Modelling Transmission of Hexavalent Chromium Concentration and Its Health Cost with a Water Quality Analysis Simulation Program

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1 **Modelling Transmission of Hexavalent Chromium Concentration and Its Health**
2 **Cost with a Water Quality Analysis Simulation Program**

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18

19 **Abstract**

20 In this study the Water Quality Analysis Simulation Program (WASP7) was used
21 to evaluate the transmission of hexavalent chromium (Cr(VI)) contamination in a
22 water–sediment system and its flux into cultivated soils. The agricultural areas adjacent
23 to the Wu River in Taiwan were taken as the study area, as these soils were found to be
24 heavily polluted with Cr(VI) concentrations of 3,271–16,799 µg/kg. It was found that
25 the rates of accumulation of Cr(VI) are affected by the distance from the source of

contamination and the size and type of cultivated areas. Specifically, maximum concentrations of Cr(VI) most rapidly accumulated in the smallest cultivated areas. The highest concentrations of Cr(VI) (4.27 mg/kg) were detected in soil from Changhua city, and correlated with the greater risk of gastric cancer in residents from this area. Specifically, the risk of gastric cancer due to Cr(VI) contamination of agricultural soil was 3×10^{-7} – 15.2×10^{-6} in Taichung city (upstream) and 1.3×10^{-6} – 76.3×10^{-6} in Changhua county (downstream). More worryingly, young children had a threefold greater risk of gastric cancer than adults. The values of statistical life-years (VSLYs) were US\$6.2–10 million for rice, US\$42–60 million for corn, and US\$360–580 million for other vegetables, respectively, each year. It is critical that techniques other than source reduction are used to reduce health costs associated with human exposure to Cr(VI), such as chemical oxidation or ion-exchange treatment to remove Cr(VI) from factory wastewaters, prior to their discharge into rivers.

Keywords: chromium; health cost; loss of life expectancy; WASP; water

51 **List of Abbreviations**

AR	Attributable risk
CSF	Cancer slope factor
EPA	Environmental Protection Agency
LADD	Lifetime average daily dose
LLE	Loss of life expectancy
VSLY	Value of statistical life-year

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1. Introduction

Heavy metal contamination of sediment and water is one of the greatest threats to the environment, especially the hydrological cycle (Liu et al. 2019). It was first reported in Japan in 1973 that heavy metal pollution in water and sediments of rivers could cause disease; in this case, it was mercury contamination that caused Minamata disease (Forstner and Muller 1973). Since then, there has been extensive research interest in heavy metal contamination of the environment. Such contamination of the environment depends on natural environmental and anthropogenic factors. Rivers accumulate heavy metals as they flow through areas where different anthropogenic activities are taking place, and sediments act as both carriers and sinks for contaminants (Sundaray et al. 2011). More than 97% of the mass of heavy metals transported to oceans is known to be derived from river sediments (Jain and Sharma 2001). Scientific interest in and sustainable management of agricultural areas and water have become fundamental to ensuring global food security, due to the increasing demand for agricultural and water resources (Kesstra et al. 2016; Key et al. 2016).

Intensive cultivation of land has resulted in critical problems for the agricultural economy and increasing healthcare costs (Raptis et al. 2018). The high density of electroplating factories had a potential link with high blood chromium concentrations in a sample of Taiwanese people (Chang et al. 2006a,b). A possible association between chromium and oral cancer was also identified in Taiwan (Yuan et al. 2011). Chromium exists in aqueous environments with its two stable oxidation states being Cr(III) and Cr(VI). They may be transformed by various physicochemical processes, such as precipitation/dissolution and adsorption and desorption reactions (Kotaś et al. 2000). In comparison, Cr(VI) is more harmful and can be generated by the oxidation of Cr(III) during chemical processes (Pradhan et al. 2017).

Our previous study showed that there was a link between Cr(VI) contamination in rivers and gastric cancer (Tseng et al. 2018 & 2019). High concentrations of Cr(VI) in plants have also been shown to induce changes in their physiology and morphology, and Cr(VI) has been identified as a potential carcinogen in human and animal studies (Haney et al. 2014). Taiwan established a maximum total chromium contaminant concentration of 50 µg/L; in contrast the U.S. EPA extends limits of 100 µg/L (U.S. EPA 2003). General public may ingest water and/or food contaminated with Cr(VI). Exposure to chromium has been determined to have severe ill effects on health, including gastric and lung carcinoma (Zhang and Li 1997); liver, kidney, and urinary dysfunction (Linos et al. 2011); and eye and skin irritations (Costa 2003; Mohanty et al. 2005). It has been reported that reducing discharge of Cr(VI) into the environment may minimize risks to human health (Akinremi et al. 2000; Gustafsson et al. 2014; Keesstra et al. 2016).

The speciation of chromium in water and sediment and its subsequent passage into the environment has been investigated (Chow et al. 2018). The water–sediment system generates different mixing conditions, leading to an oxygenated medium that favors the formation of Cr(VI) (Kotaś and Stasicka 2000). Furthermore, sediments with negatively charged clay minerals predominate in water–sediment systems, and thus Cr(VI) cations are repelled by these soil particles, resulting in Cr(VI) being more mobile and consequently more bioavailable than Cr(III) in the water–sediment system (Unceta et al. 2010). The concentration of total chromium in rivers and lakes is usually 0.5 to 100 nM (Kotaś et al. 2000). Accumulation of heavy metals from the water may occur due to sediment settling (Ghrefat and Yusuf 2006).

In this study, the Water Quality Analysis Simulation Program (WASP7) was used to investigate Cr(VI) contamination in the Wu River watershed in Taiwan, extending

our previous research (Tseng et al. 2018 & 2019) to delineate the transmission of Cr(VI) contamination in a water–sediment system and its flux into cultivated soils. The WASP program has been applied in the development of watershed management strategies in many studies (Lin et al. 2011). For example, it was used to show that sediments in the Saltwater River watershed in southern Taiwan were polluted by chromium and other heavy metals (Liu et al. 2011). The WASP has also been widely used to simulate toxicant concentrations in surface waters and sediments over a range of complexities and temporal and spatial scales (Knights et al. 2019). The health risks associated with Cr(VI) contamination of cultivated soils have been monetized into loss of life expectancy (LLE). In turn, this allows evaluation of the capability of different types of wastewater treatment plants to remove Cr(VI) contamination.

2. Materials and Methods

2.1. System boundaries used in this study

As shown in Fig. 1, the Wu River is 119 km long and enters the area in Taichung city (upstream) and flows into irrigation channels in Changhua county (downstream). The watershed belongs to the fourth largest river in Taiwan in terms of drainage area. The agricultural areas in Changhua city (marked as “C”) and Homei district (“H”) in Changhua county are heavily polluted by heavy metals, including Cr(VI), and are irrigated mostly with water drawn from the Wu River watershed. The Dali district (marked as “D”) and Wufen district (marked as “W”) in Taichung city contain many factories, which are source of pollutants. Rice cultivation in Changhua county accounts for 18–20% of national rice production, and is therefore a key to food supply in Taiwan. The effects of Cr(VI) on rice, corn and vegetable cultivation are compared, as these may lead to different health risks.

Figure 1 here.

2.2. Description of the WASP7

The development of the WASP started in 1970s, supported by the U.S. EPA. The WASP7 is the latest version of the WASP, and links hydrodynamic and sediment transport models that be used to solve problems such as eutrophication or heavy metal pollution. To establish our model for analysis by the WASP7, the waterbody of the river was divided into 12 superficial segments of equal size in each city/district; this two-dimensional (2-D) horizontal distribution was established to take account of aquaculture (Fig. 1). Due to a scarcity of data on actual river depth, it was reasonably assumed that the Wu River is a shallow river, containing 0.1 m (sediment) and 0.047 m (soil cultivated for rice) in its vertical section. These measurements are consistent with those found in many studies of samples of surface sediments (<10 cm in depth) from contaminated sites (Fang et al. 2019). Table 1 presents the model's core elements; a step-size time variable of 1 year is used for each interaction.

As mentioned before, total chromium concentrations in water have been restricted by the Taiwanese EPA. Water quality of total chromium were acquired from the regular monitoring stations in the Wu River. The Taiwanese EPA regularly conducts manual water quality sampling every month to compare data from regular monitoring stations. The Cr(VI) concentration was assumed to account for 7.2% of the total chromium concentration in water based on the assumption that $\leq 7.2\%$ of the total chromium in water is in the hexavalent (Cr(VI)) form (Proctor et al. 2002). In addition, the Cr(VI) concentration was assumed to account for 1% of the total chromium concentration in soil (Hsu et al., 2012). The effects of chemical reactions with particulate materials in water were excluded from this study.

2.3. Verification of the WASP7

The mean absolute percentage error (MAPE) is a common measure for verification and calibration, and is also used in this study. It can be calculated as the average absolute percentage error for each time period minus actual values, divided by actual values as follows, where A_t is the actual value and F_t is the forecast value:

$$MAPE = \frac{1}{n} \sum_{t=1}^n \left| \frac{A_t - F_t}{A_t} \right| \quad (1)$$

2.4. Calculation of ingestion exposure dose

We used Eq. (2) to calculate the cancer risk from Cr(VI) contamination:

$$\text{Risk} = \frac{C \times IR \times ED}{BW} \times CSF \quad (2)$$

where the lifetime average daily dose (LADD) value (in mg/kg/day) was used in this study to calculate exposure from ingestion. C is the ingested Cr(VI) concentration (mg/kg), I is the intake volume (kg/day) of rice of the Taiwanese population, ED is the exposure duration with regard to the age range of population, and BW is the average body weight in Taiwan (kg). Table S1 tabulates the body weights of Taiwanese males and females, and their intake volume of different plants at various ages. CSF is the cancer slope factor (0.5/mg/kg/day) (U.S. EPA 2010).

Therefore, an estimation of the death toll from gastric cancer (G_{cancer} , per capita) for this study was formulated as follows (Eq. (3)):

$$G_{cancer} = \text{Risk} \times F \times P \quad (3)$$

Where P is total exposure population (capita) and F is the mortality rate of gastric cancer in Taichung city is 51.16%, and the mortality rate of gastric cancer in Changhua county is 48.31% (Tsuang 2016).

2.5. Calculation of value of loss of life

The value of statistical life year (VSLY) method was applied to recognize that any change in an individual's mortality risk. The VSLY method uses attributable risk (AR), which is the ratio of the estimated excess population who died of gastric cancer (per capita) as a result of Cr(VI) exposure and the actual population (P) who died of gastric cancer without Cr(VI) exposure (per capita). The AR value was then applied to an established life table to calculate the LLE in the study area. The estimated years of economic loss after estimation of the AR of Cr(VI) pollutants; it can be calculated as follows (Eq. (5)):

$$\text{Value of loss life} = \frac{LLE \times P \times VSLY}{Y} \quad (4)$$

where is Y is the life expectancy (y). Detailed calculations can be referred to our previous research (Tseng et al. 2018 & 2019).

2.6. Calculation of pollution prevention cost

The costs of wastewater treatment were considered to include construction and operation and maintenance (O&M) costs. These were calculated using Eq. (5):

$$C_{ww} = C_{construct} + C_{O\&M} \quad (5)$$

The construction costs for chemical oxidation treatment and ion-exchange treatment in Taiwan were roughly US\$350,000 and US\$83,000, and these treatments can last for 25 and 5 years, respectively (Industrial Development Bureau 2004). The O&M costs evaluated in this study comprised electricity costs, medical costs, general maintenance costs, and personnel expenses.

3. Results and discussion

3.1. Cr(VI) accumulation in soil

Figure 2 shows the Cr(VI) accumulation in cultivated soils in the four city/districts. The soil in Wufen district was heavily polluted, containing 16,799 $\mu\text{g/kg}$ Cr(VI), followed by Changhua city (4,273 $\mu\text{g/kg}$), Homei district (3,977 $\mu\text{g/kg}$), and Dali district (3,271 $\mu\text{g/kg}$). The Cr(VI) accumulation in soils was higher than original expects and results of other counties with similar conditions. In India, it was found that 79.5 $\mu\text{g/kg}$ and 45.8 $\mu\text{g/kg}$ Cr(VI) was accumulated in cultivated rice and corn (Mishra et al., 1995 & 1997), while oats and lettuce plants respectively accumulated 941.1 $\mu\text{g/kg}$ and 63.9 $\mu\text{g/kg}$ Cr(VI) (Asfaw et al 2017; Wyszowski and Radziemska 2013). A case study of Taiwan indicated that partition coefficients of heavy metal species and particulate settling rates play an important role in the distribution of dissolved heavy metals (Liu et al. 2012).

The accumulation rates were also different over the course of a year. The Cr(VI) contamination was highest on day 72 in Wufen district, on day 234 in Dali district, on day 310 in Homei district, and on day 325 in Changhua city. A previous study showed that the residence time of chromium in water ranges from 4.6 to 18 years (Schmidt and Andren 1984). Cr(VI) accumulation may be affected by the distribution distances, and the area and type of cultivation of the land under study. It was found in this study that the anthropogenic Cr(VI) was mainly discharged from factories located in Taichung city, and thus higher concentrations of Cr(VI) accumulated in the cultivated soils of Taichung city than in those of Changhua city. Impressively, smaller cultivated areas accumulated maximum concentrations of Cr(VI) in short time. Cr(VI) transmission throughout the environment involves multicomponent linked actions and critical processes specific to the sediment–water system.

Figure 2 here.

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245 **3.2. Verification of the WASP7 model**

246 The results of the verification of the WASP7 model are shown in Table 2. The
247 MAPE results were 28.5% and 28.8% in Taichung city and Changhua county,
248 respectively. According to the statistical data from Taiwan EPA (2019), the monitored
249 Cr(VI) concentrations in cultivated soil were 1.7–2.5 mg/kg in Taichung city and 2.9–
250 3.5 mg/kg in Changhua county. The simulated results of the WASP7 model were
251 consistent with these results. In particular, the high concentrations of Cr(VI)
252 contamination in the soil can be attributed to the high density of electroplating factories
253 in Wu river watershed (Chiang et al. 2010). When Cr(VI) accumulates in cultivated soil,
254 it is absorbed by food crops and thus ultimately ingested by humans via the food chain.
255 Cr(VI) accumulation in soil was the greatest in Changhua city (4.3 mg/kg), followed
256 by Homei district (4.0 mg/kg), Dali district (3.3 mg/kg), and Wufen district (2.2 mg/kg).
257 An acceptable soil chromium concentration of 64 mg/kg was established to protect
258 environmental and human health (CCME 2015). These findings suggest that rice plants
259 irrigated with downstream river water carry a greater carcinogenic risk.

260 Table 2 here.

261

262 **3.3. Health costs associated with Cr(VI) contamination**

263 The risk of gastric cancer caused by Cr(VI) contamination was an average of
264 3×10^{-7} – 15.2×10^{-6} in Taichung city and 1.3×10^{-6} – 76.3×10^{-6} in
265 Changhua county (Table 3). Specifically, eating vegetables was associated with the
266 greatest risks of causing gastric cancer, followed by eating corn and rice. More seriously,
267 young children had a threefold greater risk than adults. This is due to the relative intake
268 rate being higher for young children, as a result of their lower body weight. Therefore,

the LADD values were higher for young children. Female children and adults had higher cancer risks than their male counterparts, due to females' lower body weights. The AR values were 2.2×10^{-5} – 1.3×10^{-3} in Taichung city and 7.8×10^{-5} – 4.5×10^{-3} in Changhua county. The exposure population in Taichung city and Changhua county was 278 million and 128 million in 2017. Therefore, the estimated values for the population who died of gastric cancer are 1 for rice, 3–6 for corn, and 22–47 for vegetables each year.

The LLE due to Cr(VI) pollution was 0.002–0.103 years in Taichung city and 0.006–0.361 years in Changhua county. That is, the average life expectancy in Taichung city decreased from 80.3 to 80.2 years, whereas the average life expectancy in Changhua county decreased from 79.9 to 79.5 years. The VSLYs in Taichung city were US\$6.2 million for rice, US\$42 million for corn, and US\$360 million for vegetables each year, respectively (Fig. 3). In addition, the VSLYs in Changhua county were US\$10 million for rice, US\$68 million for corn, and US\$587 million for vegetables each year. The VSLY was an average of 1.5 times higher in Changhua county than in Taichung city. Although most Cr(VI) was discharged from Taichung city, the increased health risks and losses occurred downstream in Changhua county.

3.4. Cost of pollution prevention

In this study the costs of the two most common applied treatments to control Cr(VI) contamination were evaluated, namely chemical oxidation and ion-exchange treatments. The key advantages of ion exchange are that it enables recovery of valuable metals, its selectivity and that it produces low volumes of sludge (Jachuła and Hubicki 2013). On average, 706,210 m³ of sludge containing 0.1 mg/kg Cr(VI) contamination is discharged from factories into the Wu River each year (Taiwan EPA 2019) and, as

shown in Fig. 4, the chemical oxidation and ion-exchange treatments to remove Cr(VI) contamination from the river cost US\$71,800 and US\$65,367, annually. The medical cost for the ion-exchange treatment was 10 times greater than that for chemical oxidation treatment (US\$1,320/y vs. US\$112/y). However, the O&M cost of the chemical oxidation treatment was US\$10,500/y, greater than that for ion-exchange treatment (US\$2,500/y). When divided by their respective life expectancies, the construction costs of both treatments were similar (US\$14,000–16,667).

These results show that it is critical for the government to use either of these treatments to reduce Cr(VI) pollution in the Wu River and thus reduce health costs, given that the VSLY caused by the health risks of crops irrigated with Wu River water was US\$16.3 million/y for rice, US\$110.5 million/y for corn, and US\$947.3 million/y for other vegetables. It is recommended that the factories near the Taichung and Fuma channels reduce their discharge of Cr(VI), as this at-source action would be more effective than end-pipe treatment.

4. Conclusions

In this study a WASP7 model was used to explore the transmission of Cr(VI) contamination in a water–sediment system and its flux into cultivated soils in the Wu River watershed. The Wu River watershed enters the area in Taichung city (upstream) and flows into irrigation channels in Changhua county (downstream). The soil in Wufen district was found to be heavily polluted, containing 16,799 $\mu\text{g/kg}$ Cr(VI), followed by Changhua city (4,273 $\mu\text{g/kg}$), Homei district (3,977 $\mu\text{g/kg}$), and Dali district (3,271 $\mu\text{g/kg}$). Cr(VI) accumulation may be affected by distribution distances, cultivated areas, and types of cultivations. The MAPE results were 28.5% and 28.8% in Taichung city and Changhua county, consistent with the simulated results from the WASP7 model.

The risk of gastric cancer caused by Cr(VI) contamination was an average of 3×10^{-7} – 15.2×10^{-6} in Taichung city and 1.3×10^{-6} – 76.3×10^{-6} in Changhua county. The VSLYs were US\$6.2–10 million for rice, US\$42–60 million for corn, and US\$360–580 million for vegetables each year, respectively. The VSLY were on average 1.5 times higher in Changhua county than in Taichung city.

It was calculated that chemical oxidation and ion-exchange treatment to remove Cr(VI) contamination from the Wu river would annually cost an average of US\$71,800 and US\$65,367, respectively. It is therefore critical that the government uses these or other treatments to reduce Cr(VI) contamination in the Wu River, and thus reduce gastric cancer-associated health costs. Importantly, the use of these at-source reduction techniques will be more efficient than those that remove Cr(VI) once it has been released into the river.

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Declarations

- Conflicts of interest/Competing interests: The authors declare that they have no conflict of interest.
- Availability of data and material (data transparency): Data available in a public

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 345 (non-mandated deposition)
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 351 Ying-Chu Chen

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Fig. 1. Locations of Changhua county, Taichung city, and the Wu River watershed.

Fig. 2 Cr(VI) contamination in different cities/districts simulated by the WASP model

Fig. 3. The VSLY results from different cultivated crops in the Taichung city and Changhua county.

Fig. 4. The pollution-prevention costs for chemical oxidation and ion-exchange treatment in the Wu River.

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Table 1. Kinetic constants for the WASP7 model in this study.

Parameter	Unit	City/district				Source
		H	C	D	W	
Segment length	m	1,348	1,452	1,211	34.96	Taichung Irrigation Association, 2008
Segment width	m	4,045	4,357	3,635	104.88	
River depth	m	0.047				Council of Agriculture, 2015
Soil depth	m	0.1				Tseng et al., 2018
Soil infiltration coefficient	cm/s	2×10^{-5}				Department of Irrigation and Engineering, 2018
Water flow rate	m ³ /s	3.498	4.058	1.99	0.0016	Taichung Irrigation Association, 2008
Cr(VI) concentration in water	μg/L	48	56	18	14	Taiwan EPA, 2008
Settling rate	m/d	6.56				Water Resources Agency, 2011
Partition coefficients	log KD in L/kg	1.1				Allison and Allison, 2005
Suspended solids in water	mg/L	20	11.8	18	6.1	Taichung Irrigation Association, 2008

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Table 2 Results of MAPE for the WASP7 model in this study.

City/district	D	W	H	C
Total Cr concentration in water (mg/L) ^a	0.25	0.2	0.67	0.76
Cr(VI) concentration (mg/L) ^b	0.018	0.014	0.048	0.054
Average Cr(VI) concentration (mg/L)	0.016		0.051	
Monitored Cr(VI) concentration in soil (mg/kg)	2.5	1.71	2.93	3.50
Simulated Cr(VI) concentration (mg/kg)	3.27	2.18	3/97	4.27
Average simulated Cr(VI) concentration (mg/kg)	2.73		4.12	
MAPE (%)	28.5		28.8	

561 ^a The Cr(VI) concentration was assumed to account for 7.2% of the total chromium
562 concentration in water.

563 ^b the Cr(VI) concentration was assumed to account for 1% of the total chromium
564 concentration in soil.

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Table 3 Health risks from intake of different cultivated plants for different ages of
sexes in the Taichung city and Changhua county.

Cultivation Age (y) Sex		Rice		Corn		Vegetable	
		Taichung city	Changhua county	Taichung city	Changhua county	Taichung city	Changhua county
1-6	M	0.4	2.1	2.9	14.6	24.7	124.4
	F	0.4	2.2	3.0	14.9	25.3	127.5
7-12	M	0.3	1.5	2.0	10.3	17.5	88.0
	F	0.3	1.5	2.0	10.3	17.6	88.4
13-18	M	0.2	0.8	1.1	5.4	9.2	46.5
	F	0.2	0.9	1.2	6.2	10.6	53.2
>19	M	0.1	0.6	0.9	4.4	7.4	37.3
	F	0.2	0.8	1.1	5.3	9.0	45.1
Average		0.3	1.3	1.8	8.9	15.2	76.3

M = male, F = female

Figures

Figure 1

Locations of Changhua county, Taichung city, and the Wu River watershed. Note: The designations employed and the presentation of the material on this map do not imply the expression of any opinion whatsoever on the part of Research Square concerning the legal status of any country, territory, city or area or of its authorities, or concerning the delimitation of its frontiers or boundaries. This map has been provided by the authors.

Figure 2

Cr(VI) contamination in different cities/districts simulated by the WASP model

Figure 3

The VSLY results from different cultivated crops in the Taichung city and Changhua county.

Figure 4

The pollution-prevention costs for chemical oxidation and ion-exchange treatment in the Wu River.

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