Research Article

Paraquat in Surface Water of Some Streams in Mai Chau Province, the Northern Vietnam: Concentrations, Profiles, and Human Risk Assessments

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The concentrations and profiles of paraquat, a kind of herbicide, were studied in water samples taken from a stream flowing through five villages of Mai Chau province, the Northern Vietnam, during dry and rainy seasons. In this study, paraquat was found at almost the sampling sites and showed an average concentration for paraquat to be 30.69 $\mu\text{g/L}$ and with a maximum of 134.08 $\mu\text{g/L}$. The herbicide concentration tended to be the highest on the dry season because of the least rainfall and also the highest evaporation rate of water in the stream. For risk assessment of human health, a hazard index (HI) value was calculated for estimating the risk towards the residents. HQ in the dry and rainy season is from 0.0001 to 0.2448 and from 0.0001 to 0.0279, respectively. The results showed a minimum risk; however, there are concerns toward the danger of long-term exposure to the residents from the stream that could affect their life quality.

1. Introduction

Paraquat (1,1'-dimethyl-4,4'-bipyridylium dichloride), a kind of a nonselective, contact, broad-spectrum herbicide, has been widely used as a herbicide for many decades [1–3]. The herbicide is sold in about 130 countries for use on large and small farms, paddies, and nonagricultural weed control. Gramoxone is the commercial name for paraquat. The use of paraquat has been discussed for decades in international, national, and nongovernmental organizations [4–16].

The herbicide under aerobic conditions is rapidly reoxidized by accepting an electron from nicotine adenine dinucleotide phosphate (NADPH) with resulting formation of the superoxide or other oxygen radicals. Both the superoxide and the radicals damage cell membranes and cytoplasm from NADPH depletion, free radical generation, and lipid peroxidation, or some combination of these factors [17–19]. It is poorly absorbed through intact skin, but penetration is considerably increased by damage to the skin, which is of particular concern because paraquat itself is a skin irritant [20].

With damage mentioned above, using paraquat may pose potential environmental hazards to humans or surrounding ecosystems [21, 22]. Its acute toxicity at oral LD50 ranges from 4 to 40 mg kg^{-1} [23]. However, up to 3.5 mg kg^{-1} can be absorbed through the skin or respiratory route without acute toxic damage. Besides irreversible human lung damage [24], recent concern has been raised about an association of Parkinson's disease to paraquat or a combination of paraquat and fungicide exposures [25]. There have been few studies on their appearance, persistence, and distribution in the environment [17, 26], and none has been performed in the Southeastern Asia area. However, the results that have been obtained are alarming. In water environment, paraquat is adsorbed onto particles and sediment with a half-life time to be between 2 and 820 years depending on sunlight and depth of water, and it has been found in both surface waters and groundwater [27]. Paraquat is not easily degraded chemically or microbiologically and demonstrates long-term persistence in river waters with more than 80% remaining after 56 days of incubation [28]. In water, this compound is completely

FIGURE 1: Map of the sampling sites in Mai Chau province, the Northern Vietnam.

ionized and shows an octanol-water partition coefficient (K_{ow}) of 1.8, which means that the 64.4% of paraquat appears in the octanol phase [28]. The toxicity effect of paraquat is from low to medium for fish but high for birds [29].

The aim of this study was to evaluate the impact of paraquat on pollution in water environment. For this purpose, the concentration of this herbicide was studied in a stream flowing through five villages of Mai Chau province. This area is characteristic of the Northern Vietnam geography. The analytical methodology used is based on the separation and concentration of the herbicides by solid phase extraction and determination by means of HPLC with UV [30, 31]. The risk assessment of human health was estimated from a hazard index (HI) value. These results may provide useful information for establishing paraquat control and give initial guidance on the health risk.

2. Materials and Methods

2.1. Taking Sample. In this study, the samples were collected from a stream flowing through five villages (Cun Pheo, Pieng Ve, Mai Hich, Xam Khoe, and Van Mai) at Mai

Chau province, the Northern Vietnam, during dry and rainy seasons (Figure 1). Samples of 2.5 L were taken into polypropylene containers because paraquat adheres to glass [32, 33]. Afterward, they were filtrated through a Whatman 0.45 μ m membrane filter and stored in a refrigerator at 4°C until analyzed.

2.2. Analytical Procedure. The analytical procedure of paraquat in the water samples has been described by Ibanez et al. [30]. Commercial silica Sep-Pak cartridges (Waters) were successively washed with 2.5 mL 0.5 M sulphuric acid, 5 mL distilled water, and 2.5 mL 2% (v/v) ammonium hydroxide solution and again with 5 mL distilled water. Samples (250 mL) were passed through the Sep-Pak cartridges [31]. Then they were inverted and eluted with 5 mL of a mixture composed of 2 g tetramethylammonium hydroxide pentahydrate (TMAOH) and 30 g ammonium sulphate adjusted to pH 3 with 5 M sulphuric acid and methanol (90 : 10). A 20 μ L aliquot of the eluted sample was injected onto the HPLC system.

The HPLC set-up comprised a pump system Alltech 426 HPLC, a programmable variable-wavelength detector Flexar

UV/VIS LC (PerkinElmer, USA). The analytical column was a Inertsil ODS-3 (GL Sciences, Japan) (15 cm × 2.2 mm ID filled with 3 µm particle size silica) with a silica guard cartridge (1 cm × 4.6 mm ID, 5 µm) was used to prevent damage of the analytical column.

The separation of the selected herbicides was performed by gradient elution. The initial mobile-phase composition was a 100% solution of 2 g TMAOH with 30 g ammonium sulphate in 1 L of water and adjusted to pH 3 with 5 M sulphuric acid solution. It was linearly programmed to mix with 50% methanol after 15 min and hold up during 10 min. The flow rate of the mobile-phase was 0.5 mL/min. The eluted compounds were monitored with a UV detector, set initially at 260 nm after 6 min for paraquat.

The concentrations of paraquat in the final extract were calculated by interpolating the peak area value in the linear equation obtained by least square adjustment of four points which cover a concentration range of 0.1–10 µg/L.

The mean accuracy, precision, and method detection limits (MDLs) were calculated for data acquired from 10 replicate analysis of distilled water and from samples of the three studied areas. Samples were spiked at 0.4 µg/L. The mean recovery for paraquat ranged from 85 to 102%. The recovery depends on the water sample characteristics like pH, inorganic salts, humic acids, and other organic contaminants presents. The precision of the measurements ranged from 3.5 to 12.7%. MDLs ranged from 0.03 to 0.09 µg/L depending on the recovery obtained.

2.3. Dermal Exposure Assessment. Based on the information of the residents using the stream water and the results of paraquat concentrations, dermal absorbed dose (DAD) value was done after which the hazard quotient (HQ) was calculated. To indicate risk, HQ have to be more than one (HQ > 1). DEA was obtained based on exposure of residents through certain body surfaces, for example, arms, hands, legs, and feet [34]

$$\text{DAD} = \frac{\text{DA}_{\text{event}} \times \text{EV} \times \text{ED} \times \text{EF} \times \text{SA}}{\text{BW} \times \text{AT}}, \quad (1)$$

where DAD is dermal absorbed dose (mg/kg-day), DA_{event} represents the absorbed dose per event (mg/cm²-event), SA is surface area available contact (cm²), EV is event frequency (events/day), EF is exposure frequency (days/year), and ED is the exposure duration (years). Additionally, BW represented body weight (kg), and AT is the averaging time (days), where for noncarcinogenic effects AT value is equal to the ED × 365 days/year. For carcinogenic effects, AT is stated as 70 years × 365 days/year. As for absorbed dose per event, it was calculated as follows [35]:

$$\text{DA}_{\text{event}} = K_p \times C_w \times t_{\text{event}}, \quad (2)$$

where the DA_{event} is the absorbed dose per event (mg/cm²-event), K_p is interpreted as the dermal permeability coefficient of compound in water (cm/hour), and C_w is related to chemical concentration in water (mg/cm³), while t_{event} is the event duration (hour/event) that was gathered through

FIGURE 2: Concentration of paraquat (µg/L) in four villages (Cun Pheo, Pieng Ve, Xam Khoe, and Mai Hich) at Mai Chau province, the Northern Vietnam.

questionnaire. After DAD value had been obtained, it was used to calculate HQ by dividing the DAD value with reference dose value ($\text{RfD}_{\text{ABS}} = 0.0045 \text{ mg/kg/day}$) [35] (see (3)). For the absorption of this chemical compound through different body encounter, HQ values were combined to form hazard index (HI), with the assumption that the effects of different route of entry for paraquat compound and effects could be additive. Thus, this formula was used to estimate the volume possessed by the entry of paraquat compound through several parts of the body [36]

$$\text{Hazard quotient (HQ)} = \frac{\text{DAD}}{\text{RfD}_{\text{ABS}}}, \quad (3)$$

$$\text{Hazard index (HI)} = \sum \text{HQ}_{\text{Arms, Hands, Legs, Feet}}, \quad (4)$$

where it was stated that HI value that exceeds one would most probably indicate risk towards the target organ of related compound vice versa.

3. Results and Discussion

3.1. Contamination Characteristic of Paraquat in Surface Water. The bipyridylum herbicide was found in the studied areas. Concentrations of paraquat in the stream water were given in Table 1 with range from 4.70 to 134.08 µg/L. Median paraquat concentrations in groundwater in Cun Pheo, Pieng Ve, Xam Khoe, and Mai Hich were 81.14, 34.95, 30.51, and 13.28 µg/L, respectively (Table 1). Of the 68 samples analyzed from Mai Chau province, paraquat was found in 67.7% of the samples. However, about 42.6% of these samples which contained paraquat concentrations exceeded Canada drinking water guideline of 10 µg/L [37]. Of the 16 samples taken in Van Mai village in dry and rainy seasons, paraquat was not detected in 100% of the samples.

The results from Table 1 and Figure 2 show that general patterns of paraquat accumulation in stream water in Mai Chau were Cun Pheo > Pieng Ve > Mai Hich > Xam Khoe >

TABLE 1: Concentrations of paraquat ($\mu\text{g/L}$) in the surface water samples in five villages (Cun Pheo, Pieng Ve, Xam Khoe, Mai Hich, and Van Mai) at Mai Chau province in the Northern Vietnam.

	Paraquat	
	Dry season	Rainy season
<i>Cun Pheo village</i>		
Mean	68.55	20.70
SD	40.37	17.98
Min	7.80	4.70
Max	134.08	55.74
Median	64.63	10.95
N ^a	9/9	9/9
<i>Pieng Ve village</i>		
Mean	34.95	21.11
SD	19.80	11.08
Min	5.30	9.74
Max	52.74	43.62
Median	40.98	18.46
N	8/8	5/8
<i>Xam Khoe village</i>		
Mean	30.51	2.62
SD	5.38	0.82
Min	23.65	1.72
Max	36.78	3.68
Median	30.09	2.64
N	5/5	5/5
<i>Mai Hich village</i>		
Mean	13.28	4.5
SD	9.39	-
Min	4.70	4.5
Max	21.57	4.5
Median	13.43	4.5
N	4/4	1/4
<i>Van Mai village</i>		
Mean	-	-
SD	-	-
Min	-	-
Max	-	-
Median	-	-
N	0/8	0/8
Drinking water guideline (Canada, 1991)	10	

^aNumber of samples with detectable concentration.

Van Mai village. It may be explained that amounts of the herbicide used for agriculture by the residents show very high difference between the studied villages. The herbicide concentration tended to be the highest on the dry season because these are the months with the least rainfall and the highest evaporation rates.

In the comparison with some previous studies, concentrations of paraquat in Mai Chau are significantly higher than those in some areas reported like in water from marsh of the Valencian community, Spain 0.19–3.95 $\mu\text{g/L}$ [17], and rivers and dams in Saint Lucia, range of paraquat is from 0.5 to 1.0 $\mu\text{g/L}$ [38], and mean concentration of paraquat in surface waters in Elechi Creek, Niger delta, Nigeria, is 0.01 $\mu\text{g/L}$ [39]. From Warri river basin, Niger delta, Nigeria ranges from 0.02 to 4.17 $\mu\text{g/L}$ [40]. In the drainage canals of the Kerian paddy fields, Malaysia ranges from 0.6 to 6.92 $\mu\text{g/L}$ [41] (Figure 3).

3.2. The Information of Residents Using the Stream Water at the Studied Area. Table 2 showed characteristics of the 60 residents using the stream water at the studied area. The result indicates that the ages of residents below 40, from 41 to 50, from 51 to 60, from 61 to 70, and above 70 are 48.33%, 36.67%, 8.33%, 5%, and 1.67%, respectively. Additionally, the number of male residents at 19 persons (31.67%) is approximately 2 times smaller than the female residents of 41 persons (68.33%). This study has shown that the residents, aged between 40 and 70 years, were predominantly females rather than male residents. 36.7% of the residents weighed from 51 kg to 60 kg, followed by the weight group of 61 kg to 70 kg at 30% and 41 kg to 50 kg at 21.7%, those who weigh below 40 kg at 8.3%, and those who are above 70 kg at 3.3%. The people were also asked regarding duration in which they have been using paraquat-containing stream water. This is to estimate their exposure duration with the paraquat. It was indicated that more than half of the residents involved in this survey (53.3%) were exposed to this compound for approximately three years. In addition, 30% were exposed for two years and 8.3% for one year, with only 1.7% for both four-year and five-year exposure. On the other hand, 5% of residents indicated no usage of paraquat at all; therefore, for the purpose of this study, they were considered as to have no direct exposure to the compound.

3.3. Dermal Exposure Assessment Analysis. It can be seen from Figure 4 that the HI values for the residents living five studied villages decreased in the order Cun Pheo > Pieng Ve, Mai Hich > Xam Khoe > Van Mai village. The HQ values for the residents are much smaller than 1 (where HQ > 1 indicates at risk [36]); therefore, the risk of paraquat to the health of the residents living in studied areas is insignificant (Table 3 and Figure 4). HQ in the dry season is from 0.0001 to 0.2448 and mean is 0.013; HQ in the rainy season is from 0.0001 to 0.0279 and mean is 0.0052. The HQ mean value of paraquat in the dry season is approximately 9 times higher than that in the rainy season.

4. Conclusions

The studied areas showed the presence of paraquat. Paraquat was detected in a number of sites at levels sometimes considerably above of 10 $\mu\text{g/L}$ value established by Canada drinking water guideline. The average concentration of paraquat was 30.69 $\mu\text{g/L}$, and these values tended to be the highest during

FIGURE 3: Paraquat concentrations ($\mu\text{g/L}$) in surface water in paraquat-contaminated areas. Bar and line indicate range and median, respectively.

FIGURE 4: Hazard index of residents living in Mai Chau in dry and rainy seasons.

TABLE 2: Characteristics of residents living in the studied areas in Mai Chau province.

Characteristics	Number (<i>n</i> = 60)	Percentage (%)
<i>Sex</i>		
Male	19	31.67
Female	41	68.33
<i>Age (years)</i>		
<40	29	48.33
41–50	22	36.67
51–60	5	8.33
61–70	3	5.00
>70	1	1.67
<i>Weight (kg)</i>		
<40	5	8.30
41–50	13	21.70
51–60	22	36.70
61–70	18	30.00
>70	2	3.30
<i>Hours (used the stream water) per day</i>		
0.5	20	33.30
1	37	61.70
2	1	1.70
3	2	3.30
<i>Days (used the stream water) per week</i>		
3	1	1.70
4	7	11.70
5	3	5.00
6	17	28.30
7	32	53.30
<i>Exposure to paraquat (year(s))</i>		
None	3	5.00
1	5	8.30
2	18	30.00
3	32	53.30
4	1	1.70
5	1	1.70

the dry season. These are the months with the least rainfall and highest temperatures. The results obtained show the ubiquitous presence of paraquat in the studied villages at the Mai Chau environment, where their accumulation in streams gives rise to situations that could affect the food chain and human health. The risk assessment of human health showed the insignificant effect of paraquat ($HQ < 1$). However, the use of paraquat for agriculture at this area should be controlled and adjusted to real needs. The presence of these herbicides in the environment should be monitored in order to establish the real impact on the flora and fauna. Further studies are needed to evaluate the potential health effects of paraquat from stream. Furthermore, neurological tests should be conducted to examine the

prevalence of chronic paraquat poisoning symptoms to be studied for the residents in Mai Chau province, the Northern Vietnam.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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TABLE 3: The risk assessment values of the residents using stream water in the studied areas in the dry and rainy seasons.

	Dry season					Rainy season				
	HQ _{Arm}	HQ _{Hand}	HQ _{Leg}	HQ _{Feet}	HI	HQ _{Arm}	HQ _{Hand}	HQ _{Leg}	HQ _{Feet}	HI
<i>Cun Pheo village</i>										
Mean	0.0065	0.0342	0.0121	0.0685	0.1209	0.0127	0.0070	-	-	0.0197
SD	0.0068	0.0376	0.0136	0.0751	0.1330	0.0086	0.0099	-	-	0.0158
Min	0.0008	0.0002	0.0001	0.0004	0.0008	0.0000	0.0000	-	-	0.0000
Max	0.0229	0.1224	0.0459	0.2448	0.4360	0.0279	0.0262	-	-	0.0465
Median	0.0029	0.0170	0.0058	0.0339	0.0596	0.0123	0.0017	-	-	0.0135
N ^a	14									
<i>Pieng Ve village</i>										
Mean	0.0016	0.0088	0.0032	0.0177	0.0313	0.0043	0.0033	-	-	0.0076
SD	0.0010	0.0052	0.0019	0.0104	0.0184	0.0062	0.0046	-	-	0.0108
Min	0.0001	0.0004	0.0002	0.0008	0.0016	0.0000	0.0000	-	-	0.0000
Max	0.0031	0.0157	0.0062	0.0313	0.0558	0.0170	0.0123	-	-	0.0293
Median	0.0015	0.0092	0.0031	0.0184	0.0321	0.0002	0.0004	-	-	0.0006
N	13									
<i>Xam Khoe village</i>										
Mean	0.0022	0.0107	0.0044	0.0214	0.0387	0.0007	0.0001	-	-	0.0007
SD	0.0012	0.0063	0.0024	0.0125	0.0221	0.0004	0.0001	-	-	0.0004
Min	0.0009	0.0036	0.0018	0.0073	0.0137	0.0002	0.0000	-	-	0.0002
Max	0.0045	0.0268	0.0089	0.0536	0.0938	0.0013	0.0002	-	-	0.0015
Median	0.0020	0.0087	0.0039	0.0174	0.0316	0.0007	0.0001	-	-	0.0008
N	12									
<i>Mai Hich village</i>										
Mean	0.0009	0.0044	0.0018	0.0088	0.0159	0.0001	0.0000	-	-	0.0001
SD	0.0005	0.0029	0.0011	0.0059	0.0103	0.0002	0.0000	-	-	0.0002
Min	0.0004	0.0021	0.0008	0.0042	0.0075	0.0000	0.0000	-	-	0.0000
Max	0.0020	0.0117	0.0039	0.0235	0.0411	0.0004	0.0001	-	-	0.0005
Median	0.0008	0.0030	0.0017	0.0060	0.0110	0.0000	0.0000	-	-	0.0001
N	11									
<i>Van Mai village</i>										
Mean	-	-	-	-	-	-	-	-	-	-
SD	-	-	-	-	-	-	-	-	-	-
Min	-	-	-	-	-	-	-	-	-	-
Max	-	-	-	-	-	-	-	-	-	-
Median	-	-	-	-	-	-	-	-	-	-
N	10									

^aNumber of residents with using the stream water at studied sites.

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