

ORIGINAL ARTICLE

## Heavy metal contamination at highland agricultural soil at Dan Sai District, Loei Province, Thailand

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### ABSTRACT

Although soil quality of highland was proper for cultivation, most farmers still used chemical substances i.e. pesticide, herbicide, and fertilizer. Chemical utilization could cause hazard for human health because toxic contamination can transfer through food chain. Therefore, this study evaluated heavy metal contamination in agricultural soil at highland during dry season at Dan Sai District, Loei Province. Soil samples were randomly collected at 15 cm depth from top soil at paddy fields, pepper fields, banana fields, pineapple fields, and para tree fields because the heavy metal could be transferred from the soil to crop production. The results showed that maximum heavy metal contamination was iron (Fe 85.92–593.98 mg/kg), followed by magnesium (Mg 6.22–23.67 mg/kg), copper (Cu 3.25–17.04 mg/kg), manganese (Mn 0.62–7.12 mg/kg) and zinc (Zn 0.70–2.30 mg/kg), respectively. The pineapple field at high slope highland (HPi) was contaminated by Zn (2.30 mg/kg), Mg (23.67 mg/kg), and Fe (593.98 mg/kg). The soil of banana field at low slope highland (LB) was contaminated by Mg 7.12 mg/kg. The soil of paddy field at low slope highland (LR) was contaminated by Cu 17.04 mg/kg. Average results of soil organic material were 0.08%–3.75%. The soil was under extreme and moderate acidic condition (pH 4.67–5.67). The acidic condition can extract Cu, Zn, Mg, and Fe from soils to leachate in form of bioavailability. The outcome of metal monitoring at agricultural areas is useful for evaluating hazardous index and health risk assessment.

**Keywords:** food safety, heavy metal, highland cultivation, soil contamination.

## การปนเปื้อนโลหะหนักในดินการเกษตรที่สูง อำเภอด่านซ้าย จังหวัดเลย ประเทศไทย

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### บทคัดย่อ

ถึงแม้ว่าคุณภาพดินบนที่สูงเหมาะสมต่อการเพาะปลูก แต่มีการใช้สารเคมี เช่น ยาฆ่าแมลง ยาปราบวัชพืช และปุ๋ย ซึ่งอาจอันตรายต่อสุขภาพเนื่องจากการปนเปื้อนสารพิษสามารถถ่ายทอดผ่านห่วงโซ่อาหาร ดังนั้น งานวิจัยนี้จึงประเมินการปนเปื้อนโลหะหนักในดินเกษตรที่สูงในช่วงฤดูแล้ง อำเภอด่านซ้าย จังหวัดเลย โดยสุ่มเก็บตัวอย่างดินที่ความลึก 15 เซนติเมตรจากผิวดิน ในแปลงนาข้าว พริกไทย กล้วย สับปะรด และสวนยางพารา เนื่องจากโลหะหนักอาจส่งผ่านจากดินสู่ผลผลิตการเกษตรส่งผลต่อสุขภาพของผู้บริโภค ผลการศึกษาพบว่า การปนเปื้อนโลหะหนักสูงสุด (85.92–593.98 mg/kg) รองลงมาคือ แมกนีเซียม (6.22–23.67 mg/kg) ทองแดง (3.25–17.04 mg/kg) แมงกานีส (0.62–7.12 mg/kg) และสังกะสี (0.70–2.30 mg/kg) โดยแปลงสับปะรดพื้นที่ลาดชันบนเขา (HPi) มีการปนเปื้อนของสังกะสี แมกนีเซียม และเหล็กมีค่าเฉลี่ยสูงสุดเท่ากับ 2.30, 23.67 และ 593.98 mg/kg ตามลำดับ ส่วนแปลงกล้วยพื้นที่ลาดชันต่ำ (LB) พบแมงกานีส 7.12 mg/kg และแปลงข้าวพื้นที่ลาดชันต่ำ (LR) พบทองแดง 17.04 mg/kg สำหรับอินทรีย์วัตถุในดินทุกแปลงมีค่า 0.08%–3.75% และดินมีสภาพเป็นกรดจัดมากถึงกรดปานกลาง (pH 4.67–5.67) สามารถละลายโลหะทองแดง สังกะสี แมงกานีส และเหล็กในดินได้ดี และอยู่ในรูปที่สามารถเข้าสู่สิ่งมีชีวิตได้ ผลลัพธ์ของการตรวจวัดโลหะหนักเป็นประโยชน์สำหรับการประเมินดัชนีอันตรายและการประเมินความเสี่ยงต่อสุขภาพ

**คำสำคัญ :** ความปลอดภัยของอาหาร, เกษตรที่สูง, โลหะหนัก, การปนเปื้อนในดิน

## INTRODUCTION

Food safety has been integral to sustainable development goals to ensure healthy lives and promote well-being. From the FAO statistic report, every year more than 600 million people get sick and 420,000 die because of eating contaminated food with bacteria, viruses, parasites, toxins or chemicals<sup>1</sup>. Food products from one country may effect on human health of other countries because of trading among countries so food safety is a global important issue that people have to realize in every country, including Thailand that export agricultural products for 252,957 million USD<sup>2</sup>. Thailand exports approximately 80% of total agricultural products. Major export agricultural products were rice, maize, beans, tapioca, pepper, rubber, coffee, tobacco, cereals, preserved fruits (i.e. canned pineapple, lychee, longan, mango), fruit juices (i.e. pineapple juices, orange juices, mixed fruit juices) and various types of meat. Therefore, Thailand is an important agricultural food exporter of the world. Thus, agricultural practice of Thai farmers is important to global food safety.

Heavy metal has spread through agricultural areas by utilization of herbicide, pesticide, and chemical fertilizer<sup>3</sup>. The list of toxic substance portal includes heavy metal (Al, As, Cd, Cu, Mn, Ni, Pb, and Zn) in the list of dangerous substances by the United States Environmental Protection Agency and the Agency for Toxic Substances and Disease Registry (ATSDR)<sup>4</sup>. The heavy metal from soil will be up-taken to the plant and finally transferred into human food chain through various pathways. Pathway of the toxic substances in the environment could be entered via aerial deposition, surface water, or soil by metal-containing pesticide and fertilizer applying<sup>5,6</sup>. When the heavy contaminated into the soil, it

would be accumulated for years, posing long term risks. It can be transferred from soil to plants, microbes, vertebrates, and mammals<sup>7</sup>. Contamination of these hazardous chemical into the water could be by direct and indirect ways. The acidic condition, such as acid rain, able to enhance metal mobilization in soil ecosystems to the water ecosystems by replacing cation by the H<sup>+</sup> ion in the acidic water, reducing cation exchange capacity, and increasing concentrations of the cations (Cu, Pb and Cd) in the soil-water system<sup>8,9</sup>. The contaminants in soil can be transferred from a stabilized matrix to liquid medium (such as water or other solutions) through leaching<sup>9</sup>. If people consume contaminated crops for a long time, their health will be deleterious because heavy metals are highly reactive and toxic at low concentrations on human health and ecosystem<sup>7,10,11</sup>.

Cultivation areas at highland, which is farming practices at 500 m above mean sea level covers 100,432 square kilometers or 53 percent of 20 provinces in Thailand<sup>12</sup>. Most areas are located at upstream so the leachate from upstream can have effect on downstream as Namman River. The Namman River extends from Phuhinrongkla National Park to Namhoueng Rivers which is a sub-catchment of the Mekong River Basin in the Northeast of Thailand. Most people at highland (88 percent) are poor because they earn low income. The land were cleared for cultivation by cut the trees and open burning to cultivate crops such as pepper, corn, rice, para rubber, pineapple, orange, cabbage, and dragon fruit. These crops are suitable for cultivate in highland area and produce high products. Because geography of highland is mountain, the weather condition is cold through the year with average temperature 27°C, and average rainfall 1,572.5 mm annually<sup>13</sup>.

Problems of highland are important and need sustainable solution. Not only poor problems are important, but also not properly chemical utilization in agricultural practice. The improper use of chemical substances resulted in pesticide residues in products and environment that can be found in soil and water. Lacking of knowledges and skill in good agricultural practice, chemical substances had effect in water resources and community at lowland. Soil degradation problem in highland covered the damage area around 96 percent of total area because the high slope increased risk of soil erosion, especially at monoculture and open burning area. The bare areas were leached directly by dropping of rainfall. The runoff flew down along the slope without barrier so the top soil was damage around 7.5-12.5 kg/m<sup>2</sup> annually<sup>12</sup>.

The contamination of heavy metal in soil was originated from chemical utilization in agricultural practices such as pesticide, herbicide, chemical fertilizer, and organic fertilizer<sup>14</sup>. Concentration of heavy metal in soil at highland area was high so it had effect on product quality, ecosystem, and environment. One of the effects of this contamination was poor quality and not fertile of soil. The soil condition became acidic that could extract heavy metal in the soil into soluble form. The acidic condition also had influence on nutrient and compound of soil. Moreover, leachate that contained heavy metal flew down to natural surface water and ground water<sup>14</sup>. Properties of each type of heavy metal were different. Some of them remained in cell organisms and transferred to other livings through food chain. Human was the last consumer so the amount of heavy metal accumulation in food chain was large enough to be toxic on body, neuro system, muscle system, and

respiratory system. Therefore, objective of this study is to monitor heavy metal contamination of soil at highland area during dry season in Amphoe Dan Sai, Loei Province, Thailand. In this area, main land use was agriculture and household. There is no industry or mining so contamination of heavy metal is originated from chemical substance from agricultural activities. Information of this study were useful for people and administrative in highland area to realize on the adverse effect of chemical utilization in agricultural practices. Moreover, the results of this case study could be baseline information for highland area in tropical region that has the same climate and geography

## METHODOLOGY

Soil samples were randomly collected at 15 cm depth from top soil at agricultural highland area at Prawat canal (sub branch of Mun River Amphoe Dan Sai, Loei Province). The sampling sites were selected by stratified random sampling in agricultural area. There were 9 sampling sites: rice field on high slope highland, Namyen village (HR), rice field on low slope highland, Namyen village (LR), mix pepper and soya field on high slope highland, Namyen village (HPS), pepper field low slope highland, Namyen village (LPe), high slope highland pineapple field (HPi), low slope highland banana field (LB), low slope highland banana mixed with pineapple field (LBP), low slope highland para rubber tree field (LL), and low slope highland banana field at linkage area between Mun river and Namhoueng River estuary (RB). Summary of sampling sites is presented in Table 1.

**Table 1:** Sampling points and coordinates at Dan Sai district, Loei province

Sampling Point Symbols	Coordinates	
	Longitude (degree)	Latitude (degree)
Hpi	101.12464E	17.14325N
HPS	101.10447E	17.14310N
HR	101.10588E	17.14376N
LB	101.17744E	17.49492N
LBP	101.13420E	17.14217N
LL	101.17744E	17.49492N
LPe	101.10466E	17.14318N
LR	101.10447E	17.14310N
RB	101.13493E	17.14338N

**Note:**

*Hpi means pineapple field on high slope highland, Nava village*

*HPS means mix pepper and soya field on high slope highland, Namyen village*

*HR means rice field on high slope highland, Namyen village*

*LB means banana field on low slope highland, Nava village*

*LBP means mix banana and pineapple field on low slope highland, Namyen village*

*LL means rubber tree field on low slope highland, Nava village*

*LPe means pepper field low slope highland, Namyen village*

*LR means rice field on low slope highland, Namyen village*

*RB means banana field on high slope highland, inter-point between Pakman and Namhoueng Rivers*

The soil sampling collection was conducted during 18-21 December 2017 in winter season of Thailand. <sup>15</sup>The samples were kept at 4°C during transportation from the fields to the laboratory. The samples were dried, ground, and sieved by 2 and 0.5 mm filters. The pH of sieved soil was measured by electrometric method. <sup>16</sup>Organic matter in soil was measured by Walk & Black Modified Acid-dichromate Digestion FeSO<sub>4</sub> Titration Method. <sup>17</sup>Analysis of heavy metal (Cu, Fe, Zn, Na, and Mg) was conducted by digesting soil with HClO<sub>4</sub>:HNO<sub>3</sub> at proportion of 2:1 and then measured heavy metal concentration by Atomic Absorption Spectrophotometer, brand Perkin Elmer, model PinAAcle 900T, United States of America.

The results were analyzed by statistical method One-Way ANOVA. Then the data was compared with average data at each sampling point by Least Significant Difference (LSD) at confidential level 95% (p<0.05) by Statistic 8 Software (Version 8, USA).

## RESULTS AND DISCUSSION

Results of pH analysis and soil organic matter (SOM) analysis at 9 agricultural sampling sites are presented in Table 2.

**Table 2:** Results of heavy metal concentrations and soil chemistry in highland agricultural soil

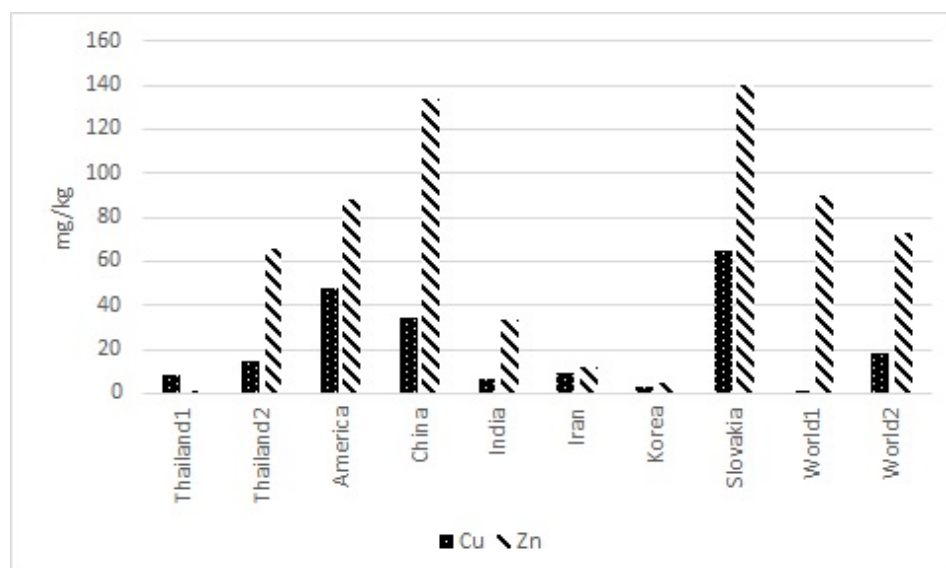
Sampling points	Soil chemistry		Heavy metal concentrations (mg/kg soil-dry weight, n=3*)					
	pH	SOM (%)	Cu	Fe	Mg	Mn	Na	Zn
HR	5.00	1.32	3.25	85.92	10.83	1.07	89.56**	0.78
LR	4.67	1.60	17.04**	382.06	6.22	0.62	43.77	1.13
HPS	5.00	1.99	10.30	400.63	7.52	0.86	33.97	0.70
LPe	5.00	3.56	4.22	247.21	9.35	2.30	63.70	0.76
HPi	5.67**	3.36	5.67	23.67**	593.98**	3.24	24.99	2.30**
LB	5.00	3.75**	6.14	18.34	414.82	7.12**	37.28	1.89
LBP	4.67	0.08	12.80	372.48	12.11	1.44	22.22	2.22
LL	5.33	0.93	5.89	539.40	11.28	5.00	44.12	0.83
RB	5.00	1.04	6.50	374.06	7.82	0.82	38.66	1.05
Mean (n=9)	5.04	1.96	7.98	271.53	119.33	2.50	44.25	1.30
%CV	10.11	8.43	0.88	0.40	0.71	10.37	5.97	5.27

**Note:** \* n=3 means triplicate analysis, \*\* showed significantly different from sampling points ( $p \leq 0.0$ )

The main results found soil condition was under extreme and moderate acidic condition (pH 4.67-5.67). The soil organic matter (SOM) was low level at 0.08 – 3.75%. Total forms of metal in the soil are presented in Table 2. The soil consisted of Fe 85.92-593.98 mg/kg, Na 5.97-89.56 mg/kg, Mg 6.22-23.67 mg/kg, Cu 3.25-17.04 mg/kg, Mn 0.62-7.12 mg/kg, and Zn 0.70-2.30 mg/kg. The LR and LB areas found maximum contamination of Cu 17.04 mg/kg and Mn 7.12 mg/kg. The HR and HPi area found contamination of Na 89.56 mg/kg, Zn 2.30 mg/kg, Fe 593.98 mg/kg, and Mg 23.67 mg/kg, respectively. The acidic condition of cultivated soil can extract Cu, Zn, Mg, and Fe from soils to leachate in form of bioavailability, especially Cu and Zn was

classified as high toxicity.<sup>12</sup> Although Na, Mg, and Fe were found at high amount, the toxicity was not in seriously danger.<sup>13</sup> However, slightly acidic soil (pH 5.0-6.5) was suitable for cultivation of rice, para rubber, pineapple, pepper, and banana because it can extract nutrient from soil under this condition. These crop types were the same as cultivated crops at highland area of the studied areas at Namyen village and Nava village.

For the results of heavy metal Cu and Zn, which are included in substance priority list of ASTDR that could be harmful for human health when they consume agricultural products. The results of this study were compared with other studies in Fig.1.



**Figure 1** Mean value of Cu and Zn in soil

**Note:**

Thailand 1 means results of this study

<sup>20</sup>Thailand 2 means Mangrove sediment Tha Chin Estuary, Thailand

<sup>21</sup>America means Agricultural soil in America

China means mean value of agricultural soil in <sup>22</sup>Beijing, <sup>23</sup>Guangzhou, <sup>24</sup>Yangzhou, <sup>25</sup>Wuxi, <sup>26</sup>Chengdu, <sup>27</sup>Xuzhou, and <sup>28</sup>Kunshan.

<sup>29, 30, 31</sup>India means mean value of agricultural soil in India

<sup>32</sup>Iran means mean value of agricultural soil in Iran

<sup>33</sup>Korea means mean value of agricultural soil in Korea

<sup>34</sup>Slovakia means mean value of agricultural soil in Slovakia

<sup>35</sup>World1 means mean value of agricultural soil in the world

<sup>36</sup>World2 means mean value of agricultural soil in the world

From Fig. 1, the contamination of Cu and Zn at agricultural area at Amphoe Dan Sai, Loei province is low when compare with the sediment in the river and agricultural soil from research results of other studies, and lower than the soil quality standard<sup>37</sup> (Cu 37 mg/kg and Zn 400 mg/kg). The reason of results in this study were lower than other sites because duration of sampling period was dry season so leaching of heavy metals was originated from spraying water, not from

run off. At highland, there was no cover crop to protect top soil from leaching so it could be moved to low land, surface water, or ground water. The amount of heavy metal could be detected, but it was lower than low land and sediment that accumulated heavy metal from leachate or run off from highland or upstream to downstream. The low amount of heavy metal at highland could be exposure through crop production consumption and accumulated in human body, which could

have chronic adverse health effect in long term. This data has been evaluated to identify toxic metals of significance with respect to potential effects on the soil ecosystem and uptake via the food chain. The outcome of this study will enable the development of more focused monitoring for soil toxicity at highland agricultural areas to evaluate the health risk. These are ranked by using the hazard index.

## CONCLUSION

Agricultural soil of highland area at 15 cm depth found contamination of heavy metal: Fe, Mg, Na, Cu, Mn, and Zn, respectively. The pineapple field on high slope highland, Nava village (HPi) was found the highest contamination of Mg, Fe, and Zn statistically with significantly difference ( $P < 0.01$ ). At moderate acidic soil (pH 5.67) and high fertility (SOM 3.36%). At rice field on low slope highland, Namyen village (LR) found maximum contamination of Cu. At high acidic soil (pH 4.67) and moderate fertility (SOM 1.60%), the acidic soil leached useful nutrient for plant but it caused high contamination of heavy metal. The acidity changed form of heavy metal to be toxic on living organisms. The contaminated agricultural products will not be safe for the consumers. Therefore, information of heavy metal contamination at highland made local people realized on effect of chemical utilization for cultivation of farmers.

The results of this study are basic information for farmers at Nava village and Namyen village, including other highland area in tropical region that use chemical substances for cultivation. Further study should be done such as health risk analysis and environmental impact assessment of chemical substances release to environmental ecology of agricultural area. Moreover,

environmental quality monitoring and health checking of farmers should be done to find prevention, measure, and mitigation to reduce environmental impact and improve health quality of people at highland area.

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