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Naturally occurring thallium: a hidden geoenvironmental health hazard?

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Abstract

This paper illustrates a real environmental concern and draws attention to the fact that natural processes can mobilize thallium (Tl), a highly toxic metal, which may enter the food chain as a "hidden health killer" with severe health impacts on local human population. Natural processes may be exacerbated by human activities such as mining and farming, and may cause enrichment of Tl in the environment. In geochemically anomalous areas with concentrated levels of Tl in the surface environment (bedrocks, waters, soils, and crops), such as the Lanmuchang area in southwestern Guizhou Province, China, it is essential to establish base-level values and to pay heed to the geological context of "natural contamination," as high concentrations of Tl in bedrocks/ores (6–35,000 mg/kg) can lead to enrichment of Tl in the aquatic system (0.005–1100 μg/l in groundwaters and 0.07–31 μg/l in surface waters) and soil layers (1.5–124 mg/kg). In sensitive areas such as the Yanshang area of southwestern Guizhou, elevated natural levels of Tl from bedrocks may also cause higher concentrations of Tl in the surface environment, and thus more attention must be paid to geoenvironmental management of human activities if socio-economic catastrophes are to be avoided. Due to high uptake of Tl by crops, Tl can be transferred from soils to crops and remarkably concentrated in food crops. Concentrations of 1–500 mg/kg Tl based on dry weight (DW) were determined in many food crops growing on Tl-contaminated arable soils from the Lanmuchang area. The daily intake of 1.9 mg of Tl from consumed food crops was estimated for the local adult inhabitant of Lanmuchang. Thus, Tl is regarded as a latent health hazard with potential risk of toxicity in humans within areas of "natural" contamination by Tl.

Keywords: Thallium; Geoenvironment; Health hazard; Contamination; Chronic poisoning; Guizhou; China

1. Introduction

The fact that "nature" contributes to pollution is often overlooked or ignored when searching for the source of problems involving metal pollution. Human activities can accelerate natural processes. Thus, mining operations in sensitive areas can generate toxic pollution, by either the liberation of natural elements within the rocks, or by addition of artificial compounds such as cyanide leachates or hydrocarbons. To provide reliable identification of the cause of pollution, it is important to ask what kinds of base-level data exist concerning the natural migration of toxic metals prior to the onset of mining activities. Such data are essential if the impact of pollution related to metal dispersion induced by human activities is to be adequately measured. This involves the concept of geoenvironment.

The term *geoenvironment* is defined here as a geoscientific regime, which can play a role, in conjunction with other parameters [such as mineral resources, soil, topography, (sub)surface water, crops, socio-economic development, etc.], in defining environmental impacts (Guha, 2003). These impacts can be chemical or physical, and they will greatly influence the choice of parameters required to make a clear diagnosis of assessment criteria needed for proper management.

The scientific and medical literature documents negative effects of numerous mildly to highly toxic metals on human health (Christensen, 1995; Nixon and Moyer, 1996; Janssen

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et al., 2003). Thallium (Tl)'s role as a toxic health threat, which mainly affects the central nervous system, causes visual disorders (with failing eyesight or total blindness), hair loss, and even death, and has been investigated extensively (Feldman and Levisohn, 1993; Meggs et al., 1994) but mostly within the context of industrial pollution (Dolgner et al., 1983; Wells, 2001). Nevertheless, numerous studies show the presence of Tl in ore deposits, rocks, soils, water, and plants. Kuroko-type volcanogenic massive sulphide ores are known to carry significant Tl levels, with much of the metal hosted by pyrite (Murao and Itoh, 1992). Arsenical gold deposits may also display a Tl-enriched signature (Percival and Radtke, 1993), as may some Mississippi Valley-type Pb–Zn deposits (Leach et al., 1995). Experiments with bioaccumulator plant species indicate that, under favorable circumstances, plant tissues may develop dry weight (DW) Tl contents of several tenths of 1% (Leblanc et al., 1999). The importance of trace elements in soil for human health has been established for elements such as F, As, and Se, as well as heavy metals such as Cd, Hg, and Pb (Abrahams, 2002). The analysis of Tl in soil has recently been discussed by Castle (2000). However, no previous comprehensive study has provided evidence of natural contamination processes of Tl that may have serious consequences for human health. Recognition of this problem prompted a comprehensive investigation in a rural area with Tl–Hg–As sulfide mineralization in southwestern Guizhou Province, China (Fig. 1). In this rural area, symptoms related to thallotoxicosis were detected in the early 1960s with 200 cases of Tl poisoning, resulting in a number of deaths. Most of the early investigations focused on local mining activities as the cause (Zhou and Liu 1985; Zhang et al., 1998). High Tl concentrations in drinking water were found to be the principal cause of Tl poisoning. In the early 1990s, the drinking water problem was resolved by piping Tl-free groundwater from outside areas, yet high quantities of Tl (153–2668 μg/l) are still being detected in urines of the local population (Xiao, unpublished data). Thus, a further study on the health impact of Tl on the local population is urgently needed.

2. Materials and methods

2.1. Study area

The study area, centered on Lanmuchang, a small town with approximately 1000 inhabitants, was chosen for a pilot study for Tl's potential impact on the environment and the local inhabitants during both natural process and man-made disturbance. The area presents a karstic topography, with a generally higher elevation in the northwest and lower elevation in the southeast. The average altitude is 1400 m above sea level, with a relative relief of 100–200 m.

The Tl mineralization within a metallogenetic belt in the study area is associated with either independent Tl mineralization (e.g., the Lanmuchang Hg–Tl–As deposit), gold mineralization (e.g., the Yanshang Au deposit), or hydrocarbon accumulation (the Lanmuchang coal deposits) (Fig. 1). The area exhibits a karstic topography and the deposits show some similarities to epithermal Carlin-type disseminated Au mineralization, hosted by strata of Permian to

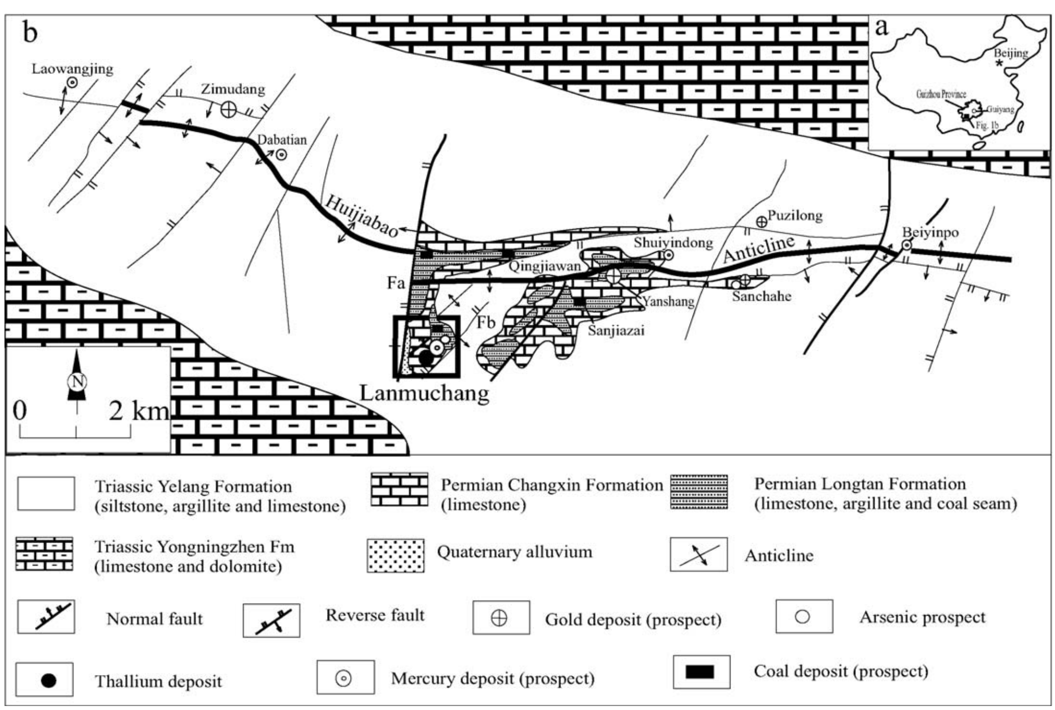

Fig. 1. (a) Sketch map showing the study area and (b) Geological sketch map showing the Huijiabao anticline metallogenic belt in southwest Guizhou, China.

Triassic age. The local history of mining for coal and mercury dates back to more than 350 years.

A detailed description of the local geology has been recently reported and published (Xiao, 2001; Xiao et al, 2003a,b). Therefore, only a brief summary is provided here. The Lanmuchang Hg–Tl–As deposit area is underlain by Permo-Triassic sediments and overlain by Quaternary alluvium. The exposed rocks include limestones, argillites, and coal seams. Thallium enrichment is associated with sulfide minerals of Tl–As–Hg with a reserve of about 500 metric tons of Tl (Chen, 1989; Xiao, 2001). The deposit has a long mining history for Hg and coals, and it was mined for Tl in the 1990s. Thallium is also present in coals. Sporadic artisanal mining of coal seams is undertaken by local residents to augment their fuel supply for domestic heating. Thallium mineralization outcrops in the hills, where it is susceptible to weathering and dispersion by natural processes.

2.2. Sampling and analysis

Extensive suites of rocks (46 samples), groundwaters and surface waters (58 samples), soils (38 samples), and edible crops (47 samples) were collected and analyzed using sampling and analytical protocols established by the Geological Survey of Canada (Boyle and Xiao, 2003). Care was taken to include sampling of old mine workings, mineralized zones, and barren host rocks, as well as control samples remote from the influence of mineralization. When sampling vegetables and cereals, a corresponding soil sample was collected for correlation purposes. To further test for natural dispersion processes, another known mineralized area was chosen at Yanshang, a site 6 km from Lanmuchang, which has no known mining tradition. All samples were determined for Tl and other metals by inductively coupled plasma mass spectrometry (ICP-MS) at the Geological Survey of Canada (Ottawa, for rock and water samples), Norwest Laboratories (Edmonton, Canada, for soils), and Activation Laboratories (Ancaster, Ontario, for crops). Detailed information on sampling and analyis procedures has been presented recently (Xiao et al., 2003a,b,c). The minimum detection limit for Tl by the above methods is 0.02 mg/kg in rocks/ores, 0.005 μg/l in waters, 0.2 mg/kg in soils, and 0.05 mg/kg in crop samples. Analytical precision, determined by quality assurance/quality control procedures, using duplicates (at every 10th sample), reagent blanks, internal standards (Rh solution) and international reference samples (Ottawa 96, GBPG-1, OU-6, NBS-1663A, GSR-5, CAL-5, etc.), was better than ± 10%.

3. Results

3.1. Thallium dispersion in the Lanmuchang area

The analytical data for Tl in various geological media from the Lanmuchang area are listed in Table 1.

Table 1
Concentrations of thallium in various sampling media from the Lanmuchang area

	Sampling media	Numbers of samples	Tl concentrations	
			Range	Mean
Rocks/ores (mg/kg)	Sulfide ores	9	100–35 000	4400
	Coals	2	12–46	29
	Secondary minerals	3	25–1100	89
	Mine wastes	3	32–2600	136
	Altered host rocks	6	39–490	124
	Outcropping rocks	6	6–330	39
	Bedrocks in background area	3	0.06–0.2	0.16
Waters (μg/l)	Deep groundwater	5	13–1100	62
	Shallow groundwater	15	0.005–0.75	0.04
	Well water	5	0.01–0.38	0.2
	Background groundwater	2	< 0.005	< 0.005
	Stream water (base-flow regime)	12	0.09–31	1.8
	Stream water (flood-flow regime)	12	0.07–4.5	1.5
Soils (mg/kg)	Soils in mine area	10	40–124	63
	Natural slope wash materials	2	20–28	24
	Alluvial deposited soils	12	14–62	31
	Undisturbed natural soils	5	1.5–6.9	4.2
	Background area	3	< 0.2–0.5	0.3
Crops (mg/kg, DW)	Green cabbage	6	15–495	120
	Carrot	1	22	22
	Chili	3	0.8–5.3	2.9
	Rice	4	1–5.2	1.7
	Chinese cabbage	9	0.87–5.4	1.3
	Corn	8	0.78–3.1	1.3
	Background area	7	0.05–0.35	0.27

Thallium concentrations in the Lanmuchang area range from 100 to 35,000 mg/kg in sulfide ores, and from 12 to 46 mg/kg in coals. Secondary minerals, produced by weathering, contain 25–1100 mg/kg, with 32–2600 mg/kg in mine wastes. Altered host rocks contain 39–490 mg/kg Tl, with 6–330 mg/kg in outcropping rocks. Unaltered country rocks from the background area, barren of metallic mineralization, contain only 0.06–0.2 mg/kg Tl (Xiao et al., 2003b).

The small karstic watershed of Lanmuchang (area about 3 km^2) exhibits an enrichment of Tl level in groundwaters and related stream waters (Xiao et al., 2003a). This affords an excellent demonstration of the natural processes of Tl dispersion, and the resultant impact on the local ecosystem. The concentration of Tl in the water system decreases from deep groundwater to stream water to shallow groundwater. Thallium shows high levels (13–1100 μg/l) in deep groundwater within the Tl-mineralized area, decreasing with distance away from the mineralized area to background levels (< 0.005 μg/l). The occurrence of Tl in the water system is dictated by Tl mineralization, water–rock interactions, and hydrogeological conditions. Thallium concentrations in waters generally correlate with concentrations of total dissolved solids, sulphate, calcium, and pH values. This indicates water–rock interactions driven by weathering of Tl-bearing sulfides, which decreases pH values in groundwater, and by dissolution of limestones enhanced by acid fluids. Thallium in stream water in both the base-flow regime (0.09–31 μg/l) and flood-flow regime (0.07–4.5 μg/l) shows higher concentrations than those found in shallow groundwater (0.005–0.75 μg/l), which serves as the stream's source (mainly springs, dug-well flows, and karstic cave waters). This can be, in part, ascribed to Tl release into the stream through soil–water interaction. Due to dilution effects, concentrations of Tl in stream water in the flood-flow regime (mean value 1.5 μg/l) are generally lower than those in the base-flow regime (mean value 1.8 μg/l). Interestingly, Tl concentrations in stream water in both regimes are remarkably higher (2- to 30-fold) downstream than in upstream and midstream proximity to the Tl-mineralized area (Xiao et al., 2003a). The marked increases of Tl concentration are likely caused by unidentified natural discharges of Tl-rich deep groundwater through fracture zones to the watershed downstream of the mining area.

Thallium levels in soils of the Lanmuchang area range from 40 to 124 mg/kg in soils from the mining area, from 20 to 28 mg/kg in natural slope wash materials, from 14 to 62 mg/kg in alluvial deposits downstream, from 1.5 to 6.9 mg/kg in undisturbed natural soils, and from <0.2 to 0.5 mg/kg in soils from the background area devoid of sulfide mineralization (Table 1). These values demonstrate that the erosion of natural soils from the Tl-mineralized area and the local mining activity are both responsible for the dispersion of high Tl values in soils (Xiao et al., 2003c).

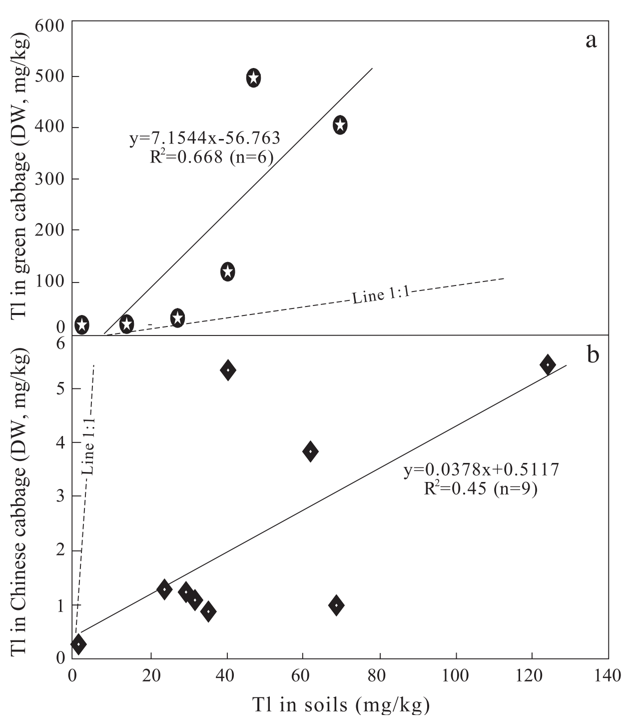

Fig. 2. Concentrations of Tl in (a) green cabbages and (b) Chinese cabbages vs. Tl concentrations in soils.

Table 2
Concentrations of thallium in various sampling media from the Yanshang area

	Sampling media	Numbers of samples	Tl concentrations	
			Range	Mean
Rocks/ores (mg/kg)	Au ores	7	0.22–16	2.5
	Coals	4	0.3–8.4	3.5
	Background area	3	0.06–0.2	0.16
Waters (μg/l)	Groundwater	7	0.006–0.095	0.016
Soils (mg/kg)	Natural soils	6	0.9–1.4	1.13
Crops (mg/kg, DW)	Vegetables and cereals	9	0.07–1.4	0.21

Thallium concentrations in crops are species-dependent (Xiao et al., 2003c). The enrichment of Tl in edible parts of crop species decreases in the following order with regards to mean values: green cabbage>carrot>chili>rice>Chinese cabbage≅corn (Table 1). The highest level of Tl in green cabbage is up to 500 mg/kg (DW), surpassing the values of Tl (14–124 mg/kg) in the soils in which the green cabbages grow. The enrichment factor [i.e., the ratio of metal concentration (DW) in crops to that in soils] for Tl in green cabbage is up to 1–10 (Fig. 2), indicating that green cabbage is a high accumulator for Tl.

3.2. Thallium dispersion in the Yanshang area

The analytical data for Tl in various geological media from the Yanshang area are listed in Table 2.

In the Yanshang area, where the gold orebody is at a depth of 300–400 m, drill core samples from the primary gold zone show a range of Tl levels from 0.22 to 16 mg/kg, with a mean value of 2.5 mg/kg. Natural soils from the area contain 0.9–1.4 mg/kg, groundwater contains 0.006–0.095 μg/l, and edible parts of crops contain a 0.07–1.4 mg/kg Tl (DW). It is important to note that the mean concentration in soils is slightly above the Canadian guideline of 1 mg/kg (CCME, 2003) for arable soils, and edible parts of crops are higher than the worldwide limits of 0.03–0.3 mg/kg (DW) (Table 3).

Table 3
Environmentally safe limits for thallium

	Tl	Sources
Drinking water	2 μg/l	USEPA (2003)
Arable soils	1 mg/kg	CCME (2003)
World land plants	0.008–1.0 mg/kg (DW)	Kabata-Pendias and Pendias (1992)
World edible plants	0.03–0.3 mg/kg (DW)	Kabata-Pendias and Pendias (1992)
World average daily intake	2 μg/day	Sabbioni et al. (1984)
Oral reference dose	0.056 mg/day	RAIS (2003)

4. Discussion

4.1. Environmental implication

The high concentrations of naturally occurring Tl in the Lanmuchang area, as well as lower concentrations in the Yanshang area, show that Tl disperses into waters, soils, and, ultimately, crops at levels that are above the health guidelines set by many countries (Tables 1–3). This dispersion is not necessarily related to mining activities at Yanshang; in contrast to Lanmuchang, it has seen no mining activity. Groundwater-related Tl transfer processes affect the ecosystem through contamination of water supply and arable soil, diffusing up the food chain to pose an undoubted threat to human health.

The endemic studies of Li (1963) and Zhou and Liu (1985) indicated the prevalence of thallotoxicosis in the Lanmuchang area in the period between 1960 and 1970, which was attributed to increased farming in the mined area. However, these studies did not detect the widespread dispersion of Tl beyond the immediate environments of mine workings. Drinking water showed high Tl concentrations, which were thought to be the major agent of Tl poisoning (Zhou and Liu, 1985). Although drinking water with Tl contents below international limits (2 μg/l) in both the well waters (0.01–0.38 μg/l) and the Tl-free piped groundwaters (< 0.005 μg/l) (Tables 1 and 3) is now being supplied, Tl levels in urines of local villagers are still quite high, ranging from 153 to 2668 μg/l (Xiao, unpublished data). These urinary Tl levels are several orders of magnitude higher than the world urinary Tl levels of < 1 μg/l for "nonexposed" humans (Brockhaus et al., 1981; Minoia et al., 1990; CDC, 2003).

Local villagers in the Lanmuchang Tl mineralized area consume the crops growing in the Tl-contaminated soils during the entire year. Chinese cabbage, green cabbage, and chili are often freshly consumed, whereas corn and rice are consumed after air drying. The yield of Tl in vegetables from fresh to dried samples was determined by dividing the dry weight by the fresh weight. Thus, using the values of dry yield, consumption amounts of fresh vegetables, and dry cereals, the amount of Tl from each crop type consumed by the local villagers can be quantitatively estimated. Accordingly, the average daily intake of Tl by villagers of the Lanmuchang area through consumption of locally planted crops has been estimated at 1.9 mg per person (Xiao et al., 2003c). The calculated average human ingestion rate of Tl (given a mean adult body weight of 70 kg) is up to 27 μg/kg/day in the Lanmuchang area, 50 times the ingestion rate of the metal (0.04 mg/day) in people of Tl-free background area. This high ingestion rate of Tl is 1000 times higher than the world average daily intake (2 μg/day) as indicated by Sabbioni et al. (1984), and also far above the element's "oral reference dose" of 0.056 mg/day (RAIS, 2003). This clearly indicates that Tl in the contaminated soils related to natural Tl mineralization is being readily transferred to the human body through the food chain. It represents both a significant threat to the health of the local villagers and a hidden geoenvironmental hazard.

4.2. Geoenvironmental management concern

This study illustrates a real environmental concern related to land use and human health in areas containing high concentrations of Tl in soils associated with the natural occurrence of Tl-rich sulfides and coals, with or without mining activities. Thallium contamination in rocks and soils, and underground and surface drainage patterns should be critical parameters for proper land use and health-related environmental planning and regulation. This research in Guizhou and earlier interesting studies have identified the capacity of certain plants to superaccumulate Tl (green cabbage in the present case), so even a value slightly higher than the accepted Canadian guideline (1 mg/kg) for Tl in arable soils can be a risk factor for such plants, if consumed.

Proper land management practice should not necessarily be restricted to mining activities, and procedures such as the use of mine tailings for terracing should be banned. However, it is also important to note that without proper knowledge of the base-level values of Tl (and also other toxic metals) in rocks, waters, and soils, and their dispersion patterns, unsafe practices can compound the problem of Tl dispersion and increase the risk to the biosphere. Potentially hazardous activities include foundation excavations, agricultural expansion based upon irrigation with waters tainted with Tl, lining of irrigation channels with rocks containing Tl minerals, and artisanal mining of coal seams in terraced hillsides.

Although proper nutrition can offset or retard Tl poisoning in Lanmuchang (Xiao, 2001), it still needs strict monitoring for the geoenvironment to safeguard the health of the population. Simple solutions for immediate implementation include elimination of the planting of green cabbage, a species that hyperaccumulates Tl. Although the scope of this study did not permit an assessment of the human intake of Tl through livestock, it is inevitable that animals fed with local crops and other plants may also transfer Tl to humans.

4.3. Impact beyond the study area

Thallium deposits are rare in nature. However, the occurrence of Tl in a number of hydrothermal ore deposits and alteration zones is a well-established fact and the presence of Tl has been used as tracer for geochemical exploration (Ikramuddin et al., 1983). For instance, hydrothermal precipitates in the Rotokawa geothermal system of New Zealand have Tl levels as high as 5000 mg/kg (Krupp and Seward, 1987). The Allchar Sb–As–Tl deposit in the former Yugoslavia also boasts of an extremely high natural Tl content (Percival and Radtke, 1994), as does the Lengenbach Pb–Zn–As–Ba–Tl in Switzerland (Hofmann and

Knill, 1996). Some Chinese iron sulfide deposits and Pb–Zn deposits also show high Tl levels (Chen et al., 2001).

From the pilot study in the Lanmuchang area (with high baseline values of Tl) and the Yanshang area (with low baseline values of this element), it is clear that Tl can be dispersed beyond a mineralized zone, and its abundances in water, soils, and crops can rise above the permissible levels to a health-threatening extent. Thallium is associated with hydrothermal mineralization in many mining areas of the world, in quantities similar to Lanmuchang or Yanshang, and is concentrated within rocks susceptible to weathering. Soils developed in situ on such rocks would have a higher background of Tl concentration, which, coupled with a favourable hydrological regime, could potentially contaminate the environment and ultimately affect the biosphere. Thallium contamination in soils poses a significant threat to human health due to the high toxicity of the element and its ready assimilation by crops and other plants. This study points to the fact that environmental management related to Tl does not concern industrial contamination alone, and indicates that more attention must be paid to natural processes of contamination. From the current study, it is easy to visualize how a geoenvironment amenable to dispersion of naturally occurring Tl may create environmental health hazards. Without proper knowledge of the background values in an area, this hidden health hazard may become worse if coupled with mining or agricultural activities.

5. Conclusions

Thallium may pose high-potential health risks on humans through the food chain due to both natural processes and human activities. In terms of geoenvironmental concern, proper planning would be far less costly to the well-being of the population. To do so, it is essential that the distribution pattern of Tl be established. The base-level data from bedrocks, waters, and soils will provide guidelines for the safe use of lands, and serve to minimize or prevent cases of Tl poisoning. The present study underlines the fact that ignorance regarding surface geological processes creates health hazards due to improper land use.

Similar contexts, with high primary content of Tl, can be found in other areas of China and the world. Therefore, a thorough geoscientific study must be the primary step in any environmental monitoring in these areas in order to manage land use, whether for mining, agriculture, or habitation. Without establishing specific geoenvironmental guides, Tl poisoning can reach epidemic proportions.

Acknowledgements

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