Comparative Assessment of Pesticide Exposures in Domestic Dogs and Their Owners Using Silicone Passive Samplers and Biomonitoring

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Table S1. Urine pesticide biomarker analyte summary (μg/L).

Biomarker Name	Biomarker Abbreviation	Parent chemical	Chemical class	Analytical LOD*	Adult Reference range	Literature references
3-(diethylcarbamoyl)benzoic acid	DCBA	N,N-diethyl-meta-toluamide (DEET)	Insect repellent	0.2	2.93-145	3
3-(ethylcarbamoyl)benzoic acid	ECBA	N,N-diethyl-meta-toluamide (DEET)	Insect repellent	0.2	1.08-50.5	3
Imidacloprid	IMI	Imidacloprid	Neonicotinoid	0.4	<LOD	2,3
5-Hydroxy imidacloprid	5-OH-IMI	Imidacloprid	Neonicotinoid	0.4	<LOD-1.38	2,3
Thiacloprid	THIAC	Thiacloprid	Neonicotinoid	0.03	<LOD	2,3
Clothianidin	CLOTH	Clothianidin, Thiamethoxam	Neonicotinoid	0.2	<LOD-0.393	2,3
Acetamiprid	ACET	Acetamiprid	Neonicotinoid	0.3	<LOD	2,3
Acetamiprid-N-desmethyl	ACET-Ndes	Acetamiprid	Neonicotinoid	0.2	<LOD-1.27	2,3
2-isopropyl-4-methyl-6-hydroxypyrimidine	IMPY	Diazinon	Organophosphate	0.1	<LOD-0.496†	3
3,5,6-Trichloro-2-pyridinol	TCPY	Chlorpyrifos, Chlorpyrifos-methyl	Organophosphate	0.1	0.97–4.18‡	1
para-Nitrophenol	PNP	Parathion, methyl parathion, ethyl parathion, nitrobenzene, O-ethyl-O-(4-nitrophenyl) phenylphosphonothioate,	Organophosphate	0.1	0.572–3.21†	3
2,4-Dichlorophenoxyacetic acid	2,4-D	2,4-Dichlorophenoxyacetic acid (and it's esters)	Phenoxy acid	0.15	0.272–1.32†	3
3-phenoxybenzoic acid	3-PBA	Cyhalothrin, Cypermethrin, Deltamethrin, Fenpropathrin, Permethrin, Tralomethrin	Pyrethroid	0.1	0.62–7.36†	3
4-fluoro-3-phenoxybenzoic acid	4-F-3-PBA	Cyfluthrin, Flumethrin	Pyrethroid	0.1	<LOD-0.177†	3
trans-3-(2,2-Dichlorovinyl)-2,2-dimethylcyclopropane carboxylic acid	*trans*-DCCA	Permethrin; Cypermethrin; Cyfluthrin	Pyrethroid	0.6	<LOD–6.3†	3

*Results near the limit of detection (LOD) are subject to greater uncertainty. The LOD =3*SD (the standard deviation of the concentration derived from the measurement process as the concentration approaches zero). Limit of detection (LOD) and reference ranges are in μg/L. Reference ranges are limited to the 50th and 95th percentile intervals for the U.S. population 20 years of age and older from NHANES 2015-2016 unless otherwise noted. ‡ Values limited to the 50th to the 95th percentile intervals for the U.S. population ages 20-59 from NHANES 2009-2010. † Values limited to the 50th to the 95th percentile intervals for the U.S. population 20 years of age and older from NHANES 2013-2014.

Table S2. Information collected on flea and tick products used on the participating dog by study participants. Also included are the corresponding specific gravity corrected urinary metabolite concentrations (μg/L) in human and dog samples.

ID #	Imidacloprid	5-OH-IMI human	IMI human	5-OH-IMI dog	IMI dog	Flumethrin	4-F-3-PBA human	4-F-3-PBA dog
CEA0001	No	<LOD	<LOD	13.09	0.77	No	<LOD	<LOD
CEA0002	No	‡	0.59	<LOD	<LOD	No	<LOD	<LOD
CEA0003	No	<LOD	0.55	1.46	<LOD	No	1.25	<LOD
CEA0004	No	‡	0.74	<LOD	<LOD	No	<LOD	<LOD
CEA0005	Yes	‡	1.50	79.10	9.26	Yes	0.38	0.55
CEA0006	No	<LOD	0.49	<LOD	<LOD	No	<LOD	<LOD
CEA0007	Yes	‡	‡	782.27	158.34	Yes	0.76	1.33
CEA0008	Yes	23.85	‡	95.16	23.33	Yes	0.62	0.24
CEA0009	Yes	‡	‡	215.40	29.83	Yes	0.33	1.17
CEA0010	No	‡	0.50	1.06	‡	No	<LOD	<LOD
CEA0011	No	<LOD	<LOD	<LOD	‡	No	<LOD	<LOD
CEA0012	No	1.18	0.92	<LOD	<LOD	No	<LOD	<LOD
CEA0013	Yes	1.70	<LOD	‡	<LOD	Yes	0.23	<LOD
CEA0014	No	<LOD	<LOD	<LOD	<LOD	No	<LOD	<LOD
CEA0015	No	‡	0.49	‡	0.41	No	<LOD	<LOD
CEA0016	Historical	‡	<LOD	40.71	‡	No	<LOD	<LOD
CEA0017	No	<LOD	<LOD	<LOD	<LOD	No	<LOD	<LOD
CEA0018	No	<LOD	<LOD	<LOD	<LOD	No	<LOD	<LOD
CEA0019	No	<LOD	<LOD	‡	‡	No	<LOD	<LOD
CEA0020	Yes	‡	<LOD	24.42	3.89	No	<LOD	<LOD
CEA0021	No	‡	0.70	<LOD	<LOD	No	<LOD	<LOD
CEA0022	No	‡	<LOD	<LOD	<LOD	No	<LOD	<LOD
CEA0023	No	‡	‡	147.65	25.17	No	<LOD	<LOD
CEA0025	No	1.03	<LOD	7.96	‡	No	0.26	<LOD
CEA0026	Yes	‡	‡	‡	7.47	Yes	0.16	0.22
CEA0027	No	<LOD	<LOD	<LOD	<LOD	No	<LOD	<LOD
CEA0028	No	<LOD	0.42	<LOD	<LOD	No	<LOD	<LOD
CEA0029	No	‡	0.53	9.23	‡	No	0.49	<LOD
CEA0030	No	2.66	<LOD	<LOD	<LOD	No	<LOD	<LOD
CEA0031	Historical	11.97	‡	150.11	21.44	Historical	0.23	<LOD

Questionnaire responses indicated each participant did (Yes) or did not (No) report use of flea and tick products containing specific pesticides (imidacloprid/flumethrin). “Historical” indicates that the dog has been treated with a flea and tick product with that particular active ingredient within the past year but was not using that product during the time of the study period. Concentrations of associated urinary biomarkers, 5-OH-IMI and IMI for imidacloprid and 4-F-3-PBA for flumethrin, were measured in human and dog urine. ‡Some samples had mass spectrometry interferences that prevented accurate quantification of some biomarkers, those samples were excluded from statistical analysis. <LOD means that concentrations were below the limit of detection.

Table S3: Matrix spike recoveries for pesticides extracted from wristbands. Recoveries represent the average recovery from three replicates. Each replicate involved spiking anywhere from 50 – 250 ng onto a 1-gram piece of silicone.

COMPOUND	CAS #	% RECOVERY OF MATRIX SPIKE
N,N-DIETHYL-META-TOLUAMIDE (DEET)	134-62-3	104%
LINDANE	58-89-9	84%
CHLORPYRIFOS	2921-88-2	101%
FIPRONIL	120068-37-3	91%
TRANS-CHLORDANE	5103-74-2	99%
CIS-CHLORDANE	5103-71-9	99%
CIS-PERMETHRIN	61949-76-6	105%
TRANS-PERMETHRIN	61949-77-7	112%
CYPERMETHRIN	52315-07-8	89%
AZOXYSTROBIN	131860-33-8	115%

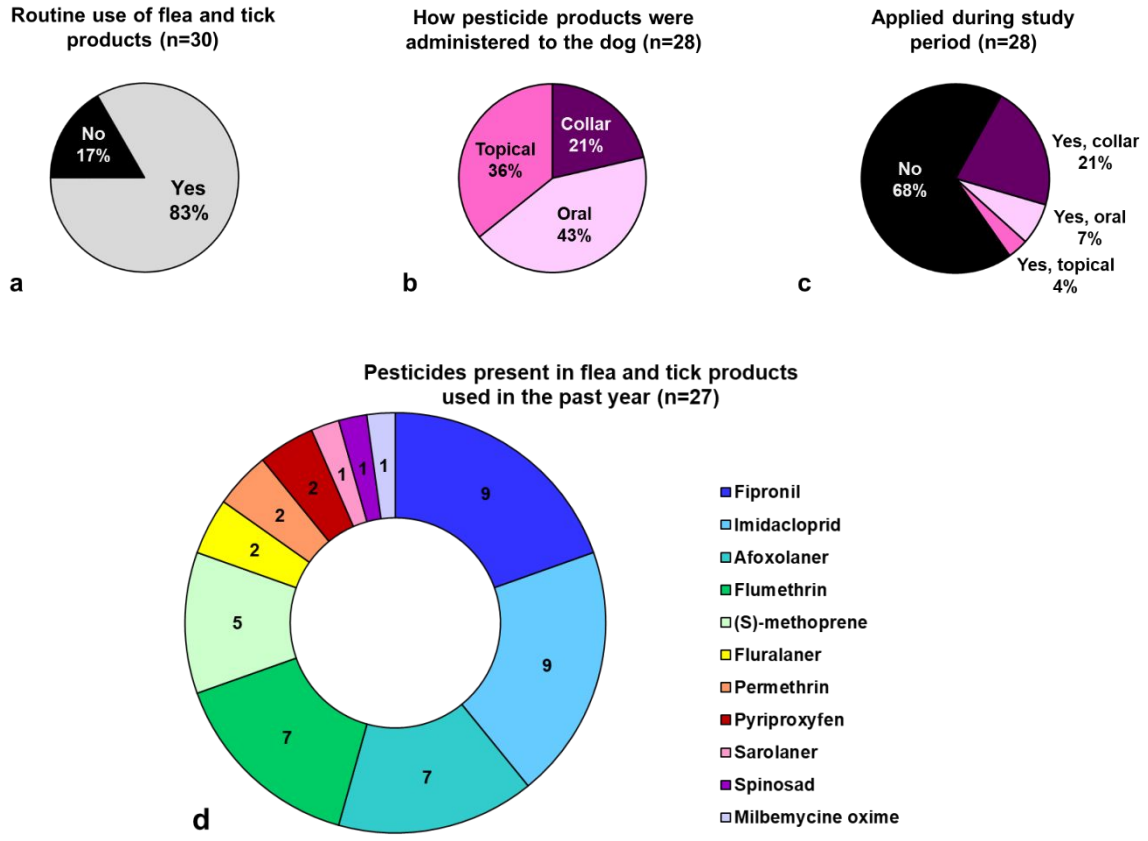

Figure S1. Reported use of pesticide containing flea and tick preventatives.
A majority of study participants routinely use pesticide containing flea and tick products on their dog (a). Two owners reported they did not use any flea and tick preventatives on their dog within the past year. Participants reported how the most recent application of the pesticide products were administered (b) and whether the application occurring during the study period (c). In total, owners reported 11 different pesticides listed as active ingredients in the flea and tick preventatives used for their dogs during the previous year (d). One owner did not indicate which product was used. Numbers reported in the chart are total number of dogs exposed to each pesticide.

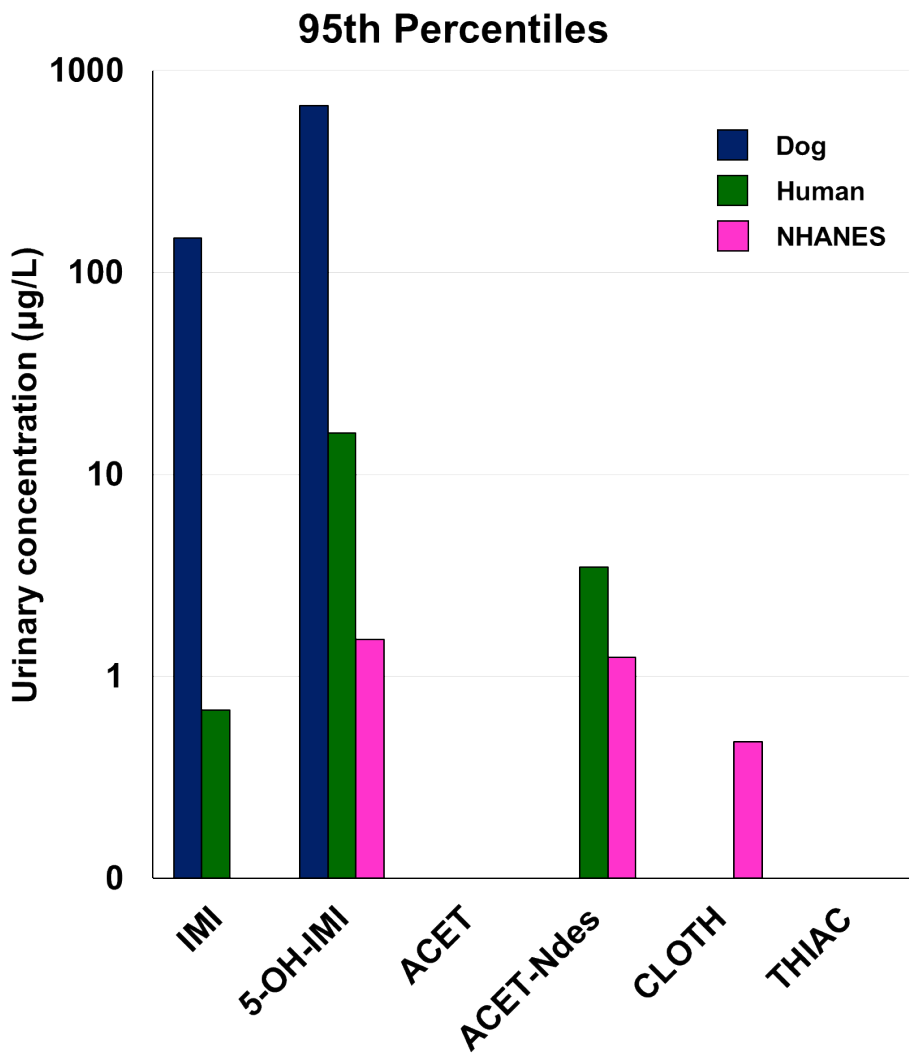

Figure S2. A comparison of the neonicotinoid pesticide urinary biomarker 95th percentile concentrations for chemicals with a low detection frequency. Absence of a bar indicates the 95th percentile for that set of samples was <LOD.

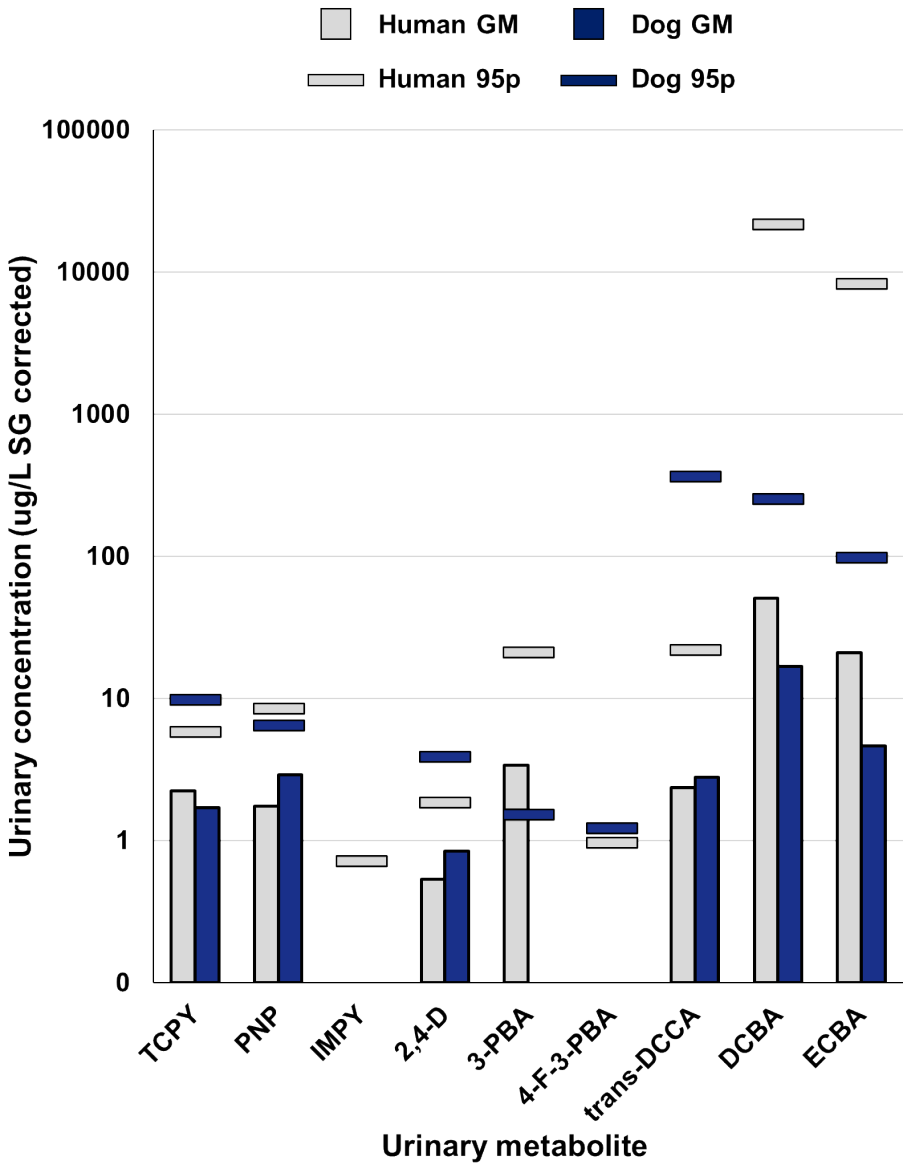

Figure S3. A comparison of pesticide biomarker concentrations in urine samples from humans and dogs. Comparisons are shown for the geometric mean (GM) and 95th percentiles (95p) based on SG corrected concentrations of frequently detected metabolites measured in human and dog urine in our study. Comparisons were not included for analytes with a high proportion of samples with concentrations <LOD. IMPY was not detected in any dog urine samples.

Supplemental Information References:

1. Fourth National Report on human Exposure to Environmental Chemicals, Updated Tables, Volume One, March 2018, DLS, NCEH, CDC, https://www.cdc.gov/exposurereport/pdf/FourthReport_UpdatedTables_Volume1_Mar2018.pdf

2. Ospina, M.; Wong, L.Y.; Baker, S.E.; Bishop Serafim, A.; Morales-Agudelo, P. and Calafat, A.M. Exposure to Neonicotinoid Insecticides in the U.S. General Population: Data from the 2015–2016 National Health and Nutrition Examination Survey. *Env. Res.* **2019,** *176,* 108555; DOI 10.1016/j.envres.2019.108555.

3. Fourth National Report on Human Exposure to Environmental Chemicals, Updated Tables, March 2021, DLS, NCEH, CDC. https://www.cdc.gov/exposurereport/