

Supplementary Information

Multiresidue Method for Quantification of Nitroimidazoles, Anthelmintics, Amphenicols, and Dyes in Meat, Shrimp, and Fish by Using a Modified QuEChERS-type Extraction and Isotope Dilution Ultra-High Performance Liquid Chromatography-Tandem Mass Spectrometry

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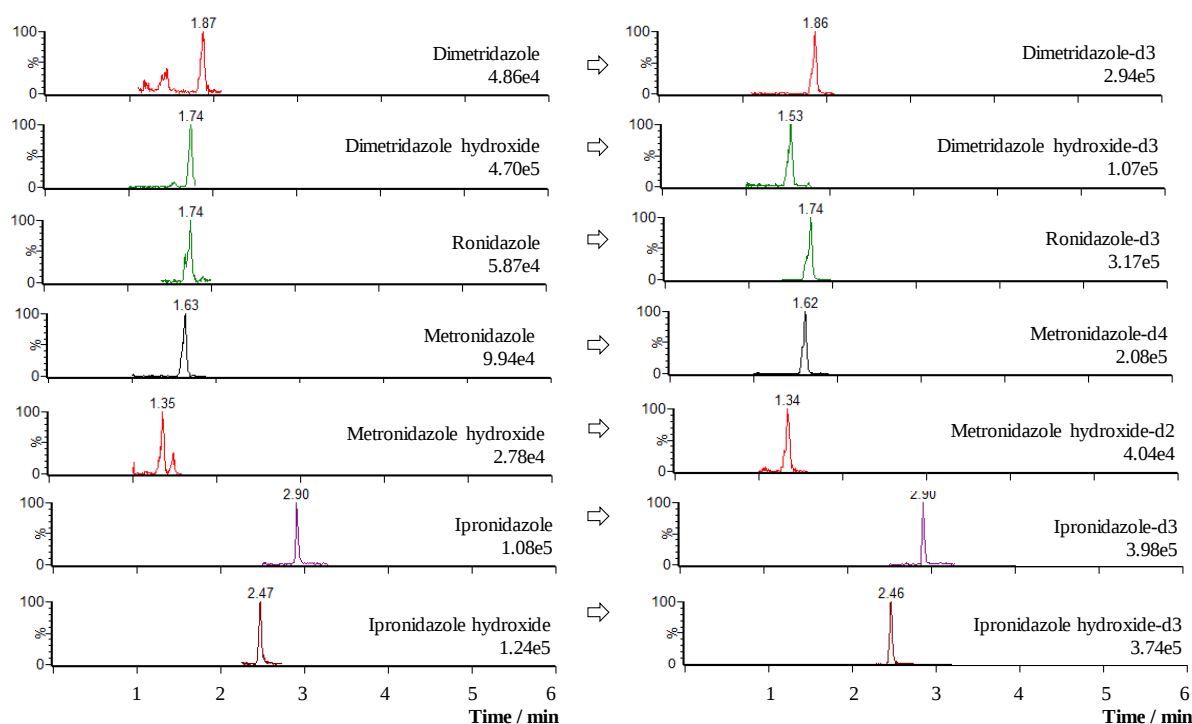


Figure S1. UHPLC-MS/MS TIC for a fish matrix blank fortified at the LOQ with nitroimidazoles and their corresponding IS.

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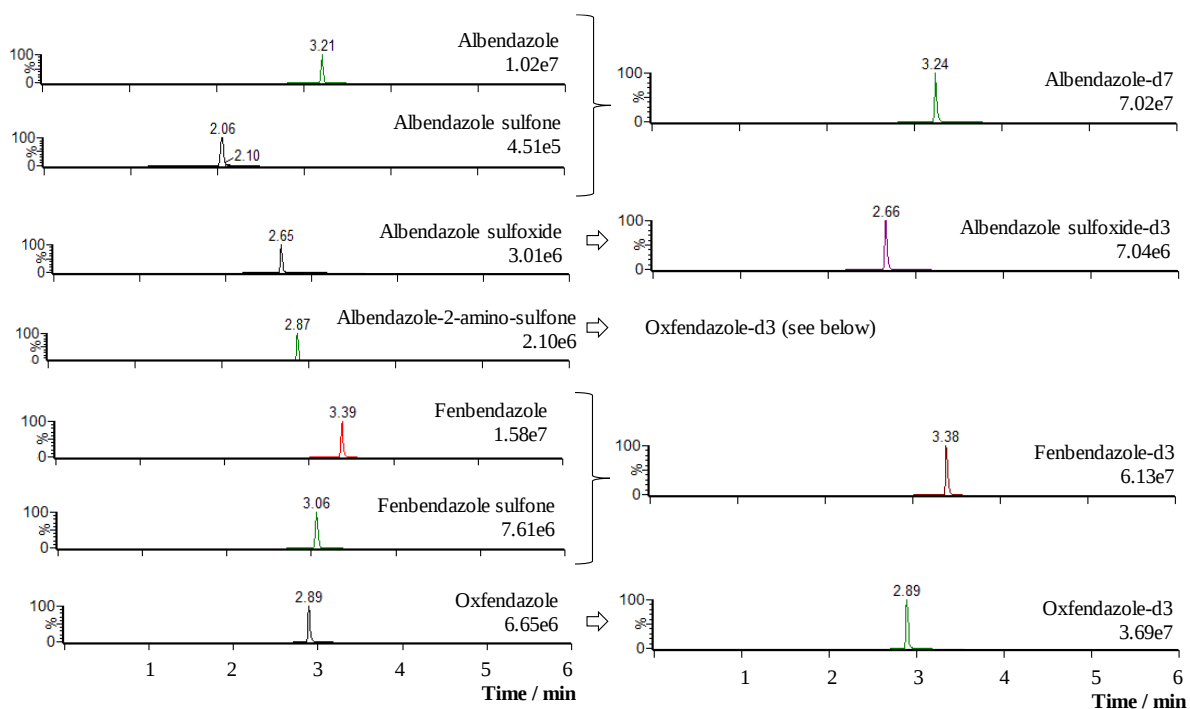


Figure S2. UHPLC-MS/MS TIC for a fish matrix blank fortified at the LOQ with benzimidazoles and their corresponding IS (part 1).

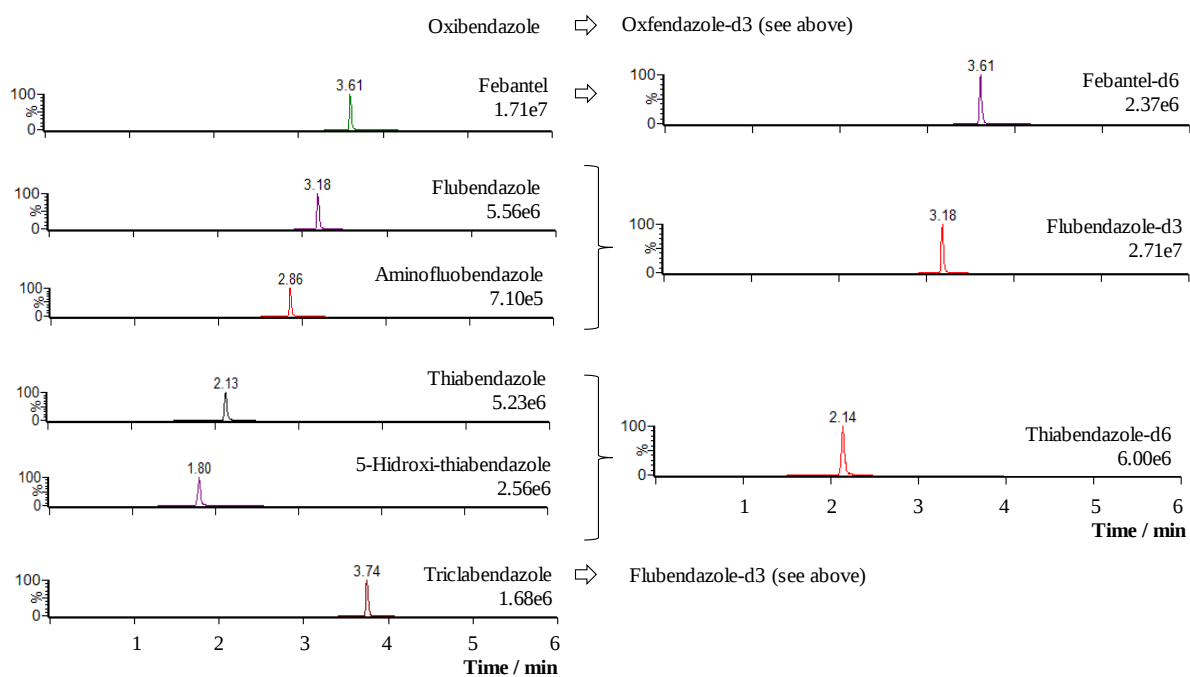


Figure S3. UHPLC-MS/MS TIC for a fish matrix blank fortified at the LOQ with benzimidazoles and their corresponding IS (part 2).

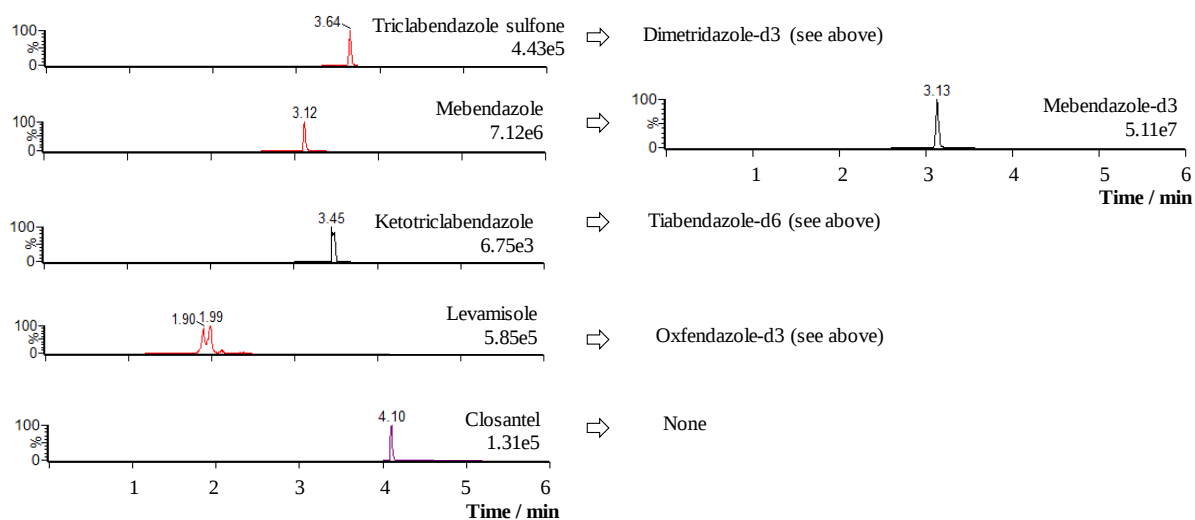


Figure S4. UHPLC-MS/MS TIC for a fish matrix blank fortified at the LOQ with benzimidazoles, levamisole, closantel, and their corresponding IS.

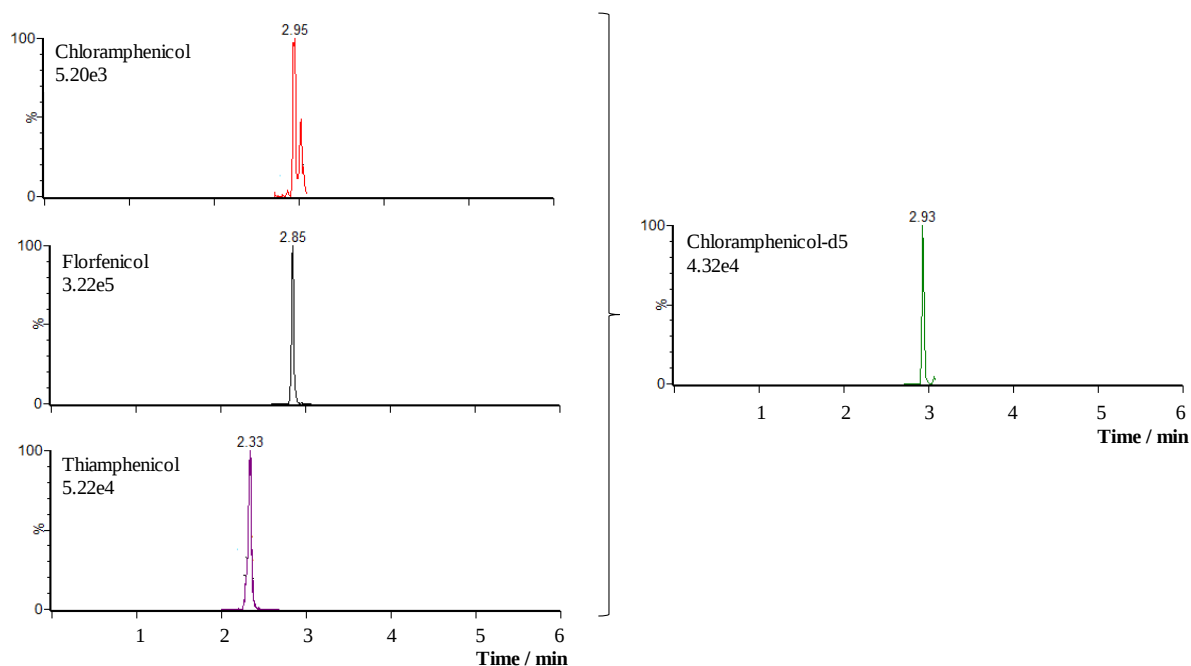


Figure S5. UHPLC-MS/MS TIC for a fish matrix blank fortified at the LOQ with amphenicols and their corresponding IS.

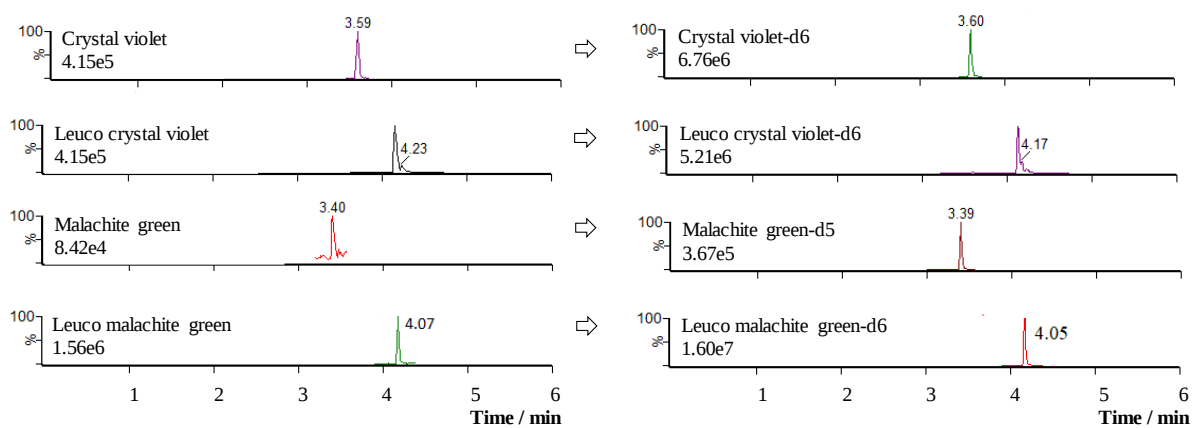


Figure S6. UHPLC-MS/MS TIC for a fish matrix blank fortified at the LOQ with dyes and their corresponding IS.

Table S1. LOQ and linear ranges of matrix-fortified calibration curves for nitroimidazoles, benzimidazoles, levamisole, closantel, amphenicols, and dyes

Analyte	Poultry, pork, beef, horse		Shrimp, fish	
	LOQ / ($\mu\text{g kg}^{-1}$)	Upper level / ($\mu\text{g kg}^{-1}$)	LOQ / ($\mu\text{g kg}^{-1}$)	Upper level / ($\mu\text{g kg}^{-1}$)
Dimetridazole	1.50	9.0	1.50	9.0
Dimetridazole hydroxide	1.50	9.0	1.50	9.0
Ronidazole	1.50	9.0	1.50	9.0
Metronidazole	1.50	9.0	1.50	9.0
Metronidazole hydroxide	1.50	9.0	1.50	9.0
Iprnidazole	1.50	9.0	1.50	9.0
Iprnidazole hydroxide	1.50	9.0	1.50	9.0
Albendazole	5.0	30.0	2.50	30.0
Albendazole sulfone	5.0	30.0	2.50	30.0
Albendazole sulfoxide	5.0	30.0	2.50	30.0
Albendazole 2-amino-sulfone	5.0	30.0	2.50	30.0
Fenbendazole	5.0	30.0	2.50	30.0
Fenbendazole sulfone	5.0	30.0	2.50	30.0
Oxfendazole	5.0	30.0	2.50	30.0
Oxibendazole	5.0	30.0	2.50	30.0
Febantel	5.0	30.0	2.50	30.0
Flubendazole	5.0	30.0	2.50	30.0
Aminoflubendazole	5.0	30.0	2.50	30.0
Thiabendazole	5.0	30.0	2.50	30.0
5-Hydroxy-thiabendazole	5.0	30.0	2.50	30.0
Triclabendazole	5.0	30.0	2.50	30.0
Triclabendazole sulfone	5.0	30.0	2.50	30.0
Mebendazole	5.0	30.0	2.50	30.0
Ketotriclabendazole	5.0	30.0	–	–
Levamisole	5.0	30.0	2.50	30.0
Closantel	5.0	30.0	2.50	30.0
Chloramphenicol	0.10	0.60	–	–
Florfenicol	5.0	30.0	–	–
Thiamphenicol	5.0	30.0	–	–
Malachite green	–	–	0.50	3.0
Leuco malachite green	–	–	0.50	3.0
Crystal violet	–	–	0.50	3.0
Leuco crystal violet	–	–	0.50	3.0

Table S2. Determination coefficients (R^2) of matrix-fortified calibration curves for nitroimidazoles, benzimidazoles, levamisole, closantel, amphenicols, and dyes

Analyte	R^2					
	Poultry	Pork	Beef	Horse	Shrimp	Fish
Dimetridazole	0.9810	0.9973	0.9868	0.9410	0.9907	0.9900
Dimetridazole hydroxide	0.9628	0.9789	0.9596	0.9910	0.9869	0.9698
Ronidazole	0.9800	0.9848	0.9880	0.9980	0.9930	0.9855
Metronidazole	0.9743	0.9927	0.9873	0.9890	0.9933	0.9895
Metronidazole hydroxide	0.9330	0.9799	0.9861	0.9970	0.9833	0.9844
Iprnidazole	0.9521	0.9934	0.9802	0.9860	0.9810	0.9687
Iprnidazole hydroxide	0.9552	0.9893	0.9910	0.9970	0.9864	0.9726
Albendazole	0.9815	0.9902	0.9969	1.0000	0.9597	0.9431
Albendazole sulfone	0.9826	0.9927	0.9951	1.0000	0.9762	0.9644
Albendazole sulfoxide	0.9800	0.9702	0.9708	0.9840	0.9854	0.9813
Albendazole 2-amino-sulfone	0.9659	0.9806	0.9891	0.9850	0.9715	0.9725
Fenbendazole	0.9872	0.9950	0.9966	0.9960	0.9923	0.9841
Fenbendazole sulfone	0.9867	0.9984	0.9950	0.9980	0.9923	0.9862
Oxfendazole	0.9860	0.9963	0.9939	0.9970	0.9755	0.9106
Oxibendazole	0.9813	0.9949	0.9875	0.9970	0.9590	0.9579
Febantel	0.9888	0.9980	0.9939	0.9750	0.9932	0.9953
Flubendazole	0.9849	0.9952	0.9798	0.9990	0.9799	0.9097
Aminoflubendazole	0.9637	0.9863	0.9894	0.9840	0.9695	0.9482
Thiabendazole	0.9885	0.9914	0.9858	0.9910	0.9879	0.9893
5-Hydroxy-thiabendazole	–	0.9838	0.9906	0.9860	0.9742	0.9571
Triclabendazole	0.9538	0.9939	0.9980	0.9940	0.9844	0.9785
Triclabendazole sulfone	–	0.9393	0.9790	0.9910	0.9376	0.9667
Mebendazole	0.9530	0.9952	0.9821	0.9990	0.9760	0.9933
Ketotriclabendazole	–	–	0.9930	0.9290	–	–
Levamisole	0.9799	0.9756	0.9594	0.9770	0.9623	0.9900
Closantel	0.9752	0.9052	0.9075	0.9880	0.9808	0.9846
Chloramphenicol	0.9820	0.9340	0.9490	0.9500	–	–
Florfenicol	0.9590	0.9670	0.9770	0.9260	–	–
Thiamphenicol	0.9345	0.9485	0.9625	0.9155	–	–
Malachite green	–	–	–	–	0.9970	0.9940
Leuco malachite green	–	–	–	–	0.9970	0.9980
Crystal violet	–	–	–	–	0.9885	0.9755
Leuco crystal violet	–	–	–	–	0.9895	0.9885

Table S3. Analyte recovery (f_{REC}) determined from fortified matrix blanks with nitroimidazoles, benzimidazoles, levamisole, closantel, amphenicols, and dyes

Analyte	f_{REC} in meat						f_{REC} in shrimp and fish			
	Conc. / ($\mu\text{g kg}^{-1}$)	Poultry / %	Pork / %	Beef / %	Horse / %	Criteria / %	Conc. / ($\mu\text{g kg}^{-1}$)	Shrimp / %	Fish / %	Criteria / %
Dimetridazole	1.5	104.60	96.00	97.30	89.00	70 to 120	1.5	108.50	96.40	70 to 120
Dimetridazole hydroxide	1.5	113.30	93.90	85.50	104.70	70 to 120	1.5	92.80	100.40	70 to 120
Ronidazole	1.5	100.30	104.80	99.50	89.70	70 to 120	1.5	107.60	105.20	70 to 120
Metronidazole	1.5	96.80	106.40	100.30	90.50	70 to 120	1.5	102.90	93.50	70 to 120
Metronidazole hydroxide	1.5	112.50	93.10	101.40	93.40	70 to 120	1.5	96.10	83.90	70 to 120
Iprnidazole	1.5	96.30	111.70	94.90	113.90	70 to 120	1.5	99.30	75.00	70 to 120
Iprnidazole hydroxide	1.5	104.10	105.00	100.40	93.40	70 to 120	1.5	116.60	90.40	70 to 120
Albendazole	100.0	95.40	110.00	103.30	101.50	80 to 120	5.0	102.00	94.80	70 to 120
Albendazole sulfone	100.0	98.30	105.70	99.90	102.20	80 to 120	5.0	91.20	95.80	70 to 120
Albendazole sulfoxide	100.0	96.90	103.40	101.60	89.20	80 to 120	5.0	98.20	96.70	70 to 120
Albendazole 2-amino-sulfone	100.0	96.10	105.30	99.50	114.60	80 to 120	5.0	96.90	93.90	70 to 120
Fenbendazole	50.0	99.20	101.70	96.90	96.90	80 to 120	5.0	94.10	98.30	70 to 120
Fenbendazole sulfone	100.0	96.50	102.00	94.90	104.00	80 to 120	10.0	95.10	93.00	80 to 120
Oxfendazole	50.0	98.50	103.30	96.70	97.90	80 to 120	5.0	101.10	103.10	70 to 120
Oxibendazole	50.0	99.50	107.80	102.10	95.80	80 to 120	5.0	107.50	105.30	70 to 120
Febantel	50.0	96.90	107.70	104.60	94.10	80 to 120	5.0	100.10	96.20	70 to 120
Flubendazole	10.0	97.60	101.30	96.70	98.10	80 to 120	5.0	97.10	99.90	70 to 120
Aminoflubendazole	10.0	96.10	106.80	104.20	104.70	80 to 120	5.0	109.60	101.40	70 to 120
Thiabendazole	10.0	96.80	99.80	101.90	97.60	80 to 120	5.0	98.10	101.60	70 to 120
5-Hydroxy-thiabendazole	10.0	–	94.80	106.70	–	80 to 120	5.0	101.70	97.40	70 to 120
Triclabendazole	10.0	100.30	90.70	90.80	94.60	80 to 120	5.0	115.70	113.10	70 to 120
Triclabendazole-sulfone	10 (5 for pork)	–	91.10	101.10	88.30	80 to 120	5.0	106.60	105.60	70 to 120
Mebendazole	10.0	94.40	102.70	92.60	101.50	80 to 120	5.0	97.20	98.10	70 to 120
Ketotriclabendazole	10.0	–	–	102.90	107.10	80 to 120	5.0	–	–	70 to 120
Levamisole	10.0	93.90	–	104.40	94.90	80 to 120	5.0	108.20	115.90	70 to 120
Closantel	10 (5 for pork)	100.90	106.40	103.20	101.50	80 to 120	5.0	89.40	87.80	70 to 120
Chloramphenicol	0.10	96.20	95.60	99.20	175.50	50 to 120	–	–	–	–
Florfenicol	5.0	94.70	104.70	109.00	100.40	70 to 120	–	–	–	–
Thiamphenicol	5.0	93.70	102.10	81.70	110.90	70 to 120	–	–	–	–
Malachite green	0.5	–	–	–	–	50 to 120	0.50	80.90	107.80	50 to 120
Leuco malachite green	0.5	–	–	–	–	50 to 120	0.50	101.10	104.30	50 to 120
Crystal violet	0.5	–	–	–	–	50 to 120	0.50	103.00	101.30	50 to 120
Leuco crystal violet	0.5	–	–	–	–	50 to 120	0.50	107.20	102.70	50 to 120

Table S4. Method's within-laboratory reproducibility (CV)

Analyte	CV in meat						CV in shrimp and fish			
	Conc. / ($\mu\text{g kg}^{-1}$)	Poultry / %	Pork / %	Beef / %	Horse / %	Criteria / %	Conc. / ($\mu\text{g kg}^{-1}$)	Shrimp / %	Fish / %	Criteria / %
Dimetridazole	1.5	11.70	13.60	8.60	13.50	30	1.5	8.95	16.05	30
Dimetridazole hydroxide	1.5	7.80	12.00	12.40	6.60	30	1.5	15.82	11.58	30
Ronidazole	1.5	11.60	10.10	8.00	5.80	30	1.5	7.57	10.03	30
Metronidazole	1.5	7.00	8.00	7.00	7.20	30	1.5	5.87	7.78	30
Metronidazole hydroxide	1.5	12.00	17.00	10.90	12.00	30	1.5	21.70	17.76	30
Iprnidazole	1.5	15.00	15.90	15.30	5.60	30	1.5	18.90	13.71	30
Iprnidazole hydroxide	1.5	10.30	12.00	8.40	7.10	30	1.5	14.28	14.91	30
Albendazole	100.0	5.40	18.10	13.70	2.70	25	5.0	8.53	5.03	30
Albendazole sulfone	100.0	3.50	5.80	2.80	5.40	25	5.0	16.43	16.89	30
Albendazole sulfoxide	100.0	4.20	8.10	4.60	3.30	25	5.0	4.38	6.76	30
Albendazole 2-amino-sulfone	100.0	5.80	10.50	5.90	2.80	25	5.0	12.53	9.74	30
Fenbendazole	50.0	5.90	5.10	4.90	1.20	25	5.0	7.73	9.08	30
Fenbendazole sulfone	100.0	3.30	6.70	5.80	3.30	25	10.0	7.74	10.73	25
Oxfendazole	50.0	5.20	5.70	3.80	4.20	25	5.0	15.97	26.08	30
Oxibendazole	50.0	5.00	9.70	5.10	9.20	25	5.0	20.83	25.35	30
Febantel	50.0	1.80	6.60	5.40	2.60	25	5.0	7.59	6.12	30
Flubendazole	10.0	2.70	6.10	4.00	5.40	25	5.0	18.07	17.43	30
Amino-flubendazole	10.0	9.80	18.10	9.10	3.40	25	5.0	20.82	23.26	30
Thiabendazole	10.0	4.80	5.80	10.70	5.50	25	5.0	8.58	13.07	30
5-Hydroxy-thiabendazole	10.0	—	22.30	11.70	—	25	5.0	12.19	9.48	30
Triclabendazole	10.0	14.70	19.70	6.80	8.50	25	5.0	12.26	12.30	30
Triclabendazole sulfone	10 (5 for pork)	—	19.50	22.20	15.00	25/30	5.0	12.57	8.53	30
Mebendazole	10.0	8.20	5.40	3.40	2.60	25	5.0	12.16	9.70	30
Ketotriclabendazole	10.0	—	—	11.30	15.60	25	5.0	—	—	30
Levamisole	10.0	6.30	7.40	6.50	5.20	25	5.0	17.23	27.39	30
Closantel	10 (5 for beef)	22.70	12.30	12.30	14.40	25/30	5.0	12.65	13.95	30
Chloramphenicol	0.10	13.00	23.90	29.90	54.30	30	—	—	—	—
Florfenicol	5.0	24.80	24.30	18.30	26.50	30	—	—	—	—
Thiamphenicol	5.0	16.00	22.40	17.80	10.30	30	—	—	—	—
Malachite green	—	—	—	—	—	—	0.5	27.20	24.80	30
Leuco malachite green	—	—	—	—	—	—	0.5	5.20	8.00	30
Crystal violet	—	—	—	—	—	—	0.5	17.30	14.20	30
Leuco crystal violet	—	—	—	—	—	—	0.5	11.70	12.70	30

