

Supplementary table 1: Baseline characteristics of the participants

Characteristics	Non-cases (n=34,050)	Hyperthyroidism (n=271)	
	n (%)	n (%)	OR (95% CI) <sup>a</sup>
<b>Age</b>			
≤ 45 years	17009 (50)	109 (40.2)	Ref
46-55 years	8086 (23.7)	66 (24.4)	1.09 (0.79, 1.50)
56-65 years	6226 (18.3)	70 (25.8)	1.46 (1.05, 2.02)
> 65 years	2729 (8)	26 (9.6)	1.15 (0.72, 1.85)
<b>Gender</b>			
Women	706 (2.1)	21 (7.7)	Ref
Men	33344 (97.9)	250 (92.3)	0.32 (0.19, 0.53)
<b>State of residence</b>			
Iowa	22223 (65.3)	147 (54.2)	Ref
North Carolina	11827 (34.7)	124 (45.8)	1.18 (0.89, 1.56)
<b>Education</b>			
≤ High school graduate	19165 (56.3)	158 (58.3)	Ref
1–3 years beyond high school	8585 (25.2)	69 (25.5)	1.13 (0.84, 1.52)
College graduate or more	6300 (18.5)	44 (16.2)	0.94 (0.66, 1.34)
<b>Smoking status</b>			
Never smoker	18810 (55.2)	134 (49.4)	Ref
Former smoker	10315 (30.3)	93 (34.3)	1.20 (0.90, 1.60)
Current smoker	4925 (14.5)	44 (16.2)	1.25 (0.87, 1.81)
<b>Alcohol consumption (past year)<sup>b</sup></b>			
No	10933 (33.3)	120 (46.9)	Ref
Yes	21907 (66.7)	136 (53.1)	0.64 (0.49, 0.85)

Abbreviations: OR, Odds Ratio; CI, Confidence Intervals

<sup>a</sup>Estimated using logistic regression with all covariates in the model

<sup>b</sup>n=1210 missing for non-cases, and 15 missing in hyperthyroidism

Supplementary table 2: Ever-use of pesticides and hyperthyroidism risk excluding female applicators (n= 34,375)<sup>a</sup>

Pesticides	Exposed cases	HR (95% CI) <sup>b</sup>
Organochlorine insecticide		
Aldrin	43	0.83 (0.58, 1.19)
Chlordane	67	0.95 (0.70, 1.28)
DDT	72	1.00 (0.73, 1.37)
Dieldrin	18	1.02 (0.62, 1.68)
Heptachlor	37	0.91 (0.62, 1.34)
Lindane	45	0.99 (0.71, 1.37)
Toxaphene	31	0.74 (0.50, 1.09)
Carbamate insecticide		
Aldicarb	26	0.85 (0.55, 1.32)
Carbaryl	133	0.79 (0.59, 1.05)
Carbofuran ≤ 62 years <sup>c</sup>	28	<b>0.65 (0.42, 0.98)</b>
> 62 years	28	1.00 (0.64, 1.58)
Organophosphate insecticide		
Chlorpyrifos	93	0.84 (0.65, 1.09)
Coumaphos	15	0.73 (0.43, 1.23)
Diazinon	84	1.10 (0.83, 1.47)
Dichlorvos	23	1.00 (0.64, 1.56)
Fonofos	51	1.16 (0.83, 1.62)
Malathion	149	<b>0.66 (0.50, 0.87)</b>
Parathion	39	0.95 (0.66, 1.36)
Phorate	61	0.75 (0.55, 1.02)
Terbufos	82	0.91 (0.69, 1.21)
Pyrethroid insecticide		
Permethrin (animals)	29	1.06 (0.71, 1.59)
Permethrin (crops)	24	0.78 (0.51, 1.20)
Fumigant		
Carbon tetrachloride/ Carbon disulphide 80/20 mix	13	0.92 (0.52, 1.62)
Aluminum Phosphide	11	0.98 (0.54, 1.81)
Ethylene Dibromide	11	1.15 (0.62, 2.14)
Methyl Bromide	47	0.87 (0.60, 1.25)
Fungicide		
Benomyl	20	0.66 (0.41, 1.08)
Chlorothalonil	15	0.60 (0.35, 1.04)
Maneb	17	<b>0.54 (0.32, 0.91)</b>
Metalaxyl	60	0.90 (0.64, 1.26)
Herbicide		
Alachlor	113	0.82 (0.63, 1.07)
Butylate	61	0.76 (0.56, 1.03)
Chlorimuron Ethyl ≤ 62 years <sup>c</sup>	42	<b>0.66 (0.46, 0.95)</b>
> 62 years	29	1.23 (0.78, 1.94)
Dicamba	98	0.76 (0.56, 1.03)

Pesticides	Exposed cases	HR (95% CI) <sup>b</sup>
EPTC	35	0.83 (0.57, 1.20)
Glyphosate	197	1.10 (0.80, 1.51)
Imazethapyr	89	1.04 (0.76, 1.43)
Metolachlor	82	<b>0.70 (0.53, 0.92)</b>
Paraquat	48	<b>0.71 (0.50, 1.00)</b>
Pendimethalin	95	0.90 (0.69, 1.18)
Petroleum Oil	100	0.91 (0.69, 1.19)
Trifluralin	118	1.04 (0.79, 1.38)
2,4-D	182	0.87 (0.65, 1.17)
2,4,5-T	56	1.03 (0.75, 1.41)
2,4,5-T P	22	0.94 (0.60, 1.47)
Atrazine	166	0.77 (0.59, 1.02)
Cyanazine	84	0.87 (0.64, 1.17)
Metribuzin	99	0.97 (0.73, 1.29)

Abbreviation: 2,4-D, 2,4-Dichlorophenoxyacetic acid; 2,4,5-T, 2,4,5-Trichlorophenoxyacetic acid; 2,4,5-T,P, 2-(2,4,5-trichlorophenoxy) propionic acid; CI, Confidence Intervals; DDT,

Dichlorodiphenyltrichloroethane; EPTC, S-Ethyl dipropylthiocarbamate; HR, Hazard Ratio

<sup>a</sup>Of 34375, 33,344 were non-cases, 250 were hyperthyroidism cases, and 781 were hypothyroidism cases.

<sup>b</sup>Adjusted for education, state, and smoking

<sup>c</sup>Hazard ratio allowed to vary by the median age (i.e., 62 years) for pesticides for which proportional hazards assumptions were not met ( $p \leq 0.10$ )

Note: bold-face indicates  $p$ -value  $\leq 0.05$ .

Supplementary table 3: Ever-use of pesticides and hyperthyroidism risk adjusting for pesticides with Spearman correlation coefficient  $\geq 0.40$ .

Pesticide	HR (95% CI) <sup>a</sup>	Correlated pesticides
Organochlorine insecticide		
Aldrin	0.78 (0.50, 1.20)	DDT, Dieldrin, Heptachlor
Chlordane	0.92 (0.67, 1.27)	DDT
DDT	0.98 (0.69, 1.39)	Aldrin, Chlordane
Dieldrin	1.18 (0.67, 2.07)	Aldrin, Heptachlor
Heptachlor	1.02 (0.65, 1.59)	Aldrin, Dieldrin
Carbamate insecticide		
Aldicarb	0.94 (0.58, 1.52)	Benomyl, Chlorothalonil
Carbaryl	0.81 (0.60, 1.10)	Diazinon
Organophosphate insecticide		
Diazinon	1.13 (0.84, 1.52)	Carbaryl
Fumigant		
Methyl Bromide	0.90 (0.60, 1.36)	Metalaxyl
Fungicide		
Benomyl	0.96 (0.54, 1.69)	Maneb, Chlorothalonil, Aldicarb
Chlorothalonil	0.65 (0.33, 1.25)	Aldicarb, Benomyl
Maneb	<b>0.49 (0.27, 0.87)</b>	Benomyl
Metalaxyl	0.97 (0.69, 1.38)	Methyl Bromide
Herbicide		
Butylate	0.75 (0.54, 1.03)	Metribuzin
Dicamba	<b>0.72 (0.53, 0.98)</b>	Imazethapyr
Imazethapyr $\leq 62$ years <sup>b</sup>	0.83 (0.56, 1.23)	Dicamba
$> 62$ years	<b>1.90 (1.10, 3.10)</b>	
Trifluralin	1.03 (0.76, 1.40)	Metribuzin
2,4,5-T	1.04 (0.72, 1.50)	2,4,5-TP
2,4,5-TP	0.89 (0.53, 1.49)	2,4,5-T
Metribuzin	1.03 (0.75, 1.43)	Butylate, Trifluralin

Abbreviation: 2,4,5-T, 2,4,5-Trichlorophenoxyacetic acid; 2,4,5-T,P, 2-(2,4,5-trichlorophenoxy) propionic acid; CI, Confidence Intervals; DDT, Dichlorodiphenyltrichloroethane; HR, Hazard Ratio

<sup>a</sup> Adjusted for sex, education, state, smoking, and correlated pesticides

<sup>b</sup> Hazard ratio allowed to vary by the median age (i.e., 62 years) for pesticides for which proportional hazards assumptions were not met ( $p \leq 0.10$ )

Note: bold-face indicates  $p$ -value  $\leq 0.05$ .

Supplementary table 4: Intensity-weighted lifetime days of use of pesticides and hyperthyroidism risk

Pesticide	Days	Hyperthyroidism cases	HR (95% CI) <sup>a</sup>	P-trend <sup>b</sup>
Organochlorine insecticide				
Chlordane	Unexposed	99	Ref	0.81
	>0–≤350	13	0.91 (0.50, 1.64)	
	>350	16	1.07 (0.62, 1.84)	
DDT	Unexposed	93	Ref	0.23
	>0–≤770	20	1.23 (0.74, 2.04)	
	>770	13	0.71 (0.39, 1.31)	
Carbamate insecticide				
Carbofuran	Unexposed	177	Ref	<b>0.01</b>
	>0–≤775	36	1.06 (0.74, 1.52)	
	>775	20	<b>0.55 (0.34, 0.87)</b>	
Chlorpyrifos	Unexposed	145	Ref	<b>&lt;0.01</b>
	>0–≤894	64	1.15 (0.86, 1.55)	
	>894	33	<b>0.59 (0.40, 0.86)</b>	
Organophosphate insecticide				
Fonofos	Unexposed	182	Ref	0.71
	>0–≤882	29	1.25 (0.83, 1.88)	
	>882	20	0.92 (0.57, 1.48)	
Malathion	Unexposed	50	Ref	0.34
	>0–≤735	39	0.92 (0.60, 1.41)	
	>735	41	0.81 (0.53, 1.23)	
Phorate	Unexposed	97	Ref	0.22
	>0–≤662	19	1.04 (0.62, 1.76)	
	>662	13	0.69 (0.38, 1.25)	
Terbufos	Unexposed	156	Ref	0.48
	>0–≤1206	41	0.92 (0.65, 1.32)	
	>1206	40	0.87 (0.61, 1.25)	
Pyrethroid insecticide				
Permethrin (animals)	Unexposed	204	Ref	0.60
	>0–≤740	11	0.78 (0.42, 1.43)	
	>740	16	1.16 (0.69, 1.94)	
Permethrin (crops)	Unexposed	208	Ref	0.25
	>0–≤490	10	0.66 (0.35, 1.25)	
	>490	12	0.72 (0.40, 1.29)	
Fumigant				
Methyl Bromide	Unexposed	216	Ref	0.57
	>0–≤682	18	0.71 (0.43, 1.18)	
	>682	29	1.08 (0.70, 1.67)	
Fungicide				
Metalaxyl	Unexposed	100	Ref	0.51
	>0–≤756	14	0.97 (0.55, 1.73)	
	>756	14	0.82 (0.44, 1.5)	
Herbicide				
Alachlor	Unexposed	118	Ref	0.36
	>0–≤1550	57	0.88 (0.64, 1.22)	

Pesticide	Days	Hyperthyroidism cases	HR (95% CI) <sup>a</sup>	P-trend <sup>b</sup>
Butylate	>1550	56	0.84 (0.61, 1.16)	0.18
	Unexposed	107	Ref	
	>0-≤882	10	0.54 (0.28, 1.04)	
Chlorimuron Ethyl	>882	12	0.66 (0.36, 1.21)	0.93
	Unexposed	91	Ref	
	>0-≤354	20	1.15 (0.70, 1.88)	
Dicamba	>354	19	1.02 (0.62, 1.68)	0.29
	Unexposed	138	Ref	
	>0-≤1078	48	0.72 (0.50, 1.04)	
Glyphosate	>1078	49	0.76 (0.53, 1.08)	0.47
	Unexposed	58	Ref	
	>0-≤1232	98	1.03 (0.74, 1.43)	
Imazethapyr	>1232	110	1.12 (0.80, 1.55)	0.82
	Unexposed	142	Ref	
	>0-≤637	49	1.12 (0.77, 1.62)	
Metolachlor	>637	41	0.98 (0.67, 1.44)	0.09
	Unexposed	146	Ref	
	>0-≤1344	45	0.8 (0.56, 1.12)	
Pendimethalin	>1344	40	0.71 (0.50, 1.02)	0.09
	Unexposed	91	Ref	
	>0-≤725	21	0.90 (0.56, 1.45)	
Petroleum	>725	16	0.62 (0.36, 1.07)	0.49
	Unexposed	103	Ref	
	>0-≤1029	15	1.23 (0.71, 2.13)	
Trifluralin	>1029	10	0.80 (0.42, 1.53)	0.96
	Unexposed	115	Ref	
	>0-≤2016	61	1.07 (0.77, 1.48)	
2,4-D	>2016	58	1.03 (0.73, 1.43)	0.27
	Unexposed	78	Ref	
	>0-≤2552	97	0.91 (0.66, 1.25)	
Atrazine	>2552	89	0.82 (0.59, 1.13)	0.22
	Unexposed	100	Ref	
	>0-≤2240	82	0.77 (0.56, 1.05)	
Cyanazine	>2240	83	0.75 (0.55, 1.02)	0.29
	Unexposed	151	Ref	
	>0-≤1062	44	0.89 (0.62, 1.27)	
Metribuzin	>1062	39	0.80 (0.55, 1.17)	0.22
	Unexposed	91	Ref	
	>0-≤588	22	0.92 (0.56, 1.51)	
	>588	17	0.71 (0.42, 1.22)	

Abbreviation: 2,4-D, 2,4-Dichlorophenoxyacetic acid; CI, Confidence Intervals; DDT, Dichlorodiphenyltrichloroethane; HR, Hazard Ratio

<sup>a</sup> Adjusted for sex, education, state, and smoking

<sup>b</sup> P-trend values were obtained using an ordinal variable coded with median values for each category

Note: bold-face indicates p-value ≤ 0.05, and intensity-weighted analyses are presented only for the pesticides with adequate sample size