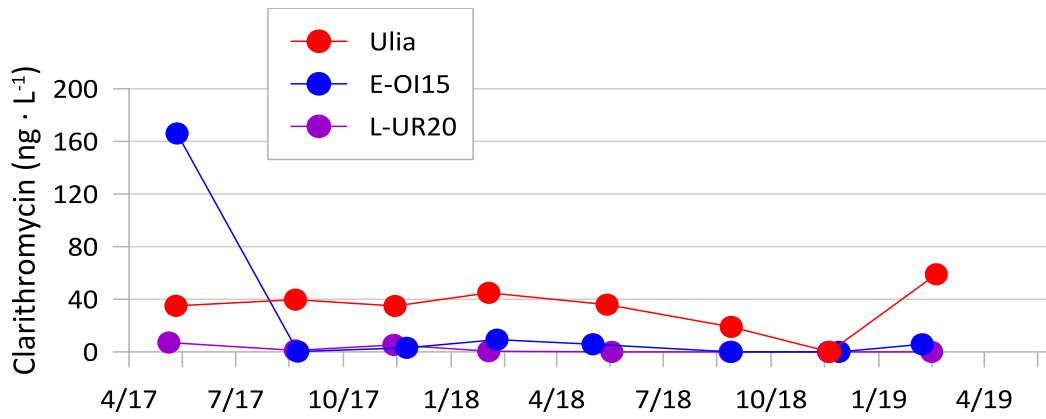




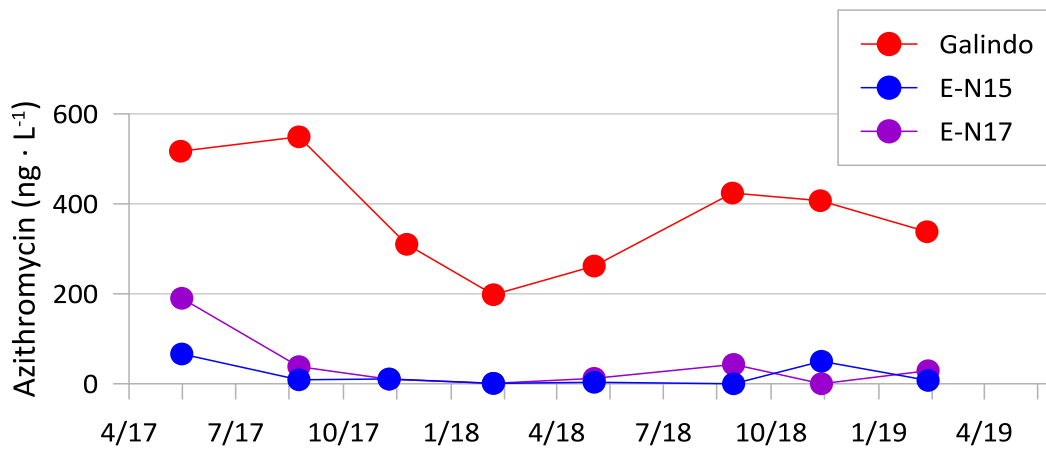
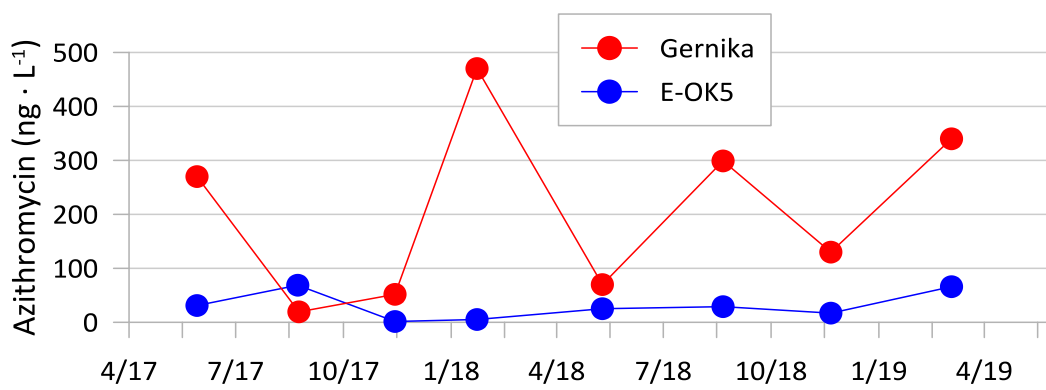
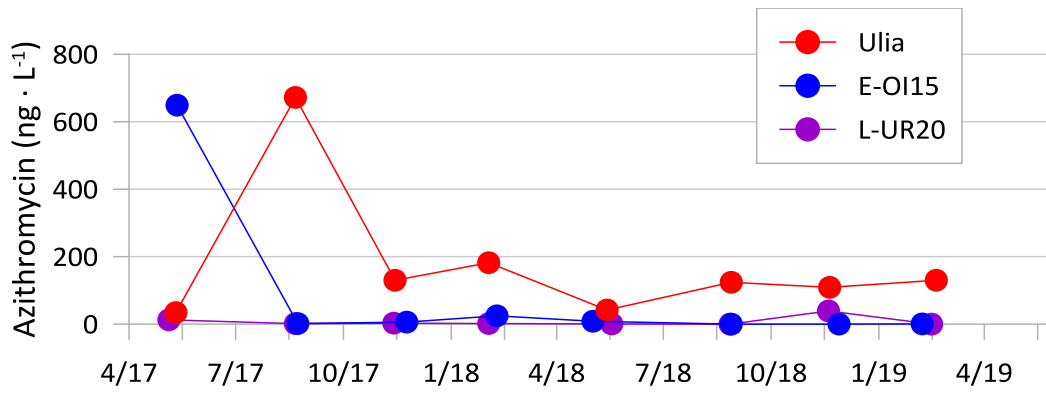
**Figure S4.** Temporal variability of the concentration of EHMC in the Basque coast, from May 2017 to March 2019. Note: the values below the limit of quantification have been considered as zero.



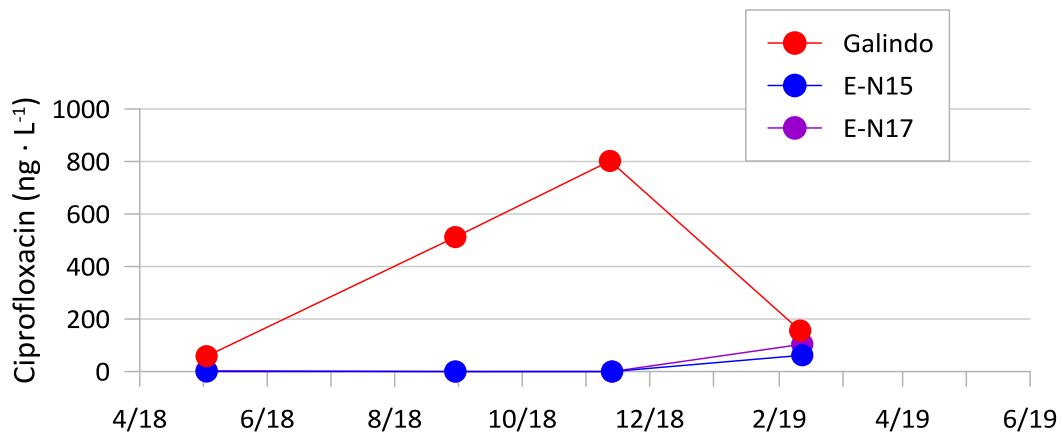
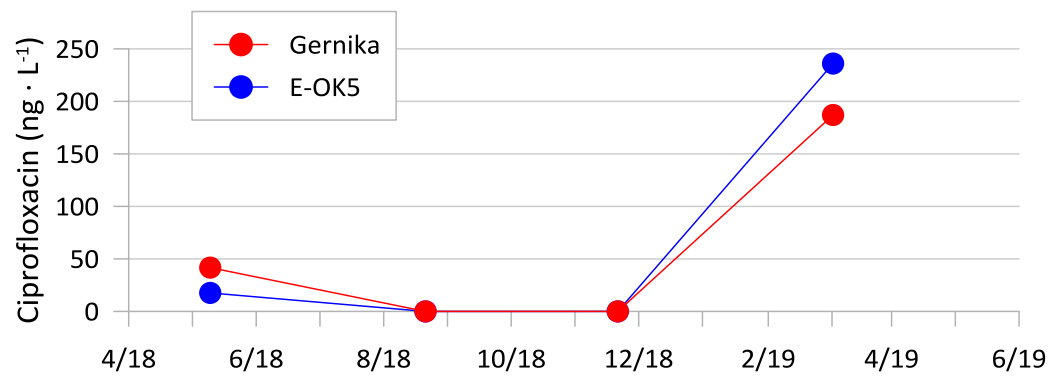
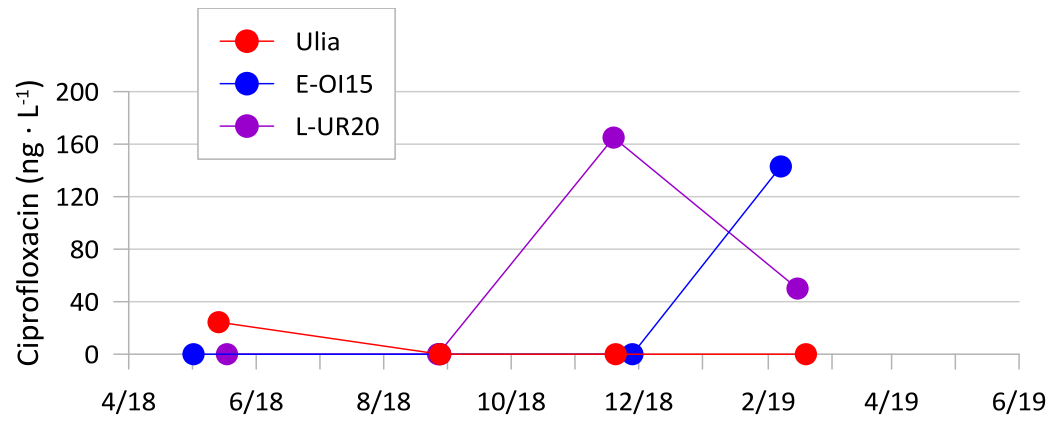
**Figure S5.** Temporal variability of the concentration of erythromycin in the Basque coast, from May 2017 to March 2019. Note: the values below the limit of quantification have been considered as zero.



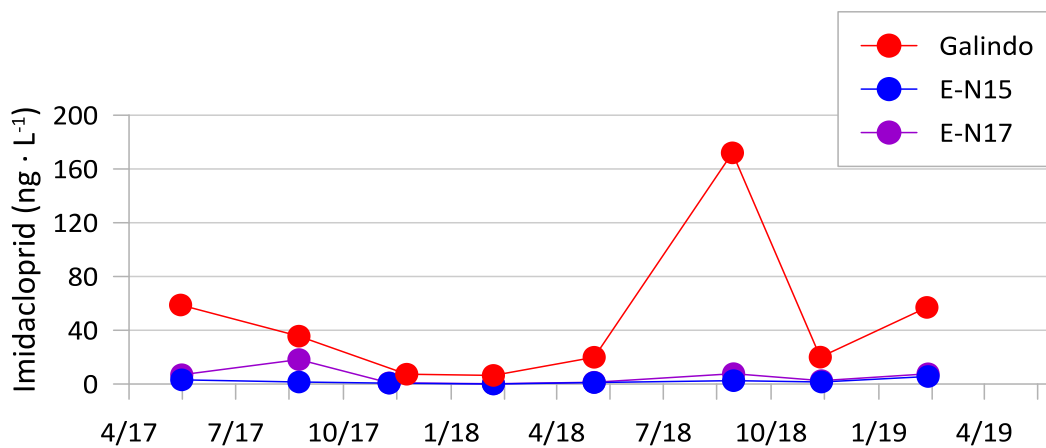
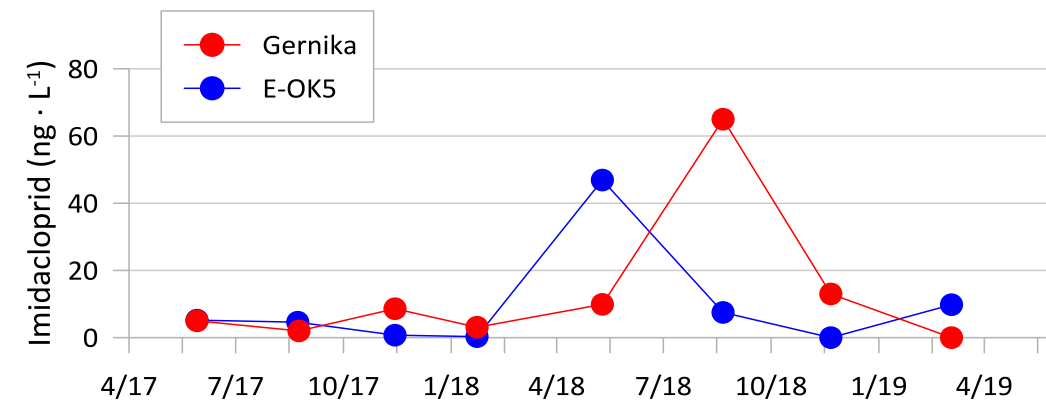
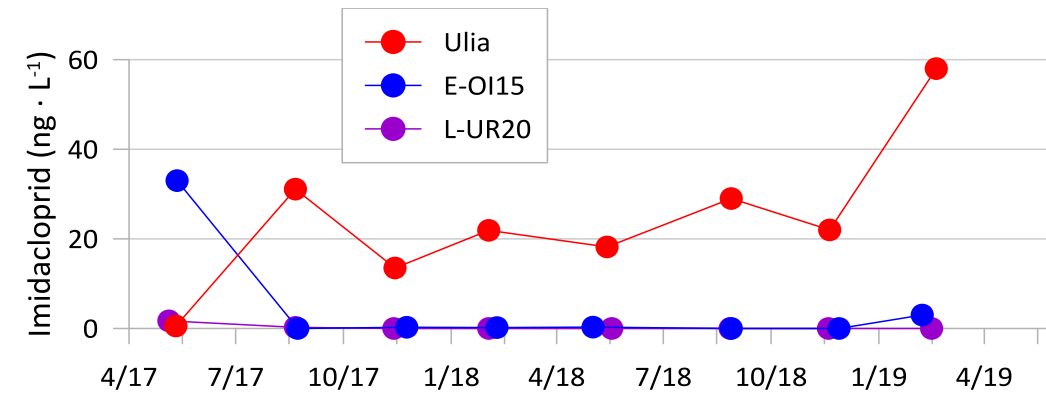
**Figure S6.** Temporal variability of the concentration of clarithromycin in the Basque coast, from May 2017 to March 2019. Note: the values below the limit of quantification have been considered as zero.



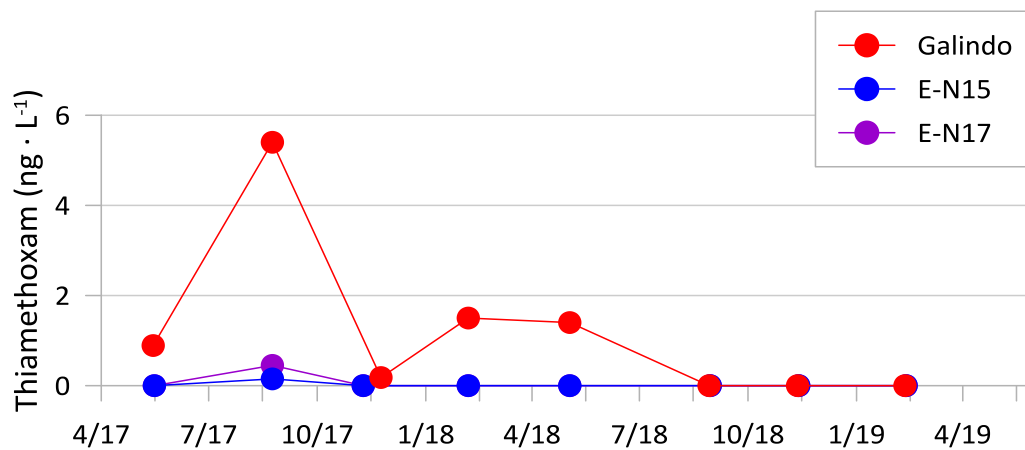
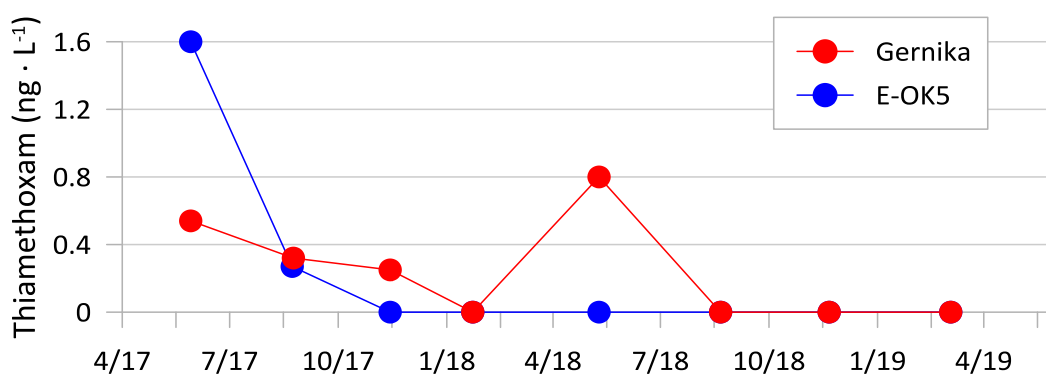
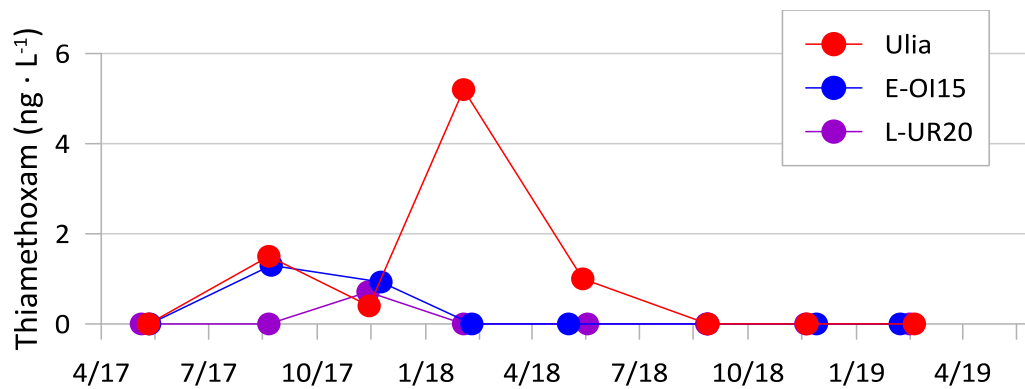
**Figure S7.** Temporal variability of the concentration of azithromycin in the Basque coast, from May 2017 to March 2019. Note: the values below the limit of quantification have been considered as zero.



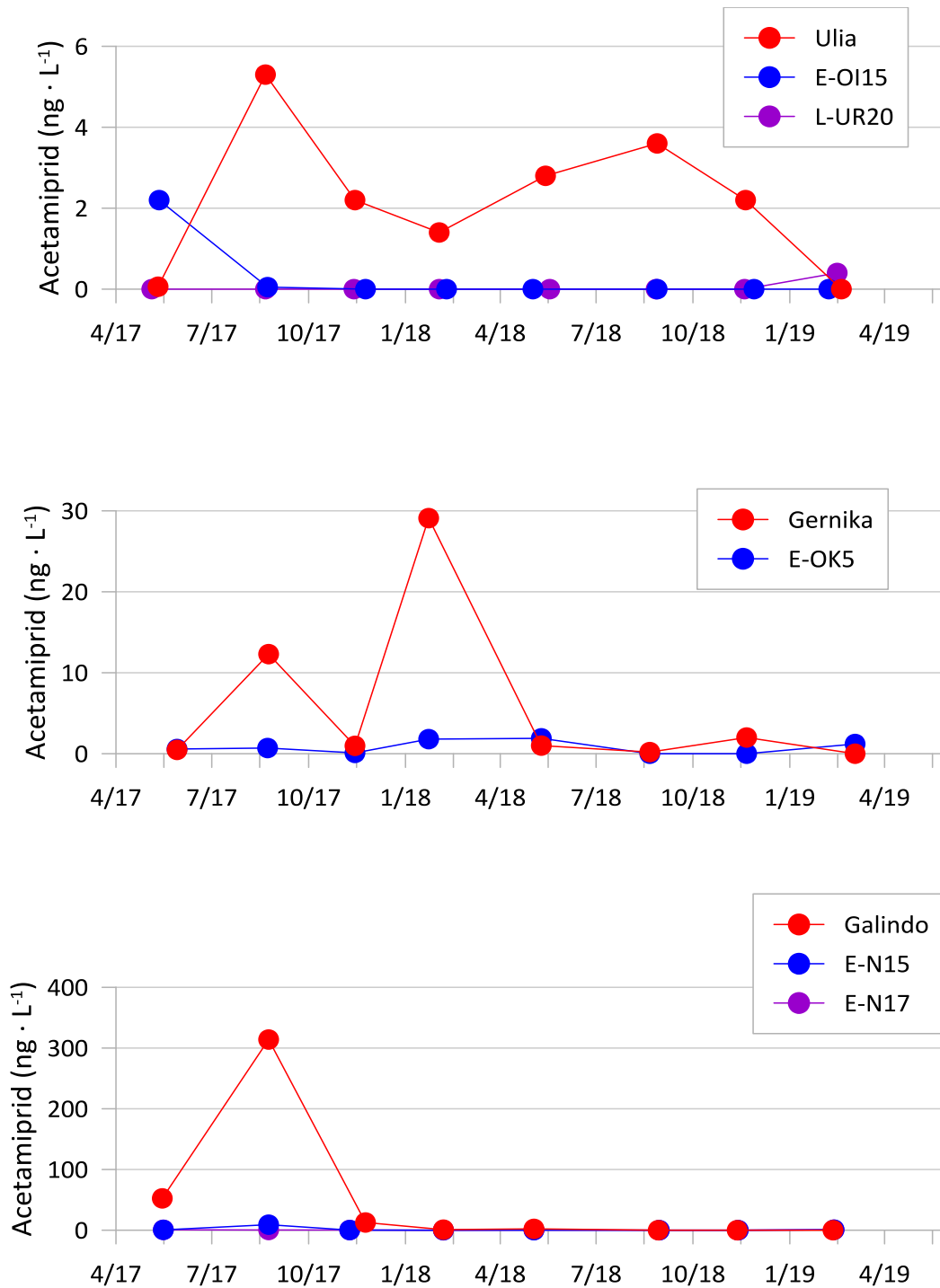
**Figure S8.** Temporal variability of the concentration of ciprofloxacin in the Basque coast, from May 2017 to March 2019. Note: the values below the limit of quantification have been considered as zero.



**Figure S9.** Temporal variability of the concentration of imidacloprid in the Basque coast, from May 2017 to March 2019. Note: the values below the limit of quantification have been considered as zero.

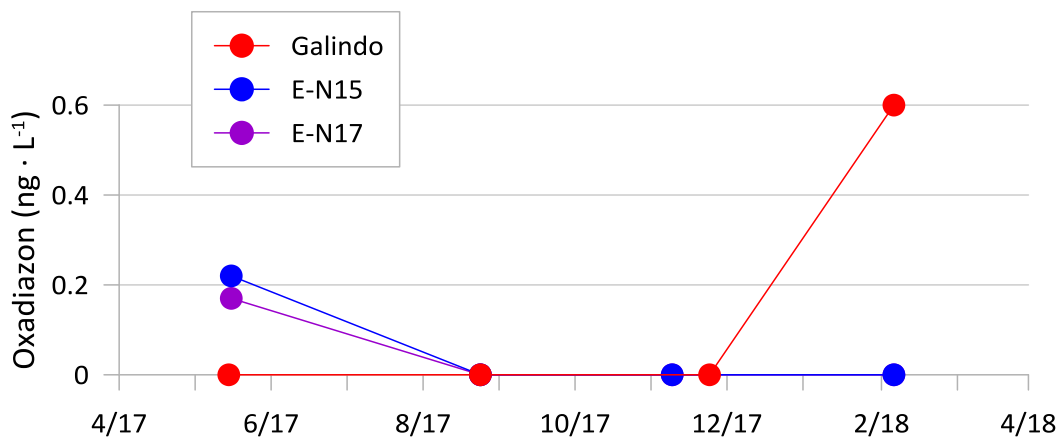
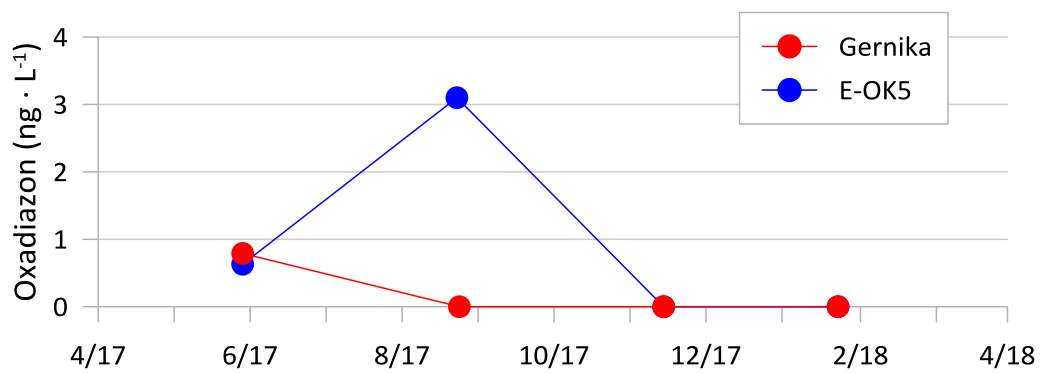
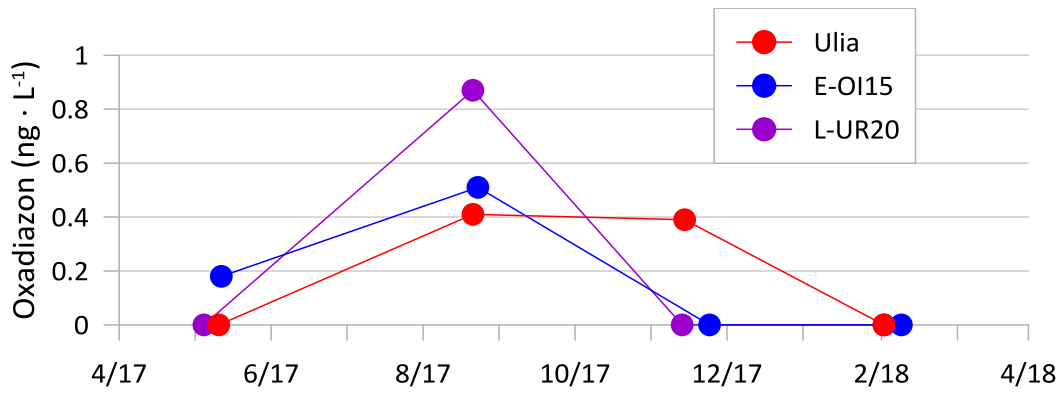


**Figure S10.** Temporal variability of the concentration of thiamethoxam in the Basque coast, from May 2017 to March 2019. Note: the values below the limit of quantification have been considered as zero.

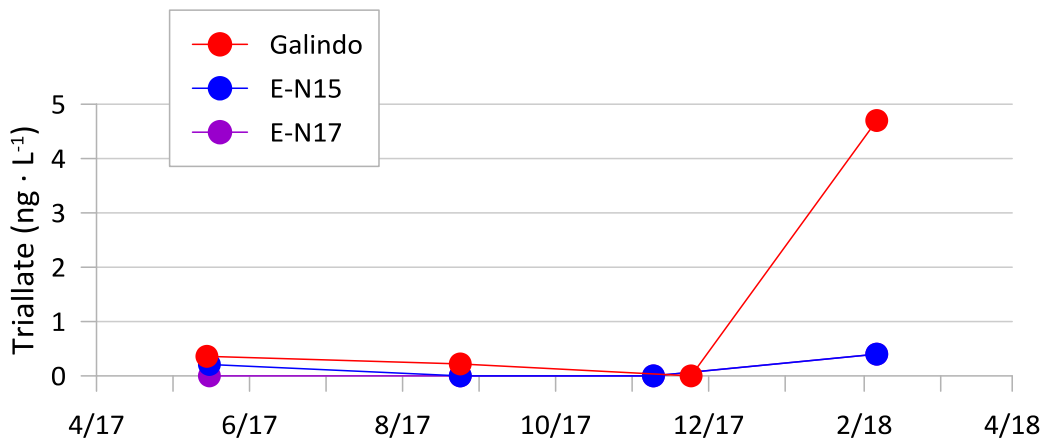
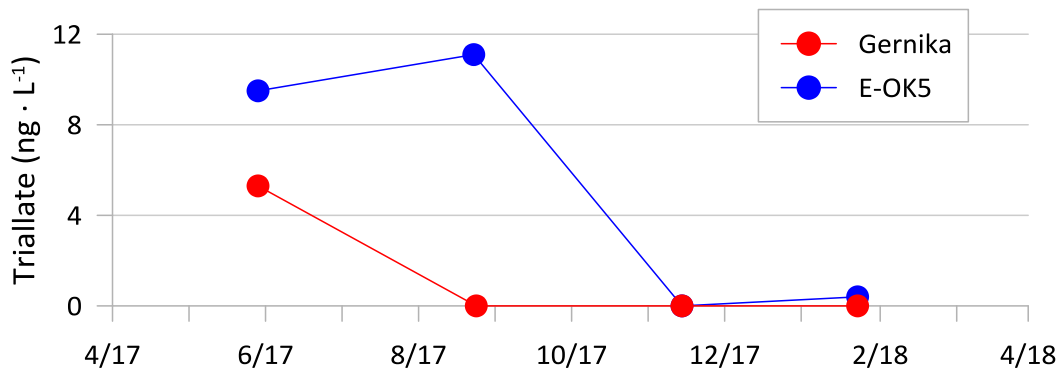
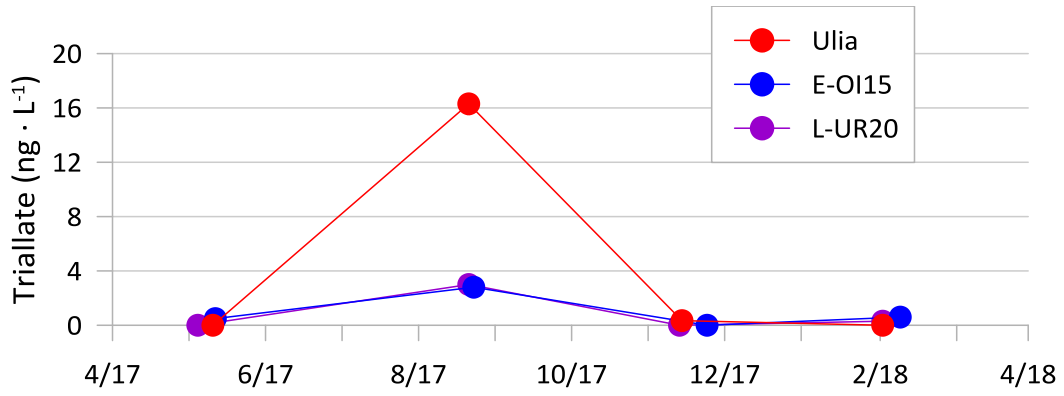


**Figure S11.** Temporal variability of the concentration of acetaminiprid in the Basque coast, from May 2017 to March 2019. Note: the values below the limit of quantification have been considered as zero.





**Figure S12.** Temporal variability of the concentration of oxadiazon in the Basque coast, from May 2017 to March 2019. Note: the values below the limit of quantification have been considered as zero.



**Figure S13.** Temporal variability of the concentration of triallate in the Basque coast, from May 2017 to March 2019. Note: the values below the limit of quantification have been considered as zero.

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