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Metal pollution and intoxication from artisanal gold mining in Kamituga, Eastern Congo

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Summary (English)

Metal pollution and intoxication from artisanal gold mining in Kamituga, Eastern Congo

This thesis investigates the use of mercury (Hg) in artisanal small-scale gold mining (ASGM), the pollution of aquatic ecosystem by Hg and other metals (Au, Cd, Co, Cr, Cu, Fe, Hg, Mn, Ni, Pb, U, V and Zn), as well as human intoxication by these metals. It is based on research carried out in Kamituga, a mining town in eastern Democratic Republic of Congo (DRC) with about 150,000 inhabitants, 16,000 of which are gold miners. This thesis is built around five chapters.

The first chapter presents an overview of the connections between ASGM and Hg both worldwide and in Congo; the negative outcomes of Hg and other metals on aquatic ecosystems and human health; their dynamics and assessment in aquatics ecosystems as well as the exposure of human to these metals. I had a look at the theses of the others from the group and they usually give a summary of their introduction here; something a like short abstract.

The second chapter assesses perceptions on mercury, making use of qualitative data - individual and focus group interviews - as well as a quantitative survey. In this chapter we found that despite existing laws banning Hg use in ASGM, a lack of enforcement makes these laws ineffective, leading to widespread use of Hg in the mines. However, miners in Kamituga use Hg only on ore concentrates, thereby limiting the level of pollution their activity causes. But at the same time, miners use Hg in residential areas and within watersheds of streams, thus exposing both the communities and aquatic ecosystems. The study also found that people are poorly informed on effects of Hg on environment and health. Finally, the findings include some promising signs as well, since our respondents prioritize environment and health protection more than economic profit from ASGM.

The third chapter analyses samples collected on a tributary of the Congo river (Zalya) and its network. It investigates the concentration of metals in water, sediment, indigenous labeo fish (*Labeo sp.*) captured in selected streams as well as in tilapia (*Oreochromis niloticus*) that were experimentally exposed to these streams' waters for 28 days. It also analyses the bioavailability of these metals from sediments, the relationship between accumulated metals and fish condition and potential risks for humans of water and fish consumption from these streams. We found

that metal concentrations in water were within drinking water quality standards and environmental quality standards. Metal concentration in sediments, however, often exceeded the TEC (threshold effect concentrations) and sometimes even exceeded the PEC (probable effect concentration) in the case of Hg, Cr and Ni. This is potentially endangering aquatic life, since these metals can be released to benthic organisms and to the overlaying water. Indigenous fish from mining-affected streams had higher metal concentration in their muscle compared to fish from non-affected streams. For experimentally exposed fish, an overall high mortality was observed but no significant differences were found between sites, in terms of metal accumulation concentrations in surviving fish after 28-day of exposure. Fish condition was negatively related to elevated Hg, Pb, Ni and Zn concentrations in local fish muscle and with Cd concentration for experimentally exposed fish, but it was not related to the total metal load of either species. Fish consumers may be at risk of Hg, Cd and Cr intoxication if their daily consumption exceeds 77, 145 g and 138 g of fish respectively, for an average 60 kg person.

The fourth chapter assesses whether dietary and occupational exposure of miners and non-miners lead to metal intoxications and how metal intoxication is reflected in the health symptoms people experience. The chapter analyses the diet, health status and symptoms relatable to metal poisoning of miners and non-miners and compared to metal levels in their blood, urine, nails and hair. It shows that more than 80% of the participants had blood Hg above reference values, and nearly 25% had alert or high values. It was found that many people also had Hg levels in their urine, nail and hair as well as concentration of other metals in different tissues above normal range. Miners had higher nail Hg than the rest of the community; while blood, urine and hair had similar Hg levels for miners and non-miners. Prevalence of potential Hg poisoning was equally spread in the community disregarding occupations, gender, age and education, but much higher for people with fish-rich diet. Indeed, tilapia being farmed in waters exposed to metal pollution, increased prevalence of potential Hg poisoning on its consumers. Most symptoms were not correlated to higher Hg levels. Neither was the total number of symptoms correlated to higher levels of one of the assessed metals. Symptoms were correlated to other factors such as age and undernourishment.

This thesis recommends, in its fifth and final chapter, raising awareness in the community about the ongoing dietary and occupational exposure, risks of mercury and quantities of fish that are safe to consume from polluted streams. It further recommends increasing formalization of the ASGM sector to reduce Hg use. Furthermore, it suggests increasing the capacity of law-enforcement agencies to enable them to supervise Hg use in mines. It finally recommends the monitoring of potentially polluted streams and sharing the results with communities and local medical personnel.

Samenvatting (Nederlands)

Metaalverontreiniging en intoxicatie door artisanale goudwinning in Kamituga, Oost-Congo

Dit proefschrift onderzoekt het gebruik van kwik (Hg) in artisanale kleinschalige goudwinning (ASGM - *Artisanal and small-scale gold mining*), de verontreiniging van aquatische ecosystemen door Hg en andere metalen (Au, Cd, Co, Cr, Cu, Fe, Hg, Mn, Ni, Pb, U, V and Zn), evenals menselijke intoxicatie door deze metalen. Het omvat onderzoek dat is uitgevoerd in Kamituga, een mijnstadje in het oosten van de Democratische Republiek Congo (DRC) met ongeveer 150.000 inwoners, waarvan 16.000 goudzoekers. Dit proefschrift omvat vijf hoofdstukken.

Het eerste hoofdstuk geeft een overzicht van de relatie tussen ASGM en Hg, zowel wereldwijd als in Congo. In dit hoofdstuk werden eveneens de negatieve effecten van Hg en andere metalen voor aquatische ecosystemen en de menselijke gezondheid, hun dynamiek en distributie in aquatische ecosystemen, evenals de blootstelling van de mens aan deze metalen onderzocht.

In het tweede hoofdstuk werd de perceptie van het gebruik en de risico’s van kwik beoordeeld, gebruik makend van kwalitatieve gegevens - individuele en doelgroep interviews - en een kwantitatief onderzoek. Ons onderzoek toonde aan dat de uitvoering en handhaving van de bestaande wetten, die het gebruik van Hg in ASGM verbieden, gebrekkig is, wat deze wetten ondoeltreffend maakt en leidt tot een wijdverbreid gebruik van Hg in de mijnen. Mijnwerkers in Kamituga gebruiken Hg echter alleen op concentraten van goudertsen, waardoor de vervuiling die hun activiteit veroorzaakt, wordt beperkt. Maar tegelijkertijd gebruiken mijnwerkers Hg in woongebieden en binnen stroomgebieden van waterlopen, waardoor zowel de gemeenschappen als de aquatische ecosystemen worden blootgesteld. Uit het onderzoek bleek ook dat mensen slecht geïnformeerd zijn over de effecten van kwik op het milieu en hun gezondheid. Ten slotte bevatten de bevindingen ook enkele veelbelovende signalen, aangezien onze respondenten milieu- en gezondheidsbescherming meer prioriteit geven dan economische winst van ASGM.

Het derde hoofdstuk onderzoekt de concentratie van metalen in water, sediment, inheemse labeo-vissen (*Labeo sp.*) gevangen in geselecteerde beekjes op een zijrivier van de Congo-rivier (Zalya) en zijn netwerk. Bovendien werden metaalconcentraties onderzocht in tilapia (*Oreochromis niloticus*), die 28 dagen experimenteel werden blootgesteld aan het water van deze waterlopen. Daarnaast werd de biologische beschikbaarheid van deze metalen uit

sedimenten, de relatie tussen geaccumuleerde metalen en de toestand van vissen, en het mogelijke risico voor mensen bij water- en visconsumptie afkomstig uit deze waterlopen geanalyseerd. We ontdekten dat de metaalconcentraties in het water binnen de drinkwater kwaliteitsnormen en milieukwaliteitsnormen lagen. De metaalconcentraties in sedimenten overschreden echter vaak de TEC- (Threshold Effect Concentrations; drempelwaarde) en soms zelfs de PEC- (Probable Effect Concentration; waarschijnlijke effect-concentratie) waarde in het geval van Hg, Cr en Ni. Dit brengt mogelijk het aquatische leven in gevaar, aangezien deze metalen kunnen worden afgegeven aan benthische organismen en aan het water. Inheemse vissen uit, door de mijnbouw beïnvloede waterlopen hadden een hogere metaalconcentratie in hun spierweefsel in vergelijking met vissen uit niet-beïnvloede waterlopen. Voor experimenteel blootgestelde vissen werd een hoge mortaliteit waargenomen, maar er werden geen significante verschillen gevonden tussen locaties in termen van metaalconcentraties geaccumuleerd in de overlevende vissen na 28 dagen blootstelling. De conditie van de vissen was negatief gerelateerd aan de concentraties van Hg, Cd, Pb, Ni en Zn in het spierweefsel van de vissen, maar niet aan de totale metaalbelasting. Mensen lopen mogelijk risico op Hg-, Cd- en Cr-vergiftiging als hun dagelijkse visconsumptie hoger is dan respectievelijk 77, 145 en 138 g vis, voor een persoon van gemiddeld 60 kg.

In het vierde hoofdstuk wordt nagegaan of de voeding en beroepsmatige blootstelling van mijnwerkers en niet-mijnwerkers tot metaalvergiftiging kan leiden en hoe metaalvergiftiging wordt weerspiegeld in de gezondheidssymptomen die mensen ervaren. Dit hoofdstuk analyseert het dieet, de gezondheidstoestand en de symptomen, die verband houden met metaalvergiftiging van mijnwerkers en niet-mijnwerkers en vergelijkt deze met het metaalgehalte in hun bloed, urine, nagels en haar. Het toont aan dat bij meer dan 80% van de deelnemers de bloed-Hg concentraties boven de referentiewaarden lagen. Bijna 25% had een alerte of sterk verhoogde bloed concentratie. Er werd vastgesteld dat veel mensen ook verhoogde Hg concentraties in hun urine, nagels en haar hadden, evenals verhoogde concentraties van andere metalen in verschillende weefsels. De Hg concentraties in nagels van de mijnwerkers was hoger dan bij de rest van de gemeenschap; terwijl bloed, urine en haar vergelijkbare Hg-concentraties hadden voor mijnwerkers en niet-mijnwerkers. De prevalentie van mogelijke Hg-vergiftiging was gelijkmatig verspreid in de gemeenschap, ongeacht beroep, geslacht, leeftijd en opleiding, maar veel hoger voor mensen met een visrijk dieet. Inderdaad, consumptie van tilapia, die wordt gekweekt in wateren blootgesteld aan metaalverontreiniging, verhoogde de prevalentie van potentiële Hg-vergiftiging bij de consumenten. De meeste symptomen waren niet gecorreleerd aan een verhoogde Hg concentratie. Evenmin was het totale aantal symptomen een weerspiegeling van hogere concentraties van een van de andere onderzochte metalen. De symptomen waren waarschijnlijk te wijten aan andere factoren, zoals leeftijd en ondervoeding.

Op basis van dit proefschrift wordt in het vijfde en laatste hoofdstuk aangeraden om het bewustmaking, over de risico's van Hg en de voortdurende blootstelling aan Hg door voedsel of beroepsmatige activiteiten, in de gemeenschap te verhogen om zo het algemene gebruik van Hg te verminderen en een veilige visconsumptie te realiseren. Verder wordt er aanbevolen om de formalisering van de ASGM-sector te vergroten om het gebruik van Hg te verminderen. Er wordt voorgesteld om de capaciteit van wetshandhavingsinstanties te vergroten om hen in staat te stellen toezicht te houden op het gebruik van Hg in de mijnen. Ten slotte wordt aanbevolen n om waterlopen die mogelijk verontreinigd zijn te monitoren en de resultaten te delen met gemeenschappen en lokaal medisch personeel.

Résumé (Français)

Pollution et intoxication métalliques par l’extraction artisanale d’or à Kamituga, à l’Est de la République Démocratique du Congo

Cette thèse étudie l'utilisation du mercure (Hg) dans l'extraction artisanale de l'or à petite échelle (ASGM – *Artisanal and small-scale gold mining*), la pollution des écosystèmes aquatiques par le Hg et d'autres métaux, ainsi que l'intoxication humaine par ces métaux. Elle est basé sur des recherches menées à Kamituga, une ville minière de l'est de la République démocratique du Congo (RDC) comptant environ 150 000 habitants, dont 16 000 creuseurs d'or. Cette thèse est construite autour de cinq chapitres.

Le premier chapitre présente un aperçu des liens entre l'ASGM et le Hg à la fois dans le monde et au Congo; les effets négatifs du mercure et autres métaux sur les écosystèmes aquatiques et la santé humaine; leur dynamique et leur évaluation dans les écosystèmes aquatiques ainsi que l'exposition de l'homme à ces métaux.

Le deuxième chapitre évalue les perceptions locales sur le Hg, en utilisant des données qualitatives - entretiens individuels et de groupes de discussion - ainsi qu'une enquête quantitative. Dans ce chapitre, nous avons constaté que malgré les lois existantes interdisant l'utilisation du Hg dans l'ASGM, le manque d'application de ces lois les rend inefficaces, conduisant à une utilisation généralisée du Hg dans les mines. Cependant, les creuseurs de Kamituga n'utilisent le Hg que sur les concentrés, ce qui réduit considérablement le niveau de pollution que cause leur activité. Cependant, les creuseurs utilisent du Hg dans/près de leurs résidences et dans les bassins versants des cours d'eau, exposant ainsi à la fois les communautés et les écosystèmes aquatiques. L'étude a également révélé que les gens sont mal informés des effets du mercure sur l'environnement et la santé. Enfin, les résultats incluent également des signes prometteurs, puisque nos répondants accordent la priorité à la protection de l'environnement et de la santé plus qu'au profit économique de l'ASGM.

Le troisième chapitre analyse des échantillons prélevés sur un affluent du fleuve Congo (Zalya) et son réseau. Ce chapitre étudie la concentration de métaux (Hg, Cd, Co, Cr, Cu, Fe, Mn, Ni, Pb, U, V, Zn et Au) dans l'eau, les sédiments, chez les poissons labeo indigènes (*Labeo sp*) capturés dans les rivières sélectionnées ainsi que dans le tilapia (*Oreochromis niloticus*) qui ont été exposés expérimentalement aux eaux de ces rivières pendant 28 jours. Il analyse également la biodisponibilité de ces métaux à partir des sédiments, la relation entre les métaux

accumulés et l'état des poissons et le risque potentiel pour les humains de la consommation d'eau et de poisson de ces cours d'eau. Nous avons constaté que les concentrations de métaux dans l'eau respectaient les normes de qualité de l'eau potable et les normes de qualité environnementale. Cependant, la concentration de métaux dans les sédiments dépassait souvent la TEC (Threshold Effect Concentrations ; seuil des concentrations à effet) et dépassait parfois même la PEC (Probable Effect Concentration; concentration à effets probable) dans le cas du Hg, Cr et Ni. Cela met potentiellement en danger la vie aquatique puisque ces métaux peuvent être rejetés dans les organismes benthiques et dans l'eau de recouvrement. Les poissons indigènes des rivières affectés par l'exploitation minière avaient une concentration de métaux plus élevée dans leurs muscles que les poissons des rivières non affectées. Pour les poissons exposés expérimentalement, une mortalité élevée a été observée, mais des différences significatives n’ont pas été observées entre les sites, en termes de concentrations d'accumulation de métaux chez les poissons survivants après 28 jours d'exposition. L'état du poisson (CF) était négativement lié aux niveaux de Hg, Pb, Ni et Zn dans les muscles des poissons locaux et Cd pour les poissons exposés expérimentalement, mais pas à la charge totale de métal. Les consommateurs de poisson locaux peuvent être exposés à un risque d'intoxication au Hg, au Cd et au Cr si leur consommation quotidienne dépasse respectivement 77, 145 et 138 g de poisson, pour une personne moyenne pesant 60 kg.

Le quatrième chapitre évalue si l'exposition alimentaire et professionnelle des creuseurs et autres membres de la communauté conduit à des intoxications aux métaux et comment l'intoxication aux métaux se reflète dans les symptômes de santé des personnes. Ce chapitre analyse le régime alimentaire, l'état de santé et les symptômes liés à l'empoisonnement aux métaux des mineurs et des non-mineurs et comparé aux niveaux de métaux dans leur sang, leur urine, leurs ongles et leurs cheveux. Il a montré que plus de 80% des personnes avaient un Hg dans le sang supérieur aux valeurs de référence, et près de 25% avaient des valeurs d'alerte ou élevées. Il a été constaté que de nombreuses personnes avaient également des niveaux de Hg dans leur urine, leurs ongles et leurs cheveux ainsi qu'une concentration d'autres métaux dans différents tissus au-dessus de la normale. Les creuseurs avaient dans les ongles un niveau de Hg plus élevés que le reste de la communauté ; tandis que le sang, l'urine et les cheveux, il y avait des niveaux de Hg similaires pour les creuseurs et autres membres de la communauté. La prévalence de l'intoxication potentielle au Hg était également répandue dans la communauté sans tenir compte des professions, du sexe, de l'âge et de l'éducation, mais beaucoup plus élevée pour les personnes ayant un régime alimentaire riche en poissons. En effet, le tilapia étant élevé dans des eaux exposées à la pollution par les métaux, une prévalence accrue de l'intoxication potentielle au Hg chez ses consommateurs. La plupart des symptômes ne reflétaient pas un niveau plus élevé de Hg chez les personnes qui en souffraient. Le nombre total de symptômes ne reflétait pas non plus des niveaux plus élevés de l'un des métaux évalués. Les symptômes étaient probablement dus à d'autres facteurs tels que l'âge et la sous-alimentation.

Cette thèse recommande, dans son cinquième et dernier chapitre, de sensibiliser la communauté à l'exposition alimentaire et professionnelle en cours, aux risques de mercure et aux quantités de poisson pouvant être consommées sans danger dans les cours d'eau pollués. Il recommande en outre d'accroître la formalisation du secteur de l'ASGM pour réduire l'utilisation de Hg. Il suggère d'augmenter la capacité des organismes d'application de la loi pour leur permettre de superviser l'utilisation du mercure dans la mine. Il recommande enfin de surveiller les cours d'eau potentiellement pollués et de partager les résultats avec les communautés et le personnel médical local.

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Chapter 1 : Introduction

1.1. Gold and mercury: the bright and the ugly

For millennia gold has been the embodiment of preciousness and been used as currency or guarantee for paper printed currencies. Golden decorations of tombs in Egypt can be dated around 1800-1550 BC (Troalen et al, 2014), with its mining in the Nubia and Egypt kingdoms since 3000 BC (Klemm *et al.*, 2001). Currently, gold production continues both at industrial and artisanal scale. While industrial gold production has the highest production, artisanal and small-scale gold mining (ASGM) still produces 500-800 tons of gold annually (20-30% global gold production), spread across nearly 70 countries. It provides in many developing nations a primary or an additional source of income, in rural economies where economic alternatives to agriculture are limited. There is an estimate of 10-19 million people mining gold, with around 50 million more people working in ASGM and supporting 80-100 million people (Esdaile and Chalker, 2018; Zolnikov and Ortiz, 2018; Veiga *et al.*, 2006). Despite, all available mining technologies today, in ASGM, there is usually no prior geological exploration or proven reserves, no estimate of ore tonnage output. The major driving force in ASGM is survival of miners and their families or supplementing their other (often seasonal) meagre sources of income (Veiga *et al.*, 2006).

Like in other economic structures, miners aim for maximum profit with minimum investment. Thus, despite being illegal in many countries, mercury (Hg) amalgamation remains the preferred method for ASGM miners to extract fine gold (Esdaile and Chalker, 2018; Veiga *et al.*, 2006). Mercury has actually been closely associated with gold mining since the antiquity, with high soil, air, biota and sediment Hg contaminations dating in the 5th century BC (Wiener *et al.*, 2003). Gold miners use Hg, since at ambient temperature its elemental form (zero-oxidation state Hg^0) is liquid (from -39 to 357°C) and able to form an amalgam with gold, then be easily evaporated from the amalgam at temperatures (357°C) where gold is still solid (Clarkson, 1997, Schroeder and Munthe, 1998). Mercury is very easy to use for gold miners since it does not form an alloy with gold, but undergoes a phenomenon of deep sorption, involving some interpenetration of the two elements. The sorption of Hg on gold particle is due to Hg's surface tension that is greater than that of water and less than that of gold. Gold then sinks in the Hg leaving its lighter gangue material floating overtop (Veiga *et al.*, 2006). There are two typical methods of amalgamations used in ASGM: whole ore amalgamation in which Hg is mixed with whole ores without any concentration; and amalgamation of gravity concentrates in which miners first concentrate their ore, dispose-off the tailings and apply mercury only to the concentrate. In both cases, the amalgam is then heated, vaporizing the Hg into the atmosphere while isolating the gold (Esdaile and Chalker, 2018; Veiga *et al.*, 2006). The difference between the effects of these two methods lies in the total amount of Hg each method loses outside of the heating process. Indeed, aside from the Hg being released to the atmosphere when heating the amalgam (35% of global ASGM Hg emissions), the remaining ore (tailing) is usually disposed-off while still containing important quantities of Hg, most of which ending in water systems (65% of global ASGM Hg emissions). Whole ore amalgamation loses as high as 100 g of Hg per g of gold produced while amalgamation of concentrates loses as low 1g of Hg per g of gold produced. It is estimated that 650 to 1350 tons of Hg are released to the environment from ASGM worldwide (of which China releases 200-250 tons, Indonesia 100-150 tons/year and most other countries releasing less than 30 tons/year) (Esdaile and Chalker, 2018; Telmer and Veiga, 2009; Veiga *et al.*, 2006).

However, once in aquatic ecosystems (figure 1.1), Hg oxidises into Hg^{2+} and makes inorganic and organic combinations (Clarkson and Magos, 2006), including methylmercury (CH_3Hg^+), a highly toxic molecule able to bio-concentrate (bioaccumulate and biomagnify) up to more than a million folds, affect almost any organ of the human body (Kim *et al.*, 2015; Schroeder and Munthe, 1998). A case of Hg release, for example, occurred in the 1950s and 1960s in Minamata and Niigata (Japan), poisoning thousands of people (Ministry of Environment – Government of

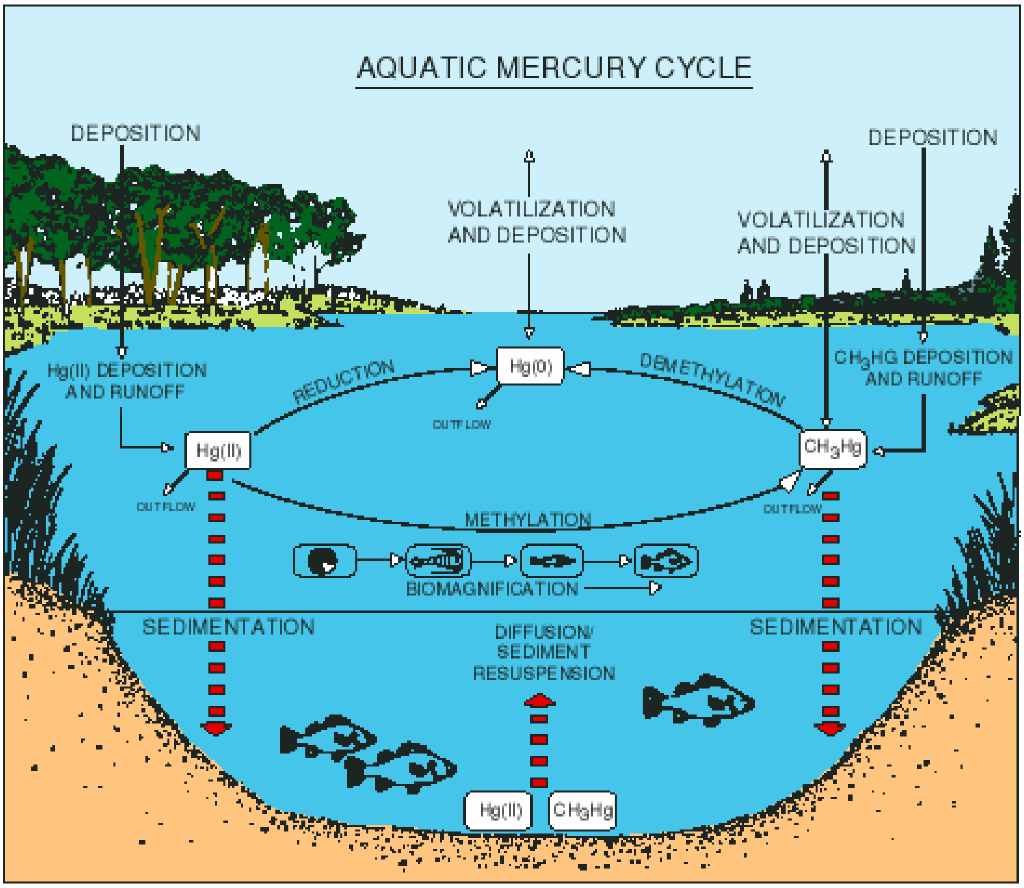

Figure 1.1 : Mercury biogeochemical cycle pathways (www.usgs.gov)

Japan, 2002). In 2013, the "Minamata Convention", a global treaty aimed at reducing and if possible, eliminating Hg pollution and named after the above-mentioned incident was initiated (UNEP, 2019a). As of 2020, the convention has been signed by 128 countries and ratified by 124 of them (www.minamataconvention.com). The convention is implemented by [1]UNEP, UNDP and UNIDO with funding from the GEF, and executed in each country by national agencies of environment with the support of UNITAR, AGC or other non-governmental agencies. The implementation includes a national action plan (NAP) to reduce or eliminate Hg use. This should be paired with broader efforts to better organize and assist the ASGM sector so as to render the reduction of Hg feasible and sustainable. Despite all this, 2000 to 3000 tons of mercury are still emitted into the atmosphere every year, with ASGM representing 37.7% of the global emission and 80% of Sub-Saharan Africa's emission (UNEP, 2019b).

Few studies have tried to understand why, despite everything we know about mercury's detrimental effects, miners and communities continue to use it, even though alternatives ways of extraction are possible (such as centralized processing centres, techniques to reduce mercury release and alternative techniques to mercury amalgamation (Zolnikov and Ortiz, 2018)).

Our study analyses why Hg use and other polluting techniques are widespread despite both laws and risks. Moreover, we assess to what extent mining communities are aware of the risks they face from ASGM-induced pollution, but also how they perceive the trade-off between ASGM income on which they rely for survival and its environmental and health impact from which their community are likely to suffer. Understanding this may increase the chances for a successful implementation of the Minamata convention (or similar initiatives) through consideration of the local community's perspective, instead of enforcing top-down decisions that do not align to the community's vision of the problem and its priorities in addressing the problem.

1.2. Congo's gold mining and mercury

Although some traces of precolonial mining have been found in the Democratic Republic of the Congo (DRC), gold mining properly started in the early 20th century during DRC's colonization by Belgium, with European prospectors discovering gold deposits in the Eastern part of the country, and Belgian colonial investors starting to invest in a (semi)industrial

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[1] UNEP (UN Environment Program), UNDP (UN Development Program), UNIDO (UN Industrial Development Organization), GEF (Global Environment Funds), UNITAR (UN Institute of Training and Research), AGC (Artisanal Gold Council)

exploitation. During the colonial period, gold mining concentrated in Kivu province around the Kamituga mine, and in Oriental Province around the Kilo Moto mines. After the independence, gold mining continued with foreign, largely Belgian, capital, while many important mining companies were nationalized by President Mobutu from 1973 onwards. Fluctuating commodity prices combined with bad management and deteriorating infrastructure produced a deep crisis from the 1970s onwards. In 1982, President Mobutu 'liberalized' gold mining (outside industrial concessions) in an attempt to curb the effects of this economic crisis, which provoked a major rush to the mining sites, and incited the population to 'fend for itself' in an expanding informal economy. In the 1990s industrial production came to a complete standstill, and informal artisanal mining boomed. Between 1996 and 2003, it became enmeshed in a regional war economy that heavily depended on exploitation and trade of mineral resources. A new Mining Code adopted in 2002 (RDC, 2002) promoted foreign direct investment; and many concessions were taken over by multinational mining corporations. These mining corporations played an important role in the country's economy. In 2005, the corporation that had acquired most North-Eastern Congo's mining rights, AngloGold Ashanti, reported gold income representing 37% of the country's GDP (Prosansky, 2007; Geenen et al, 2013). However, alongside those companies, artisanal miners are also extracting gold all over the country (Geenen, 2015). Due to their informal status estimates of the total ASGM population or gold production are difficult to make. Steckling *et al.* (2017) estimates DRC having one of the highest ASGM population in the world (but acknowledges a large variation in estimates of the ASGM populations from 0.6 to 2.9 million). Despite the large body of evidence on Hg pollution from ASGM (Moreno-Brush *et al.*, 2019), very little has been done in the DRC, which is one of the largest ASGM spots on the planet. This limited research on environmental impact of ASM in DRC was most likely due to security concerns during the First and Second Congo War (1996-1997 and 1998-2003), the remaining conflicts that occur in many areas where mining is ongoing (Butsic *et al.*, 2015) and the larger attention awarded to socio-economic problems of the ASM sectors, including the connections between ASM and ongoing conflicts (Nkuba *et al.*, 2019). The DRC is however involved in the international effort to reduce/eliminate Hg use. Though it has not yet ratified the Minamata convention, it has been actively involved in the establishment of the Convention. It has conducted a baseline study on mercury use (Nkuba *et al.*, 2018) and put in place a National Action Plan (NAP) to reduce this mercury usage and to develop and formalize the ASGM sector (Nkuba et al, 2020). Nkuba *et al.* (2018) found in the five provinces producing 80% of the country's gold (Ituri, South-Kivu, Haut-Uele, Tanganyika and Tshopo), that miners use mercury in 25-30% of their production, releasing more than 3 tons of mercury in the environment per year. The large involvement in ASGM all over the country combined to the widespread use of mercury leads to an estimated total of 52,320 to 366,841 years lived with disabilities (YLD) from Hg intoxication (Steckling *et al.*, 2017). The NAP (Nkuba *et al.*, 2020) thus includes activities to (1) eliminate worst mercury use practices, (2) promote practices using less or no mercury, (3) facilitate formalization and regulation of ASGM, (4) manage mercury trade, (5) capacity-building of miners and informing affected communities, (6) collaborate with stakeholders in implementation, (7) improve public health in ASGM areas and (8) protect vulnerable people (children and women in child-bearing age).

Current industrial gold extraction in DRC as well as ASGM, like in the colonial period, revolves around two major hotspots: the East (formerly Kivu Province) and the North-East (formerly Oriental Province); and in both regions mercury use is widespread (Nkuba *et al.*, 2018). Our study focuses on Kamituga, South Kivu's largest gold mining town, built as a colonial mining town starting in the 1920s. The company MGL (Minière des Grands Lacs) started industrial operations here in the 1930s and was present until 1976. In 1976, several mining companies in the region merged into a new company, SOMINKI (Société Minière et Industrielle du Kivu, which continued exploiting until 1996. In the 1980s, artisanal mining boomed in the region becoming a pillar of the local economy (Geenen, 2015). The town currently has 150,000 inhabitants (based on the 2019 Kamituga Health Zone report), of which about 16,000 are miners (Nkuba *et al.*, 2017) and the rest of the population depending directly or indirectly on ASGM for their livelihood. Indeed, in 2010, after ban of ASGM activities, cases of school drop-outs, malnutrition, disease spread with no treatment due to lack of means to afford health cost, migration of people back to their place of birth, etc. (Geenen, 2012). Hg is commonly used there for gold processing, despite the origin of the ore (see figure 1.2). Indeed, in Kamituga there four ore extraction practices : mountain underground, riverbeds, mountains eroded soils or reprocessing of soils from shops and other place where miners operate. After extraction all ores go through a gravity concentration in primary or secondary processing pits (locally called *lutra* or *domaines*), before they can be treated with Hg. Though it's mostly used on concentrates rather than on whole ores, Hg is still a threat since it is used in/near people's homes and in the watershed of a tributary of the biodiversity-rich Congo river (Nkuba *et al.*, 2017). Nkuba *et al.* (2017) also found that the country has many laws forbidding mercury use in ASGM, but several institutional challenges hinders the ability of governmental structures to implement these laws. Lassen *et al.* (2016), analysing Sub-Saharan Africa's mercury trade, noticed that in the DRC mercury is easily accessed in gold mines since it is smuggled in from neighbouring eastern countries (Uganda, Rwanda, Burundi, Kenya and Tanzania) (see figures 1.3a and 1.3b), through the same channels that are used to smuggle gold out; and is usually given to miners on credit by gold traders as an incentive for them to later sell the gold they extract to the traders the owe money for mercury. Most of the gold is smuggled out of the country through illegal trade. In 2001, artisanal miners in North-Eastern Congo, who smuggle their gold production to Uganda, where they can find better prices, raised

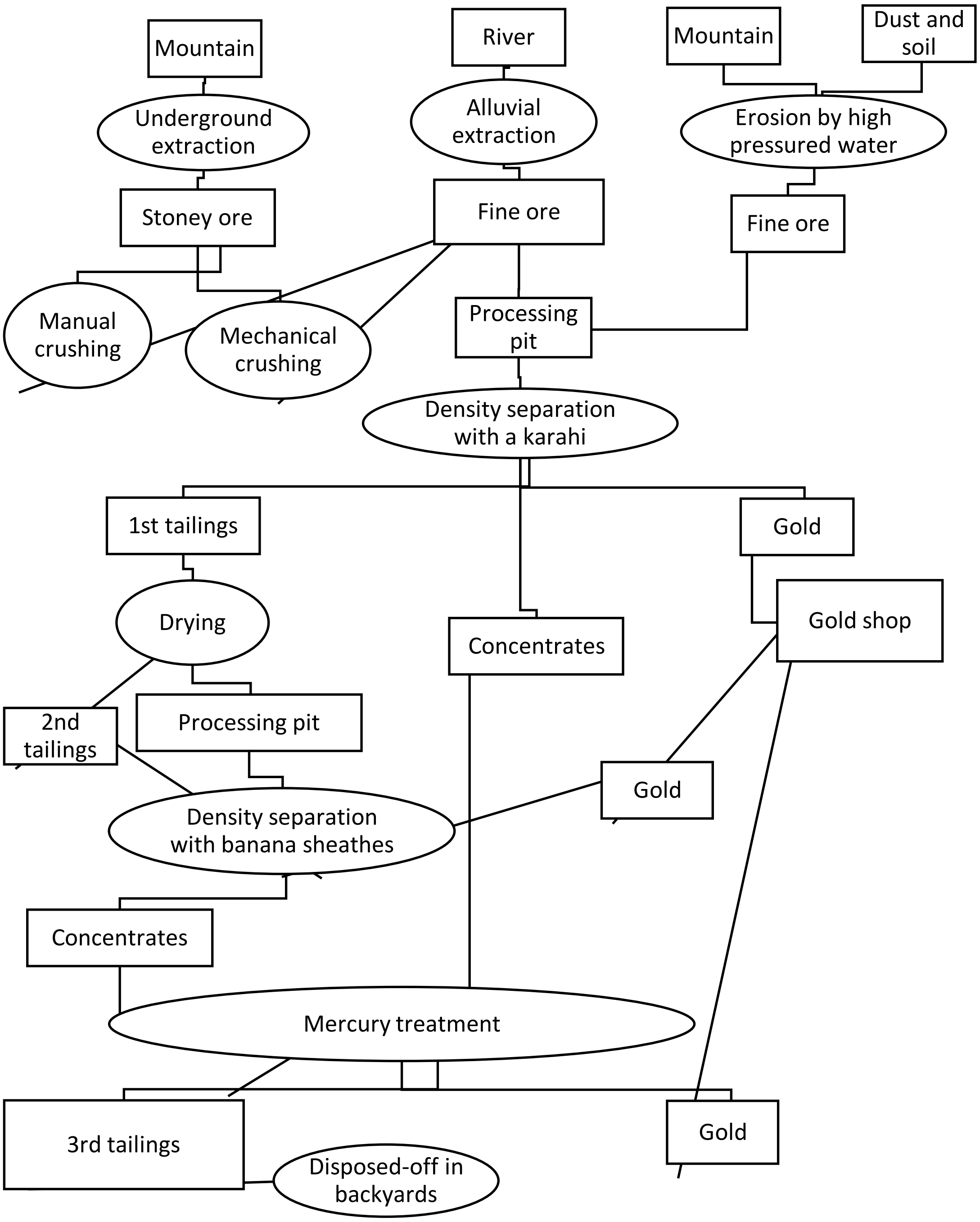

Figure 1.2: Gold extraction and processing, and mercury use in Kamituga (Nkuba et al., 2017)

Ugandan gold exportation from less than one kilogram of national production to more than six tons exported that year (Veiga *et al.*, 2006).

With around 12 tons of gold produced (nearly 600 million USD based on the average gold price between 2010 and 2020 (https://www.gold.org)), ASGM currently employs most of the 0.8-2 million ASM actors who support 10 million Congolese (16% of the country's population) (Nkuba *et al.*, 2018; Nkuba *et al.*, 2017; World Bank, 2008). Both the ease of trade of Hg in ASGM mines and the institutional incapacity to fight its use, increase Hg use and likelihood of mercury pollution in the area. We consider, in such conditions, the community to be an even more important player, and its perspective becoming paramount to the effort to promote mercury-free techniques and other practices that reduces pollution.

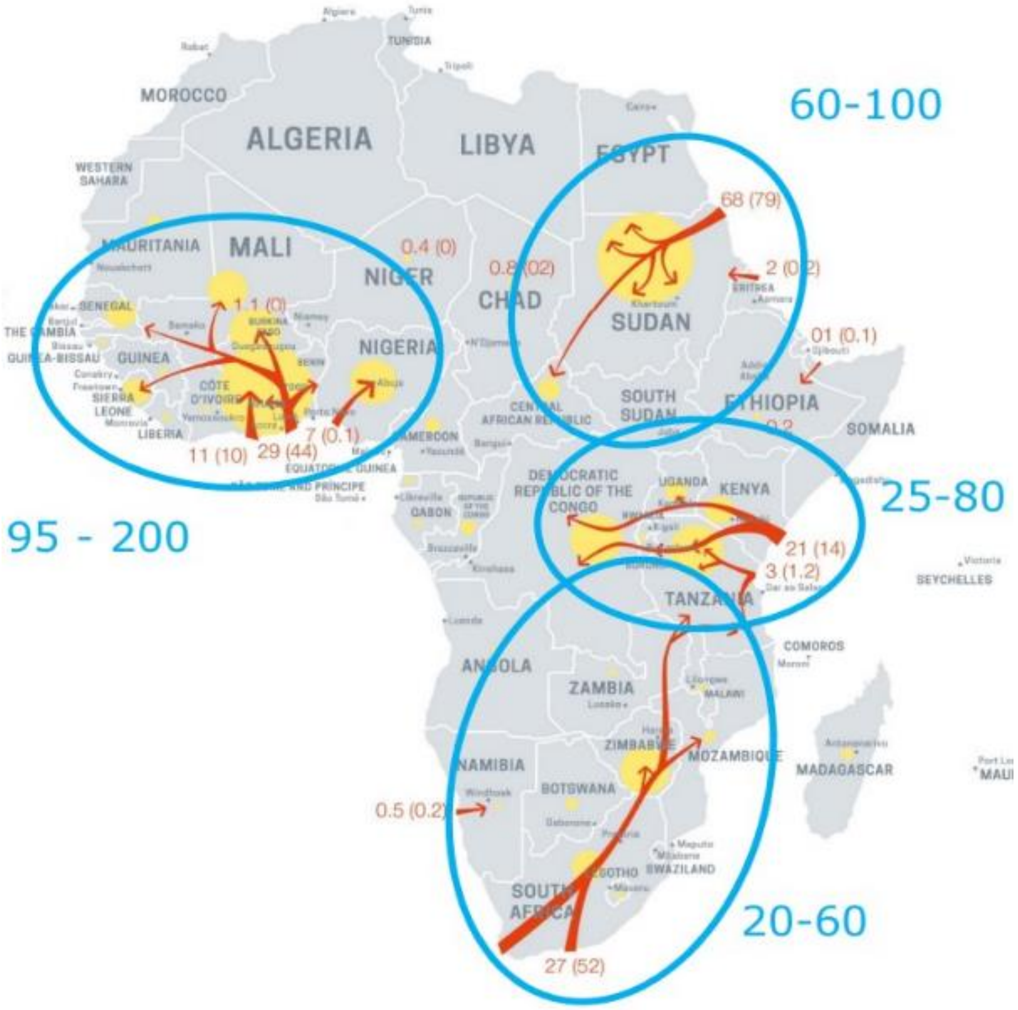

Figure 1.3a. African major trade route (red lines) for Hg (Lassen et al., 2016)

1.3. Metals: essential and non-essential

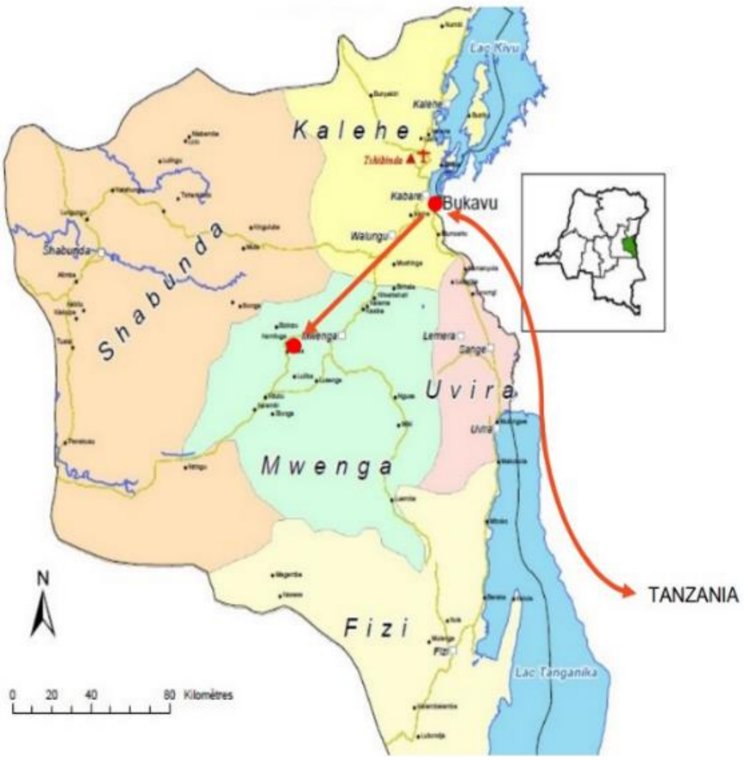

Figure 1.3b. Major trade route (red lines) for gold and Hg from (and to) Kamituga (Lassen et al., 2016)

Metals exist naturally on earth from its inner core to the atmosphere, but mining causes perturbations to equilibrium and cycling of these metals in terrestrial and aquatic ecosystems (Deb and Fukushima, 1999; Allan, 1997; Thorton, 1996). Among the 35 naturally existing metals and metalloids, 23 are known for their direct or indirect adverse effects on ecosystems and organisms: cadmium (Cd), cobalt (Co), chrome (Cr), copper (Cu), iron (Fe), gold (Au), manganese (Mn), Hg, lead (Pb), nickel (Ni), uranium (U), vanadium (V), zinc (Zn) and others (Engwa *et al.*, 2019).

They can be categorized in two groups: the first group includes metals (Co, Cr, Cu, Fe, Mn, Mo, Ni, Zn, magnesium and selenium) that are essential for various physiological and biochemical activities in organisms, including oxidative phosphorylation, gene regulation and free-radical homeostasis (Jakimska *et al.*, 2011; Deb and Fukushima, 1999; WHO, FAO, IAEA, 1996). These metals can result in deficiency diseases or syndromes when present at very low doses. And when in excessive quantities, they can be damageable to the organism. It is thus crucial for this group to keep concentrations within the narrow window in which they are neither deficient nor excessive, but rather essential to organisms. Many biological systems naturally exist already on the margin of either toxicity or deficiency of metals. Mining and other human activities (industries, agriculture, wastewater, smelting, etc.) causing physical and chemical redistribution of these metals usually disrupt these equilibria (Masindi and Mwedi, 2018; Jakimska *et al.*, 2011; Deb and Fukushima, 1999; WHO, FAO, IAEA, 1996; Luoma, 1883). The second group includes metals (Cd, Hg, Pb, Cr, arsenic and silver), that play no known beneficial role to organisms and can have delirious effects at very low doses (Jakimska *et al.*, 2011; Deb and Fukushima, 1999; WHO, FAO, IAEA, 1996). Metals are known to not degrade and have long half-lives and to bioaccumulate in living tissues (Jakimska *et al.*, 2011). But also, when they are oxidized (Zn^{2+}, Cd^{2+}, Pb^{2+}, Hg^{2+}, etc.), metals can readily bind to biological molecules such as proteins and enzymes to form stable and strong bonds, destroying them and disrupt their cellular function. These reactions damage blood constituent, lungs, liver, kidney and other vital organs promoting several disease conditions (Jaishankar *et al.*, 2014). They may also damage nucleic acids, cause mutations, mimic hormones and eventually lead to cancer (Engwa *et al.*, 2019).

Toxicity can be attributed to dysfunction of cellular functions that are critical to metal uptake, regulation, utilization and accumulation. This dysfunction results in interactions with inappropriate cellular structures. The process through which metals accumulate in tissue varies from one species to another. In fish, metal uptake occurs via gills, digestive track and integument, from where they are transported by blood and hemolymph toward internal organs for utilization, storage and release (Deb and Fukushima, 1999). Many organisms produce metallothioneins, cysteine-rich low-molecular-mass metal-binding proteins and use them to regulate essential metals thanks to their cysteine residues and detoxify both essential and non-essential metals (Cd, Ag, Cu, Hg and Zn). Metallothioneins are present in the small intestine, liver and gills of fish. Liver and kidney are the organs most involved in detoxification, long term storage and excretion of most metals (Jakimska *et al.*, 2011; Deb and Fukushima, 1999). Despite the existence of these mechanisms of detoxification, the vast majority of metals are stored in the tissues of organisms (Jakimska *et al.*, 2011).

Our study focuses on contamination of aquatic ecosystems by 13 of the 23 metals (Hg, Cd, Co, Cr, Cu, Ni, Pb and Zn as well as Au, Fe, Mn, U[2] and V), that aside from oxidative stress and DNA damage, can cause to many species additional damaging effects such as higher production of enzymes, enzyme inhibition, disruption of cell assimilation sodium and calcium, homeostasis and ion balance; lower cardiovascular function, hemorrhage, hematological, biochemical and histological abnormalities, including fish utilizing stored energy from vital organs to cope with the toxicity stress, increased muscle and blood lactic acid, excessive excretion, reduced white cells, red cell and hemoglobin concentration, reduced blood's coagulation time, skeletal deformities, reduced reproductive success, delay in egg hatching,

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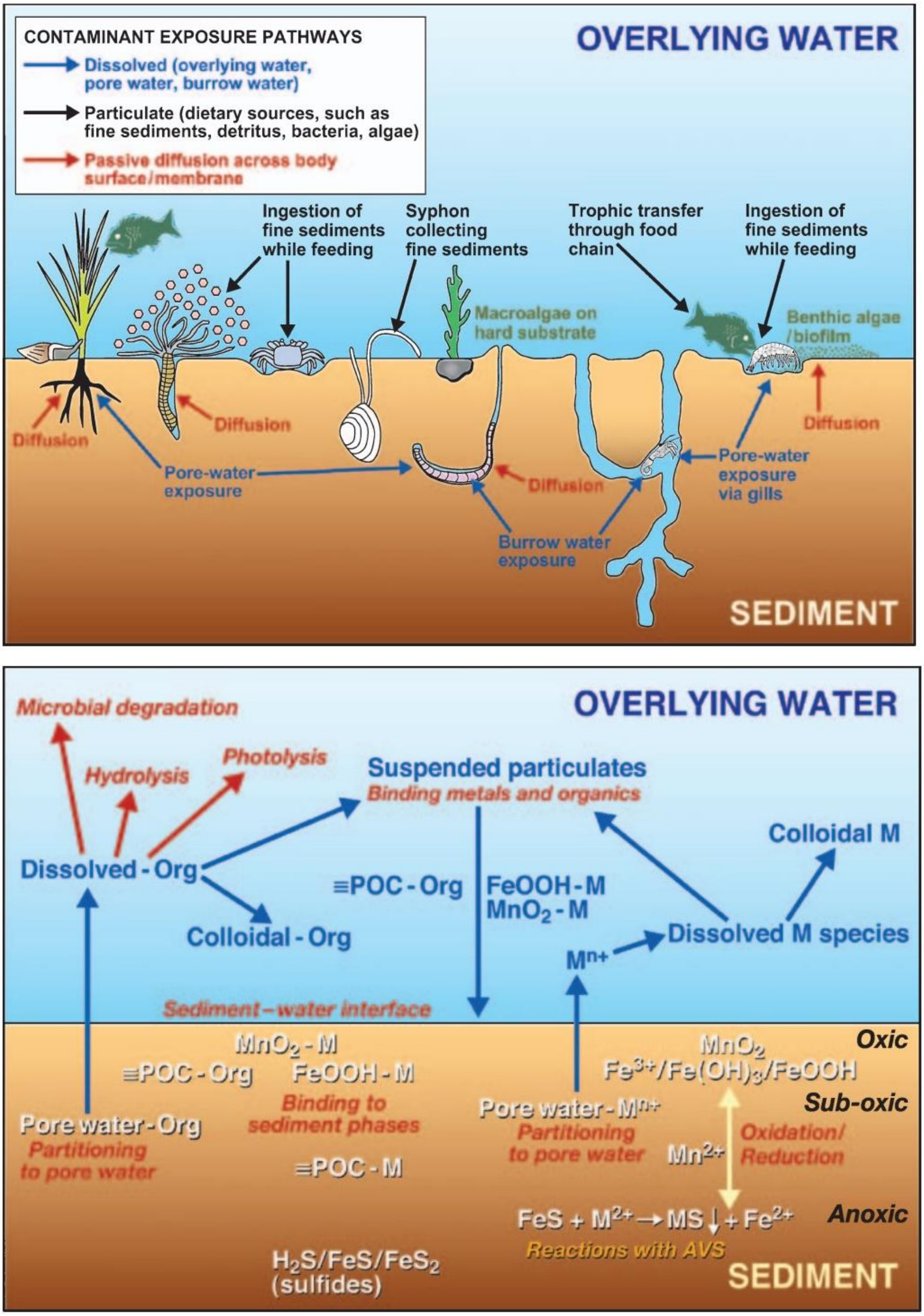
[2] Au and U were included in this study because, though they are not commonly analysed for toxicity in animal ; the study (Eastern DRC) is known for its high concentrations in both metals but studies have not analysed their presence in water systems and aquatic organisms.

increased egg mortality, liver, intestinal and gills histological alteration, immune response alteration (reduced antibody production, lymphocyte count and humoral response), reduced survival and growth rate, development abnormality, anorexia, excessive fin movement, cell degeneration, membrane damage, protein and lipid damage, behavioral and cognitive dysfunctions, neurogenerative disorders, altered olfactory sensations, cell signaling deregulation and neurotransmission impairment, swimming behavior, and others (Meina *et al.*, 2020; Sun *et al.*, 2020; Lee *et al.*, 2019; Vaz *et al.*, 2019; Simon *et al.*, 2018; Aslam & Yousafzai, 2017; Clearwater *et al.*, 2002; Boening, 2000; Vuori, 1995; Wright & Welbourn, 1994).

1.4. Metals in aquatic ecosystems: dynamics and assessment

Polluting metals have multiple origins, but once they have entered an ecosystem, they are very difficult to remove completely (Muharpawar, 2015). Sequestration and exchanges are thus key elements of metal dynamics and assessment in the quality of aquatic ecosystems. Sediments are the ultimate repository of metals in water ways (Simpson and Batley, 2016). From there, they are released to the water column by diffuse flux of dissolved chemicals in the pore waters, through physical disturbance (such as sediment resuspension by dredging, trawling wave and current actions) and biological actions (such as sediment bioturbation, mostly caused by organisms while ingesting particles) (Batley and Kumar, 2016) (see figure 1.4a). Assessment of sediment, as a source of contaminants to overlying waters and benthic organisms and food chain, can be done by comparing of measured contaminant concentrations with sediment quality guideline values (SQGVs) adopted by the EU, US and many other regions. In addition to sediment metal content, particle size, organic carbon and AVS (acid volatile sulphides) should be analysed to assess metal sequestration in sediment and release to aquatic life (Simpson *et al.*, 2016) (see figure 1.4b). Particle size of sediment determines its ability to store or release metals, since fine particles have a greater relative surface area and more binding sites for metals, while sandy sediment have a greater partitioning to pore water. Organic particles



Figures 1.4a & 1.4b: Dynamics of metals in water (Simpson and Batley, 2016)

also provide additional binding sites for metals (Batley and Simpson, 2016). Submerged sediments, through microbial respiration of organic carbon, frequently accumulate iron and manganese monosulphide (FeS, MnS), that readily react with several dissolved metals (Ag, As, Cd, Co, Cu, Hg, Ni, Pb and Zn) to form metal-sulphides precipitates. If the reactive forms of sulphides, measured as AVS, is in molar excess over simultaneously extractable metals (SEM $=\sum$Cd, Cu, Ni, Pb, Zn) (meaning $\text{SEM-AVS}<0$), the pore-water concentration of SEM is usually negligible and should not cause a direct toxicity (Ankely *et al.*, 1996; USEPA, 2005). Further refinements consider partitioning of metals to both AVS and organic carbon: $(\text{SEM-AVS})/f_{OC}$, where f_{OC} is the fraction of sediment that is organic carbon (USEPA, 2005).

Once released, metals can bioconcentrate in organisms via respiration, or direct contact with contaminated sediment or water; causing organisms to have metals in much higher concentrations than in the surrounding environment. Due to this process, organisms can be affected even when concentrations of a given metal in their environment is low. Bioaccumulation occurs through bioconcentration and additional intake by ingestion of sediment particles and food. Bioconcentration happens particularly in organisms at lower levels of the food web. Trophic transfer may be observed when these low-tropic-level organisms are consumed, but biomagnification (increase in concentration through three or more trophic levels) does not occur with most metals, except for Hg (Van Ael *et al.* 2017; Maher *et al.*, 2016; Jakimska *et al.*, 2011). When bioaccumulated, even below lethal doses, metals may cause molecular, cellular and physiological responses (biomarkers), precursors of effect on growth, reproduction or survival within the intoxicated organism (Smit *et al.*, 2009). These include subtle changes in normal physiological function such as reproductive behaviour, resilience to diseases, and prey-capture abilities, which can have profound impacts on an organism's longer-

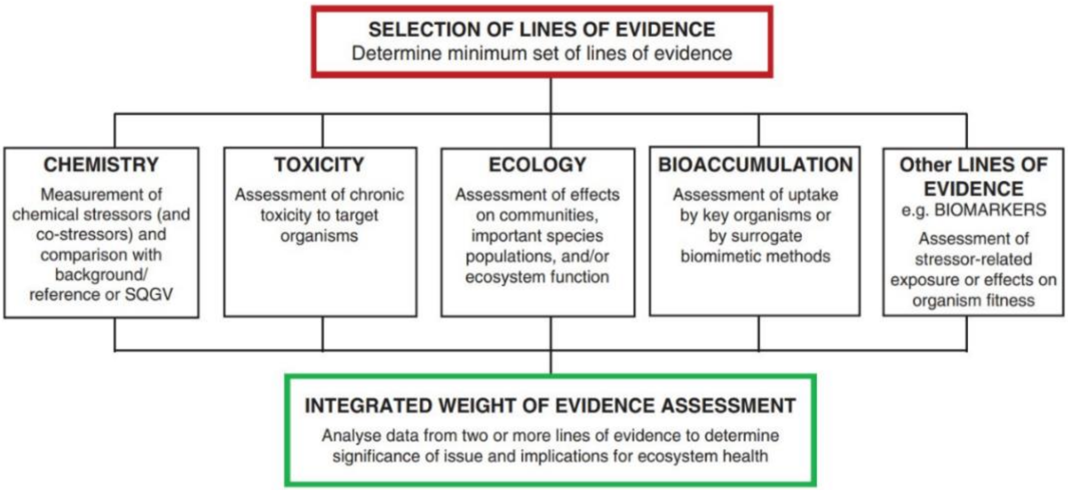


Figure 1.5: Lines of evidence in sediment and water quality (Simpson and Batley, 2016)

term survival and reproductive output; ultimately, these can affect ecosystem health (Connon *et al.*, 2009).

Multiple lines of evidence can be distinguished, including bioaccumulation and benthic ecology, biomarkers of sub-lethal exposure and effects, contaminant bioavailability tests (for example pore-water measurement, AVS, passive samplers and biomimetic approaches for hydrophobic contaminants), and toxicity testing (see figure 1.5). There is no single multiple line-of-evidence approach for sediment quality assessment, and studies should be custom-designed, and lines of evidence chosen to suit the site-specific circumstances (Simpson and Batley, 2016).

Despite the necessity of the integrated approach, very few studies include all components of aquatic ecosystems in their analysis, even fewer assess the interactions between these components. In the case of tropical ecosystem, despite more than 500 studies just on the Amazon region from the last 30 years, there is even smaller understanding of how the hydrology and biogeochemistry affects the distribution of Hg (Moreno-Bush *et al.*, 2019). In our study, we take an approach including multiple lines of evidence to have a comprehensive understanding of the distribution of metals in a tributary of the Congo river, exposed to mining activities and the threat they pose to aquatic ecosystems and human consumers. We assess metals concentrations in sediments and the sediments characteristics (such as AVS, particle size and organic carbon) that determine their sequestration or release. We then assess their concentrations in water and water characteristics determining uptake by fish and other organisms. After that, we assess metal levels in indigenous organisms who live in that ecosystem and in experimental organisms that are exposed to waters from these ecosystems under controlled conditions. And finally, we use biomarkers such as fish condition factor (CF), a change in the ratio between fish's weight and length, that may allow to understand if physiological effects are already being experienced by the fish existing in or exposed to the polluted waters.

1.5. Human exposure and health impacts

Many studies show that gold miners, gold traders and residents living in mining towns are exposed to high air concentrations of Hg when burning gold-mercury amalgams in miners' homes or in gold shops (Esdaile and Chalker, 2018; Nkuba et al., 2017; Cordy et al., 2011). In addition to airborne contamination, food consumption, especially fish, both wild and farmed, is another important source of mercury contamination in mining communities (Ha et al., 2017; Langeland et al., 2017).

Similarly, other metals are absorbed by humans through the oral or respiratory paths (see figure 1.6) and can damage and alter the functioning of the brain, kidney, liver, skin, lungs, testes, immune system, bones, hematological and cardiovascular (Engwa *et al.*, 2019; Masindi and Muedi, 2018). Though some people are primarily exposed to metals in their workplace, for most people the main route of exposure is diet (food and water) (Morais *et al.*, 2011). In mining areas, many people are likely to be exposed to the combination of these two contamination routes.

No matter through which route Hg is taken, no matter its form (either metallic, inorganic and organic) it can damage the kidney, brain and developing fetus and cause cancer (ATSDR, 1999). Indeed, inhaled Hg vapor distribute to all tissues in the body. Usually, it is well absorbed by lungs, joins the bloodstream and tissue fluids for proper distribution across the body, easily crossing the placental and blood-brain barriers (Clarkson and Magos, 2006), causing neurologic disfunctions, declined motor speed, visual scanning, visuomotor coordination, verbal and visual memory; and prenatal exposures resulting mental retardation, cerebellar symptoms, retention of primitive reflexes, malformation and other abnormalities. Inhaled Hg also causes fatigue, anorexia, weight loss and gastrointestinal disturbance (Bernhoft, 2012). Organic Hg, from different sources including fish consumption, can easily penetrate cell membranes, affects the nervous system and can alter brain functions, leading to tremors, shyness, irritability, memory problems and changes in hearing or vision (Clarkson and Magos, 2006).

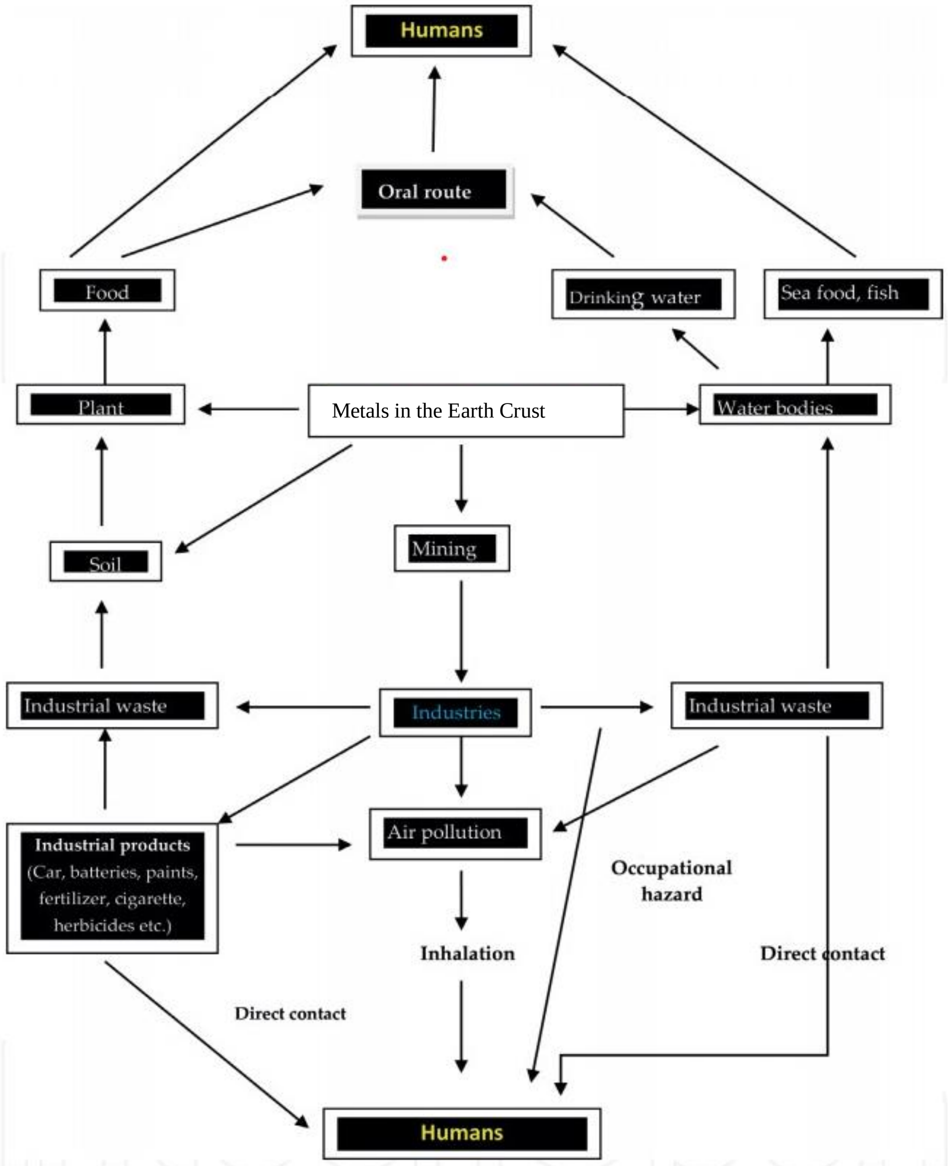


Figure 1.6: Pathways of human metal intoxication (modified from Engwa et al., 2019)

Aside from Hg, our study also analyses concentrations of Au, Cd, Co, Cr, Cu, Fe, Mn Ni, Pb, U, V and Zn in blood, urine, nails and hair. These metals are known to potentially damage human kidney, bones and lungs, cause bone mineralization and fractures, DNA damage and lung, renal and prostate cancer, causes neurological, cardiovascular and endocrine deficits, cause irritation and ulcers to the nose, stomach and small intestine, vomiting, diarrhea, gastro-intestinal bleeding, anemia, damage sperm and male reproductive system, allergic reactions, liver and kidney disease, hypotension, lethargy, hepatic necrosis, tachycardia, metabolic acidosis, allergies by contact hypersensitivity, hazardous immunological effects, neurological impairment (brain damage, mental retardation, dyslexia, autism, hyperactivity, psychosis, tremors, gait disorder, postural instability, mask-like expression, memory, mood and cognitive disorders …), birth defects, weight loss, muscular weakness, paralysis, renal and reproductive defects, asthma, rhinitis, anemia, uremia, neuronal death, coma and even cause death (Engwa *et al.*, 2019; Leyssens *et al.*, 2017; Yang *et al.*, 2017; Corlin *et al.*, 2016; Zambeli *et al.*, 2016; O’Neal and Zheng, 2015; Plum *et al.*, 2010; Martin and Griswold, 2009; Bernard, 2008; Uriu-Adams and Keen, 2005; Bhasin et al, 2002; Hillman, 2001; Markowitz, 2000).

To avoid the above undesirable health risks of excessive metals, FAO/WHO, FDA (US Food and Drugs Administration), European Union, etc. have developed a safety approach to establish tolerable intakes of substances with threshold toxicity. These include the tolerable daily intake (TDI) and provisional tolerable weekly intake (PTWI), calculated on a body weight basis ($mg.kg^{-1}$ of body weight), which all describe levels of intake of a given metal (or other toxicant) the body can be ingested over a lifetime without considerable health risks. Exposures exceeding the TDI or PTWI for short periods should not have deleterious health effects; but if the exceedance is too large, acute toxicity may occur even over short periods of time. Also, metals with long half-lives may accumulate in the body and cause health risks once critical concentrations have been exceeded in a given organ (Mudgal *et al.*, 2010).

Our study, in addition to assessing the social factors favoring pollution from mining activities, the concentrations of metals in the environment and in fish, and the risk of metal poisoning to human consuming water and fish, also analyzes on how both the oral and respiratory routes contribute to metal contamination in communities. In addition to causes and pathways, we assess the consequences of this contamination in terms of different damage it may cause. Many studies have identified effects of metal contamination, but our study focuses on determining if symptoms associated with these effects are related to the level of contamination people actually experience, or if these same effects may be due to other causes and mistaken for effects of metal contamination. In addition, our study analyzed, based on the TDI, both the hazard quotient (HQ) and the maximum edible amounts (MEA). The HQ is a ratio representing what risk does an average fish consumer take considering the metal content of the consumed fish. While the MEA represent how much fish one can consume regularly without being exposed to intoxication by the target metal.

1.6. Objectives and outlines

Our study analyzes metal pollution from gold mining in a comprehensive way, combining mining practices leading to pollution by Hg and other metals, the distribution of these pollutants in potentially aquatic ecosystem, as well as routes and effects of human contamination from these metals. It uses an interdisciplinary approach that involves social sciences methods such as qualitative and quantitative surveys, biological and analytical tools such as exposure experiments, metal analyses, and medical survey tools including analysis of nutrition survey, general health assessment and symptom analysis. The combination of points of assessment and analysis tools was necessary to identify potential entry points for future interventions and to recommend to different categories of stakeholders what they can best do to solve the threat of mercury and metal contamination to mining communities and their environment.

Aside from a general introduction (**chapter 1**) that presents gold mining as the source of livelihood for communities but also a source of metal pollution, dynamics of these metals in aquatic ecosystems and their intoxication of human communities, this study includes three research chapters and a general discussion chapter.

Chapter 2 identifies the practices that uses mercury and pollute the rivers with other metals. It analyzes the governance of mercury use, the enforcement problems, and aims to understand why, miners continue to use these harmful techniques. It finally focuses on local perspectives surrounding mercury and other threats from gold mining, bringing more light to the level of knowledge within the local communities of the threat mercury and other mining forms of pollution pose for them, as well as the trade-off between the mining income they rely on and the health and environmental impacts they are likely to suffer from.

Chapter 3 analyses the levels of these pollutants that have entered the waters (including parameters that may hinder or enhance metal uptake from water by aquatic organisms), sediments (including parameters that condition the sequestration and release of these pollutants to the overlying waters), local and experimentally exposed fish (especially in their muscle tissues which represent the highest risk for human consumers), as well as the threat mercury and other metal represents to consumers of both water and fish (assessing both the hazard quotient and maximum edible amount of fish).

Chapter 4 assesses how metals that enter the environment end-up affecting the communities that live in the mines. It looks at the dietary habits as well as the major forms of occupation within the community, focusing on how dietary and occupational exposures leads to increases level of metals (particularly mercury) in people's organs. It also analyses the metal contamination related symptoms experienced by people within the community and connects them to the metal levels and prevalence of potential metal intoxication.

Finally, a general discussion brings together the contributions of all previous chapters and provides recommendation for policymakers, researchers and the mining community itself (**chapter 5**).