Supporting Information

Using Isomeric and Metabolic Ratios of DDT to Identify the Sources and Fate of DDT in Chinese Agricultural Topsoil

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16 pages, 3 texts, 4 tables, 2 figures

Text S1. Detailed explanation for the starting year in kinetic model

In general, commercial technical DDT was used from 1951 to 1983 in China[1], but DDT's usage period and amount in different regions might be more or less different. Meanwhile, DDTs were continuously applied into the soils from 1951 to 1983 and the degradation of the related compounds took place continuously. Given the particularity of pesticide, commercial technical DDT had relatively fixed formulation and annual usage in a unit area had little variation in agricultural soil. Hence, the initial ratio of R*s,p,p′/p,p′* was approximately equal in different regions, and it is hypothesized that cumulative usage of technical DDT in a unit area increased linearly year by year in agricultural soil. Quantitative approach in kinetic model was based on the ratio of R*s,p,p′/p,p′* instead of the absolute concentrations of DDTs, it could minimize the impact on the quantitative analysis from the variation of DDT's usage period and amount in different regions. Although the application and degradation of technical DDT in soil took place continuously from 1951 to 1983, this study defined the starting year in the first-order kinetic model as the median of 1951 and 1983 which is based on the assumption that the cumulative usage of technical DDT in a unit area increased linearly year by year in agricultural soil.

Text S2. Detailed explanation for R*s,o,p′/p,p′* higher than 7.0

There are three possible reasons: 1) R*d,p,p′/p,p′* was an arithmetic mean derived from dicofol formulation of several provinces, which had an arithmetic mean 7.0 from Qiu's study[2] in China. In fact, some independent values of R*d,o,p′/p,p′* were a little higher than the arithmetic mean. 2) R*s,o,p′/p,p′* was an arithmetic mean of all soil samples for a region, if the concentration of *p,p′*-DDT was below the detection limit, and the concentration of *o,p′*-DDT was a little higher in some soil samples, the original authors usually set the half of the detection limit to calculate the concentration of *p,p′*-DDT, possibly it can increase independent value of R*d,o,p′/p,p′*. 3) *o,p′*-DDT has a little higher octanol-water partition coefficient (lgk_{ow} 6.79[3] for *o,p′*-DDT and 6.39[4] for *p,p′*-DDT), so that *o,p′*-DDT is more lipophilic than *p,p′*-DDT to adsorb on soil organic matter in topsoil.

Text S3. Detailed explanation for the influence of long-range transport

As China's topography is various and complicated, and western region is so mountainous that it possibly benefits mountain cold-trapping of POPs, such as in the mountain soils of western Sichuan of China[5]. Based on the global distribution of POPs[6], POPs evaporated from plants and soils in middle and low latitudes, then came into the atmosphere and migrated along with atmospheric flow, and deposited in the frigid zone such as Antarctic and Arctic. DDT tends to preferentially deposit and accumulate in mid-latitudes due to its relatively low mobility[6], which parts of Chinese regions are located in mid-latitudes. Furthermore, Yang et al.[7] found that there were 1~10 ng/g DDTs in primary forest soil in the southeast Tibetan Plateau of China, and the concentrations of DDTs maintained the linear correlations with the altitude. They suggested that dicofol-type DDT in the southeast Tibetan Plateau of China was derived from India Subcontinent via the Indian monsoon by the long-range transport and cold-trapped by the mountain. It is inferred that parts of western China possibly suffer from long-range transport of DDTs in the neighboring countries by current use of dicofol. However, most of our collected data were derived from central and eastern China. Hence, long-range transport has little influence on the residues and fate of DDT in our study areas.

Table S1. The database of DDTs in topsoil in China

Sample number	Sampling year	Sample quantity	Concentrations of DDTs in topsoil (ng/g)				Reference
			o,p′-DDT	*p,p′*-DDT	*p,p′*-DDE	*p,p′*-DDD	
S01	2013	1	1.30	3.91	2.45	0.74	[8]
S02	2009	25	3.09	1.47	0.76	0.59	[9]
S03	2011	33	2.38	8.58	0.85	0.50	[10]
S04	2004	544	4.93	25.00	49.00	9.85	[11]
S05	2012	26	0.07	0.16	0.24	0.07	[12]
S06	2009	44	10.64	52.74	73.38	9.26	[13]
S07	2009	60	12.51	74.27	79.08	8.57	[14]
S08	2010	48	0.86	1.48	0.32	0.01	[7]
S09	2011	18	3.44	3.16	3.44	1.32	[15]
S10	2011		2.24	5.07	1.28	1.14	[15]
S11	2009	61	3.43	7.55	12.82	6.27	[16]
S12	2009		1.44	4.44	7.97	2.73	[16]
S13	2009		0.05	0.10	0.34	0.21	[16]
S14	2009	24	3.42	49.82	14.16	3.77	[17]
S15	2006	58	21.00	9.60	21.00	20.00	[18]
S16	2009	93	2.89	0.73	1.97	2.08	[19]
S17	2009		2.13	0.67	1.62	2.18	[19]
S18	2009		4.66	0.56	3.17	4.22	[19]
S19	2009		16.90	0.45	2.99	1.74	[19]
S20	2009	5	16.11	75.53	173.22	40.44	[20]
S21	2009	17	29.40	1.66	2.95	0.64	[21]
S22	2009	8	2.31	1.01	1.65	0.28	[21]
S23	2004	544	4.90	25.00	49.00	9.90	[22]
S24	2006	12	1.66	4.85	11.93	1.88	[23]
S25	2008	87	3.01	1.84	22.50	4.57	[24]
S26	2008	31	1.40	6.60	7.80	0.60	[25]
S27	2006	245	11.47	13.50	4.35	1.69	[26]
S28	2007	36	1.66	3.26	16.14	4.56	[27]
S29	2006	29	14.02	28.99	134.24	2.60	[28]
S30	2006	5	2.28	3.25	6.13	1.29	[28]
S31	2003	43	2.91	13.46	20.82	5.25	[29]
S32	2006	10	0.90	1.76	2.19	1.34	[30]
S33	2003	131	2.32	19.06	50.10	5.71	[31]
S34	2004	7	6.92	27.78	27.90	19.02	[32]
S35	2004		2.64	8.55	22.78	11.50	[32]
S36	2004		2.67	7.96	7.51	3.78	[32]
S37	2003	63	14.02	46.50	40.79	8.42	[33]
S38	2005	7	11.77	16.70	17.20	17.67	[34]
S39	2001	188	0.90	27.50	18.80	3.40	[35]
S40	2001	8	8.39	13.66	39.72	2.68	[36]

S41	2002	37	6.34	20.90	21.90	7.22	[37]
S42	2012	8	0.05	1.08	1.03	0.30	[38]
S43	2012	37	40.04	50.41	9.27	14.06	[39]
S44	2013	33	0.11	0.32	0.52	0.30	[40]
S45	2008	106	4.92	12.00	14.00	4.50	[41]
S46	2008		0.36	1.00	8.00	4.50	[41]
S47	2009	160	2.34	7.67	19.64	7.23	[42]
S48	2009	18	4.39	0.24	0.81	0.51	[43]
S49	2009	64	5.95	41.51	24.00	2.48	[44]
S50	2009	36	46.32	44.48	2.44	8.02	[45]
S51	2010	36	19.66	48.41	23.57	3.46	[46]
S52	2009	31	5.75	10.56	157.65	2.48	[47]
S53	2008	408	79.07	184.61	67.42	33.18	[48]
S54	2009	42	4.64	9.01	3.96	1.03	[49]
S55	2010	69	6.20	13.40	24.00	12.10	[50]
S56	2008	137	3.22	34.37	74.16	17.01	[51]
S57	2005	299	4.77	6.92	10.74	5.73	[52]
S58	2007	5	3.90	15.80	12.10	2.10	[53]
S59	2005	286	0.39	0.38	1.80	0.44	[54]
S60	2003	129	1.57	2.24	6.91	0.44	[55]
S61	2006	131	*N.A.*	2.30	4.90	1.60	[56]
S62	2010	35	*N.A*	2.15	3.35	3.55	[57]
S63	2004	302	*N.A.*	12.80	48.70	2.09	[58]
S64	2005	26	*N.A.*	51.00	26.00	2.08	[59]
S65	2001	29	*N.A.*	56.43	28.25	4.98	[60]
S66	2005	75	*N.A.*	10.90	22.49	20.18	[61]
S67	2004	444	*N.A.*	2.41	5.09	2.68	[62]

Note: *N.A.* = not available.

For all original collected data in the existing literature, the data of DDT concentrations below detectable limit 0.1 ng/g were unaccepted, and the outlier more than three times the standard deviation were rejected.

Table S2. The fraction of DDT from technical DDT

Sample number	Sampling year	R*s,o,p′/p,p′*	The mean of *x*	Concentrations of DDT in topsoil (ng/g)	
				N*t*	N*d*
S01	2013	0.33	0.93	4.83	0.38
S02	2009	2.10	0.29	1.33	3.23
S03	2011	0.28	0.97	10.68	0.28
S04	2004	0.20	1.00	29.93	0
S05	2012	0.44	0.85	0.19	0.04
S06	2009	0.20	1.00	63.38	0
S07	2009	0.17	1.00	86.78	0
S08	2010	0.58	0.75	1.76	0.58
S09	2011	1.09	0.52	3.46	3.14
S10	2011	0.44	0.84	6.16	1.15
S11	2009	0.45	0.83	9.15	1.83
S12	2009	0.32	0.93	5.49	0.39
S13	2009	0.50	0.80	0.12	0.03
S14	2009	0.07	1.00	53.24	0
S15	2006	2.19	0.28	8.56	22.04
S16	2009	3.96	0.11	0.41	3.21
S17	2009	3.18	0.17	0.47	2.33
S18	2009	8.32	0	0	5.22
S19	2009	37.56	0	0	17.35
S20	2009	0.21	1.00	91.64	0
S21	2009	17.71	0	0	31.06
S22	2009	2.29	0.27	0.88	2.44
S23	2004	0.20	1.00	29.90	0
S24	2006	0.34	0.92	5.98	0.53
S25	2008	1.64	0.38	1.83	3.02
S26	2008	0.21	1.00	8.00	0
S27	2006	0.85	0.62	15.38	9.59
S28	2007	0.51	0.80	3.92	1.00
S29	2006	0.48	0.81	34.98	8.03
S30	2006	0.70	0.69	3.79	1.73
S31	2003	0.22	1.00	16.37	0
S32	2006	0.51	0.80	2.12	0.54
S33	2003	0.12	1.00	21.38	0
S34	2004	0.25	1.00	34.73	0
S35	2004	0.31	0.95	10.59	0.60
S36	2004	0.34	0.92	9.82	0.81
S37	2003	0.30	0.95	57.68	2.84
S38	2005	0.70	0.68	19.47	9.00
S39	2001	0.03	1.00	28.40	0
S40	2001	0.61	0.73	16.15	5.90

S41	2002	0.30	0.95	25.92	1.32
S42	2012	0.05	1.00	1.13	0
S43	2012	0.79	0.64	57.93	32.52
S44	2013	0.34	0.92	0.39	0.04
S45	2008	0.41	0.87	14.64	2.28
S46	2008	0.36	0.90	1.23	0.13
S47	2009	0.31	0.95	9.51	0.50
S48	2009	18.29	0	0	4.63
S49	2009	0.14	1.00	47.46	0
S50	2009	1.04	0.54	49.08	41.72
S51	2010	0.41	0.87	59.11	8.96
S52	2009	0.54	0.77	12.62	3.69
S53	2008	0.43	0.85	224.67	39.01
S54	2009	0.51	0.79	10.82	2.83
S55	2010	0.46	0.83	16.22	3.38
S56	2008	0.09	1.00	37.59	0
S57	2005	0.69	0.69	8.09	3.60
S58	2007	0.25	1.00	19.70	0
S59	2005	1.03	0.55	0.42	0.35
S60	2003	0.70	0.69	2.61	1.20

The mean of x ranges from 0.11 to 1.0 with arithmetic mean 0.74 ± 0.30 and median mean (interquartile range) 0.84 (0.34).

Table S3. The correction ratio y in agricultural topsoil in China

Sample number	Sampling year	The mean of x	The mean of y ($1.25/x \cdot R_{s,p,p'/p,p'}$)	The degradation time of t (yr, starting year 1967)
S02	2009	0.29	1.27	42
S04	2004	1.00	2.35	37
S06	2009	1.00	1.57	42
S07	2009	1.00	1.18	42
S11	2009	0.83	2.61	42
S12	2009	0.93	2.44	42
S13	2009	0.80	5.71	42
S15	2006	0.28	5.99	39
S16	2009	0.11	12.31	42
S17	2009	0.17	10.02	42
S20	2009	1.00	2.36	37
S24	2006	0.92	2.89	39
S26	2008	1.00	1.27	41
S28	2007	0.80	6.60	40
S29	2006	0.81	4.89	39
S30	2006	0.69	2.45	39
S31	2003	1.00	1.94	36
S33	2003	1.00	2.93	36
S34	2004	1.00	1.69	37
S35	2004	0.95	4.04	37
S38	2005	0.68	2.24	36
S39	2001	1.00	0.81	34
S40	2001	0.73	3.28	34
S41	2002	0.95	1.40	35
S45	2008	0.87	1.58	41
S46	2008	0.90	12.71	41
S47	2009	0.95	3.53	42
S53	2008	0.85	0.56	41
S54	2009	0.79	0.58	42
S56	2008	1.00	2.65	41
S57	2005	0.69	2.55	38
S58	2007	1.00	0.90	40
S60	2003	0.69	3.52	36

Table S4. Vertical distribution of R*s,p,p′/p,p′* in deep soil from pesticide-producing sites

Soil core number	Depth (m)	R*s,p,p′/p,p′*	$R_{DDE/DDD}$
D1	5.0 ~ 6.0	*N.D.*	**>100**
	7.0 ~ 8.0	0.2	0.1
	10.0 ~ 11.0	0.1	0.7
	13.0 ~ 14.0	0.2	0.9
	16.0 ~ 17.5	*N.D.*	**>100**
	20.0 ~21.5	0.2	0.3
D2	5.2 ~ 5.6	0.1	0.9
	6.2 ~6.7	0.1	0.8
	7.2 ~ 7.8	0.3	1.4
	8.4 ~ 9.3	0.2	0.7
	10.4 ~ 10.9	0.3	0.8
	12.0 ~ 12.5	0.3	0.3
	13.6 ~ 13.9	0.2	1.0
	15.6 ~ 16.0	0.4	5.0
D3	4.8 ~ 5.2	0.3	0.2
	6.6 ~ 7.0	0.3	**>100**
	8.0 ~ 8.3	0.3	1.4
	11.0 ~ 11.5	*N.D.*	**<0.01**
	12.7 ~ 13.0	0.3	1.4
	15.0 ~ 15.4	0.6	0.6
	16.4 ~ 16.8	0.6	0.2
D4	5.1 ~ 5.5	0.2	**<0.01**
	7.6 ~ 8.0	0.3	2.0
	11.0 ~ 11.5	0.4	0.5
	12.8 ~ 13.2	0.3	2.0
D5	5.0 ~ 5.5	0.6	0.8
	7.5 ~ 8.0	0.1	1.1
	10.5 ~ 11.0	1.1	1.3
	12.0 ~ 12.5	0.3	0.4
D6	5.0 ~ 5.5	0.4	1.4
	8.0 ~ 8.5	0.9	5.0

Note: *N.D.* = not detectable for *p,p′*-DDT or its metabolites (*p,p′*-DDE and *p,p′*-DDD); **>100**, DDD undetected; **<0.01**, DDE undetected.

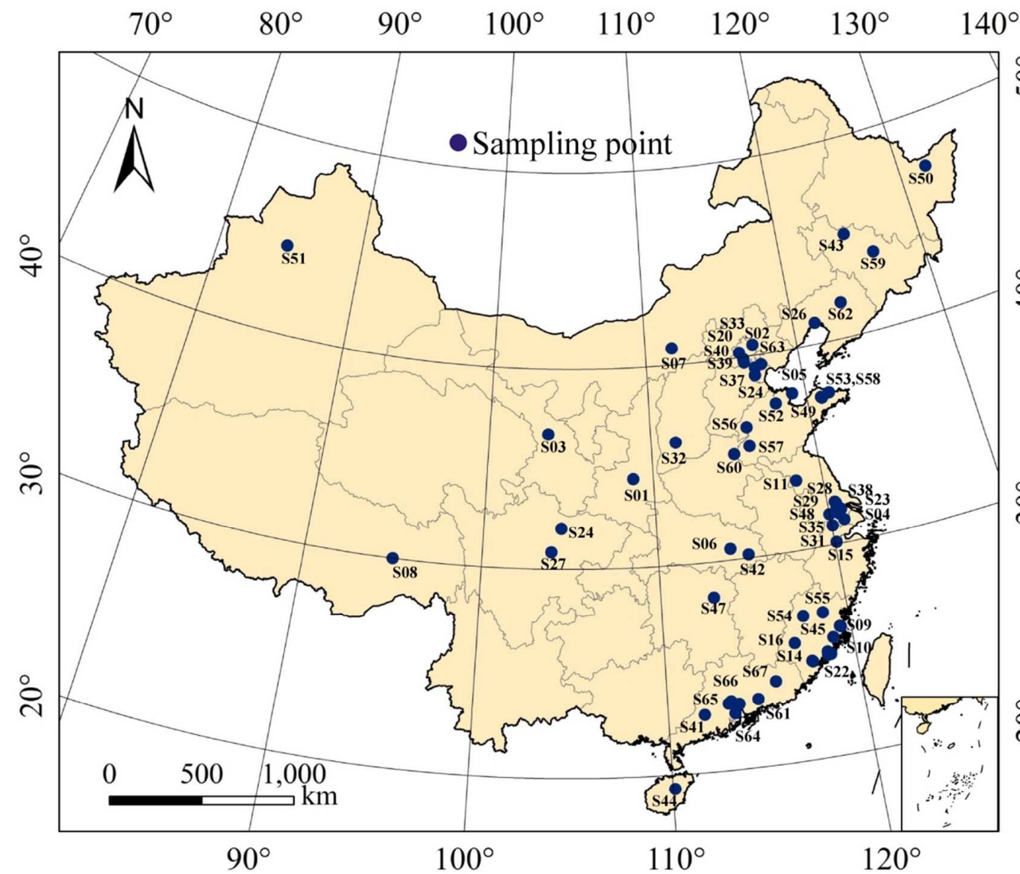

Figure S1. The location of sampling points in topsoil in China. (If a sampling point was derived from a large survey area, this point would be located in the center of this survey area.)

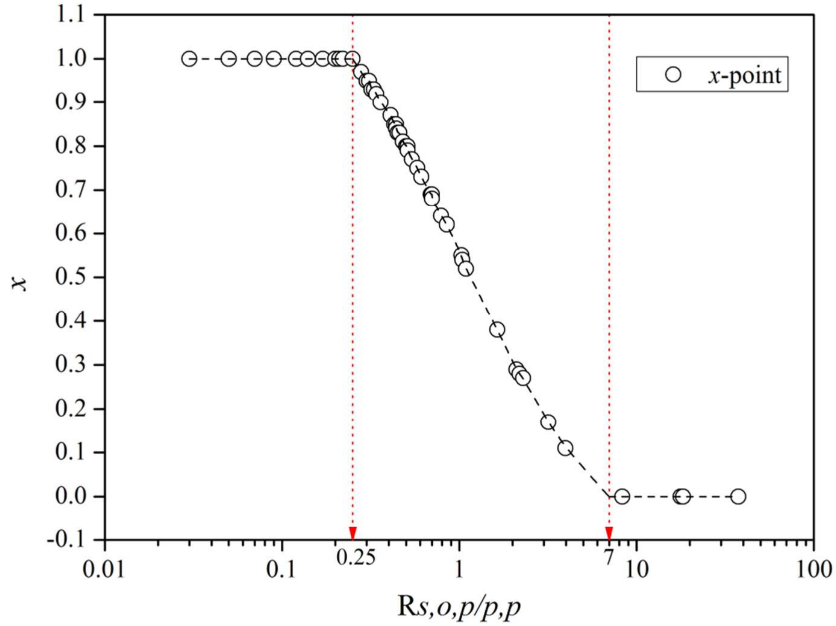

Figure S2. The variation in x *vs.* R$s,o,p'/p,p'$.

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