

Assessed health risk and uncertainty of rural drinking water sources in typical mountainous and hilly area of Sichuan Basin

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ABSTRACT

Drinking water safety in Sichuan Basin is always the focus of people. Here, Mingshan District is selected which is a typically mountainous and hilly area in Sichuan Basin as the study area, and pollution sources of drinking water and waterborne diseases are investigated. Then, tested the water quality of rural drinking water sources, and in accordance with the drinking water sanitary standard and chemical toxicity classification though Integrated Risk Information System (IRIS) of US Environment Protection Agency (USEPA) to ensure the main contaminations of drinking water sources. Finally, health risks through exposure pathway of drinking water and skin contact were evaluated by adopting USEPA Health Risk Assessment Model, and the result showed that: (1) the carcinogenic risk of both Cr(VI) and As was over the acceptable level 10⁻⁶ and they should be considered as the major contaminants in the drinking water of this area; (2) the non-carcinogenic risk was basically within the acceptable level 1. Uncertainty is inescapability in the process of risk assessment, Monte Carlo (MC) method was adopted to quantify the uncertainties, and under the 95% confidence level, carcinogenic risk and non-carcinogenic risk were all within the acceptable level.

Keywords: Drinking water sources; Non-carcinogenic risk; Carcinogenic risk; Contamination; Monte Carlo

1. Introduction

With rapid developments of social economy, industry and agriculture in China, more and more chemical substances have been discharged into water environment, giving rise to increasingly serious water environmental pollutions. Many environmental problems also have been aroused, which not only cause great damages on water environment but also severely threaten human health and their daily lives [1,2]. As is generally known, the chemical substances occupy the first place in the factors affecting human health [3,4]. These hazardous substances in water directly cause environmental pollutions. More seriously, they will exist in water for a fairly long time, and eventually find their way into human bodies when people eat, drink or breathe, producing great harms on human health. Many chemical pollutants in water are carcinogenic, teratogenic and mutagenic, such as heavy metal pollutants [1–3], polycyclic aromatic hydrocarbons [4,5], drug residues [6] and organic pesticides [7–9]. Even at a very few doses, the longterm drinking of these hazardous substances can impose toxic effects on human health. In China, the death rate of residents induced by the suffering from cerebrovascular diseases, cancers and respiratory diseases is obviously on the rise. Moreover, there are approximately 3.5 million new cancer diagnoses and 2.5 million cancer deaths every year [10], in which 90% of cancers are caused by chemical carcinogens. According to the statistics by World Health Organization,

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the death toll induced by poor drinking water quality can reach up to 1.7 million every year all over the world, that is, 80% of diseases and deaths in whole world are associated with water [11].

Health risk assessment is a process that it makes qualitative and quantificational assessment of certain environmentally deleterious factors that can harm the health of people exposed by collection and application of toxicology, epidemiology and other associated resources, such as toxicology resources, mankind epidemiology and environment and exposure [12], in compliance with assessment principles and technical methods. The objective of health risk assessment is to assess the possibility and hazard degree of the chemicals in the specified amount and the physical factors which affect by the human, animals and plants or ecosystem. Recently, MC method as a new risk assessment, one of the most effective and practical approach to resolve randomness and uncertainty issues in risk assessment, has been progressively applied, which can generate the probability distribution results based on the repetitive calculation of variation and given parameter probability distribution value [13]. It can make the fussy calculation easier through structuring a proper model and applying analysis techniques of MC method. Under the circumstance of knowing each random variation statistic distribution, MC method is easy and proper to solve the randomness and uncertainty [14]. Nowadays, probabilistic human health risk assessment has been successfully applied to assess the potential adverse health effects of contaminants in the water [15,16].

The safety of drinking water especially in rural areas, now is a bottleneck problem that imposes restrictions on rural development in China, which is also a livelihood issue that the Chinese government shows great concerns. Minshan District, which locates on the southwestern margin of the Sichuan Basin, is a typical hilly region. In Minshan District, the problem of drinking water safety is particularly acute. The investigations reveal that [17], by 2010, 118,100 people in rural areas could not get access to clean drinking water, in which 69,200 people could not drink water safely on account of the unqualified water quality, accounting for 25% of the population in whole district. Several factors including the pollutions by chemical fertilizer and pesticides, the excrements from livestock and poultry farming, the variations in primitive environment and land utilization type take primary responsibilities for this situation. Currently, the local government has come to realize the adverse impacts induced by the problem of rural drinking water safety, and simultaneously, people have paid increasing attentions on serious health problems caused by environmental pollutions. The residents want to know why the environmental pollutions do harm to human health, how serious are these health risks. To correctly answer these questions, health risk assessment is an effective tool, which is also an extremely important research field in the studies regarding environment and health issues. Accordingly, with the adoption of Minshan District as the present study object, the risk assessments were performed on rural drinking water source by means of health risk assessment method which is recommended by the USEPA and the MC method, and the reliability of the assessment results were also verified by field investigations.

2. Material

2.1. Study area

Minshan District is located on the southwestern margin of the Sichuan Basin, with the longitude ranging from 103°02' to 10°23'E and the latitude ranging from 29°58' and 30°16'N. It covers an area of 614.27 km. According to Statistical Yearbook [18], the population in this region amounted to 27.89×10^4 in 2016, in which the agricultural population was up to 23.18×10^4 . Minshan District is a typical agricultural cultivation region in China. To achieve the increases in both production and income of crops, a large quantity of fertilizers and pesticides are applied every year. The fertilizers are primarily used in summer, during which the rains are often frequent. However, the utilization ratios of nitrogenous and phosphate fertilizers as well as pesticides are fairly low. Specifically, the utilization ratios of fertilizers are only 30%-40% while only 10%-20% of the pesticides are sprayed on the plants. Unfortunately, large proportions of fertilizers and pesticides were directly discharged into soils and water [19]. On the other side, as the large-scale livestock and poultry industry develops rapidly in this region, approximately 2.4727 million tons of excrements are generated every year [20], in which few are recycled while the overwhelming majority are directly discharged without any treatments. Besides, due to the effects of parent materials and types of soils, the background values of the primitive environment in the northeast of the study area are comparatively large, and the contents of iron (Fe) and manganese (Mn) are high in underground water. All of these factors lead to the current situation that the rural residents in Minshan District cannot access to clean drinking water.

2.2. Sampling

According to the landform, rivers characteristics and drinking water sources in the study area, the samples determination sites of drinking water are shown in Fig. 1. The sample collection proceeded in the March of 2010, 2011, and 2012, respectively. The water samples collected the people's drinking water in accordance with the Standard Examination Methods for Drinking Water (GB 5750-2006) [21]. The polyethylene plastic bottles (500 mL) and bungs were washed and scrubbed by the collected water for five times, then the bottles were sealed and the labels were affixed. When collecting the underground water samples, the water pun were opened for 5 min to leave all water in the pine out and then began to sample.

2.3. Test of water quality

The test index of samples determined in accordance with the Standard for Drinking Water Quality (GB 5749-2006) [22], and it includes: pH, sulfate, total hardness, Cr(VI), As, Cd, Mn, fluoride, nitrate, Cu, nitrite, and ammonia. Testing water quality in water samples was on the basis of the Standard Examination Methods for Drinking Water(GB 5750-2006) [21]. The concentrations of all heavy metals were determined by atomic absorption spectrophotometer (WYS2000, Anhui Wayeal Technology Co., Ltd., China) and the concentrations of sulfate, fluoride, nitrate, nitrite and ammonia were determined by UV spectrophotometer (UV-8000S, Shanghai Metash Instruments Co., Ltd., China).

3. Method

In the present work, the evaluation model in Handbook of Health Assessments on Superfund Sites as enacted by USEPA was selected, which is abbreviated as USEPA model and includes four steps, namely, data collection and analysis, toxicological evaluation, exposure assessment and risk characterization [23], In the first step – data collection and analysis – on the basis of a variety of information mainly including general information of study area, rural drinking water source, local epidemiology and the residents' health conditions, the direct or potential pollutants, sources and ways of pollutions are preliminarily identified. In the next step, the toxicological evaluations are conducted in accordance with the classification standard for the chemical pollutants in IRIS database [24] of USEPA, and subsequently, the causal relationship between exposure and the impacts of exposure on health as well as the



Fig. 1. Distribution of water sampling.

Table 1

Value of parameter

relationship between doses and the occurrence probability of adverse health effects are determined.

3.1. Exposure assessment

Exposure assessment aims at estimating the degrees or probable degrees that the people in whole country or in a certain region are exposed to certain chemical pollutants. Therefore, the identification of the characteristics of exposure population and the determination of the concentrations and distributions of the chemical pollutants in environmental media, which are correlated and inseparable with each other, are two components in exposure assessment [23,25]. In health risk assessment on drinking water quality, exposure assessment mainly includes the following steps, the measurement of the concentrations of hazardous substances in drinking water; the determination of number, gender, age and living habit of the drinking population; the estimation of several parameters of the drinking population such as the ratio of drinking, time of duration, exposure frequency of and body weight; and finally the calculation of the average daily exposure doses of drinking population.

Chemical pollutants that are in drinking water entering into the body have three ways, including: oral ingestion, dermal absorption and inhalation, which is harm for human health [26]. Oral ingestion and dermal absorption would be studied, and their exposure dose per day could be calculated as follows [23,27]:

Oral ingestion:

$$CDI = \frac{C \times IR \times ABS \times EF \times ED}{BW \times AT}$$
(1)

Dermal absorption:

$$CDI = \frac{C \times SA \times K_p \times EV \times ET \times EF \times ED \times CF}{BW \times AT}$$
(2)

where the values of the different parameters are summarized in Table 1.

| Parameter | Symbol | Unit | Value | Source |
|-------------------------------|--------|------------------|---------------------------|--------|
| Drinking water ingestion rate | IR | L/d | 1.88 | [25] |
| Body weight | BW | kg | 57.6 | [25] |
| Skin surface area | SA | m ² | 1.6 | [24] |
| Exposure frequency | EF | d/a | 350 | IRIS |
| Exposure duration | ED | a | 30 | IRIS |
| Averaging time | AT | d | Carcinogenic 365 × 72 | IRIS |
| | | | non-carcinogenic 365 × 30 | |
| Bathing frequency | EV | d/event | 1 | RAIS |
| Bathing time | ET | h/d | 0.167 | RAIS |
| Volume conversion factor | CF | L/m ³ | 1 | RAIS |

Note: RAIS [28] is the abbreviation of risk assessment information system, which was established by the Agency of Environmental Protection in Tennessee, USA.

3.2. Risk characterization

Based on the data obtained in the above three steps, risk characterization refers to the estimation of the possible health risk degrees or the occurrence probability of certain human effects of population under different exposure conditions [29]. It analyzes the uncertain factors in assessment and provides the health risk information when people are exposed to hazardous substances, which can offer scientific basis for environmental management and decision. The chemical substances exhibit different degrees of toxicity, imposing different damages on human health. Consequently, the specific formula varies in the process of risk characterization.

3.2.1. Non-carcinogenic risk

In general, the non-carcinogenic risk is hazard quotient (HQ), which is defined as the ratio of the exposure to the substance of concern to the corresponding reference dose (RfD) of that substance. The reference dose is an estimate (within an order of magnitude) of a daily dose of a substance, a lifetime exposure to which would not likely result in a harmful effect.

The formula is as follows [23]:

$$HQ_{ij} = CDI_{ij} / RfD_{ij}$$
(3)

$$HI = \sum_{j=1}^{k} \sum_{i=1}^{n} HQ_{ij}$$
 (4)

where HI is the total exposure hazard index; *i* is one of the pollutants; *j* is one of the exposure way.

3.2.2. Carcinogenic risk

Equation of carcinogenic risk is as follows [21]:

When
$$R_{ii} < 0.01$$
, $R_{ii} = SF_{ii} \times CDI_{ii}$ (5)

When $R_{ij} \ge 0.01$, $R_{ij} = 1 - \exp(-SF_{ij} \times CDI_{ij})$ (6)

$$R = \sum_{j=1}^{k} \sum_{i=1}^{n} R_{ij}$$
(7)

Table 2

Test result of water samples (unit: mg/L)

where R is the carcinogenic risk; SF is slope factor of the pollutants.

3.3. Uncertainty

Uncertainty is an important component in risk. In the whole process of health risk assessment, the uncertain factors exist in each step, and moreover, the factors themselves which are responsible for the uncertainty of assessment results are uncertain. As a most common method in quantitative analysis of uncertainty, MC method can describe the characteristics of the physical experimental processes and objects with reality, and reveal the rules that the system operates by sampling randomly and simulating the function and occurrence rules of the real system [30]. Crystal Ball is a software specialized for risk analysis and assessment, which was developed based on PC Windows platform.

4. Results and discussion

4.1. Water quality

Table 2 lists the statistical and analysis results of the detections of rural drinking water quality. It should be noted that, in 2012, the detection rate of As was 39.29%, while the contents ranged from 0 to 0.003 mg/L. With regard to Cd, it was only detected in the 6^{*} and 34^{*} water source in 2012, with the contents of 0.00304 and 0.00138 mg/L, respectively.

Additionally, as shown in Table 2, the contents of eight indices in rural drinking water source in the study area vary significantly. Some indices exceed the standards as described in the Standard for Drinking Water Quality (GB 5749-2006) [22], especially the contents of Fe and Mn which seriously exceeded the standards (according to the standard, the content of Fe should be below 0.3 mg/L while the content of Mn should be below 0.1 mg/L). The regions where the water quality exceeds the quality seriously are centralized in the water sources in the northeastern of the study area. This unqualified water quality is mainly induced by the regional soil parent materials. In the northeast of the study area, the soil parent materials consist of the Quaternary Pleistocene series of glacial till and glacial outwash. In the yellow soils which were formed by weathering, a large quantity of Fe and Mn ions are released and

| Index | 2010 | | 2011 | | 2012 | |
|----------|-----------------|--------------|-----------------|-------------|-----------------|--------------|
| | Mean | Range | Mean | Range | Mean | Range |
| Fe | 0.29 ± 0.64 | 0.1 - 3.1 | 0.25 ± 0.66 | 0.01 - 3.38 | 0.32 ± 0.70 | 0.01 - 3.26 |
| Mn | - | - | - | - | 0.08 ± 0.08 | 0.01 - 0.32 |
| Cu | 0.08 ± 0.14 | 0.00 - 0.86 | 0.07 ± 0.13 | 0.00 - 0.8 | 0.1 ± 0.17 | 0 - 0.86 |
| Cr(VI) | 0.14 ± 0.04 | 0.06 - 0.26 | 0.06 ± 0.03 | 0.02 - 0.15 | 0.08 ± 0.05 | 0 - 0.17 |
| Fluoride | 0.33 ± 0.42 | 0.05 – 1.36 | 0.24 ± 0.29 | 0.01 - 1.4 | 0.21 ± 0.21 | 0.01 - 0.98 |
| Nitrate | 6.30 ± 5.91 | 0.66 - 24.49 | 9.37 ± 7.18 | 0.32 - 30.7 | 3.86 ± 5.13 | 0.14 - 25.13 |
| Ammonia | 0.11 ± 0.18 | 0.1 – 0.92 | 0.42 ± 0.34 | 0.02 - 1.42 | 0.18 ± 0.16 | 0 - 0.7 |
| Nitrite | 0.04 ± 0.05 | 0.00 - 0.22 | 0.05 ± 0.06 | 0 - 0.14 | 0.06 ± 0.07 | 0 - 0.33 |

Note: "-" indicates that there is no data.

discharged into underground water due to the effects of eluviation, leading to the increasing contents of Fe and Mn in underground water [31]. In the 22[#] water source, the content of Cu was greater than 0.8 mg/L, exceeding the standard for drinking water in China. This is mainly induced by the wastewater and waste residues from livestock and poultry farms. As regulated in the standards for drinking water in China and European Union, the content of Cr(VI) should be below 0.05 mg/L. In 2010, the content of Cr(VI) in water sources all exceeded the standards. In 2011 and 2012, the content of Cr(VI) in water sources all exceeded the standards seriously. The previous studies indicate that most of Cr(VI) ions existed in soil solutions in a free state, and only 8.5%-36.2% of Cr(VI) are absorbed and fixed by soil colloids [32]. Furthermore, the absorption capacities for Cr(VI) vary significantly among different types of soils [2,33], in which the yellow soil is poor in the absorption of Cr(VI). On the other side, the yellow soil is widely distributed in the study area. In the 16[#] water source, the content of nitrate exceed the standard for domestic drinking water quality in China (the content of nitrate should be below 10 mg/L). The possible cause is that the cage cultures exist in reservoirs and thus high-protein fodders are often thrown into reservoirs in order to promote the high yield of aquaculture, that is, the pollutions on water are caused.

Table 3

SF of toxic substance (unit: mg/[kg·d])

| Pollutant | SF _{oral} | Source |
|-----------|--------------------|--------|
| Cr(VI) | 41 | IRIS |
| As | 15 | IRIS |

Table 4

RfD of non-toxic substance (unit: mg/[kg·d])

4.2. Toxicological evaluation

Based on the classification criteria for the toxicity of chemical pollutants in IRIS database of USEPA, both Cr(VI) and As are identified as the A-type carcinogens, and Cd is identified as the B1-type suspected carcinogen, with the carcinogenic way of breathing exposure and no carcinogenesis in diets. Pb is identified as the B2-type possible carcinogen, and Fe, Mn, fluoride, nitrate, Cu, nitrite, and ammonia nitrogen are all classified as non-carcinogens.

In the present work, the data regarding the relationship of the doses of carcinogens and non-carcinogens with reaction were selected from the IRIS and Provisional Peer-Reviewed Toxicity Values (PPRTV) databases. The data of the relationship between the doses of nine chemical substances and reaction were obtained by extrapolating animal experimental data. Table 3 lists the relationship between carcinogenic doses and reaction (also known as the carcinogenic slope factor, SF) while Table 4 lists the relationship between non-carcinogenic doses and reaction (also known as reference dose, RfD).

4.3. Exposure parameters

The gastrointestinal absorption coefficient of pollutants (ABS) and the permeability coefficient of skin are listed in Table 5.

4.4. Health risk

According to Eqs. (3)–(7) for the calculations of risk characterization, the health risk values of carcinogenic factors and non-carcinogenic factors in rural drinking water source by ways of oral intake and skin exposure were calculated, respectively.

| Pollutant | RfD _{oral} | RfD _{dermal} | Source | Pollutant | RfD _{oral} | RfD _{dermal} | Source |
|-----------|---------------------|-----------------------|--------|-----------|---------------------|-----------------------|--------|
| Cr(VI) | 0.003 | 0.00006 | IRIS | Cu | 0.04 | 0.012 | IRIS |
| As | 0.0003 | 0.000123 | IRIS | Hg | 0.0003 | 0.0003ª | IRIS |
| Pb | 0.0014 | 0.000525 ^b | IRIS | Cd | 0.0005 | 0.00001 ^b | IRIS |
| Fe | 0.3 | 0.3ª | IRIS | Nitrate | 1.6 | 1.6 ^a | IRIS |
| Mn | 0.046 | 0.00184 | IRIS | Ammonia | 0.97 | 0.97ª | PPRTV |
| Fluoride | 0.06 | 0.06ª | IRIS | Nitrite | 0.1 | 0.1ª | IRIS |

^aData induced by mouth exposure rather than skin exposure.

^bData were mainly selected from the study by Li et al. [34].

Table 5

Gastrointestinal absorption factor ABS and skin permeability constant K_n of pollutants

| Pollutant | ABS | K _p | Source | Pollutant | ABS | K _p | Source |
|-----------|------|---------------------|--------|-----------|------|----------------|--------|
| Cr(VI) | 0.02 | 0.002 | IRIS | Cu | 0.3 | 0.0006 | IRIS |
| As | 0.41 | 0.0018 ^a | IRIS | Hg | 0.07 | 0.001 | IRIS |
| Pb | - | 0.000004^{a} | IRIS | Cd | 0.05 | 0.001 | IRIS |
| Fe | 0.15 | 0.001 | IRIS | Nitrate | 0.5 | 0.001 | IRIS |
| Mn | 0.04 | 0.0001ª | IRIS | Ammonia | 0.2 | 0.001 | PPRTV |
| Fluoride | - | 0.001 | IRIS | Nitrite | 0.5 | 0.001 | IRIS |

Note: "-" denotes the condition that this parameter cannot be inquired and set as '1' in calculations.

^aData are sourced from the study carried out by the USEPA [35].

4.4.1. Carcinogenic risk

Based on the database information of USEPA IRIS [36], the Cr(VI) and As are carcinogenic in case of oral intake. However, the evidence of their carcinogenicity in case of skin exposure is insufficient. As a consequence, only the carcinogenic risks of Cr(VI) and As by oral intake were calculated in the present work.

As listed in Table 6, the carcinogenic risks of the carcinogen Cr(VI) in 2010, 2011, and 2012 are $6.69 \times 10^{-5} a^{-1} - 2.9 \times 10^{-4} a^{-1}$, $2.23 \times 10^{-5} a^{-1} - 1.67 \times 10^{-4} a^{-1}$, and $4.28 \times 10^{-6} a^{-1} - 2.03 \times 10^{-4} a^{-1}$, respectively; while the carcinogenic risks of the carcinogen As in 2005 and 2012 are $8.05 \times 10^{-5} a^{-1}$ and $0-2.41 \times 10^{-5} a^{-1}$, respectively. Obviously, by way of drinking water, Cr(VI) imposes greater risks on health than As per capita every year. Liu [37] has studied the annual carcinogenic risks of Cr(VI) induced by drinking water in a certain drinking water source along the reach of a river in Guangdong. The results indicate that the risks during the period from 2005 to 2010 are $1.43 \times 10^{-4} a^{-1}$ - $3.57 \times 10^{-4} a^{-1}$, which exceed the results in the present work. Hou [38] has investigated the carcinogenic risks of As induced by drinking water in some drinking water sources in Kaifeng. In their studies, the risks are $1.73 \times 10^{-4} a^{-1}$ -2.47 × 10⁻⁴ a⁻¹, which are significantly higher than the present results.

In line with the risk management practices for many years in foreign countries, the carcinogenic risks of chemical pollutants ranging from 10^{-6} to 10^{-4} are acceptable [39]. If following the most strict risk management criteria, that is, the carcinogenic risk of 10^{-6} is set as the acceptable level, the carcinogenic risks of Cr(VI) in rural drinking water sources of the study area in 2010, 2011, and 2012 all exceed the standard, which are approximately 66.9–273 times, 22.3–167 times and 4.28–203 times greater than the standard; while the carcinogenic risks of As in 2005 and 2012 also exceed the standard, which are approximately 80.5 times and 0–24.1 times as

greater as the standard. If the carcinogenic risk of 10^{-4} is set as the acceptable level for risk management, the carcinogenic risks of Cr(VI) in some water sources in 2010, 2011 and 2012 still exceed the standard. It is well-known that Cr(VI) and As, as two kinds of pollutants, exhibit strong carcinogenic toxicity on humans. Cr(VI) and As can find their way into human bodies by ingestion, breathing and skin exposure, and accumulate in human bodies, leading to chronic poisoning. The carcinogenic risks of Cr(VI) and As in some water sources exceed the acceptable levels, and thus should be regarded as the primary pollutants in drinking water in this region. That is to say, as suggested by our studies, the residents should find new water sources and the relevant authorities should pay high attentions on this issue.

4.4.2. Non-carcinogenic risk

When calculating the total damages and non-carcinogenic risks, as listed in Tables 7–9, Pb should be included. For the 34^{\ddagger} water source in 2012, the risks of Cd for drinking water and skin exposure as well as the total damages are 9.51×10^{-3} , 1.35×10^{-2} and 2.3×10^{-2} , respectively. For the 6^{\ddagger} water source in 2012, the risks of Cd for drinking water and skin exposure as well as the total damages are 4.32×10^{-3} , 6.14×10^{-3} and 1.05×10^{-2} , respectively.

4.4.2.1. Way of drinking water On the basis of the definition of risk index, the acceptable level of the risks induced by non-carcinogenic chronic toxicity should be equal to 1.

Table 7 lists the results of health risk assessment for various ways of drinking water, from which we can observe that, in descending order of the risks, the pollutants are fluoride, nitrate, Cu, Cr(VI), As, Fe, ammonia nitrogen, nitrite, and Mn. Wu and Sun [40] indicted that the risk through drinking pathway (HQ_{oral}) is nitrate > fluoride > nitrite, and similar

Table 6

| \sim · · | • 1 | | | 1 * 1 * | | / •/ 1\ | |
|--------------|-------------|------------|--------|----------|---------|------------------|--|
| arcinogonic | rick | accorated | TA71th | drinking | TATATOR | (11111111 - 2-1) | |
| Carcinogenic | JIDN | associated | VVILLI | UTHININE | water | unut. a T | |
| | | | | 0 | | () | |

| Index | 2010 | 2011 | 2012 |
|----------|--|---|---|
| Cr(VI) | $6.69 \times 10^{-5} - 2.9 \times 10^{-4}$ | $2.23 \times 10^{-5} - 1.67 \times 10^{-4}$ | $4.28 \times 10^{-6} - 2.03 \times 10^{-4}$ |
| As | _ | _ | $0-2.41 \times 10^{-5}$ |
| Total CR | $6.69 \times 10^{-5} - 2.9 \times 10^{-4}$ | $2.23 \times 10^{-5} - 1.67 \times 10^{-4}$ | $8.29 \times 10^{-6} - 2.03 \times 10^{-4}$ |

Table 7

Hazard index through drinking water exposure pathway

| 010 | 2011 | 2012 |
|---|--|--|
| $31 \times 10^{-2} - 5.66 \times 10^{-2}$ | $4.35 \times 10^{-3} - 3.26 \times 10^{-2}$ | 8.35×10^{-4} -3.96 × 10 ⁻² |
| | _ | $0-1.28 \times 10^{-1}$ |
| $63 \times 10^{-4} - 5.06 \times 10^{-2}$ | $1.63 \times 10^{-4} - 5.29 \times 10^{-2}$ | $1.63 \times 10^{-4} - 5.1 \times 10^{-2}$ |
| | _ | $2.84 \times 10^{-4} - 9.08 \times 10^{-3}$ |
| -2.11×10^{-1} | $0-1.96 \times 10^{-1}$ | $0-2.11 \times 10^{-1}$ |
| $44 \times 10^{-3} - 7.4 \times 10^{-1}$ | $5.44 \times 10^{-3} - 7.62 \times 10^{-1}$ | $5.44 \times 10^{-3} - 5.33 \times 10^{-1}$ |
| $73 \times 10^{-3} - 2.5 \times 10^{-1}$ | $3.26 \times 10^{-3} - 3.07 \times 10^{-1}$ | $1.43 \times 10^{-3} - 2.56 \times 10^{-1}$ |
| $36 \times 10^{-4} - 3.1 \times 10^{-2}$ | $5.05 \times 10^{-4} - 4.78 \times 10^{-2}$ | $3.36 \times 10^{-4} - 3.1 \times 10^{-2}$ |
| $63 \times 10^{-4} - 3.55 \times 10^{-2}$ | $0-2.26 \times 10^{-2}$ | $2.22 \times 10^{-4} - 5.23 \times 10^{-2}$ |
| $71 \times 10^{-2} - 1.1$ | $8.21 \times 10^{-2} - 8.54 \times 10^{-1}$ | $7.6 \times 10^{-2} - 7.41 \times 10^{-1}$ |
| | $ \frac{10}{31 \times 10^{-2} - 5.66 \times 10^{-2}} $ $ \frac{33 \times 10^{-4} - 5.06 \times 10^{-2}}{2.11 \times 10^{-1}} $ $ \frac{144 \times 10^{-3} - 7.4 \times 10^{-1}}{73 \times 10^{-3} - 2.5 \times 10^{-1}} $ $ \frac{36 \times 10^{-4} - 3.1 \times 10^{-2}}{33 \times 10^{-4} - 3.55 \times 10^{-2}} $ $ \frac{100}{71 \times 10^{-2} - 1.1} $ | $\begin{array}{cccccccccccccccccccccccccccccccccccc$ |

| Index | 2010 | 2011 | 2012 |
|----------|---|--|---|
| Cr(VI) | $8.92 \times 10^{-2} - 3.86 \times 10^{-1}$ | 2.97×10^{-2} -2.22 × 10 ⁻¹ | $5.93 \times 10^{-3} - 2.82 \times 10^{-1}$ |
| As | - | - | $0-1.08 \times 10^{-3}$ |
| Fe | $1.48 \times 10^{-6} - 4.6 \times 10^{-4}$ | $1.48 \times 10^{-6} 5.01 \times 10^{-4}$ | $1.48 \times 10^{-6} 4.83 \times 10^{-4}$ |
| Mn | _ | _ | $2.42 \times 10^{-4} - 7.74 \times 10^{-3}$ |
| Cu | $0-3.19 \times 10^{-3}$ | $0-2.97 \times 10^{-3}$ | $0-3.19 \times 10^{-3}$ |
| Fluoride | $7.41 \times 10^{-6} - 1.01 \times 10^{-3}$ | $7.41 \times 10^{-6} - 1.04 \times 10^{-3}$ | $7.41 \times 10^{-6} - 7.27 \times 10^{-4}$ |
| Nitrate | $1.83 \times 10^{-5} - 6.81 \times 10^{-4}$ | $8.9 \times 10^{-6} - 8.36 \times 10^{-4}$ | $3.89 \times 10^{-6} - 6.99 \times 10^{-4}$ |
| Ammonia | $4.59 \times 10^{-7} - 4.22 \times 10^{-5}$ | $6.88 \times 10^{-7} - 6.51 \times 10^{-5}$ | $4.59 \times 10^{-7} - 4.22 \times 10^{-5}$ |
| Nitrite | $4.45 \times 10^{-7} - 9.69 \times 10^{-5}$ | $0-6.17 \times 10^{-5}$ | $6.23 \times 10^{-7} - 1.47 \times 10^{-4}$ |
| HQ | $8.96 \times 10^{-2} - 3.9 \times 10^{-1}$ | $3.01 \times 10^{-2} - 2.23 \times 10^{-1}$ | $7.16 \times 10^{-3} - 2.88 \times 10^{-4}$ |
| | | | |

Table 8 Hazard index through skin exposure pathway

Table 9

Result of non-carcinogenic risk for multi-exposure pathway

| Index | 2010 | 2011 | 2012 |
|----------|--|--|---|
| Cr(VI) | $1.02 \times 10^{-1} - 4.22 \times 10^{-1}$ | $3.4 \times 10^{-2} - 2.55 \times 10^{-1}$ | 6.77×10^{-3} - 3.21×10^{-1} |
| As | _ | _ | $0-1.29 \times 10^{-1}$ |
| Fe | $1.65 \times 10^{-4} - 5.1 \times 10^{-2}$ | $1.65 \times 10^{-4} - 5.34 \times 10^{-2}$ | $1.65 \times 10^{-4} - 5.15 \times 10^{-2}$ |
| Mn | _ | _ | $5.26 \times 10^{-4} - 1.68 \times 10^{-2}$ |
| Cu | $0-2.14 \times 10^{-1}$ | $0-1.99 \times 10^{-1}$ | $0-2.14 \times 10^{-1}$ |
| Fluoride | $5.45 \times 10^{-3} - 7.41 \times 10^{-1}$ | $5.45 \times 10^{-3} - 7.63 \times 10^{-1}$ | $5.45 \times 10^{-3} - 5.34 \times 10^{-1}$ |
| Nitrate | 6.75×10^{-3} -2.51 × 10 ⁻¹ | $3.27 \times 10^{-3} - 3.08 \times 10^{-1}$ | $1.43 \times 10^{-3} - 2.57 \times 10^{-1}$ |
| Ammonia | $3.37 \times 10^{-4} - 3.1 \times 10^{-2}$ | 5.05×10^{-4} -4.79 × 10 ⁻² | $3.37 \times 10^{-4} - 3.1 \times 10^{-2}$ |
| Nitrite | $1.64 \times 10^{-4} - 3.56 \times 10^{-2}$ | $0-2.27 \times 10^{-2}$ | $2.23 \times 10^{-4} - 5.25 \times 10^{-2}$ |
| HI | 2.3 × 10 ⁻¹ -1.38 | $1.27 \times 10^{-1} - 9 \times 10^{-1}$ | $8.86 \times 10^{-2} - 8.53 \times 10^{-1}$ |
| | | | |

order is obtained for risks posed by dermal contact according to mean risk values. Except the risk index of total damages of the 2[#] water source in 2010 by way of drinking water which exceed 1, the single-factor non- carcinogenic risk indices and the total damages of the aforementioned nine kinds of primary pollutants in water sources are all below 1 (the acceptable level), that is, these pollutants are all lower than the thresholds which do harms on human health. Accordingly, the pollutants in human bodies by way of drinking water could not produce non-carcinogenic chronic toxicity on drinking population. Blaylock et al. [41] concluded that, when HQ is below 0.1, the chemical pollutants cannot produce adverse effects on health; when HQ is greater than 0.1 but lower than 1, the further investigations are required before the measures are taken; when HQ exceed 1, the chemical pollutants are likely to produce adverse impacts and the measures of remediation should be taken immediately. Consequently, we cannot ignore the pollutions induced by chemical substances on water sources in the study area. Especially the fluoride, it can be observed that in the 2[#] water source, the calculation results of non-carcinogenic risk for four times all exceed 0.5. while in the 1[#], 2[#], and 32[#] water source, the calculation results of non-carcinogenic risk for four times are all greater than 0.1. Besides, by way of drinking, the non-carcinogenic risks of As, Cu, and nitrate in some water sources exceed 0.1. The above-described results should be attached serious attentions. These pollutants should be listed as the pollutants with emphasis on priority, and further analyses on the source of pollutions are required.

4.4.2.2. Way of skin exposure As listed in Table 8, in descending order of risk indices induced by single pollutants by way of skin exposure, the pollutants include Cr(VI), Mn, Cu, As, fluoride, nitrate, Fe, ammonia nitrogen, and nitrite. Except Cr(VI), in water resources to be detected, the calculation results of non-carcinogenic risk induced by the other eight pollutants by way of skin exposure are all below 0.01, suggesting that these eight pollutants are harmless on human health by way of skin exposure. With regard to Cr(VI), the non-carcinogenic risks by way of skin exposure are basically greater than 0.01. Even in some water sources, the non-carcinogenic risks due to skin exposure exceed 0.1; that is, the in-depth investigations on Cr(VI) are necessary.

By comparing the results in Tables 7 and 8, we can conclude that, for the primary pollutants (excluding Cr(VI)), the non-carcinogenic risks by way of skin exposure are lower compared with the risks by way of drinking water. These results, which are in good agreements well with the research results conducted by Hou [38] indicates that the pollutants in water enter human bodies mainly by way of drinking water.

4.4.2.3. Total non-carcinogenic risk Table 9 displays the health risk assessment results of nine primary pollutants

which do harm to human health, from which we can observe that, the risk of fluoride is greatest, followed by Cr(VI), nitrate, As, Cu, Fe, Mn, and ammonia nitrogen, while the risk of nitrite is lowest. In 2010, 2011, and 2012, the total damage indices of these nine primary pollutants by ways of drinking water and skin exposure are $0-7.41 \times 10^{-1}$, $0-7.63 \times 10^{-1}$, and $0-5.34 \times 10^{-1}$, while the non-carcinogenic risks of the combination of pollutants are 2.3×10^{-1} -1.38, 1.27×10^{-1} -9 × 10⁻¹, and 8.86×10^{-2} – 8.53×10^{-1} , respectively. Except the conditions in 2010, in the 2[#], 15[#], and 31[#] water source, the total risks of the combined factors exceed 1, and the calculation results of total risks in the other water sources are all less than 1 (the risk control criterion). Zhang et al. [42] assessed the groundwater quality from the perspective of human health risk in Hetao Plain of mid-north China, and they found that 87.9% of the collected samples were not suitable for drinking from the perspective of human health risk. It is easy to find from the comparison that the study area is among the most risky areas in China regarding groundwater contamination. These results indicate that, when exposed to the vast majority of water sources in the study area, no obvious non-carcinogenic risks could be produced. Overall, fluoride exhibits the highest non-carcinogenic risk, which is therefore ranked first among these non-carcinogenic risk factors. By ways of drinking water and skin exposure, the average contribution rates of fluoride on total non-carcinogenic risks in 2010, 2011, and 2012 are 72.17%, 25.8%, 29.28%, and 31.54%, respectively.

4.5. Analysis of uncertainty

4.5.1. Analysis of simulation results

In the present work, CB (version 11.1) was adopted to perform MC simulations on uncertainty [43].

First, the distributions of the values of weight, rate of drinking, exposure frequency and the concentrations of pollutants were input into the corresponding cells. The number of simulations can impact the accuracy and stability of the final results. Some studies have shown that the stimulation results can be stable with more than 4,000 simulations and that are even more accurate with 10,000 simulations [43,44]. Based on the run of 10,000 trials and the mean standard error and proper confidence intervals can be constructed [44]. With the use of CB11.1, the sampling was repeated for 10,000 times at a confidence level of 95%, and then the statistical analysis results and probability distributions of carcinogenic and non-carcinogenic risks of the pollutants at the confidence interval of [5%, 95%] were acquired, with the results presented in Fig. 2.

As shown in Fig. 2, the total carcinogenic risks of the primary pollutants in 2010, 2011, and 2012 are 5.05×10^{-5} - 3.15×10^{-4} , 1.64×10^{-5} - 1.66×10^{-4} , and 6.15×10^{-6} - 2.35×10^{-4} , with the averages of 1.55×10^{-4} , 6.84×10^{-5} , and 8.69×10^{-5} , all of which exceed the acceptable level (10^{-6}). Compared with the calculation results of total carcinogenic risks by USEPA model (as listed in Table 6), the total carcinogenic risks calculated by MC method exhibit wide ranges, that is, the samplings of risk distribution in simulations are wider.

It can be observed from Table 10 and Fig. 3 that the estimation results of total non-carcinogenic risks of the primary pollutants by MC simulation method in 2010, 2011, and 2012 are 2.19×10^{-1} –1.11, 1.23×10^{-1} – 8.17×10^{-1} , and 1.26×10^{-1} – 7.7×10^{-1} ,

respectively, the risks are all controlled within 1 on the whole. Compared with the calculation results of total non-carcinogenic risks by USEPA model (as listed in Table 9), the total non-carcinogenic risks calculated by MC method are narrower. These results depend on the setting of confidence interval.

4.5.2. Analysis of sensibility

Analysis of sensibility, as an important step in MC simulations, refers to the quantitative research on the influences and the related degrees of influence of several parameters with strong sensibility on risk assessment results. In the present work, the sensibilities of several parameters on the statistical results of carcinogenic and non-carcinogenic risks were analyzed using CB11.1 software.

As displayed in Table 11, the sensitivities of various factors on the statistical results of carcinogenic and non-carcinogenic risks are –14.5%–52.5% and –14.7%–35.2%. To be specific, the body weight (BW) is negatively sensitive to risk assessment results while the rate of drinking (IR), exposure frequency (EF) and the surface area of skin (SA) exhibit positive sensitivity to risk assessment results. Moreover, the rate of drinking and the concentration of Cr(VI) are most sensitive to risk assessment results. It should be noted that, the larger the absolute value of the sensitivity of a factor, the greater the effect of this factor on assessment results. Accordingly, the exposure parameters play critical roles in



Fig. 2. Distribution of total carcinogenic risks simulated by MC.

Table 10 Simulation results of non-carcinogenic risks

| 2010 | 2011 | 2012 |
|--|--|---|
| $[1.22 \times 10^{-1}, 4.41 \times 10^{-1}]$ | $[3.59 \times 10^{-2}, 2.38 \times 10^{-1}]$ | $[1.03 \times 10^{-2}, 3.34 \times 10^{-1}]$ |
| _ | _ | $[3.7 \times 10^{-4}, 6.16 \times 10^{-2}]$ |
| [1.18 × 10 ⁻⁴ , 3.94 × 10 ⁻²] | $[8.14 \times 10^{-5}, 5.09 \times 10^{-2}]$ | $[1.2 \times 10^{-4}, 4.43 \times 10^{-2}]$ |
| _ | _ | [3.06 × 10 ⁻⁴ , 1.47 × 10 ⁻²] |
| [6.09 × 10 ⁻³ , 5.79 × 10 ⁻²] | $[7.63 \times 10^{-3}, 4.87 \times 10^{-2}]$ | [6.38 × 10 ⁻³ , 6.96 × 10 ⁻²] |
| [6.62 × 10 ⁻³ , 5.73 × 10 ⁻¹] | $[5.06 \times 10^{-3}, 4.17 \times 10^{-1}]$ | [4.4 × 10 ⁻² , 4.73 × 10 ⁻¹] |
| [5.15 × 10 ⁻³ , 2.31 × 10 ⁻¹] | [4.04 × 10 ⁻³ , 3.29 × 10 ⁻¹] | $[1.67 \times 10^{-3}, 1.67 \times 10^{-1}]$ |
| [5.43 × 10 ⁻⁴ , 1.16 × 10 ⁻²] | $[2.16 \times 10^{-3}, 4.01 \times 10^{-2}]$ | $[7.57 \times 10^{-4}, 1.76 \times 10^{-2}]$ |
| [3.13 × 10 ⁻⁴ , 2.13 × 10 ⁻²] | [1.43 × 10 ⁻³ , 2.36 × 10 ⁻²] | [3.69 × 10 ⁻⁴ , 5.5 × 10 ⁻²] |
| $[2.19 \times 10^{-1}, 1.11]$ | $[1.23 \times 10^{-1}, 8.17 \times 10^{-1}]$ | $[1.26 \times 10^{-1}, 7.7 \times 10^{-1}]$ |
| | $\begin{array}{c} 2010 \\ [1.22 \times 10^{-1}, 4.41 \times 10^{-1}] \\ - \\ [1.18 \times 10^{-4}, 3.94 \times 10^{-2}] \\ - \\ [6.09 \times 10^{-3}, 5.79 \times 10^{-2}] \\ [6.62 \times 10^{-3}, 5.73 \times 10^{-1}] \\ [5.15 \times 10^{-3}, 2.31 \times 10^{-1}] \\ [5.43 \times 10^{-4}, 1.16 \times 10^{-2}] \\ [3.13 \times 10^{-4}, 2.13 \times 10^{-2}] \\ [2.19 \times 10^{-1}, 1.11] \end{array}$ | $\begin{array}{c ccccccccccccccccccccccccccccccccccc$ |



Fig. 3. Distribution of total non-carcinogenic risks simulated by MC.

carcinogenic risk assessment. In order to reduce the uncertainties in risk assessment results, we should enhance the accuracy and representativeness of exposure parameters. By contrast with the results of carcinogenic risks, there are more effecting factors on non-carcinogenic risks. On account of the sharing of the concentrations of several pollutants such as fluoride, As, Fe, and Cu, the absolute values of the sensitivity of the rate of drinking and Cr(VI) are reduced; that is, the more the parameters involved, the less the absolute value of the sensitivity of single factor.

5. Discussion

According to the statistical data from the Health and Family Planning Commission of Sichuan Province, the population death rate in this region was only 8.59‰ in 2015, in which the top 10 types of diseases and top 10 single diseases with high morality are listed in Table 12 [45].

As displayed in Table 12, the tumor ranks the second among the types of diseases causing death, with the death rate of 1.65‰, while the lung cancer, liver cancer, stomach cancer, and esophageal cancer rank the 3rd, 4th, 5th, and 6th among the single diseases causing death, with the death rates above 0.2‰. In other words, the cancers now have become one of the primary causes of death in the study area. The total carcinogenic risks of rural drinking water quality calculated by USEPA model and MC simulation both exceed the acceptable level (10-⁶). More seriously, the annual carcinogenic risks of some water sources even exceed the risk management criteria by USEPA (10⁻⁴). Considering that the annual death rate induced by cancer in the study area is 165.1/10⁵ (i.e., approximately 165.1 people die with cancer among 100,000 people), the carcinogenic risks of water quality calculated by these two methods are fairly low and thus acceptable. Meanwhile, the non-carcinogenic damages in rural drinking water quality are acceptable on the whole. As a consequence, for the current concentrations of chemical pollutants in drinking water, the carcinogenic risk of Cr(VI) imposes the greatest threat on human health.

6. Conclusion

USEPA Health Risk Assessment Model was improved in response to the drinking habit of the residents in this research area, and thus a health risk assessment of water quality is carried out against concerned contaminants. The main achievements were as follows:

- 1. The carcinogenic risk of both Cr(VI) and As was over the acceptable level 10⁻⁶ and they should be considered as the major contaminants in the drinking water of this area.
- 2. As for non-carcinogenic risk, excepted that the total non-carcinogenic risk of the drinking water in Matou Village, Yongxing Town in 2010 was greater than 1, that of others was all within the acceptable level 1. Skin exposure route: the non-carcinogenic risk of skin exposure to Cr(VI) was basically greater than 0.01, while that of skin

| Table 11 | | |
|----------|----------------|-------------|
| Results | of parameter's | sensitivity |

| Index | IR | Cr(VI) | Fluoride | BW | Nitrate | As | Fe | Cu | SA | Ammonia | EF | Nitrite | Mn |
|-------------|-----------|-----------|----------|------------|---------|-----|-----|-----|-----|---------|-----|---------|-----|
| Sensitivity | 52.5(CR) | 32.7(CR) | 17.7 | -14.5(CR) | 5.6 | 3.0 | 1.7 | 1.4 | 0.6 | 0.6 | 0.3 | 0.2 | 0.1 |
| (%) | 18.9(NCR) | 35.2(NCR) | | -14.7(NCR) | | | | | | | | | |

CR, carcinogenic risk; NCR, non-carcinogenic risk.

Table 12

Death rate of 10 major diseases categories and 10 single-species major diseases

| Order | Type of disease | | Singe disease | | | | |
|-------|-----------------------------------|---------------------------------|-----------------------------|---------------------------------|--|--|--|
| | Name | Death rate (1/10 ⁵) | Name | Death rate (1/10 ⁵) | | | |
| 1 | Circulatory System | 176.1 | Chronic respiratory disease | 125.7 | | | |
| 2 | Cancer | 165.1 | Cerebrovascular disease | 109.2 | | | |
| 3 | Respiratory system | 143.6 | Lung cancer | 41.8 | | | |
| 4 | Toxic | 48.2 | Liver cancer | 29.8 | | | |
| 5 | Digestive system | 22 | Stomach cancer | 22.9 | | | |
| 6 | Endocrine and immunity | 11.8 | Esophagus cancer | 21.1 | | | |
| 7 | Infectious and parasitic diseases | 6.8 | Acute myocardial infarction | 20.6 | | | |
| 8 | Genito-urinary system | 6.6 | Heart disease | 15.7 | | | |
| 9 | Diseases of nervous system | 3.6 | Pneumonia | 12.9 | | | |
| 10 | Mental disorders | 2.1 | Traffic accidents | 11.5 | | | |

exposure to all the remaining eight major contaminants are less than 0.01.

3. The non-carcinogenic risk was basically within the acceptable level 1. Uncertainty is inescapability in the process of risk assessment, so MC was adopted to quantify the uncertainties, and under the 95% confidence level, carcinogenic risk and non-carcinogenic risk were all within the acceptable level.

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