

Supplementary Material

Input Flux and the Risk of Heavy Metal(Loid) of Agricultural Soil in China: Based on Spatiotemporal Heterogeneity from 2000 to 2021

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2.1 Data collection and database building

With comprehensive consideration, the following factors need to be taken into account in our study: atmospheric deposition, fertilizer, livestock and poultry manure, irrigation water, while factors that have a low impact such as agrochemical or are not applied universally such as industrial waste and sludge are not considered for the fewer contributions to the quantities of total input.

The relevant publications were collected from Web of Science (WOS) and China National Knowledge Infrastructure (CNKI) with the following search term: *soil AND (agricultural OR farmland OR cropland) AND (heavy metal* OR trace element* OR As OR Cd OR Cr OR Cu OR Hg OR Ni OR Pb OR Zn) AND (flux* OR inventory OR input* OR source*) AND (atmospheric deposition OR fertilizer OR irrigation OR manure) AND (China OR Chinese).*

The 109 publications used to create the database are listed in the references[1-109].

Table S1. Descriptive statistics of heavy metal input fluxes in China (mg/m²/yr)

Source		As	Cd	Cr	Cu	Hg	Ni	Pb	Zn	Total
Atmospheric deposition	N	164	235	149	143	125	91	199	141	271
	n	1516	1947	1024	924	1455	777	1621	903	
	Min	0.04	0.03	0.14	0.56	0.003	0.19	0.42	3.4	
	Max	8.69	1.73	34.34	34.3	0.144	15.51	59.3	231.52	
	Mean	2.73	0.49	10.62	11.49	0.037	4.99	19.93	63.82	
	SD	2.09	0.38	8.08	7.68	0.031	3.49	13.53	45.79	
Fertilization	N	91	149	72	78	96	18	110	75	156
	n	764	1450	477	458	770	274	869	444	
	Min	0.051	0.001	0.14	0	0	0.35	0.0054	0.27	
	Max	2.04	0.319	3.24	5.45	0.09	3.57	10.78	22	
	Mean	0.58	0.05	0.99	1.03	0.012	0.82	0.75	5.18	
	SD	0.44	0.07	0.76	1.07	0.018	0.8	1.32	5	
Irrigation	N	81	146	68	78	78	39	111	80	167
	n	3360	1780	870	893	804	682	1000	931	
	Min	0	0	0	0	0	0	0	0.021	
	Max	8.82	0.47	13	7.55	0.468	5.31	7.03	55.35	
	Mean	1.82	0.09	2.27	1.68	0.05	0.79	1.29	8.21	
	SD	2.1	0.12	3.29	1.69	0.086	1.44	1.75	11.4	

Note: N (Sites); n (Samples)

Table S2. Comparison of researches on atmospheric deposition fluxes of heavy metals in China
(mg/m²/yr)

	This study (2000-2020)			Peng [110] (2008-2018)			Ni and Ma [111] (2006-2015)			Wang [112] (1995-2015)		
	n	WM	RSD	n	Mean	RSD	n	Mean	RSD	n	Mean	RSD
As	1516	2.67	77%	574	3.39	68%	755	3.37	67%	254	2.54	99%
Cd	1947	0.49	78%	625	0.59	80%	762	0.41	88%	310	0.37	256%
Cr	1024	10.77	76%	577	22.32	76%	763	16.14	72%	226	10.38	264%
Cu	924	11.54	67%	527	16.23	141%	764	11.56	74%	232	10.99	90%
Hg	1455	0.039	84%	553	0.07	71%	750	0.09	202%	262	0.07	230%
Ni	777	5.06	70%	364	8.61	133%	376	8.08	122%	159	4.79	170%
Pb	1621	20.64	68%	608	26.29	67%	768	19.19	72%	319	21.81	175%
Zn	903	64	72%	617	108.75	88%	766	72.90	124%	279	78.87	206%

Note: n (Samples); WM (Weighted mean); RSD (Relative standard deviation)

2.2 Weighted average calculation

Table S3. Agricultural utilization rate of livestock and poultry manures in China

Region	Utilization rate	References
Hubei	30%	[113]
Lanzhou, Gansu	40%	[114]
Cenxi, Guangxi	60%	[115]
Guizhou	40%	[116]
Jilin, Hebei, Sichuan, Zhejiang, Anhui	80%	[117]
Zhengzhou, Henan	61%	[118]
Yinchuan, Ningxia	60%	[119]
Liaozhong, Liaoning	64%	[120]
Jinzhou, Liaoning	80%	[121]
Taixing, Jiangsu	50%	[122]
Luochuan, Shanxi	60%	[123]
Inner Mongolia, Liaoning, Jilin, Heilongjiang	50%	[124]
Jilin, Hebei, Sichuan, Zhejiang, Anhui	63%	[125]
Average	57%	
Utilization rate used in this study	21%	

The planting area of grain crops accounts for 70% of the total arable land in China, and the manure is applied more to cash crops, protected vegetable fields, and orchards than to grain crop fields. Therefore, according to the amount of manure applied per unit area and the total amount of excrement produced, the utilization rate of grain crop fields was calculated as 21%.

Table S4. Parameters of livestock and poultry excrements

	Excretion parameters [126] (kg/day)	Water contents [127] (%)
Chicken	0.37	53.64
Pig	3.14~3.65	84.2
Sheep	2.25	61.1
Cattle	18.71~25.015	81.1
Horse	16.16	75.1
Donkey	13.9	71.4
Mule	13.9	72.1
Rabbit	0.37	76.74

2.4 Soil environment risk assessment

Table S5. Assessment standards of soil environmental capacity

Level	P_i	Risk level	Description
I	$P_i > 1$	No risk	High capacity
II	$0.7 \leq P_i < 1$	Mild risk	Medium capacity
III	$0.3 \leq P_i < 0.7$	Moderate risk	Low capacity
IV	$0 \leq P_i < 0.3$	Severe risk	Alert
V	$P_i < 0$	Extreme risk	Overload

Table S6. Risk screening values for soil contamination of agricultural land (mg/kg) [128]

Element	$pH \leq 5.5$	$5.5 < pH \leq 6.5$	$6.5 < pH \leq 7.5$	$pH > 7.5$
As	30	30	25	20
Cd	0.3	0.3	0.3	0.6
Cr	150	150	200	250
Cu	50	50	100	100
Hg	0.5	0.5	0.6	1
Ni	60	70	100	190
Pb	70	90	120	170
Zn	200	200	250	300

3.1 Zoning of heavy metal input flux

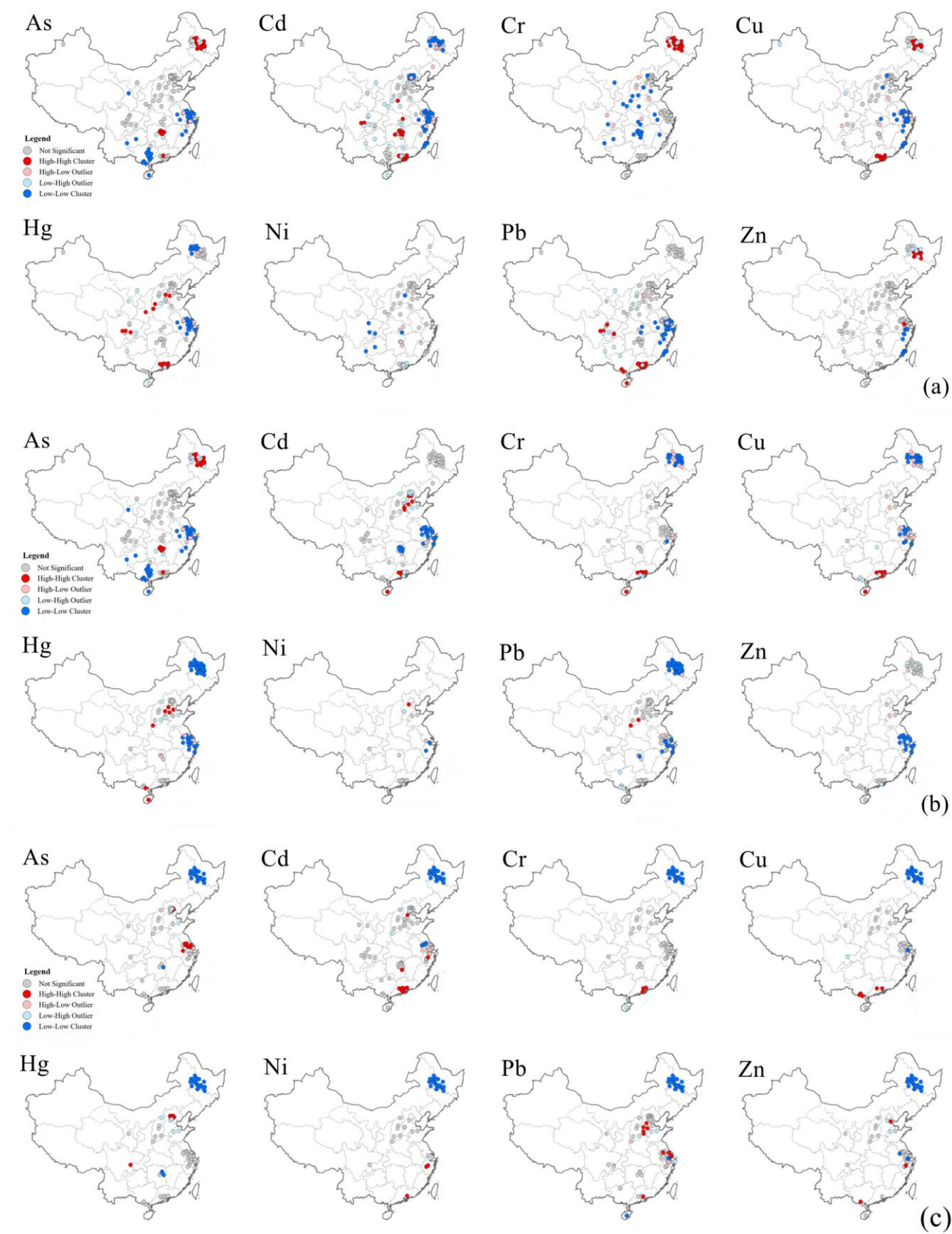


Figure S1. The LISA of heavy metal input flux via atmospheric deposition (a), fertilizer (b), irrigation (c)

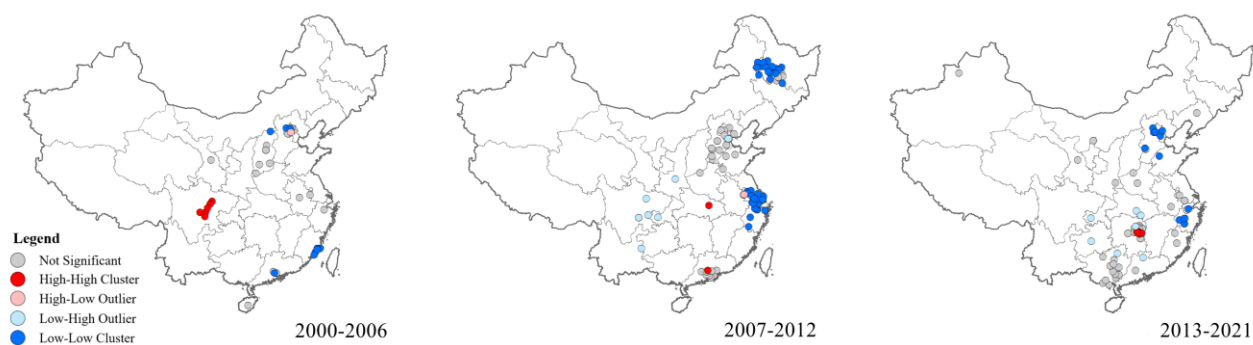


Figure S2. The LISA of Cd input flux via atmospheric deposition in different periods

3.2.3 Manure

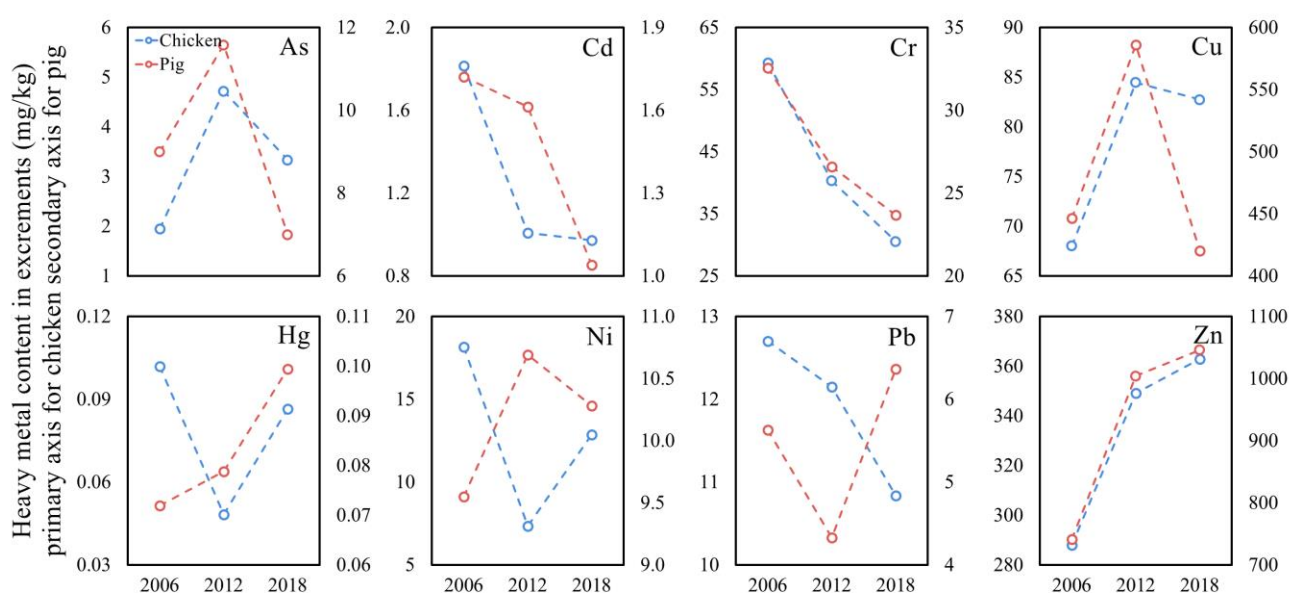


Figure S3. Heavy metal content in livestock and poultry excrements (2000-2021)

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