



## RESEARCH ARTICLE

## Is There any Relationship between Worldwide Pesticide Standard Values in Residential Soil, Drinking Water, and Agricultural Commodity Exposures?

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### Abstract

Human could exposure to pesticides via various exposure pathways. Worldwide jurisdictions are making efforts to manage human health risk by regulating pesticide standards in residential soil, drinking water, and agricultural commodity. An assumption is made that if the pesticide standards were derived by human health risk model with essential toxic data, a relationship may exist between the standard values from different exposures. Implied Dose Limit (IDL) was introduced in this research to convert pesticide standards in different exposure pathways to pesticide exposure mass loading. Comparing all the IDLs of pesticides, it illustrates that pesticide standards in soil are probably more conservative than those in drinking water and food for most largely used pesticides worldwide. Result also shows that there is no significant relationship between any two IDLs computed from these exposure pathways, suggesting that worldwide jurisdictions derived the pesticide standards independently without considering the impacts of other major exposures.

### Keywords

Pesticide standard values, Implied dose limit, Human exposure, Exposure assessment, Largely used pesticides, POPs

### Introduction

Pesticide impact is a worldwide problem due to its toxicity and ecotoxicity potential and ubiquitous presence in the environment [1]. After applied, pesticides can be transported into surrounding environments such as soil, water, air, and living organisms. In addition, pesticides can get into human body by various exposure pathways [2] including ingestion, inhalation, and dermal contact [3]. Some Persistent Organic Pollutants (POPs) such as DDT can accumulate in human body over

entire lifetime. Therefore, worldwide regulatory jurisdictions are making efforts to manage human health risk against pesticides by regulating Pesticide Standard Values (PSVs) in all possible exposure pathways. Developing PSVs regulations is a systematic and comprehensive work by considering all major exposures and applying the human health risk model. Previous studies [4-6] show that worldwide pesticide soil Regulatory Guidance Values (RGVs) are not harmonized, which could vary in five, six, or even seven orders of magnitude. Some RGVs seem too large to prevent human health impact and others are too small to achieve the remediation goal technically. A similar result was found for worldwide pesticide drinking water Maximum Concentration Levels (MCLs) [7]. In Li's [7] study of pesticide agricultural commodity Maximum Residue Limits (MRLs) analysis, it shows that the MRLs do not vary as large orders of magnitude as soil RGVs and drinking water MCLs, but some extreme values are still not conservative enough to protect human health. Joon Chuah C, et al. [8], Rachel Baum, et al. [9], Winter and Jara [10], and Chen, et al. [11] have studied the PSVs in one of major exposure pathways as well. These studies have discussed and compared PSVs at different nations in a single exposure pathway. Li [7] introduced Implied Maximum Dose Limits (IMDLs) computed from all major exposures to analyze PSVs in a more comprehensive approach. The analysis of IMDLs may help worldwide jurisdictions on the study of PSVs because human exposures to pesticide always have various pathways. In this research, the relationship between PSVs was investigated among different exposure pathways. Moreover, this research

has examined whether a nation regulated PSVs for major exposure pathways systematically by comparing the PSVs among different exposures.

## Materials

### Worldwide PSVs

The materials for this research are PSVs from worldwide jurisdictions in residential soil, drinking water, air, and agricultural commodity. Those PSVs were obtained from online data base with full references and web addresses listed in the [Supplementary Material](#). When internet addresses and online documents become invalid and out of date, key words from the jurisdictions titles would be used to address the new web location. At least 54 (28% of the worldwide nations) nations have regulated pesticide soil RGVs, 102 (52%) nations have provided drinking water MCLs, 90 (46%) nations had pesticide agricultural commodity MRLs, and only U.S. systematically provided pesticide air MCLs. Each pesticide was identified by a Chemical Abstracts Service Registry Number (CAS No.) instead of chemical nomenclature conventions, common names, and trade names because of the complex chemical structure.

### Commonly used pesticides

A total of 14 current largely used pesticides and 11 largely used pesticides in the past were selected for analysis. The current largely used pesticides are 2,4-D, Aldicarb, Atrazine, Chlorothalonil, Chlorpyrifos, Diazinon, Dicamba, Diuron, Glyphosate, Malathion, Mancozeb, MCPA, Metolachlor, and Trifluralin. The historical largely used pesticides which have been banned in most countries are Aldrin, Bromomethane, Chlordane, DDT, Dieldrin, Endosulfan, Endrin, Heptachlor, Lindane, Pentachlorophenol, and Toxaphene. Also those 11 historical largely used pesticides are the Stockholm Convention POPs.

## Methods

### Implied dose limit

Since PSVs in different exposure pathways have different units (e.g. mg/kg for soil RGV, and mg/l for drinking water), IDL (mg/kg-day) was introduced to convert the PSVs in each exposure to the pesticide exposure mass loading, which assessed the total human mass burden implied by drinking water, soil, and agricultural food PSVs of regulatory jurisdictions. A relationship of the PSVs from different exposure pathways was expected if PSVs from different nations were developed in each exposure systematically and all based on human health risk model. Because few jurisdictions regulated PSVs in air, only IDLs computed from soil, drinking water, and agricultural commodity PSVs were conducted and expressed as follows.

For drinking water:

$$IDL_{dw} = \left( \frac{EF}{HW} \right) (MCL)(V) \quad (1)$$

For residential soil:

$$IDL_{soil} = \left( \frac{EF}{HW} \right) [(RGV)(CF)(IR) + (RGV)(CF)(ABS_d)(GIABS)] \quad (2)$$

For agricultural commodities:

$$IDL_{food} = \left( \frac{EF}{HW} \right) \sum_{i=1}^n (MRL_i)(IR_i) \quad (3)$$

All IDLs are based on the following set of exposure scenario coefficient values.

EF - Exposure Factor (1, unit less) [12]

HW - Human Weight (70 kg) [12]

V - Volume of water intake rate (2 L/day) [12]

CF - Convert Factor ( $10^6$  mg/kg)

IR - Intake Rate for soil (mg/kg-day) [13]

$ABS_d$  - Absorption Factor (unit less) [13]

GIABS - GastroIntestinal Absorption Factor (unit less) [13]

$IR_i$  - Intake Rate for food  $i$  (kg/day)

### Cumulative Distribution Function (CDF) analysis

The CDF analysis was used to plot the IDLs computed from soil, drinking water, and agricultural commodity exposures (Equation 4) and compared with the cumulative distribution of a lognormal random variable with identical statistics. Pearson correlation coefficients ( $r$ ) between the empirical cumulative distribution and a theoretical normal or log-normal cumulative distribution with identical statistics were computed for PSVs in major exposures (Equation 5).

$$P(IDL_r \leq IDL_i) \approx \frac{n_i}{N}; \quad \forall i = 1, \dots, N \quad (4)$$

$IDL_r$  - a random IDL

$IDL_i$  - a known IDL

$n_i$  - integer rank of IDL in  $N$  known values

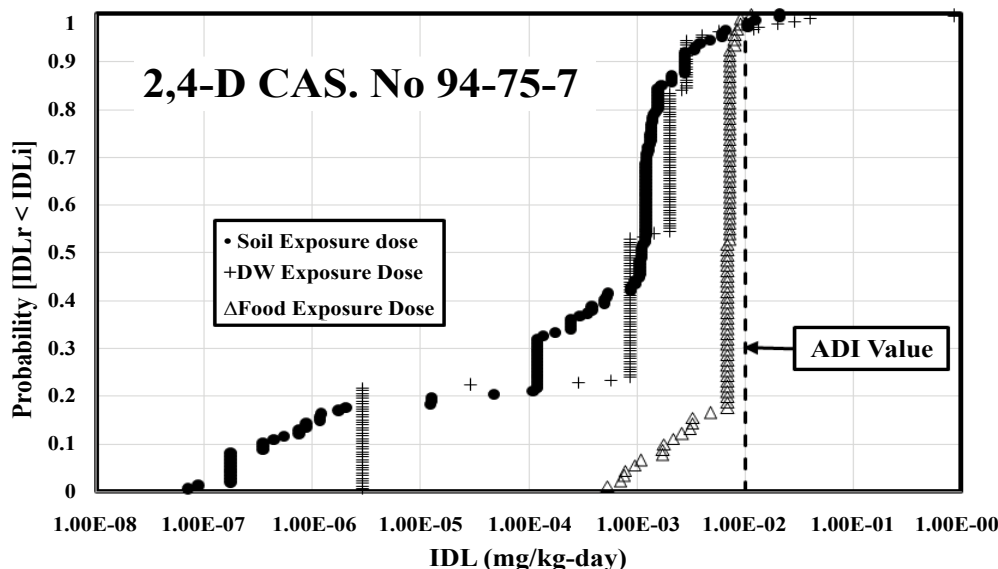
$$r = \frac{N[\sum (IDL_i * F(IDL_i))] - [\sum E(IDL_i)][\sum F(IDL_i)]}{\sqrt{[N\sum E(IDL_i)^2 - (\sum E(IDL_i))^2][N\sum F(IDL_i)^2 - (\sum F(IDL_i))^2]}} \quad (5)$$

$E(IDL_i)$  - probability computed from IDL empirical cumulative distribution

$F(IDL_i)$  - probability computed from IDL theoretical normal or log-normal cumulative distribution

## Results

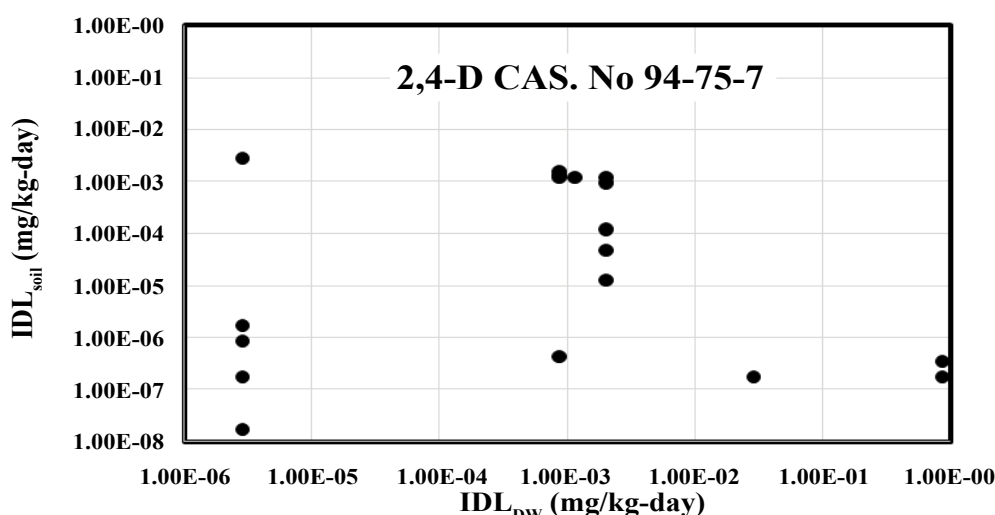
For those 14 current largely used pesticides and 11 Stockholm Convention POPs, IDLs computed from major exposures were illustrated as CDF and relationships between IDLs in two major exposures were explored. 2,4-D, Chlorpyrifos, and Lindane IDLs analyses were conducted in this study.



**Figure 1:** Worldwide computed 2,4-D IDLs from residential soil RGVs, drinking water MCLs, and commonly consumed agricultural commodities MRLs plotted in the form of CDF.

**Table 1:** Statistic summary of 2,4-D IDLs.

	Soil	Drinking water	Agricultural commodity
Number of IDLs	147	180	91
Arithmetic mean (mg/kg-day)	1.50E - 03	1.14E - 02	6.37E - 03
Median (mg/kg-day)	1.09E - 03	8.57E - 04	6.95E - 03
Geometric mean (mg/kg-day)	2.11E - 04	4.33E - 04	5.63E - 03
Standard deviation (mg/kg-day)	2.93E - 03	9.00E - 02	2.11E - 03
Max IDL (mg/kg-day)	2.08E - 02	8.57E - 01	1.14E - 02
Min IDL (mg/kg-day)	6.92E - 08	2.86E - 06	5.38E - 04
Order of magnitude variation	5.48E + 00	5.48E + 00	1.33E + 00
Correlation coefficient	0.907	0.861	0.761



**Figure 2:** Relationship between computed 2,4-D IDLs from residential soil RGVs and drinking water MCLs.

### Relationship among 2,4-D PSVs in soil, drinking water, and agricultural commodity

A total of 147, 180, and 91 computed 2,4-D IDLs from 2,4-D soil, drinking water, and agricultural commodity PSVs were illustrated as CDF in [Figure 1](#) respectively. Both the IDLs computed from 2,4-D soil and water ranged in a span of 5.48 orders of magnitude. However, the IDLs computed from commonly consumed agri-

cultural commodities only has a span of 1.33 orders of magnitude. In addition, the Pearson correlation coefficients for these IDLs listed in [Table 1](#) suggest that 2,4-D soil and water IDLs are better dispersed over the data spans. The median and geometric means of these IDLs suggests that worldwide 2,4-D soil RGV and drinking water MCL sets are probably more conservative than MRL set.

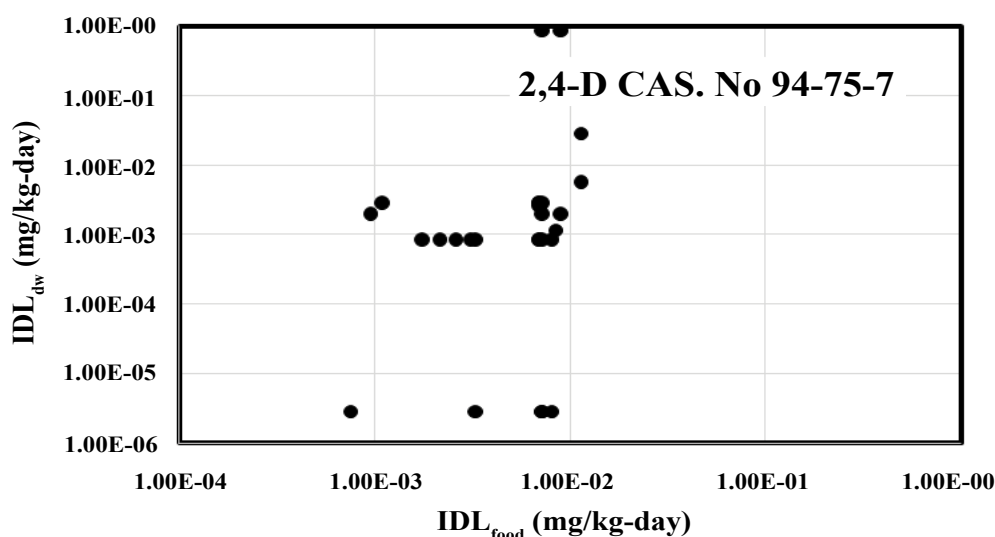
A total of 20 national jurisdictions from the same nation regulated both 2,4-D soil RGVs and drinking water MCLs. Figure 2 illustrates that there is no significant relationship between computed 2,4-D IDLs from soil and water. Similarly, Figure 3 illustrates that there is no significant relationship between 20 computed 2,4-D IDLs from drinking water MCLs and commonly consumed agricultural commodities MRLs.

A total of 65 national jurisdictions (some jurisdictions share same PSVs) from the same nation regulated both 2,4-D soil RGVs and agricultural commodity MRLs. Figure 4 illustrates that there might be a linear relationship between the IDLs. 2,4-D soil IDL decreases when agricultural commodity increases. Some nations such as Peru, United Kingdom, and Philippines had very high IDLs (0.0072 mg/kg-day) computed from food jurisdictions but relatively low IDLs computed from soil jurisdictions (i.e. 1.73E-07 mg/kg-day for Peru, 8.65E-08 mg/kg-day for United Kingdom, and 1.73E-08 mg/kg-day for

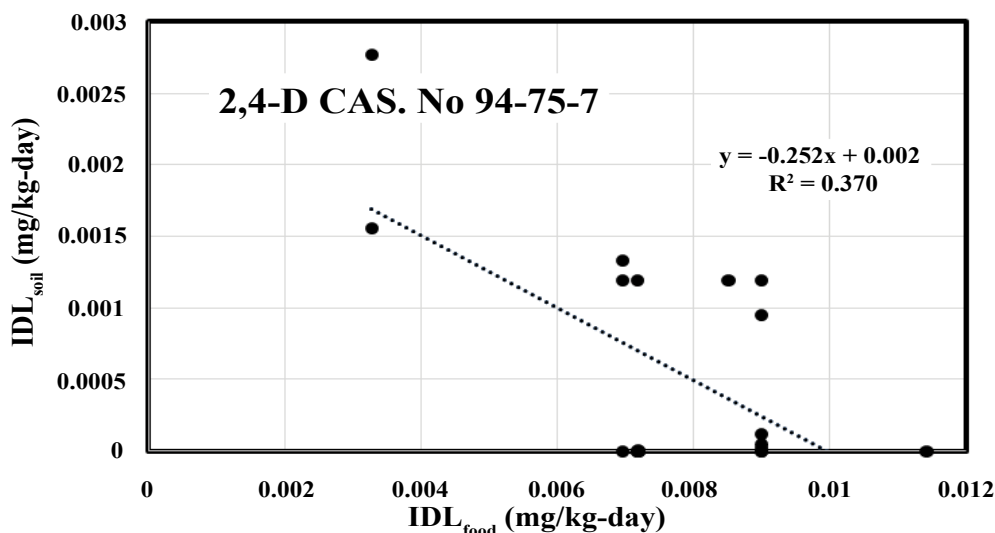
Philippines). On the other hand, the soil IDLs computed from the nations such as Chile and Ecuador were high but the food IDLs were low.

### Relationship among Chlorpyrifos PSVs in soil, drinking water, and agricultural commodity

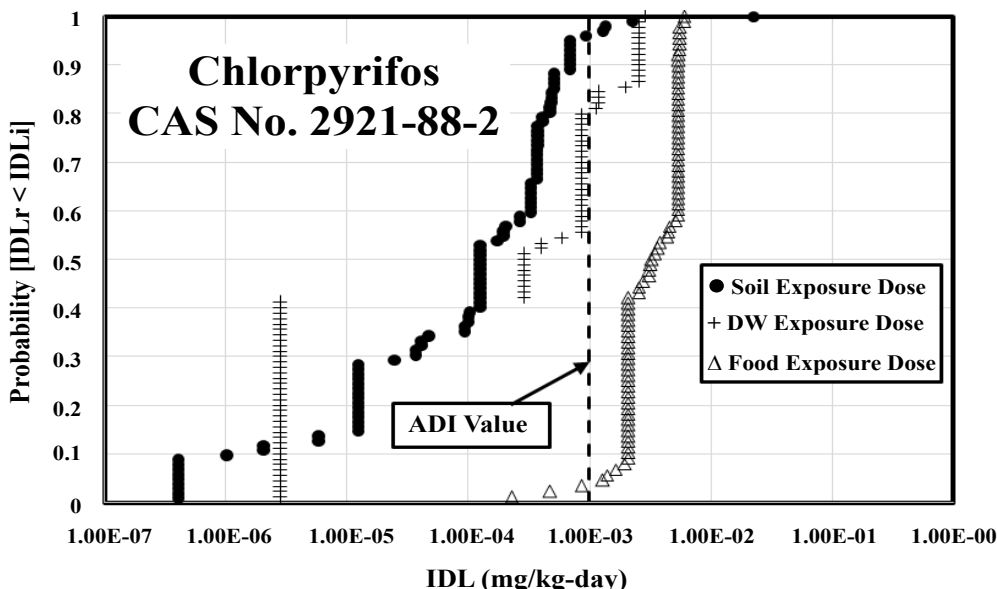
A total of 102, 90, and 91 computed Chlorpyrifos IDLs from Chlorpyrifos soil, drinking water, and agricultural commodity PSVs respectively were illustrated as CDFs in Figure 5. The IDLs computed from Chlorpyrifos soil RGVs span 4.74 orders of magnitude and the drinking water IDLs span 3 orders of magnitude. However, the IDLs computed from commonly consumed agricultural commodities only span 1.42 orders of magnitude. Pearson correlation coefficients for these IDLs listed in Table 2 suggest all of the Chlorpyrifos IDLs are well dispersed over the data spans. The median and geometric means of these IDLs suggest that worldwide Chlorpyrifos soil RGV and drinking water MCL sets are probably more conservative than MRL set.



**Figure 3:** Relationship between computed 2,4-D IDLs from drinking water MCLs and commonly consumed agricultural commodities MRLs.



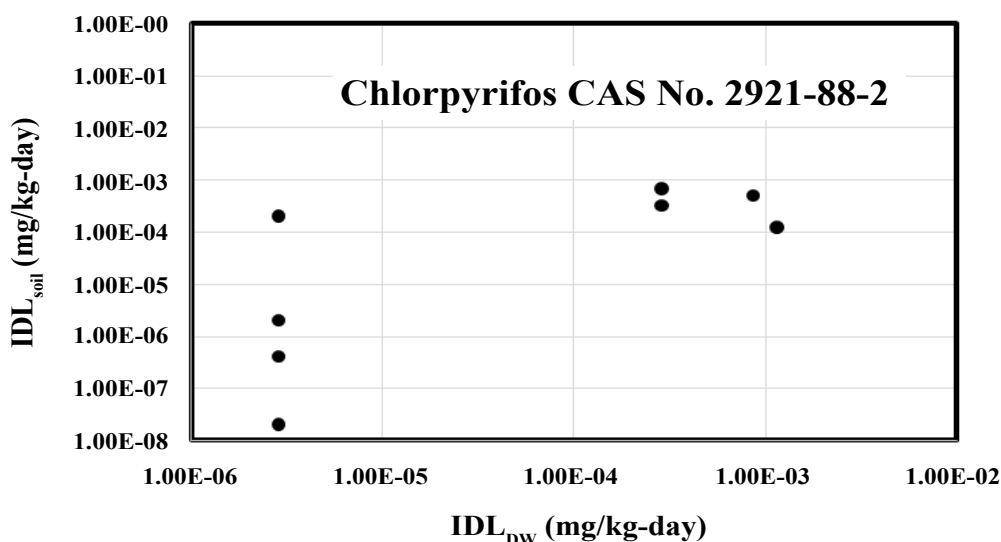
**Figure 4:** Relationship between computed 2,4-D IDLs from soil RGVs and commonly consumed agricultural commodities MRLs.



**Figure 5:** Worldwide computed Chlorpyrifos IDLs from residential soil RGVs, drinking water MCLs, and commonly consumed agricultural commodities MRLs plotted in the form of CDF.

**Table 2:** Statistic summary of Chlorpyrifos IDLs.

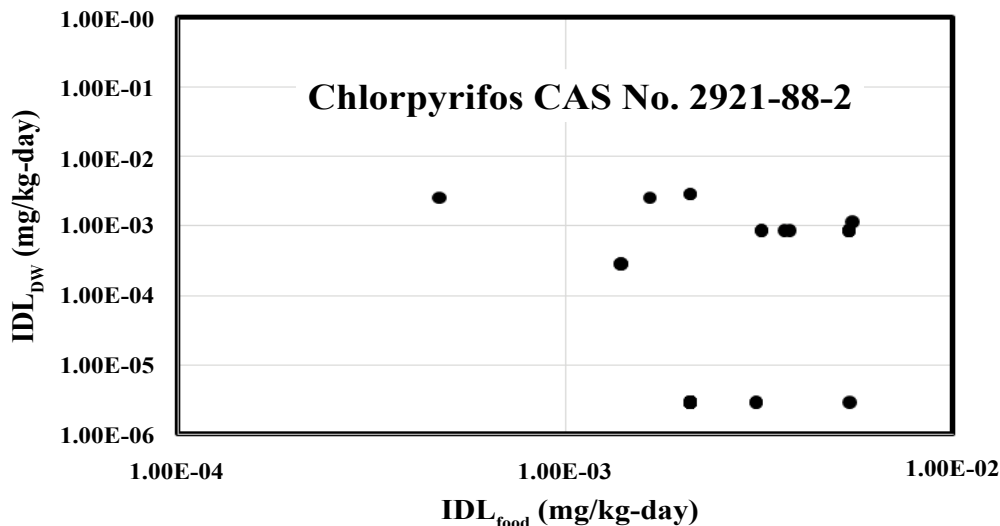
	Soil	Drinking water	Agricultural commodity
Number of IDLs	102	90	91
Arithmetic mean (mg/kg-day)	4.79E - 04	7.14E - 04	3.66E - 03
Median (mg/kg-day)	1.24E - 04	2.86E - 04	3.37E - 03
Geometric mean (mg/kg-day)	7.67E - 05	8.66E - 05	3.18E - 03
Standard deviation (mg/kg-day)	2.21E - 03	8.84E - 04	1.69E - 03
Max IDL (mg/kg-day)	2.24E - 02	2.86E - 03	6.01E - 03
Min IDL (mg/kg-day)	4.06E - 07	2.86E - 06	2.29E - 04
Order of magnitude variation	4.74E + 00	3.00E + 00	1.42E + 00
Correlation coefficient	0.96	0.911	0.929



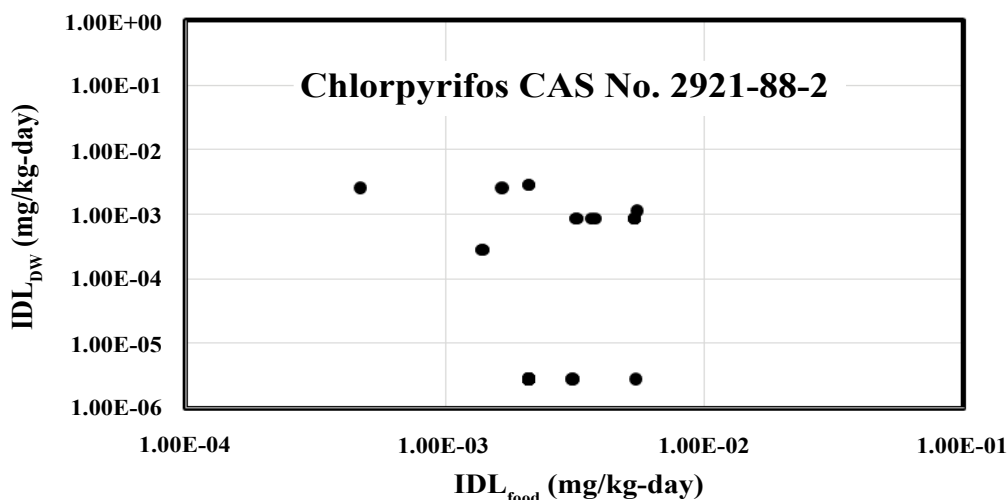
**Figure 6:** Relationship between computed Chlorpyrifos IDLs from drinking water MCLs and soil RGVs.

A total of eight national jurisdictions from the same nation regulated both Chlorpyrifos soil RGVs and drinking water MCLs. Figure 6 illustrates that there is no significant relationship between computed Chlorpyrifos IDLs from soil and water. Figure 7 illustrates that there is no significant relationship between 42 computed

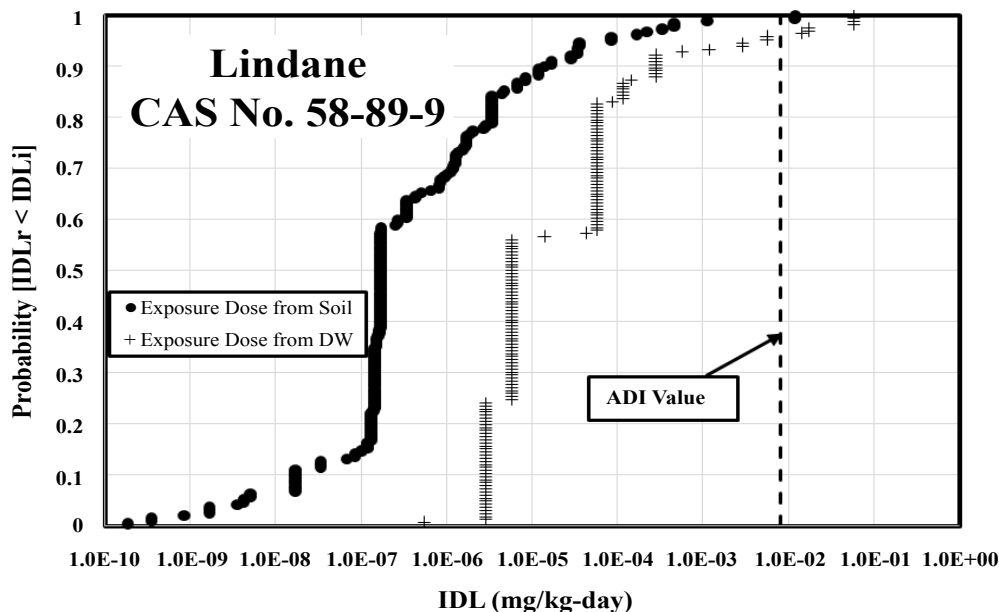
Chlorpyrifos IDLs (some jurisdictions share same PSVs) from drinking water MCLs and commonly consumed agricultural commodities MRLs. Moreover, there is no a significant relationship between the Chlorpyrifos IDLs computed from soil RGVs and agricultural commodity MRLs (Figure 8).



**Figure 7:** Relationship between computed Chlorpyrifos IDLs from drinking water MCLs and commonly consumed agricultural commodities MRLs.



**Figure 8:** Relationship between computed Chlorpyrifos IDLs from soil RGVs and commonly consumed agricultural commodities MRLs.



**Figure 9:** Worldwide computed Lindane IDLs from residential soil RGVs and drinking water MCLs plotted in the form of CDF.

## Relationship among Lindane PSVs in soil and drinking water

The Lindane IDLs were computed in soil and drinking water and there is no Lindane MRL regulated in agricultural commodity. A total of 190 and 66 computed Lindane IDLs from Lindane soil and drinking water PSVs respectively were illustrated as CDFs in Figure 9. The IDLs computed from Lindane soil RGVs span 7.81 orders of

magnitude and the drinking water IDLs span 5.02 orders of magnitude. Pearson correlation coefficients for these IDLs listed in Table 3 suggest all of the Lindane IDLs are well dispersed over the data spans. The median and geometric means of these IDLs suggest that worldwide Lindane soil RGV set is probably more conservative than drinking water MCL set.

A total of eight national jurisdictions from the same nation regulated both Lindane soil RGVs and drinking water MCLs. Figure 10 illustrates that there is no a significant relationship between computed Lindane IDLs from soil and water.

**Table 3:** Statistic summary of Lindane IDLs.

	Soil	Drinking water
Number of IDLs	190	166
Arithmetic mean (mg/kg-day)	1.41E - 04	1.82E - 03
Median (mg/kg-day)	1.21E - 03	9.01E - 03
Geometric mean (mg/kg-day)	1.67E - 07	5.71E - 06
Standard deviation (mg/kg-day)	4.08E - 07	2.18E - 05
Max IDL (mg/kg-day)	1.84E - 10	5.43E - 07
Min IDL (mg/kg-day)	1.19E - 02	5.71E - 02
Order of magnitude variation	7.81	5.02
Correlation coefficient	0.952	0.935

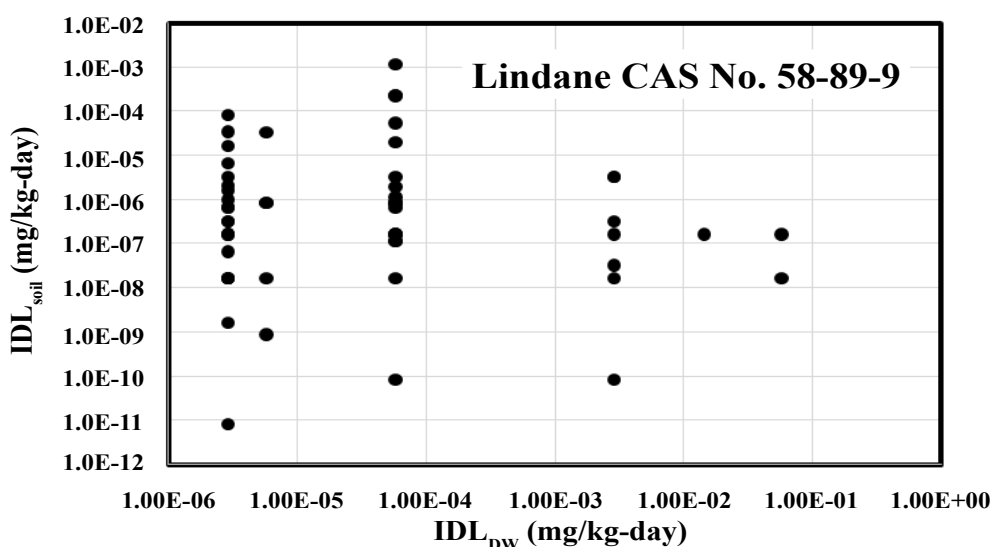
## Summary and Conclusions

### Current largely used pesticides

Table 4 provided the statistical summary of the IDLs computed from current largely used pesticides (Dicamba was omitted since few nations regulated PSVs for Dicamba in soil and drinking water). The means of the numbers of these pesticides IDLs computed from soil,

**Table 4:** IDLs statistics summary for current largely used pesticides.

Pesticide	Number of IDLs			Pearson correlation coefficient			Order of variance			IDLs median (mg/kg-day)			IDLs geometric mean (mg/kg-day)		
	Soil	Water	Food	Soil	Water	Food	Soil	Water	Food	Soil	Water	Food	Soil	Water	Food
2,4-D	147	180	91	0.907	0.861	0.761	5.48	5.48	1.33	1.09E - 03	8.57E - 04	6.95E - 03	2.11E - 04	4.33E - 04	5.63E - 03
Aldicarb	78	103	91	0.922	0.910	0.763	3.94	5.00	1.63	1.24E - 04	1.14E - 04	9.75E - 05	3.45E - 05	4.47E - 05	7.64E - 05
Atrazine	144	163	91	0.983	0.948	0.830	8.38	4.30	2.25	4.46E - 06	8.57E - 05	3.79E - 04	7.34E - 06	7.65E - 05	2.28E - 04
Chlorothalonil	67	54	91	0.903	0.816	0.897	5.80	3.45	2.06	3.25E - 04	2.86E - 06	2.02E - 03	1.89E - 04	2.01E - 05	2.68E - 03
Chlorpyrifos	102	90	91	0.960	0.911	0.929	4.74	3.00	1.42	1.24E - 04	2.86E - 04	3.37E - 03	7.67E - 05	8.66E - 05	3.18E - 03
Diazinon	86	71	95	0.945	0.901	0.949	3.93	3.48	1.59	7.21E - 05	2.86E - 06	2.59E - 04	2.05E - 05	3.41E - 05	1.59E - 04
Diuron	88	67	89	0.948	0.899	0.741	3.67	4.15	2.25	2.44E - 04	2.86E - 06	6.39E - 03	7.82E - 05	5.73E - 05	4.77E - 03
Glyphosate	93	122	91	0.904	0.864	0.907	6.51	5.45	3.67	3.05E - 03	2.00E - 02	6.53E - 02	1.11E - 03	1.21E - 03	5.17E - 02
Malathion	85	68	94	0.933	0.885	0.951	3.94	3.95	1.37	2.44E - 03	2.86E - 06	4.82E - 02	3.98E - 04	7.61E - 05	4.32E - 02
Mancozeb	54	47	90	0.589	0.711	0.949	5.34	3.62	2.22	3.66E - 03	2.86E - 06	7.04E - 03	6.95E - 04	8.52E - 06	6.43E - 03
MCPA	98	94	86	0.965	0.948	0.616	7.31	4.30	1.70	6.30E - 05	5.71E - 05	6.38E - 04	2.86E - 05	4.74E - 05	5.80E - 04
Metolachlor	83	77	51	0.908	0.930	0.703	6.42	4.45	1.25	4.67E - 03	2.86E - 04	7.57E - 05	1.29E - 03	8.69E - 05	7.98E - 05
Trifluralin	95	98	53	0.900	0.907	0.807	4.85	5.30	1.85	1.28E - 04	5.71E - 04	2.11E - 04	9.56E - 05	1.12E - 04	1.81E - 04



**Figure 10:** Relationship between computed Lindane IDLs from drinking water MCLs and soil RGVs.

**Table 5:** IDLs statistics summary for historical largely used pesticides.

Pesticide	Number of IDLs		Pearson correlation coefficient		Order of variance		IDLs median (mg/kg-day)		IDLs geometric mean (mg/kg-day)	
	Soil	Drinking water	Soil	Drinking water	Soil	Drinking water	Soil	Drinking water	Soil	Drinking water
Aldrin	241	110	0.977	0.817	7.22	6.31	2.03E - 07	8.57E - 07	4.97E - 07	2.03E - 06
Chlordane	224	163	0.990	0.939	7.52	5.40	4.68E - 06	1.52E - 05	4.98E - 06	5.71E - 06
DDT	318	115	0.977	0.965	7.53	6.96	3.22E - 06	2.86E - 05	5.44E - 06	3.10E - 05
Dieldrin	247	109	0.986	0.803	9.89	6.28	3.05E - 07	8.57E - 07	5.73E - 07	2.11E - 06
Endosulfan	189	52	0.965	0.792	8.48	3.00	7.52E - 05	2.86E - 06	3.64E - 05	1.36E - 05
Endrin	218	135	0.969	0.945	8.03	4.30	9.35E - 06	1.71E - 05	5.34E - 06	1.71E - 05
Heptachlor	212	137	0.978	0.940	6.52	6.10	4.06E - 07	8.57E - 06	9.21E - 07	5.87E - 06
Lindane	190	166	0.952	0.935	0.95	0.94	1.67E - 07	5.71E - 06	4.08E - 07	2.18E - 05
Pentachlorophenol	191	153	0.994	0.957	6.11	4.95	8.81E - 06	2.86E - 05	1.24E - 05	5.08E - 05

drinking water, and food jurisdictions are 94, 95, and 85 respectively. In some nations, there is more than one jurisdiction to regulated soil and water standards, for example, in U.S., each state, city, and county could develop its own PSVs. For agricultural commodity, each nation has only one jurisdiction probably because of international trade need. There are 106 soil 2,4-D RGVs from U.S.-related jurisdictions and 41 from elsewhere from the world. A total of 56 drinking water Glyphosate MCLs are from U.S.-related jurisdictions and 66 MCLs are from international jurisdictions. However, only single 2,4-D and Glyphosate MRL was found in each agricultural commodity. The weighted average Pearson correlation coefficients for these current largely used pesticides soil RGVs and drinking water MCLs is 0.918 and 0.898, respectively, which are higher than agricultural commodity MRLs (0.838). A total of 12 these pesticides' soil RGVs correlation coefficients are above 0.9. The correlation coefficient of Mancozeb soil RGVs is 0.589 because there are only 54 soil RGVs and the distribution is heavily skewed by the large data cluster. In addition, the orders of variance for IDLs computed from these major exposures indicate that worldwide jurisdictions probably agree more on agricultural commodity MRLs than soil RGVs and drinking water MCLs.

Previous studies show that median and geometric mean are better measures of PSVs central tendency [4-6]. The weighted average medians for soil and drinking water IDLs are 1.09 E-03 and 2.21 E-03 mg/kg-day respectively, and that for food IDLs is 1.17 E-02 mg/kg-day. Weighted average geometric means for soil and drinking water IDLs are 2.93 E-04 and 2.31 E-04 mg/kg-day respectively and that for agricultural commodity is 9.89 E-03 mg/kg-day. These results indicate that worldwide soil RGVs and drinking water MCLs are more conservative than agricultural MRL for current largely used pesticides. For most pesticides discussed in this study, there is no relationship among the IDLs computed from any two of these major exposures, which indicates that most worldwide jurisdictions develop their PSVs independently in soil, water, and food without considering other exposure pathways.

## Historical largely used pesticides

Table 5 provided the statistical summary of the IDLs computed from historical largely used pesticides (Bromomethane and Toxaphene were omitted because of few nations regulating soil RGVs and drinking water MCLs for these two pesticide). Since there is no jurisdiction providing agricultural commodity MRLs for these pesticides, only the information of the IDLs computed from soil RGVs and drinking water MCLs was summarized. The mean of the number of soil RGVs for these pesticides is 226 and that for drinking water MCLs is 127. DDT is the most regulated pesticide among the pesticides with 318 worldwide soil RGVs and Lindane is the most regulated pesticide in drinking water with 166 MCLs. Endosulfan were only regulated by 52 worldwide drinking water MCLs, and the U.S.-related jurisdictions did not provide the drinking water MCL for Endosulfan. Worldwide nations did not regulate pesticide agricultural commodity for these historical largely used pesticides because they had been banned in most countries and territories. However, since these POPs are environmentally persistent, it is necessary to regulate them for food to protect human health. The weighted average of Pearson correlation coefficients for these pesticides soil RGVs is 0.977 and for drinking water MCLs is 0.913. These pesticides soil RGVs weighted average of orders of variance is 7.07 while for drinking water MCLs it is 4.86. It suggests that worldwide soil RGVs are probably better dispersed over data spans for these historical largely used pesticides. The weighted averages of median (8.40 E-05 mg/kg-day) and geometric mean (6.79 E-06 mg/kg-day) for IDLs computed from these pesticides soil RGVs are less than those values in drinking water, which are 1.36 E-04 mg/kg-day for median and 1.77 E-05 mg/kg-day for geometric mean. This suggests that worldwide jurisdictions regulated soil RGVs more conservatively than drinking water MCLs in historical largely used pesticides. In addition, no relationship was found between IDLs computed from soil RGVs and drinking water MCLs for these historical largely used pesticides. Regulation of PSVs in each exposures is a systematical work, and hopefully the results in this research will help world-



wide regulatory jurisdictions make their PSVs in a more comprehensive approach by considering all major exposures.

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