

REVIEW OF HEAVY METAL CONTROL STANDARDS BASED ON THE UNCERTAINTY ANALYSIS OF HUMAN HEALTH AND SOIL SUSTAINABILITY

Dai-Ming Li¹, Pao-Hsuan Huang², Sheng-Wei Wang², Ming-Der Hong³, Sheng-Hsin Hsieh³ and Chihhao Fan¹

ABSTRACT

According to the past economic development policies in Taiwan, as years went by the lack of control, contributed to the situation of factories by the side of the farmlands. In order to establish a complete environmental safety regulation for agricultural production, the government authorities take scientific and technological study plan to integrate the principles and processes of the relevant regulatory standards of heavy metals in the agricultural safety management system. To view the standards in an appropriate light, we start from the perspective on human health risks and soil sustainability. While discussing human health, we also take environmental sustainability and the limits of establishing regulations into consideration. As this could be a scientific basis for evaluating the regulatory standards in the future, to reconfirm the accuracy of current heavy metal control standards. In this study, while estimating the allowable concentration of heavy metals in rice, the non-carcinogenic risk values are calculated with the local data and the regulations of the Ministry of Health and Welfare, and other parameters like the transportation rate of soil to crop, the heavy metal concentration of soil, soil weight, amount of irrigation water, and estimate of over-allowance time, are also calculated conservatively. The allowable concentrations of heavy metals in irrigation water would be appropriate to ensure the safety of human health and the sustainability of soil.

However, the heavy metals control standards of irrigation water, soil, and agricultural products were established by the governing authorities, respectively. Due to the variety of the entering pathways of heavy metal from environmental media to food webs will eventually cause a health risk to human beings. It is necessary to consider the transport mechanisms of heavy metals in their pathways. In this study, to consider the variability of the influence of media in the agricultural production environment, the uncertain analysis is taken as a great part to meet multiple situations of the actual background value. The results from the regulations and reference show that the allowable concentrations of heavy metals in irrigation water are a lot lower than the current irrigation water quality control standards, and also lower than the actual concentration of heavy metals in the study area. The main reason is that parameter evaluation such as food intake, metabolism, and heavy metal degradation in the environment are not involved during the calculation. Therefore, it is appropriate to include complete health risk data to estimate the concentrations of heavy metals in irrigation water. On the other hand, the results calculated with the local data show that the allowable concentration of heavy metals, including copper, lead, nickel, zinc, and chromium, in irrigation water in the study area was estimated to be higher than the current irrigation water quality control standards, which means that there is no need to modify the standards, but the result of cadmium cannot be calculated due to its value of soil in the study area is too high. Nevertheless, the regulatory standards of Taiwan are strict enough as compared to other countries in the world.

¹ Department of Bioenvironmental Systems Engineering, National Taiwan University, Taiwan(R.O.C)

² Sinotech Environmental Technology, Ltd., Taiwan(R.O.C)

³ Council of Agriculture, Taiwan(R.O.C)

Keywords: Heavy metals, Environmental safety regulation for agricultural production, Human health risks, Soil sustainability.

1. INTRODUCTION

To comprehend the established process of control standards in the agricultural production environment, the approach of this study includes (1) review of prevailing regulations and information collection, (2) a pot experiment to explore the heavy metal bioavailability in soil to crops, and (3) human health risk assessment. The areas of Taoyuan, Taichung, and Changhua were chosen as the study area, where the health risk assessment of heavy metal intakes was estimated by daily intakes and average concentrations of rice samples (Figure 1).

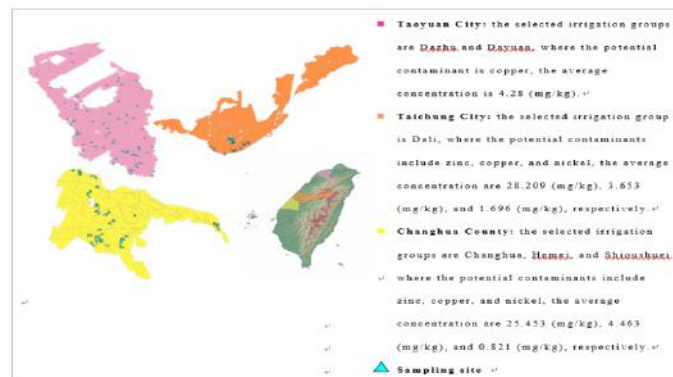


Figure 1. Study Areas and respective potential contaminations

1.1 Development of Irrigation Water Control Standards in Taiwan

In 1978, the project 'Conduct Irrigation Water Quality Monitoring Implement' in Council of Agriculture (COA) established the first criteria of irrigation water quality in Taiwan which referenced the irrigation water quality standards of United States Department of Agriculture (USDA) and the Content standards of heavy metals of United States Environmental Protection Agency (USEPA). The objective of the irrigation water quality standards aims to protect crop production and sustainable use of soil which considered heavy metal accumulations in crop and the health risk of human intakes. However, the regulation does not aim to prohibit the emission of contaminated water; it is only a criterion to distinguish the applicability of irrigation water. Based on the scientific experiment and references, the criteria was established and considered on both physical and chemical characteristics of each heavy metal, thus to prevent ionic compounds to accumulate in soil or crop root. In 2003, after the first announcement of irrigation water quality criteria, COA referred to the result of 4-year pot experiment of rice, added on two items, dissolved oxygen and iron (Fe), and relaxing the criteria of total nitrogen content in irrigation water. In addition, were tightened for mercury (Hg), nickel (Ni), vanadium (V), arsenic (As), beryllium (Be) and manganese (Mn) in reference to the criteria in Europe, United State and Japan. In Total, we revised 30 items of control standards of irrigation water.

After the revision of 2003, the control standards of irrigation water have not been revised for more than a decade. Until 2011, COA started to consult with Agriculture Technology Research Institute for further update and revise. Thus, two draft revision of irrigation water control standards were updated in 2013, and additional revision and description were suggested in 2014. However, two draft revisions are still pending (Table 1).

Table 1. Comparison of previous and current irrigation water control standards

| Items | The first irrigation water regulation, announced th on 5 July, 1978 | The present regulation, announced on 7 th December, 2003 | Differences |
|-------|--|---|-------------|
| Al | 5.0 | 5.0 | - |
| As | 1.0 | 0.05 | Tightened |
| Be | 0.50 | 0.1 | Tightened |
| B | 0.75 | 0.75 | - |
| Cd | 0.01 | 0.01 | - |
| Cr | 0.1 | 0.1 | - |
| Fe | - | 5.0 | Added |
| Co | 0.05 | 0.05 | - |
| Cu | 0.2 | 0.2 | - |
| Pb | 0.1 | 0.1 | - |
| Li | 2.5 | 2.5 | - |
| Mn | 2.0 | 0.2 | Tightened |
| Hg | 0.005 | 0.002 | Tightened |
| Mo | 0.01 | 0.01 | - |
| Ni | 0.5 | 0.2 | Tightened |
| Se | 0.02 | 0.02 | - |
| V | 10.0 | 0.1 | Tightened |
| Zn | 2.0 | 2.0 | - |

2. Revision of Irrigation Water Control Standards in 2014

From 2012, the revision of irrigation water took electrical conductivity (EC), Sodium Adsorption Ratio (SAR) and Residual Sodium Carbonate (RSC) into consideration. EC and RSC were mainly referenced from Texas State's standard and SAR was referenced from German. The comparison between EC and SAR were also consensus with the irrigation water quality classification chart from the United States Salt Institute. All items are identically in consensus with the 'Category D water bodies may be used for irrigation water, Class 2 industrial water, and environmental conservation' of the 'Surface Water Classification and Water Quality Standards', which took health risks of human intakes into considerations.

All the units were adjusted to System International d'Unités (SI), such as EC (dS/m at 25 °C), SAR (mmol(+)/L0.5) and RSC (mmol(±)/L). In 2013, there was a contamination case caused by an illegal emission from an LCD monitor industry which contained precious metals, Indium (In) and gallium (Ga). Furthermore, according to present researches about In and Ga in plant physiology revealed that could impede plant growth. Thus, to be more cautious about potential contaminants, the latest revision added In and Ga as new items. However, the draft revision is still impending for more discussion and research on both academic and practical coordination (Table 2).

3. MATERIALS AND METHODS

3.1 Human Health Risk of Heavy Metal Intake

Human health risk of heavy metal intake measured by cancer risk and non-cancer risk indices, which are taken as the first step of competing for control standards. The cancer risk and non-cancer risk indices are predicted by daily intakes and average contents of sampling rice in three areas relatively high heavy metal contamination potential, including Taoyuan, Taichung, and Changhua.

Table 2. Revision of irrigation water control standards in 2014

| Items | Units | Standard Values |
|-------------------------------|-----------------------------------|-----------------|
| Temperature | ^o c | ≤ 35 |
| Ph | - | 6.0—9.0 |
| EC | ^o Ds/M At 25 c | ≤ 0.75 |
| SS | Mg/L | ≤ 100 |
| SAR | ^{0.5 0.5} Mmol (+) /L | ≤ 0.75 |
| RSC | Mmol (±) /L | ≤ 100 |
| DO | Mg/L | ≤ 6.0 |
| Cl | Mg/L | ≤ 2.5 |
| SO ₄ ⁻² | Mg/L | ≤ 3 |
| T-N | Mg/L | ≤ 175 |
| Anionic Surfactant | Mg/L | ≤ 200 |
| Grease | Mg/L | ≤ 3.0 |
| Co | Mg/L | ≤ 5.0 |
| Cu | Mg/L | ≤ 5.0 |
| Pb | Mg/L | ≤ 0.05 |
| Li | Mg/L | ≤ 0.2 |
| Mn | Mg/L | ≤ 0.1 |
| Hg | Mg/L | ≤ 2.5 |
| Mo | Mg/L | ≤ 0.2 |
| Ni | Mg/L | ≤ 0.002 |
| Se | Mg/L | ≤ 0.01 |
| V | Mg/L | ≤ 0.2 |
| Zn | Mg/L | ≤ 0.02 |
| Al | Mg/L | ≤ 0.1 |
| As | Mg/L | ≤ 2.0 |
| Be | Mg/L | ≤ 5.0 |
| B | Mg/L | ≤ 0.05 |
| Cd | Mg/L | ≤ 0.1 |
| Cr | Mg/L | ≤ 0.75 |
| Fe | Mg/L | ≤ 0.01 |
| In | Mg/L | ≤ 0.1 |
| Ga | Mg/L | ≤ 5.0 |

3.2 Bioavailability of Heavy Metals in Crops

Bioavailability or transport mechanism of heavy metals in crops, especially in rice, is estimated by in situ sampling and pot experiment. The heavy metal contaminations of 50 paired samplings of rice and soil were measured to establish multiple regression models, as to predict the effective transport from soil to rice.

3.3 Accumulation of Heavy Metal in Soil

To better predict the accumulating volume of heavy metal in soil under current control standard of irrigation water, the local background level of heavy metals and average usage of irrigation water per year are taken into considerations.

3.4 In-situ samplings

In order to predict the human health under heavy metal intake, implementing in situ sampling fieldworks in three potentially contaminated areas. The scheme of in situ sampling sites is based on PN rank (Nemerow, Nelson L.) and major soil series. In

order to select the soil that can represent the current status and reduce the variation between different soil types in a pot experiment, taking the soil series which have the most cultivated farmland as the major soil series. The quantities of sampling sites are selected equally in 5 PN ranks of the major soil series.

The three years sampling scheme contains 303 selective sampling sites, including 100 paired samplings of rice and soil in Taoyuan, Taichung, and Changhua (Figure 2).

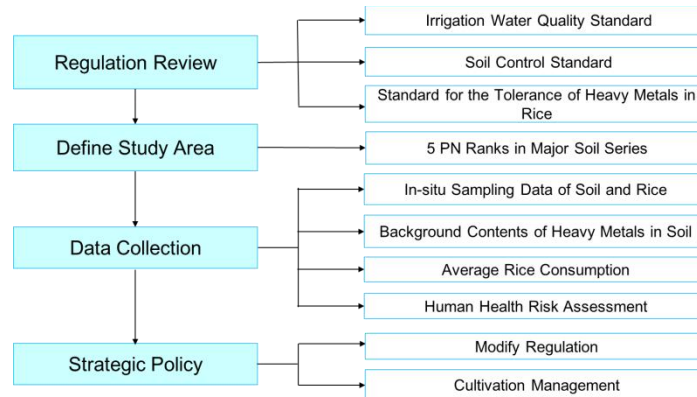


Figure 2. Study framework

4. UNCERTAINTY ANALYSIS AND SENSITIVITY ANALYSIS

Under the framework of competing for control standards in a coherent transport system, uncertainty analysis is applied to obtain distributions of control standards in certain parameters (figure 2). Within the heavy metal transport system of different environmental media, a set of chosen input parameters are passed through a model to obtain the distributions of heavy metal accumulation in soil, including background levels of heavy metal in soil and transport rate from soil to crops. The output distributions of heavy metal accumulation in the soil can describe the range of potential accumulation at some probability levels, and estimate the probability of the accumulation will be exceeded in a threshold of the target value, which can be referred to the control standards.

Sensitivity analysis aims to describe how much model output values are affected by changes in model input values. Thus, it can provide a general assessment of model precision when used to assess system performance for the alternative scenarios such as different background levels of heavy metal in soil or heavy metal content transport rate from soil to different crops (Figure 3).

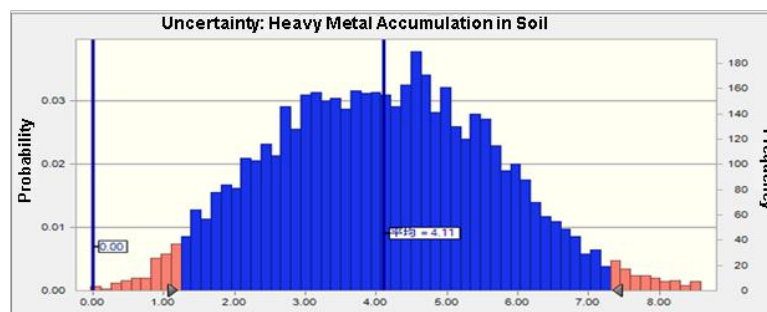


Figure 3. Uncertainty and Sensitivity Analyses for Heavy Metal Accumulation in soil

5. RESULTS AND DISCUSSION

According to the estimation of uncertainty analysis, the current irrigation water control standard is strict for both the aspects of human health risks and the present environmental background levels of irrigation water, furthermore, the current revision has already considered the newest items and regulation trends from all around the world.

Table 3. Estimation of irrigation water control standards in a competing aspect

| Estimator References | | Estimate by Current Regulations of Food and Crops | | | Estimate by Non-Cancer Risk Indices by Location | | Present Irrigation Water Control Standard |
|----------------------|-------|---|----------------------------|---|--|---|---|
| Data Resources | | Regulation and References | | | In-situ Sampling Data | | |
| Location | Items | Food Control Standards | Crops Protection Standards | Estimation A: Allowable concentration in irrigation water | Allowable concentration in rice by estimated from Human Non-Cancer Risks | Estimation B: Allowable concentration in irrigation water | |
| Units | | mg/kg | mg/kg | mg/L | mg/kg | mg/L | mg/L |
| Taoyuan City | Cu | - | 25 | 5.52E-07 | 80 | 3.621 | 0.2 |
| | Pb | 0.2 | - | 2.94E-08 | 36.3 | 26.972 | 0.1 |
| | Ni | - | 50 | 1.80E-06 | 210 | 14.908 | 0.2 |
| | Zn | - | 500 | 6.87E-06 | 940 | 24.99 | 2.0 |
| | As | 1 | - | 5.21E-08 | 0.225 | 0 | 0.05 |
| | Cd | 0.4 | - | 7.73E-10 | 0.38 | 0 | 0.01 |
| | Cr | - | 20 | 8.02E-06 | 90.4 | 71 | 0.1 |
| Taichung City | Cu | - | 25 | 5.02E-07 | 80 | 3.276 | 0.2 |
| | Pb | 0.2 | - | 3.08E-08 | 36.3 | 24.403 | 0.1 |
| | Ni | - | 50 | 1.63E-06 | 210 | 13.488 | 0.2 |
| | Zn | - | 500 | 6.20E-06 | 940 | 22.610 | 2.0 |
| | As | 1 | - | 5.06E-08 | 0.225 | 0 | 0.05 |
| | Cd | 0.4 | - | 7.33E-10 | 0.38 | 0 | 0.01 |
| | Cr | - | 20 | 7.28E-06 | 90.4 | 64.423 | 0.1 |
| Changhua County | Cu | - | 25 | 6.34E-07 | 80 | 4.299 | 0.2 |
| | Pb | 0.2 | - | 2.21E-08 | 36.3 | 32.029 | 0.1 |
| | Ni | - | 50 | 2.10E-06 | 210 | 17.703 | 0.2 |
| | Zn | - | 500 | 8.04E-06 | 940 | 29.676 | 2.0 |
| | As | 1 | - | 5.59E-08 | 0.225 | 0 | 0.05 |
| | Cd | 0.4 | - | 8.17E-10 | 0.38 | 0 | 0.01 |
| | Cr | - | 20 | 9.54E-06 | 90.4 | 84.555 | 0.1 |

The present study reviewed all the amendments of related heavy metal control standards chronologically, which were established considering different purposes by various authorities in Taiwan. In order to assess the co-competition between irrigation water, soil, sediment, and crops, the study conducted an experimental analysis regarding the distribution the heavy metal within the respective media and calculated human health risk index as a criterion to re-evaluate the rationality of all the regulated standards.

The locations of in-situ sampling were determined based on PN ranking (Nemerow, Nelson L.) of the previously-investigated area which is highly cultivated farmlands in the study areas. Such sampling framework and results may reflect the current soil background contents and reduce the variation between different soil types in a pot experiment as well. This study provides a strategic plan for modifying current control standards. The co-competition of heavy metal control standards in different environmental media, including irrigation water, soil, and crops was compared to both human health risk index and in-situ sampling data.

In order to determine the bioavailability concentration of heavy metal in the crops, especially in rice, the sampling data was utilized to calibrate a regression model. The study measured the heavy metal contaminant in 100 paired samplings of rice and soil

to verify the multiple regression model to assess the effective transfer of heavy metals from soil to rice. This study predicts the accumulation limits of heavy metals in soil under current control standard of irrigation water by collecting the soil background concentrations of heavy metals and the heavy metal discharge by industrial effluents. In order to predict the human health risk of heavy metals, this study followed a sampling scheme to investigate heavy metal concentrations via soil to food products in three potentially contaminated areas, where the contamination were leveled by 5 PN ranks. In order to predict the human health risk of heavy metal. The two-year sampling scheme contains 403 selective sampling sites, including 100 paired samplings of rice and soil in Taoyuan and Changhua Counties. A suggested plan for modifying current control standards is proposed based on human health risk assessment (Table 4).

Table 4. Suggesting modification for current control standards

| Items | Irrigation Water Quality Standard (mg/L) | Suggesting plan | Soil control standard (mg/kg) | Suggesting plan | Standard for the tolerance of heavy metal in rice (mg/kg) | Suggesting plan |
|-------|--|-----------------|-------------------------------|-----------------|---|-----------------|
| Cu | 0.2 | maintained | 200 | maintained | - | maintained |
| Pb | 0.1 | maintained | 500 | tightened | 0.2 | maintained |
| Ni | 0.2 | maintained | 200 | maintained | - | maintained |
| Zn | 2.0 | maintained | 600 | maintained | - | maintained |
| As | 0.2 | maintained | 60 | tightened | - | maintained |
| Cd | 0.01 | maintained | 5 | tightened | 0.4 | maintained |
| Cr | 0.1 | maintained | 250 | tightened | - | maintained |

6. CONCLUSION

Based on the health risk assessment in this research, arsenic in rice is higher than other heavy metals in the study areas. By calculating the allowed concentrations of irrigation water and soil from health risk indices, the current control standards of both media seem adequate to assure our environment and food safety. In a certain case, the background concentrations of arsenic in the soil are comparatively higher than other locations. Other than arsenic, other heavy metal control standards are apparently stringent enough to ensure the food security. As a result, the current strategic policy focuses on adjusting cultivation management, such as growing crops whose growth should not be affected regardless the level of heavy metal concentrations. Another possible strategy is to use water resources that meet the current standards as the alternative supplementary irrigation resources, rather than revise the current control standards.

7. REFERENCES

- Cui, H., Fan, Y., Yang, J., Xu, L., Zhou, J., and Zhu, Z. (2016). In situ phytoextraction of copper and cadmium and its biological impacts in acidic soil. *Chemosphere* 161, 233-241.
- Feng, M.-H., Shan, X.-Q., Zhang, S., and Wen, B. (2005). A comparison of the rhizosphere-based method with DTPA, EDTA, CaCl₂, and NaNO₃ extraction methods for prediction of bioavailability of metals in soil to barley. *Environmental Pollution* 137, 231-240.
- Kim, R.-Y., Yoon, J.-K., Kim, T.-S., Yang, J.E., Owens, G., and Kim, K.-R. 2015. Bioavailability of heavy metals in soils: definitions and practical implementation—a critical review. *Environmental Geochemistry and Health* 37:1041-1061.
- Lin, Y.-W., Liu, T.-S., Guo, H.-Y., Chiang, C.-M., Tang, H.-J., Chen, H.-T., and Chen, J.-H. (2015). Relationships between Cd concentrations in different vegetables and those in arable soils, and food safety evaluation of vegetables in Taiwan. *Soil Science and Plant Nutrition* 61, 983-998.