Supporting information for

Estimated daily intake of residual agricultural chemicals across general Japanese people based on the total diet study from 2019 to 2021

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Table S1 LC-MS/MS conditions for analysis by each institute

	Institute A	Institute B	Institute C	Institute D	Institute E
Instrument	LCMS-8050 (SHIMADZU Co., Kyoto, Japan)	Acquity UPLC I-class PLUS/Xevo TQ-XS (Waters Co., Milford, MA, USA)	Agilent 1260 Infinity/6460 (Agilent Technologies, Santa Clara, CA, USA)	Agilent 1290 Infinity II/6470 (Agilent Technologies)	Acquity/Xevo TQ-MS (Waters Co.)
Analytical column	Inertsil ODS-4 (2.1 mm × 150 mm, 3 µm, GL Sciences Inc., Tokyo, Japan)	Acquity UPLC BEH C18 (2.1 mm × 50 mm, 1.7 µm, Waters Co.)	Ascentis Express C18 (2.1 mm × 100 mm, 2.7 µm, Sigma-Aldrich Co., St. Louis, MO, USA)	Inert Sustain AQ-C18 HP (2.1 mm × 150 mm, 3 µm, GL Sciences Inc.)	InertSustainAQ-C18 (2.1 mm × 150 mm, 5 µm, GL Sciences Inc.)
Column oven	40ºC	40ºC	40ºC	40ºC	40ºC
Mobile phase A	5 mmol/L ammonium acetate in water	2 mmol/L ammonium acetate in water	0.05% (v/v) formic acid in water	0.1% (v/v) formic acid in water	5 mmol/L ammonium acetate in water
Mobile phase B	5 mmol/L ammonium acetate in methanol	Methanol	Methanol	0.1% (v/v) formic acid in acetonitrile	5 mmol/L ammonium acetate in methanol
Gradient elution	Time (min) / B conc. (%) 0 / 10 20 / 95 30 / 95	Time (min) / B conc. (%) 0 / 15 0.19 / 40 0.66 / 40 1.13 / 50 1.51 / 50 3.31 / 95 5.67 / 95 5.86 / 15 8.5 / 15	Time (min) / B conc. (%) 0 / 10 6 / 40 30 / 75 35 / 100 42 / 100 47 / 10 57 / 10	Time (min) / B conc. (%) 0 / 5 1 / 15 3 / 25 8 / 40 12 / 70 21 / 95 26 / 95	Time (min) / B conc. (%) 0 / 15 1 / 15 5 / 50 6 / 70 11 / 95 15 / 95
Flow rate	0.2 mL/min	0.35 mL/min	0.25 mL/min	0.2 mL/min	0.2 mL/min
Injection volume	2 µL	2 µL (5 µL was injected during the measurement of group 14, except for clothianidin)	1 µL	2 µL	5 µL
Ionization mode	ESI (±)	UniSpray (+)	ESI (+)	ESI (±)	ESI (+)
Capillary needle voltage	−3.0 kV, 4.0 kV	3.0 kV	3.0 kV	−2.0 kV, 3.0 kV	0.5 kV
Source temperature	300ºC	150ºC	300ºC	400ºC	150ºC
Desolvation temperature	250ºC	500ºC	100ºC	300ºC	400ºC
Collision gas	Ar	Ar	N_2	N_2	Ar

Table S2 GC-MS/MS conditions for analysis by each institute

<table>
<tr><th></th><th>Institute A</th><th>Institute E</th><th>Institute F</th></tr>
<tr><td>Instrument</td><td>7000D GC/MS Triple Quad (Agilent Technologies, Santa Clara, CA, USA)</td><td>SCION 456-GC/TQ (Bruker Co., Billerica, MA, USA)</td><td>7000D GC/MS Triple Quad (Agilent Technologies)</td></tr>
<tr><td>Analytical column</td><td>DB-5MS (0.25 mm × 30 m, 0.25 µm, Agilent Technologies)</td><td>BPX5 (0.25 mm × 30 m, 0.25 µm, Trajan Scientific Japan, Ltd., Kanagawa, Japan)</td><td>DB-5ms Ultra Inert (0.25 mm × 30 m, 0.25 µm, Agilent Technologies)</td></tr>
<tr><td>Column oven</td><td>Time (min) / Temperature (ºC)
0 / 50
1 / 50
4 / 125
22.5 / 310
40 / 310</td><td>Time (min) / Temperature (ºC)
0 / 50
1 / 50
4 / 125
21.5 / 300
28 / 300</td><td>Time (min) / Temperature (ºC)
4 / 80
7.6 / 170
18.85 / 260
20.85 / 310
30.85 / 310</td></tr>
<tr><td></td><td></td><td></td><td>Time (min) / Temperature (ºC)
0 / 100
0.53 / 100
1.70 / 240
2.70 / 290
22.70 / 290</td></tr>
<tr><td>Source temperature</td><td>290ºC</td><td>250ºC</td><td></td></tr>
<tr><td>Inlet temperature</td><td>250ºC</td><td>250ºC</td><td>250ºC</td></tr>
<tr><td>Carrier gas</td><td>He (1.2 mL/min)</td><td>He (1.1 mL/min)</td><td>He (1.0 mL/min)</td></tr>
<tr><td>Injection method</td><td>Splitless</td><td>Pulsed splitless</td><td>Bulk injection with using Mass inlet device for GC (LVI-S250, AiSTI SCIENCE Co.)</td></tr>
<tr><td>Injection volume</td><td>2 µL</td><td>1 µL</td><td>20 µL</td></tr>
<tr><td>Ionization mode</td><td>EI</td><td>EI</td><td>EI</td></tr>
</table>

Table S3 MS/MS transition for each analyte

Analyte	Institute	System	Ionization mode	Quantitative ion		Qualitative ion	
				m/z	Collision energy (eV)	*m/z*	Collision energy (eV)
Acephate	A	LC-MS/MS	ESI (+)	184.1 → 143.0	9	184.1 → 95.1	24
	D	LC-MS/MS	ESI (+)	184.0 → 143.0	4	184.0 → 49.1	4
	E	LC-MS/MS	ESI (+)	184.2 → 143.1	8	184.2 → 95.1	20
Acetamiprid	A	LC-MS/MS	ESI (+)	233.0 → 126.0	20	233.0 → 56.1	16
	B	LC-MS/MS	UniSpray (+)	223.0 → 126.0	20	223.0 → 56.1	15
	C	LC-MS/MS	ESI (+)	233.1 → 126	20	233.1 → 56.1	12
	D	LC-MS/MS	ESI (+)	223.1 → 126.0	24	223.1 → 56.1	24
	E	LC-MS/MS	ESI (+)	223.0 → 126.0	20	223.0 → 56.1	15
Acetamiprid metabolite	A	LC-MS/MS	ESI (+)	209.2 → 126.1	19	209.2 → 90.1	32
	E	LC-MS/MS	ESI (+)	209.2 → 126.1	16	209.2 → 90.2	28
BHC	F	GC-MS/MS	EI (−70eV)	217.0 → 181.0	6	219.0 → 183.0	6
γ-BHC	F	GC-MS/MS	EI (−70eV)	217.0 → 181.0	6	219.0 → 183.0	6
Bifenthrin	E	GC-MS/MS	EI (−70eV)	181.0 → 166.0	10	181.0 → 165.0	20
	F	GC-MS/MS	EI (−70eV)	181.0 → 165.0	28	181.0 → 166.0	12
Boscalid	A	LC-MS/MS	ESI (+)	342.9 → 307.0	20	342.9 → 140.0	21
	B	LC-MS/MS	UniSpray (+)	343.1 → 307.1	18	343.1 → 271.6	30
	C	LC-MS/MS	ESI (+)	343 → 307	16	343 → 139.9	16
	D	LC-MS/MS	ESI (+)	343.0 → 307.0	20	343.0 → 271.1	20
	E	LC-MS/MS	ESI (+)	343.0 → 140.0	20	343.0 → 307.0	23
	F	GC-MS/MS	EI (−70eV)	140.0 → 112.0	12	342.0 → 140.0	14
Buprofezin	D	LC-MS/MS	ESI (+)	306.2 → 201.1	12	306.2 → 57.1	12
	E	LC-MS/MS	ESI (+)	306.3 → 201.1	12	306.3 → 116.1	16
Chlordane (trans)	F	GC-MS/MS	EI (−70eV)	375.0 → 266.0	20	373.0 → 266.0	20
Chlordane (cis)	F	GC-MS/MS	EI (−70eV)	375.0 → 266.0	20	373.0 → 266.0	20
Chlorpyrifos	A	LC-MS/MS	ESI (+)	350.0 → 198.0	19	350.0 → 97.2	31
	B	LC-MS/MS	UniSpray (+)	350.0 → 197.9	24	350.0 → 322.0	12
	D	LC-MS/MS	ESI (+)	349.9 → 197.9	20	349.9 → 97.0	20
	E	GC-MS/MS	EI (−70eV)	314.0 → 258.0	10	314.0 → 286.0	10
	F	GC-MS/MS	EI (−70eV)	314.0 → 258.0	14	316.0 → 260.0	14
Clothianidin	A	LC-MS/MS	ESI (+)	250.1 → 132.0	15	260.1 → 169.0	13
	B	LC-MS/MS	UniSpray (+)	250.0 → 169.0	10	250.0 → 132.0	15
	C	LC-MS/MS	ESI (+)	250 → 169	8	250 → 131.9	12
	D	LC-MS/MS	ESI (+)	250.0 → 169.0	12	250.0 → 131.9	12
	E	LC-MS/MS	ESI (+)	250.0 → 169.0	10	250.0 → 132.0	10
Dinotefuran	D	LC-MS/MS	ESI (+)	203.1 → 129.0	8	203.1 → 43.1	8
Fenitrothion	A	GC-MS/MS	EI (−70eV)	277.0 → 260.0	6	277.0 → 109.0	14
	B	LC-MS/MS	UniSpray (+)	278.0 → 124.9	24	278.0 → 109.1	20
	E	GC-MS/MS	EI (−70eV)	277.0 → 260.0	10	277.0 → 109.0	10
	F	GC-MS/MS	EI (−70eV)	277.0 → 260.0	2	277.0 → 109.0	16
Fenpropathrin	F	GC-MS/MS	EI (−70eV)	265.0 → 210.0	8	265.0 → 89.0	40
Fluazifop	E	LC-MS/MS	ESI (+)	328.4 → 282.2	18	328.4 → 91.2	32
Fluazifop butyl	E	LC-MS/MS	ESI (+)	384.4 → 282.2	20	384.4 → 328.3	16

Table S3 Continued

Analyte	Institute	System	Ionization mode	Quantitative ion		Qualitative ion	
				m/z	Collision energy (eV)	*m/z*	Collision energy (eV)
Flubendiamide	A	LC-MS/MS	ESI (−)	681.1 → 254.2	28	681.1 → 274.2	17
	C	LC-MS/MS	ESI (−)	681 → 274	10	681 → 254	14
	D	LC-MS/MS	ESI (−)	681.0 → 274.0	12	681.0 → 254.0	12
Halfenprox	F	GC-MS/MS	EI (−70eV)	265.0 → 117.0	12	263.0 → 117.0	12
Heptachlor	E	GC-MS/MS	EI (−70eV)	272.0 → 237.0	30	272.0 → 141.0	30
Heptachlor epoxide (isomer A)	E	GC-MS/MS	EI (−70eV)	183.0 → 119.0	20	183.0 → 84.0	20
Heptachlor epoxide (isomer B)	E	GC-MS/MS	EI (−70eV)	353.0 → 217.0	40	353.0 → 263.0	40
Hexazinone	B	LC-MS/MS	UniSpray (+)	253.1 → 171.1	16	253.1 → 71.0	30
	C	LC-MS/MS	ESI (+)	253.2 → 171	12	253.2 → 71.1	36
	D	LC-MS/MS	ESI (+)	253.2 → 171.1	16	253.2 → 85.1	16
	E	GC-MS/MS	EI (−70eV)	171.0 → 71.0	10	171.0 → 85.0	10
Imidacloprid	D	LC-MS/MS	ESI (+)	256.1 → 209	16	256.1 → 175.1	16
Indoxacarb	E	LC-MS/MS	ESI (+)	528.3 → 150.1	22	528.3 → 150.1	44
Methamidophos	B	LC-MS/MS	UniSpray (+)	141.9 → 93.9	12	141.9 → 124.8	14
	D	LC-MS/MS	ESI (+)	142.0 → 124.9	12	142.0 → 94.0	12
	E	LC-MS/MS	ESI (+)	141.7 → 94.2	12	141.7 → 125.1	12
Nitenpyram	D	LC-MS/MS	ESI (+)	271.1 → 225.1	8	271.1 → 56.1	8
Novaluron	A	LC-MS/MS	ESI (+)	493.0 → 158.1	19	493.0 → 141.0	46
	B	LC-MS/MS	UniSpray (+)	493.0 → 158.0	15	493.0 → 141.0	30
	C	LC-MS/MS	ESI (+)	493 → 158	20	–	–
	D	LC-MS/MS	ESI (−)	491.1 → 470.9	12	491.1 → 305.0	12
	E	LC-MS/MS	ESI (+)	493.0 → 158.0	20	493.0 → 141.0	35
Pyridaben	E	LC-MS/MS	ESI (+)	365.3 → 147.3	24	365.3 → 309.2	12
	C	LC-MS/MS	ESI (+)	365.2 → 309	8	365.2 → 147.1	32
	D	LC-MS/MS	ESI (+)	365.2 → 309.0	12	365.2 → 147.1	12
Sulfoxaflor	D	LC-MS/MS	ESI (+)	278.1 → 174.0	4	278.1 → 154.0	4
Tefluthrin	F	GC-MS/MS	EI (−70eV)	177.0 → 127.0	16	177.0 → 137.0	14
Thiacloprid	A	LC-MS/MS	ESI (+)	253.1 → 126.1	22	253.1 → 90.1	38
	B	LC-MS/MS	UniSpray (+)	253.0 → 125.8	20	253.0 → 90.0	40
	C	LC-MS/MS	ESI (+)	253 → 186	12	253 → 125.9	20
	D	LC-MS/MS	ESI (+)	253.0 → 126.0	24	253.0 → 90.0	24
	E	LC-MS/MS	ESI (+)	253.0 → 126.0	23	253.0 → 90.0	35
Thiamethoxam	A	LC-MS/MS	ESI (+)	292.0 → 211.1	13	292.0 → 181.0	24
	B	LC-MS/MS	UniSpray (+)	292.0 → 211.2	10	292.0 → 132.0	20
	C	LC-MS/MS	ESI (+)	292 → 211	8	292 → 181	20
	D	LC-MS/MS	ESI (+)	292.0 → 211.0	12	292.0 → 181.0	12
	E	LC-MS/MS	ESI (+)	292.0 → 211.0	11	292.0 → 181.0	23
	F	GC-MS/MS	EI (−70eV)	247.0 → 212.0	2	247.0 → 182.0	8