

Zeitschrift: Bulletin für angewandte Geologie
Herausgeber: Schweizerische Vereinigung der Petroleum-Geologen und –Ingenieure;
Schweizerische Fachgruppe für Ingenieur-Geologie
Band: 7 (2002)
Heft: 1

Artikel: Natural arsenic-contamination of surface and ground waters in
Southern Switzerland (Ticino)
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DOI: <https://doi.org/10.5169/seals-223647>

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Natural arsenic-contamination of surface and ground waters in Southern Switzerland (Ticino)

with 12 figures and 1 table

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Abstract

Arsenic belongs together with lead, mercury and cadmium to the most toxic elements. Used extensively during the first half of the 20th century as part of pesticides, today, its industrial use has been drastically reduced, but still persists. However, the influence of natural arsenic occurrences on soil and water composition has been largely underestimated. During the last 10 years, starting in Asia, especially in Taiwan, Bangladesh and India, natural As-contamination has been discovered in many countries. The main liberation and dispersion processes are weathering of As-bearing rocks and input of deep As-rich thermal waters into shallow aquifers. The main concern is drinking water with As-contents above the internationally recommended limiting value of 10 µg/l. In Switzerland, rocks and soils containing As in elevated amounts are known from the Jura (Bohnerz formation, Fe-rich Dogger limestones) and from the Alps (sulfide ores deposits and silicate rocks relatively rich in pyrite or hematite). For the moment detailed data on drinking water is only available for southern Switzerland (Ticino area). In about a dozen communities the drinking water contains between 10 and 40 µg/l As, locally up to 80 µg/l were detected. In former mines and some isolated ground water drill holes characterized by reducing conditions, values of up to 300 µg/l As occur. These are close to levels which are known from Bangladesh and which create the typical arsenic-pathologies (e.g. "black foot disease", cancer) after a few years of exposure. Depending on the dissolved oxygen content and the oxidation potential, arsenic occurs as As(III) or As(V) or as a mixture of both. The contamination is limited to the Sotto Ceneri area and 4 types of hydro-geological situations can be distinguished: (1) Spring water and local creeks related to a fissured hard rock aquifer with an elevated As-content (gneisses, mica schists and phyllonites containing elevated amounts of As-bearing pyrite and iron oxides). (2) Spring waters related to an porous glacial aquifer which is enriched in As, due to the erosion of regional Fe-As ore deposits. (3) Mine waters issuing from abandoned addits and/or percolating through mine dump material rich in arsenic. (4) Reduced Fe- and As-rich groundwater slowly circulating or stagnating in alluvial river deposits on the valley grounds, often close to the shore line of lakes and rivers. Here, the exact origin of the As is often unknown. Actually, the state authorities monitor the critical springs and wells and some of them have recently been excluded from consumption. Water treatment procedures to reduce arsenic in drinking water to levels of about 5 µg/l exist and have been tested already, but ask for close follow up of the water treatment plant.

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Zusammenfassung

Arsen gehört zusammen mit Blei, Quecksilber und Cadmium zu den giftigsten Stoffen überhaupt. Nachdem es während der ersten Hälfte des 20. Jahrhunderts vor allem als Bestandteil von Pestiziden häufig verwendet wurde, ist seine industrielle Bedeutung heute stark zurück gegangen. Hingegen ist der Einfluss von natürlichem Arsen auf Böden und Wässer lange Zeit stark unterschätzt worden. In den letzten 10 Jahren wurden in vielen Ländern, insbesondere in Taiwan, Bangladesh und Indien, natürliche Arsen-Umweltkontaminationen entdeckt. Die wichtigsten Arsen-Eintrags- und Ausbreitungsprozesse in die Umwelt sind verbunden mit der Verwitterung von As-führenden Gesteinen und dem Aufsteigen von As-reichen Thermalwässern in oberflächennahe Grundwasserträger. Das Hauptproblem sind Trinkwässer mit Gehalten über dem international empfohlenen Grenzwert von 10 µg/l. In der Schweiz enthalten vor allem Gesteine und Böden im Jura (Bohnerz-Formation, Fe-reiche Doggerkalke) und in den Alpen erhöhte Arsengehalte (meist in Zusammenhang mit Sulfid-Erzen und Kristallingesteinen mit erhöhten Pyrit- und Hämatitgehalten). Im Moment sind detaillierte As-Daten über Trinkwasser und Böden vor allem aus dem Tessin bekannt. Dort enthält das Trinkwasser in einem Dutzend Gemeinden zwischen 10 und 40 µg/l, lokal bis 80 µg/l. In ehemaligen Minengeländen und in vereinzelt Piezometerrohren, in welchen reduzierende Bedingungen herrschen, treten Werte bis zu 300 µg/l As auf. Diese liegen nahe bei den aus Bangladesh bekannten Werten, welche mittelfristig zu den typischen Arsen-Krankheiten führen ("black foot disease", Krebs etc.). Abhängig vom Gehalt an gelöstem Sauerstoff und dem Oxidationspotential E_h tritt Arsen als As(III), As(V) oder als Gemisch beider zusammen auf. Die kontaminierten Gebiete sind auf die Gegend des Sottoceneri beschränkt, wobei 4 hydrogeologische Situationen unterschieden werden können: (1) Quell- und lokale Oberflächenwässer in Zusammenhang mit geklüfteten Festgesteinsaquiferen mit erhöhtem As-Gehalt (Gneise, Glimmerschiefer und Phyllonite mit erhöhtem Gehalt an As-führendem Pyrit und Fe-Oxiden); (2) Quellwässer aus glazialen Sedimenten, welche durch die Erosion von regionalen Fe-As-Lagerstätten entstanden; (3) Minenwässer, die aus aufgelassenen Stollen oder As-reichen Halden austreten, (4) reduzierte Fe- und As-reiche, langsam zirkulierende oder stagnierende Grundwässer in Alluvialsedimenten in Fluss- oder Seenähe. In diesen Fällen bleibt die genaue Herkunft des Arsens oft unklar. Gegenwärtig werden die kritischen Quellen und Grundwassergebiete von den Behörden überwacht und mehrere sind als Trinkwasser-Lieferanten aufgegeben worden. Auf 5 µg/l Arsen absenkende Filtermethoden für Trinkwasser existieren zwar und sind schon mehrfach getestet worden, verlangen jedoch einen verhältnismässig grossen Unterhaltsaufwand.

1. INTRODUCTION

1.1 Arsenic worldwide

Arsenic has been known for its toxic properties since antiquity (Eisler 1988). Much like other toxic elements, the industrial activities of the last two centuries, such as mining, smelting and glass manufacturing and its use as medical drug for humans, pesticide, wood preservative and growth promoter for animals, is at the origin of a widespread distribution in different environmental compartments, especially in soils and water. In 1985, the anthropogenic input of arsenic to the atmosphere was estimated to be approximately 28'000 t As/year, which corresponds to a little bit more than half of the natural input (mainly by volcanoes and forest fires: 45'000 t As/year; Chilvers & Peterson 1987).

Between 1970 and 1990 considerable efforts were undertaken worldwide, to reduce anthropogenic input and local contamination, especially the build-up of high levels in soils. But little attention was paid to natural background values in rocks, soils and waters. This can partly be explained by analytical difficulties to detect relatively small quantities of arsenic. The main natural liberation and dispersion processes are weathering of As-bearing rocks and input of deep As-rich thermal waters into shallow aquifers. While the toxic effects of elevated concentrations of arsenic were well known (0.1 g of arsenic trioxide may be lethal to humans), long term exposure

to medium and low values and accumulation effects had been underestimated. A few local studies, where people had been exposed to exceptionally high levels (e.g. in Taiwan in the sixties), showed that concentrations of 500 µg/l in drinking water lead to skin cancer or other forms of skin lesions. WHO estimates that a daily intake of water containing 20 µg/l As during 70 years will increase the number of skin or other type of cancer by 5%. For these reasons, recently, most countries, but not yet Switzerland, have reduced the maximum allowable level in drinking water from 50 µg/liter to 10 µg/l, the recommended value of WHO (1999).

This new threshold value and more and more data about millions of people in West Bengal/India and Bangladesh (Samanta et al. 1999, Hug et al. 2000) who had started in the late eighties to drink waters from deep ground water wells, instead of shallow water, and now suffer from skin cancer, hyperkeratosis and the so-called black-foot disease, lead authorities of many countries control systematically their drinking water supplies. The result of this campaign is that in many countries of the world, including the United States and part of Europe, there are many areas where the natural background of arsenic in water is higher than 10 µg/l and that thousands of local water plants touching several millions of people will need appropriate systems to remove arsenic from the water (Germany: Heinrichs & Udluft 1996, Weiss et al. 1999; Austria: Wenzel et al. 1999; France: Robert 1998; Hungary: BAW 1993, VATECH 1999; Portugal: Moreno et al. 1999, USA: Davis et al. 1994, Welch et al. 1999; Spain: Sassoon 1998, Mardones 2000; Slovenia: Svetina & Pirc 1998, Chile: Sancha 1999, Argentina: Rivero et al. 1999, Mexico: Rosas et al. 1999, Simeonova 1999; Australia: Naidu et al. 1999, India and Bangladesh: Samanta et al. 1999, Nickson et al. 2000, Vietnam: Berg et al. 2001).

1.2 Arsenic in southern Switzerland

In Switzerland, rocks and soils containing As in elevated amounts are known from the Jura (Bohnerz formation, Fe-rich Dogger limestones, Donzel 2001) and from the Alps (sulfide ores deposits and silicate rocks relatively rich in pyrite or Fe-oxides, Pfeifer & Zobrist 2002). Up to 1994, only few data on arsenic in the environment existed for Switzerland (Pfeifer et al. 1996). The only data available were on water from mineral springs (Högl 1980), a few soils and sediments from the Wallis (Woodtli et al. 1985) and a few analyses of the state laboratory of the canton Ticino from springs and wells related to new exploration work foreseen in the Malcantone area in Ticino (published in Pfeifer et al. 1996). The systematic research began with a local study of soils and creek waters in the same area (Bondietti et al. 1994, Rey 1996, Pfeifer & Rey 1998). In 1996, when one of the springs providing drinking water for the communities of Sessa and Astano was temporarily closed down, high values of arsenic were accidentally found in the other. This led to a systematic survey of the drinking waters of the whole state of Ticino (Jäggli 1996, 1997, 1999a,b; Pfeifer et al. 2000). The result of this effort was that waters containing arsenic beyond the WHO guideline value, are much more widespread than expected.

As in other countries, the presence of arsenic in the Ticino area is a natural phenomenon. The purpose of this paper is to give an overview on the occurrence and the typical composition of the arsenic-rich surface and ground waters of southern Switzerland. There are three main areas exhibiting ground waters above the WHO-guideline of 10 µg/l (Fig. 1). They are all situated in the silicate-rock dominated, so-

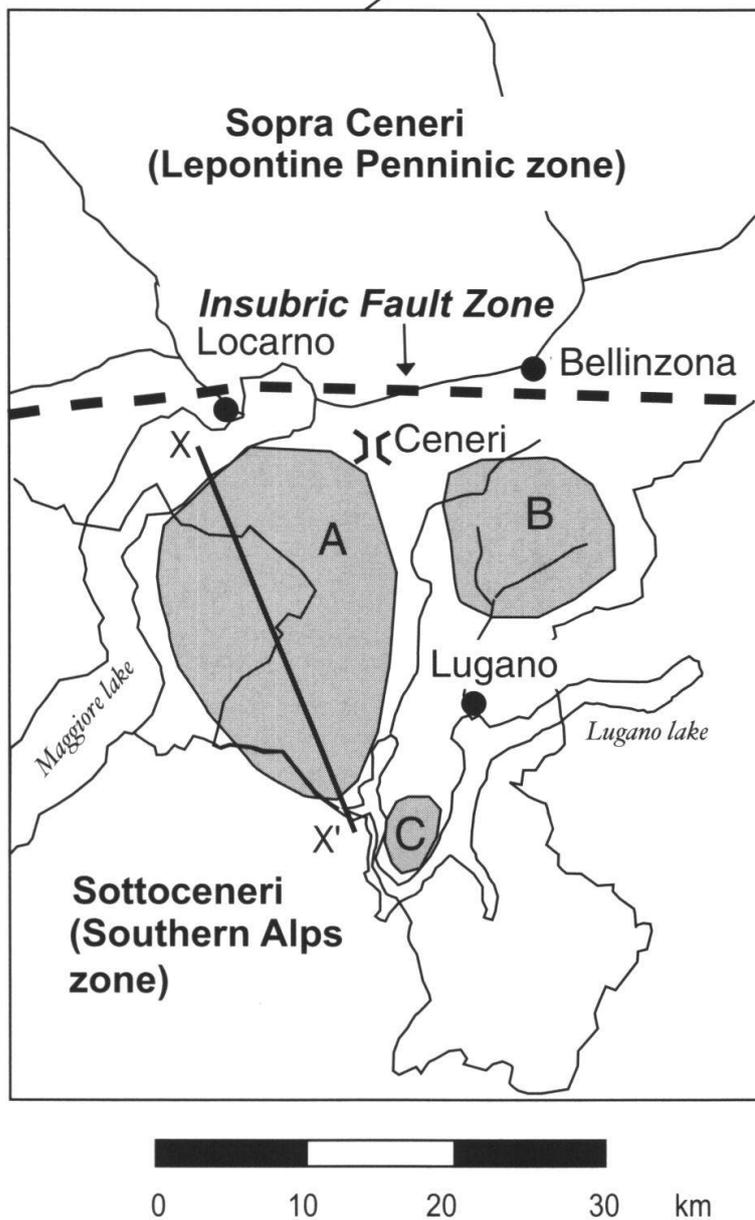
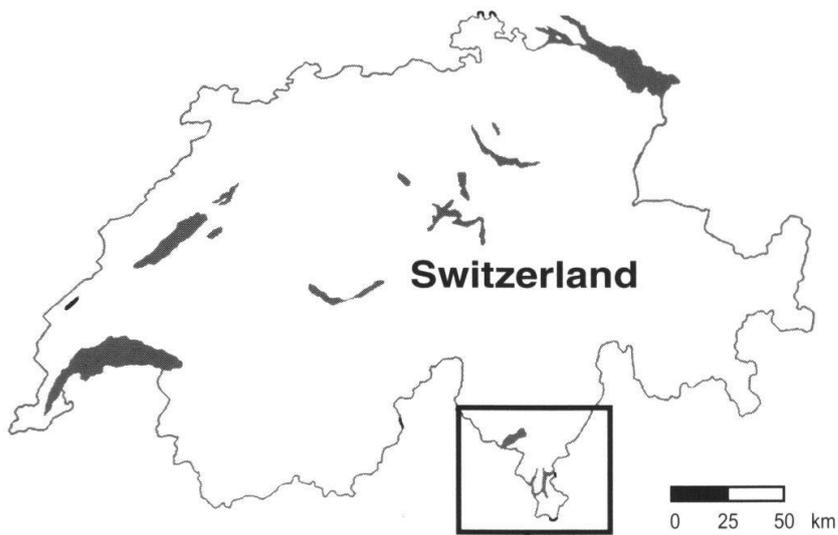


Fig. 1:

Areas in southern Switzerland with elevated natural arsenic in soils and waters. A: Gambarogno- Malcantone- Val Veddasca (Italy), B: Val Isone-Val Colla, C: Barbengo-Figino- Morcote. X-X': cross-section shown in figure 3. As additional information, the major geological units are indicated, separated by the Insubric fault zone, marking the limit between the two main regions «Sopraceneri» and «Sottoceneri».

called «Sottoceneri» area, south of Monte Ceneri, where the important geological boundary of the Insubric fault zone is located. Figure 2 shows that about a dozen communities use drinking water with arsenic levels between 20 and 80µg/l and therefore will possibly have to clean their waters, if the limiting value of the WHO will one day be adopted in Switzerland.

Arsenic shows a very complex behavior in natural systems due to its different oxidation states As(III) (arsenite, in water mainly as neutral H_3AsO_3 aq.) and As(V) (arsenate, mainly as anions $H_2AsO_4^-$ and $HAsO_4^{2-}$). This variable character influences its sorption behavior on solid phases, such as Fe- and Al-oxy-hydroxides, clay minerals and organic matter (Smith et al. 1998, Halter & Pfeifer 2001). Precipitation as oxides or as arsenate compound (so-called arsenicals) is more rare. Alkaline (high pH) and reducing conditions (low Eh) favor the mobilization of arsenic in water and soils (Deutsch 1997).

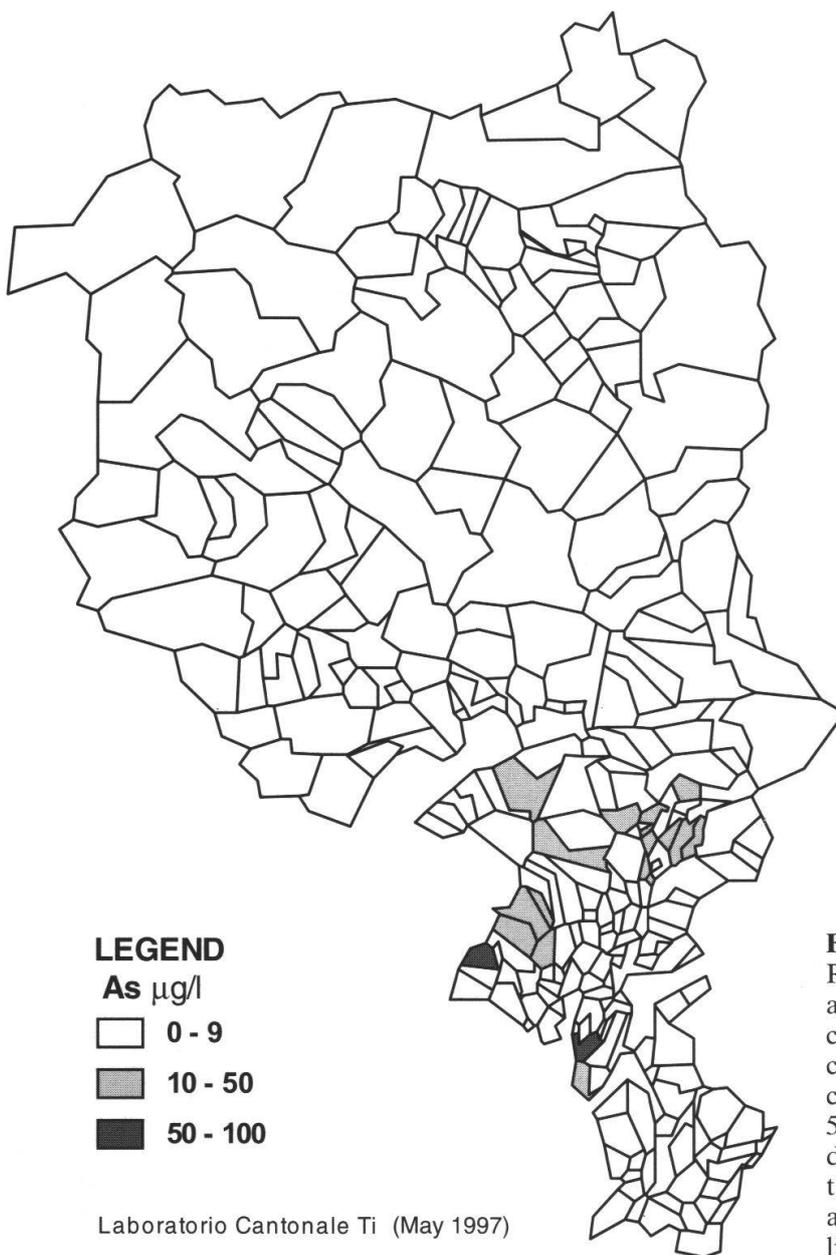


Fig. 2: Result of a systematic check of arsenic in drinking waters of all communities of the canton of Ticino (November 1996). In three cases the actual limiting value of 50 µg/L. As is trespassed, in dozen other values are between the WHO guide value of 10 µg/L and 50 µg/L. All elevated cases lie in the Sottoceneri area.

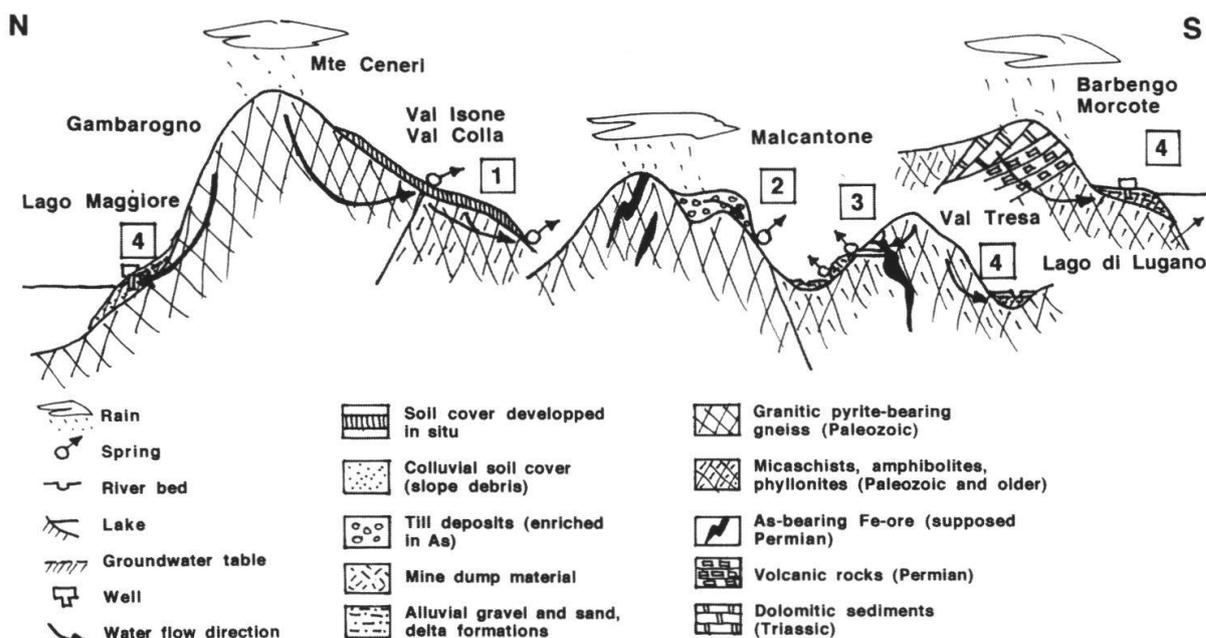


Fig. 3: Very schematic geological cross section through the Sottoceneri region, showing the different hydro-geological situations in which the waters with elevated arsenic contents occur. The four general aquifer situations indicated (with numbers 1 to 4) are explained in detail in figure 4.

2. TYPICAL HYDROGEOLOGICAL SITUATIONS

2.1 General aspects

The schematic cross-section through the Sottoceneri area of figure 3 shows the many different hydro-geological situations in which arsenic-rich waters occur in southern Switzerland. Four major situations can be distinguished (Fig.4):

- (1) Spring waters and local creeks related to fissured silicate rock aquifers
- (2) Spring waters related to porous glacial sediment aquifers
- (3) Waters issuing from abandoned mines or percolating through mine dump material
- (4) Reduced ground waters in alluvial aquifers of valley grounds

These four situations can principally occur within the same area, however, there is a certain systematic geographical distribution, which is indicated in figure 3. Type 1 is mainly encountered in the Val Isonne-Val Colla area and in the vicinity of Morcote (cf. Fig. 1). Type 2 is typical for the Malcantone- Val Veddasca area. Type 3 can be found wherever there are old mining structures (Malcantone, Medeglia/ Val Isonne) and type 4 has been found close to lakes and river borders: Barbengo-Figino (former drinking water well), Vira-Gambarogno (well for a heating pump) and along the Tresa river between Ponte Tresa and Monteggio (several piezometer holes).

Among the waters reported here, in terms of their overall chemical composition, as usual (cf. Pfeifer et al 2000), two major chemical trends can be distinguished (Tab. 1): (A) waters dominated by silicate rock interaction, characterized by low total dissolved solid contents (conductivities between 70 and 120 $\mu\text{S}/\text{cm}$, pH 7.0 to 7.5, low alkalinities: HCO_3 20 – 40 mg/l) and (B) waters influenced by carbonate rocks with

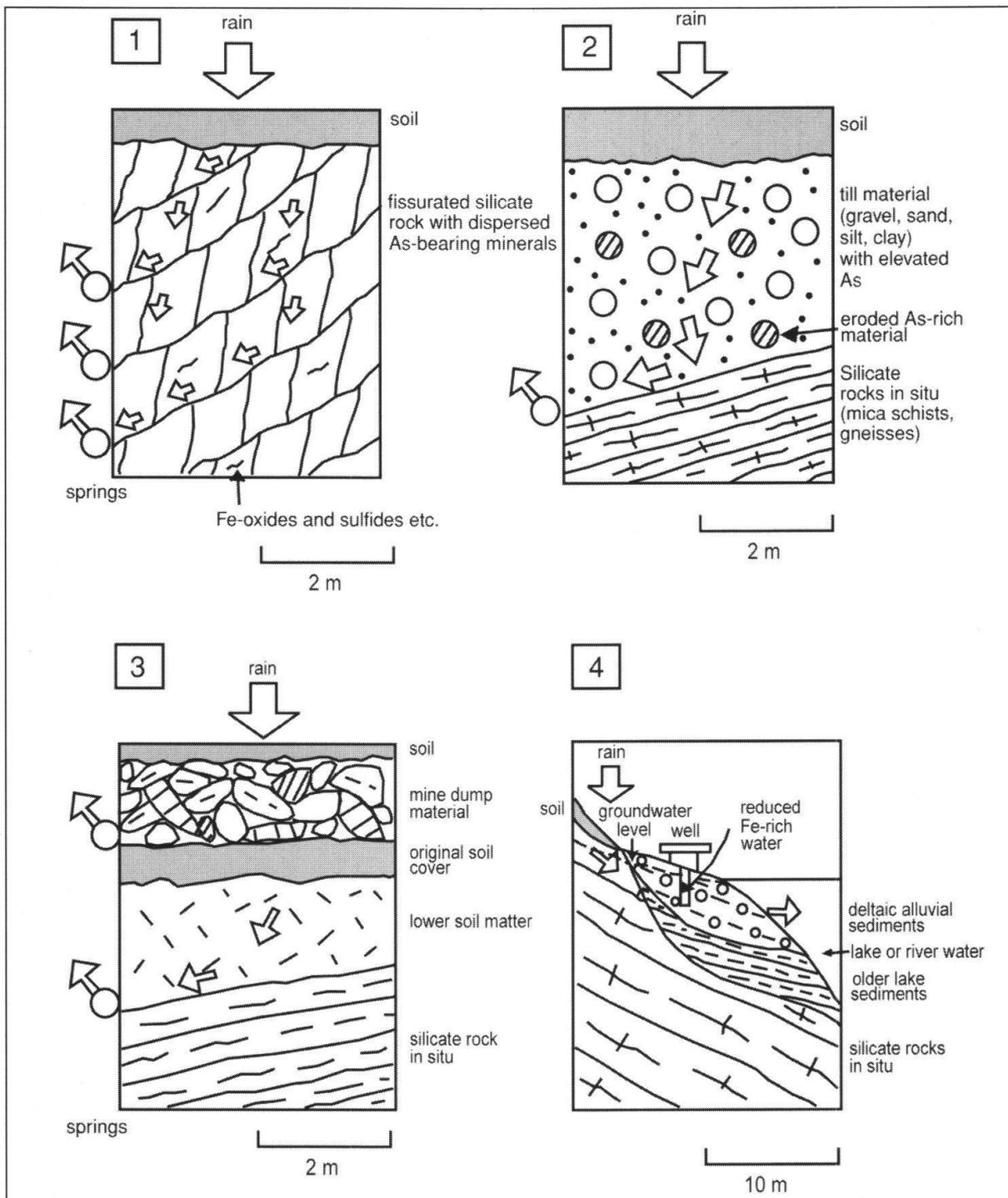


Fig. 4: The four main types of aquifers producing waters with elevated arsenic contents.

- 1: Spring water and local creeks enriched in As ($20\text{--}50\ \mu\text{g/l}$), related to a fissured hard rock aquifer with an elevated As-content (gneisses, mica schists and phyllonites containing elevated amounts of As-bearing pyrite). Example: Val Colla/Val Isonne region, Morcote area.
- 2: Spring water rich in As ($50\text{--}80\ \mu\text{g/l}$), related to an porous glacial aquifer (till deposits enriched in As, due to the presence of regional Fe-As ore deposits). Example: Malcantone area (hill slopes).
- 3: Mine water very rich in As ($100\text{--}800\ \mu\text{g/l}$) issuing from abandoned addits and/or percolating through mine dump material rich in arsenic. Examples: Astano- Costa (Malcantone), Medeglia (Val Isonne).
- 4: Reduced Fe- and As-rich groundwater ($200\text{--}300\ \mu\text{g/l}$ As), slowly circulating or stagnating in alluvial river deposits on the valley grounds, often close to the shore line of lakes and rivers. Origin of the As often unknown. Examples: Barbengo-Figino, Gambarogno area, Tresa valley.

conductivities between 250 and 300 $\mu\text{S}/\text{cm}$ and a pH often above 8. Typical As-concentrations in surface waters lie between 0.5 and max. 26 $\mu\text{g}/\text{l}$, those in normally oxidized spring waters between < 0.5 and 80 $\mu\text{g}/\text{l}$. Higher values between 150 and 300 $\mu\text{g}/\text{l}$ typically occur in the vicinity of sulfide mines (analysis no. 6 and 7) and in the stagnant, reducing ground water of the alluvial aquifers (analysis no. 9-11). Figure 5A shows the frequency of the different arsenic contents in the Sottoceneri area. The yet scarcely known partitioning between As(III) and As(V) will be discussed in section 3 and 4.

2.2 Fissured silicate-rock aquifers (Fig. 4-1)

Spring and creek waters from these aquifers typically contain between 20 and 50 $\mu\text{g}/\text{l}$ of arsenic. Soil covers are usually thin (max. 50 cm, litho- or regosoils) and springs are often situated at the bottom of cliffs or rocky areas. The arsenic contents of the aquifer silicate rocks (granitic gneisses, amphibolites and phyllonites) is very variable: 8 to 3000 $\mu\text{g}/\text{kg}$ (ppm), but values below 20 $\mu\text{g}/\text{kg}$ dominate (Fig. 5B). Direct evidence indicating the exact nature of the As-bearing mineral phase is scarce.

	1	2	3	4	5	6	7	8	9	10	11	12
T°C	10.0	13.0	10.9	6.0	10.9	10.3	10.0	7.8	-	10.0	10.0	10.0
K $\mu\text{S}/\text{cm}$	76.0	70.0	80.0	231.0	122.0	-	280.0	123.0	-	190.0	223.0	-
pH	7.8	7.7	7.5	7.8	7.5	7.5	6.2	7.9	-	8.0	7.3	8.2
Eh mV	-	-	-	-	+190.0	-	-	-	-	-9.0	+153.0	-
Na mg/L	2.6	2.5	1.6	5.2	2.9	4.2	7.1	2.5	10.3	11.3	-	-
K	1.0	0.3	0.5	1.4	9.1	1.3	1.4	1.2	2.9	1.9	-	-
Mg	3.0	2.2	0.8	8.2	0.9	14.6	4.2	1.7	26.3	3.5	-	-
Ca	10.0	5.3	12.7	37.4	16.8	41.1	19.2	18.8	61.5	22.9	-	-
HCO ₃	20.0	21.2	37.0	123.2	41.5	103.7	4.2	70.9	301.3	83.0	-	-
Cl	1.0	<0.2	4.0	6.2	9.8	2.4	<0.2	0.7	7.5	1.5	-	-
SO ₄	-	9.0	10.2	16.6	8.7	63.5	64.6	13.8	12.7	13.6	-	-
NO ₃	0.9	4.4	5.9	1.2	3.8	4.0	1.3	5.9	0.9	-	-	-
SiO ₂	7.1	3.3	7.7	1.8	-	2.3	15.4	4.3	-	-	-	-
Fe	-	<0.05	0.1	-	-	-	1.5	<0.05	-	-	-	-
As $\mu\text{g}/\text{L}$	9.0	25.8	0.5	2.0	74.0	159.0	234.0	32.5	211.0	290.0	80.0	42.0
As III	-	-	-	<0.3	1.5	4	-	-	135	220	26	<0.5
As V	-	-	-	2	72.5	155	-	-	76	70	54	42

1: River: Magliasina, Malcantone (Martini & Pillet 1996)

2: River: Fiume Bello, Val Colla (Schmit 2000)

3: River: Vedeggio, Val Isonne (Schmit 2000)

4: Lake: Lugano at Figino (Barbieri & Mosello 1992, As: this work)

5: Oxidised spring: Astano- Froda, Malcantone (AS 91): this work

6: Mine waste seepage: Beredino, Malcantone (Rey 1996 and this work)

7: Mine water: Medeglia, Val Isonne (Schmit 2000)

8: Oxidised spring: Camignolo, Val Isonne (Schmit 2000)

9: Reduced ground water: untreated drinking water well (up to 1999), Barbengo-Figino (Jäggli 1999a)

10: Reduced ground water, stagnant, Tresa river plain (Cr 6a, 2000, this work)

11: Reduced ground water, after 10 min. pumping, Tresa river plain (Cr 6d, 2000, this work)

12: Reduced ground water, after 30 min. pumping, Tresa river plain (Cr 6, 2001, this work)

Abbreviations: K: conductivity, -: not measured

Tab. 1: Typical surface and ground waters influenced by natural arsenic of the Sottoceneri area in southern Switzerland.

This type of aquifer is typical for regions without or very few potentially arsenic-bearing ore deposits. Therefore, we conclude that it must be the sulfide or Fe-oxide minerals such as pyrite or hematite, irregularly distributed in the aquifer bodies, which are responsible for the observed arsenic. Preliminary microprobe analyses point to hematite and As-bearing allanite, besides minor sulfides. While circulating in the uppermost, usually intensively fissured top zone of the silicate rock substrate, the rain and soil percolation water becomes enriched in As.

2.3 Porous aquifers (Fig. 4-2)

Spring waters related to glacial deposits (mainly till material of local or regional origin, more rarely fluvio-glacial sands and gravel, Berthoud 2001), typically have As-concentrations between 30 and 80 $\mu\text{g/l}$ (Fig. 5A). In Val Veddasca, Province of Varese, Italy, there are three springs with concentrations up to 330 $\mu\text{g/l}$ of arsenic (VAR 1998). The till material itself contains between 40 and 500 mg/kg (ppm) of arsenic. As shown on figure 4B, these high concentrations, compared to the Swiss mean of about 10 mg/kg, are due to the presence of eroded material stemming from As-ore deposits (arsenopyrite, pyrrhotine, pyrite), widespread in the Malcantone-Veddasca area (Köppel 1966), where this type of aquifer mainly occurs. In most cases, in the unconsolidated glacial sediments, arsenic is not contained in primary sulfide minerals as in type 1, but is adsorbed onto secondary alteration solids such as Fe-oxy-hydroxides (Ferrihydrite, goethite, cf. Cornell & Schwertmann 1996), clay minerals and organic soil matter (Reymond et al. 2001, Pfeifer et al. 2001). The springs are normally located at the interface between the glacial sediments and the underlying hard rock, which is little permeable (Beatrizotti 1996). However, a slight penetration in the presence of fractures cannot be excluded.

2.4 Mining areas (Fig. 4-3)

Waters issuing from addits or mine dumps are the richest in arsenic (100 to 800 $\mu\text{g/l}$). They either have been in direct contact with sulfide minerals rich in As (such as arsenopyrite, pyrite or pyrrhotine) or percolate through debris rich in arsenic (case shown in Fig. 4-3). These debris typically contain 5 to 40 % As (50 to 400 g/kg). In the known examples, the high mobility of arsenic is often favored by high pH-values, which are due to the presence of accompanying carbonate minerals (Ferdolomite). Dump materials are usually coarse grained and have a high permeability. The water either percolates down to the interface of the original soil level before mining or down to the underlying hard rock. Most of these waters occur as local seepage with low discharge. They are active after rain periods and can be followed only on short distance. They usually disappear in the ground after some tens of meters or are rapidly diluted by local creeks or rivers.

2.5 Reduced ground waters in alluvial aquifers (Fig. 4-4)

This case differs from the others by the fact that the arsenic rarely stems from the aquifer itself, but from the adjacent hills or mountains. In this situation relatively high arsenic contents have been found (200 to 300 $\mu\text{g/l}$). They are related to iron-rich waters in which Fe(III)-hydroxide (3-line ferrihydrite) forms during pumping or forms prior to pumping from corrosion of the piezometer tubes. This and measured low oxidation potentials (Eh) of -50 to -200 mV indicate reducing conditions, which favors neutral As(III)-species and de-sorption of arsenic from most

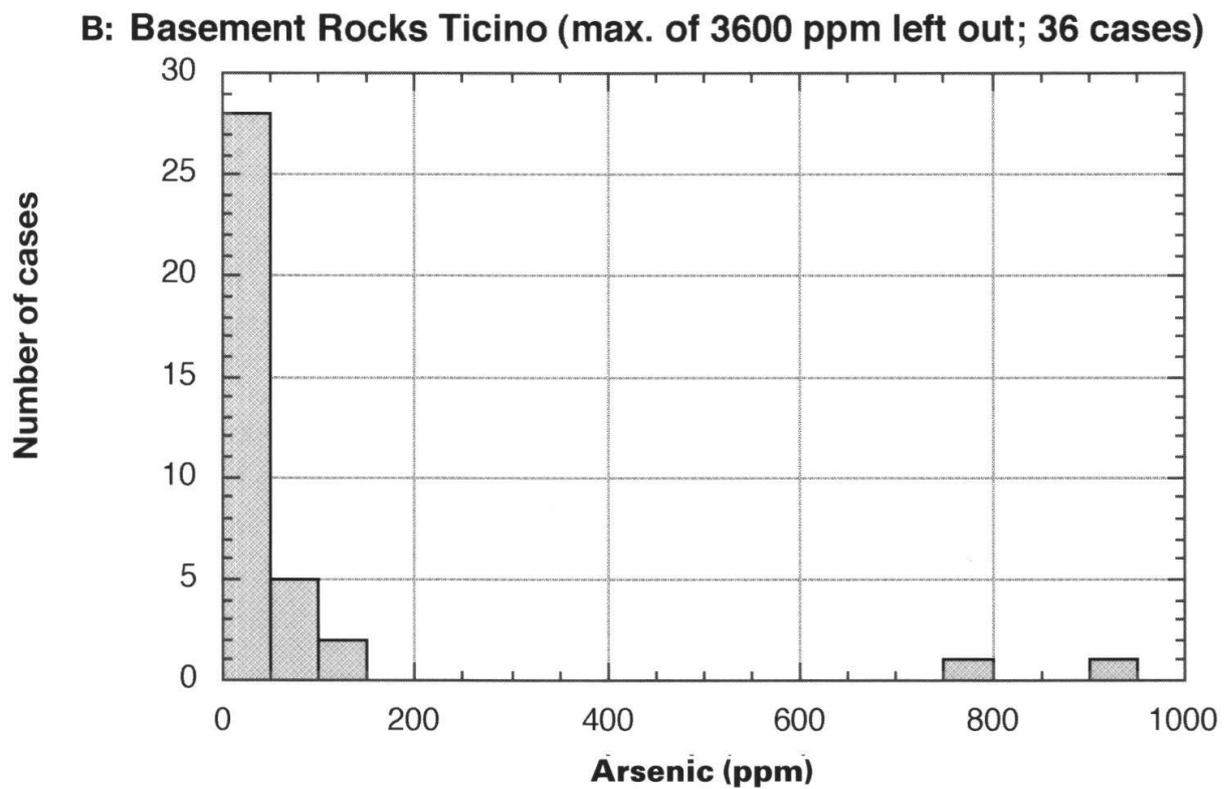
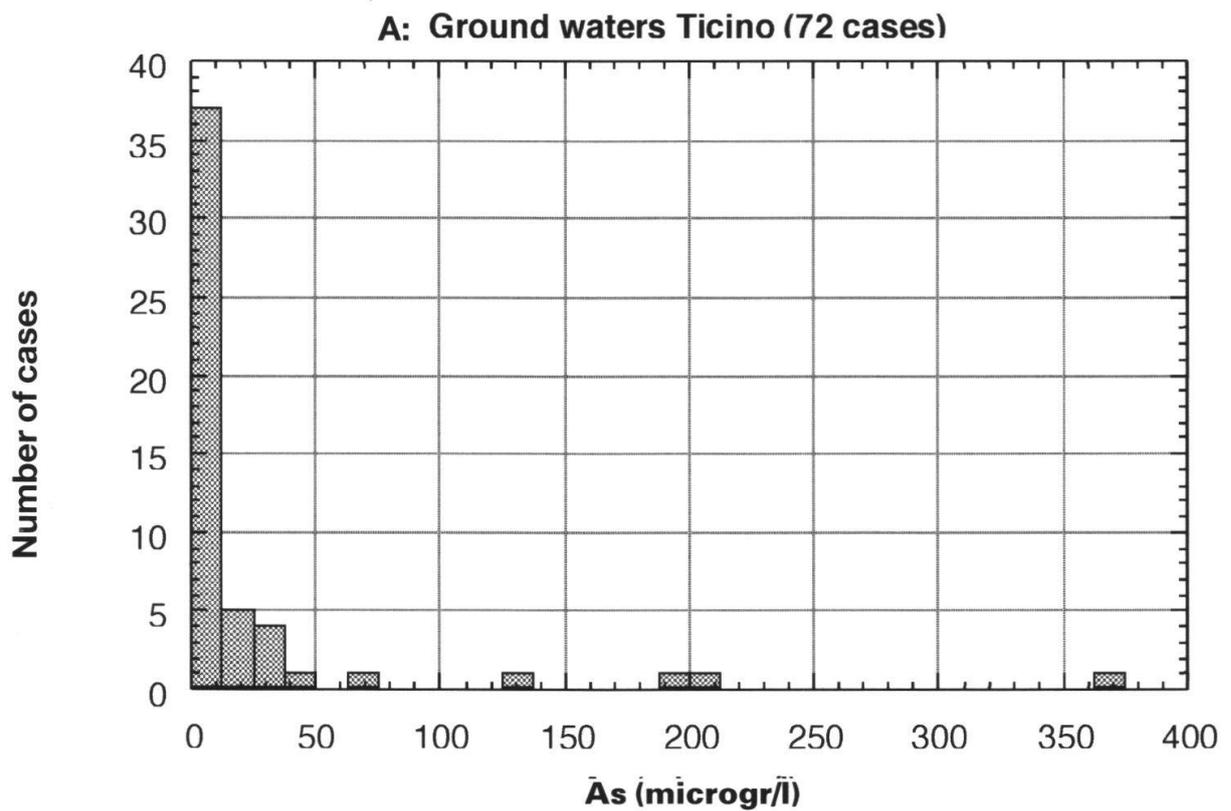


Fig. 5: Variation of arsenic in waters and rocks in the Sottoceneri area. A: Arsenic contents in surface and ground waters. Only values above 10 µg/l are shown. The complete data set is given in Jäggli (1997 and 1999): As-contents in gneisses, schists and amphibolites,

C: Soils and glacial sediments Ticino, 0 - 20 cm, 45 cases)

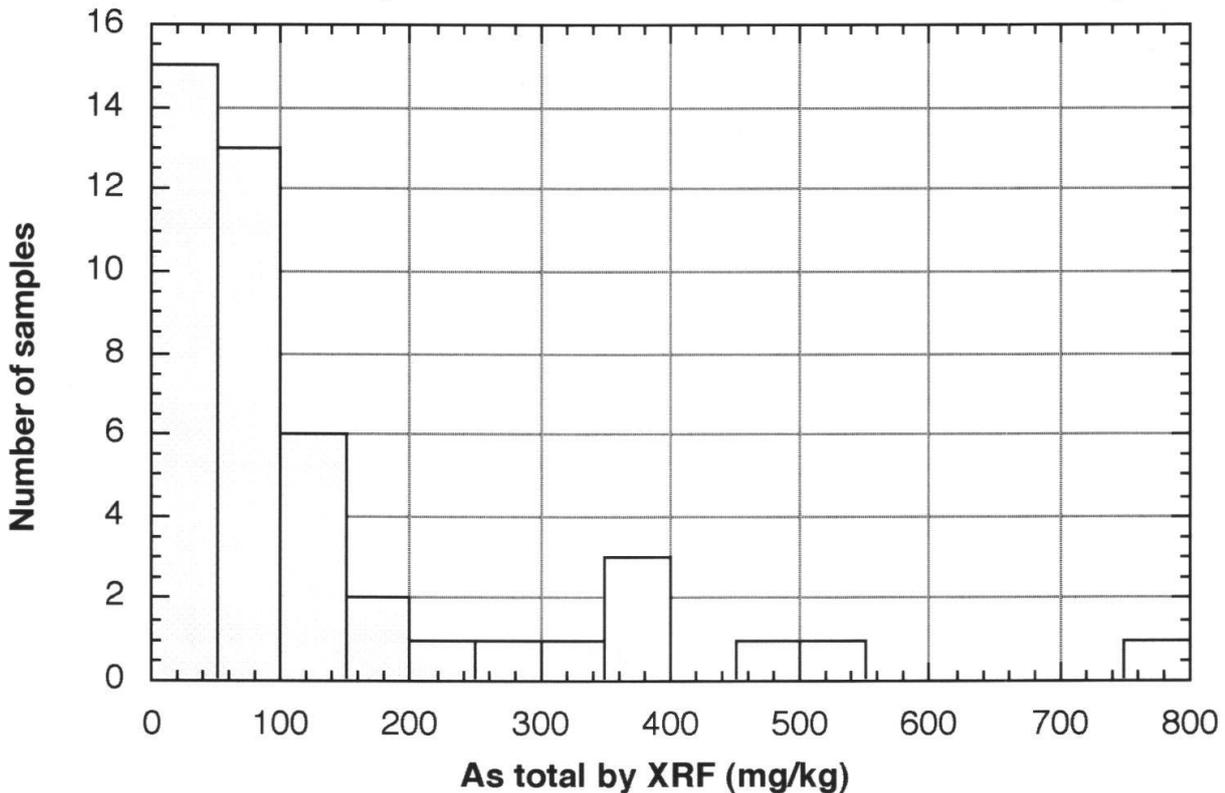


Fig. 5c: As-contents in soils and glacial sediments from the Sottoceneri area (mainly Malcantone).

solid phases. Such reducing conditions commonly prevail in slowly flowing or stagnant ground waters, possibly combined with the presence of organic material (peat or poorly decomposed wood). Inhomogeneous river and delta sediments close to lakes favor such conditions. In the following two chapters, more details are given on the arsenic-rich waters and their possible origin for the different areas.

3. MALCANTONE AREA

Situated to the west of Lugano (Fig.1) this area comprises essentially the catchments of the N-S flowing Magliasina and Lisora rivers (Fig. 6, 7). Its numerous ore deposits have locally been mined for gold and iron and have produced mine dumps of limited extension which are often enriched in arsenic. The most prominent examples are the mines of Costa- Astano and of Torri-Breno (Steiger 1999, Pfeifer et al. 2000). In their vicinity, locally high As-contents have been observed in the soils and creeks (Pfeifer & Rey 1998, Pfeifer et al. 2000, Girardet 2001). The fact that there are As-rich springs outside the mining area (e.g. N of Astano), shows that these ore deposits only have an indirect influence (Fig. 8). By and large the most important source of arsenic in soils and waters seem to be the local glacial sediments. Typical concentrations in unconsolidated glacial and river sediments vary between 40 and 150 mg/kg, the maximum lying around 500 mg/kg (cf. Fig. 5C). Locally an organic soil horizon at 60 cm depth contained up to 750 mg/kg (Girardet 2001). Köppel (1966) and Berthoud (2001) described mineralized erratic blocks from glacial sediments in the Pura area. This, together with the high arsenic contents of the fine-grained till material, indicates that the weathering and erosion by

the local glaciers during the last few 100'000 years is at the origin of this As-contamination, and that it is largely natural.

The relative percentages of As(III) and V in these waters are rather constant. Of the 15 springs checked, all were oxidizing and composed of more than 99% As(V) (cf. Tab. 1 and Jäckli 1998). For mining waters, only for two cases the partitioning of arsenic among the two oxidation forms are known (Astano-Bolle, Astano-Costa-Beredino). Since these waters often issue from sulfide containing materials which create reducing conditions (Eh below 100 mV), As(III) can make up to 10% of the total As (no. 6 in Tab. 1).

Not only soils, river sediments and waters contain arsenic, also the vegetation is slightly enriched: the water mosses in the Magliasina river contain between 10 and 50 mg/kg As, grass of the Croglio area contains up to 5 mg/kg and plants growing on the mine dump of Costa-Astano (Dandelion, birch trees, Ling heather) contain up to 10 mg/kg. Mosses growing on rotten wood in the forest of the Malcantone are also enriched in arsenic compared to their equivalents in other part of Switzerland (BUWAL 1993): they contain up to 3 mg/kg. Since mosses do not have roots, this can only be explained by the fact that also the dust, transported by the wind, is enriched in arsenic, due to the natural enrichment of the whole area. Elevated contents in rain water point in the same direction (Pfeifer et al. 2000).

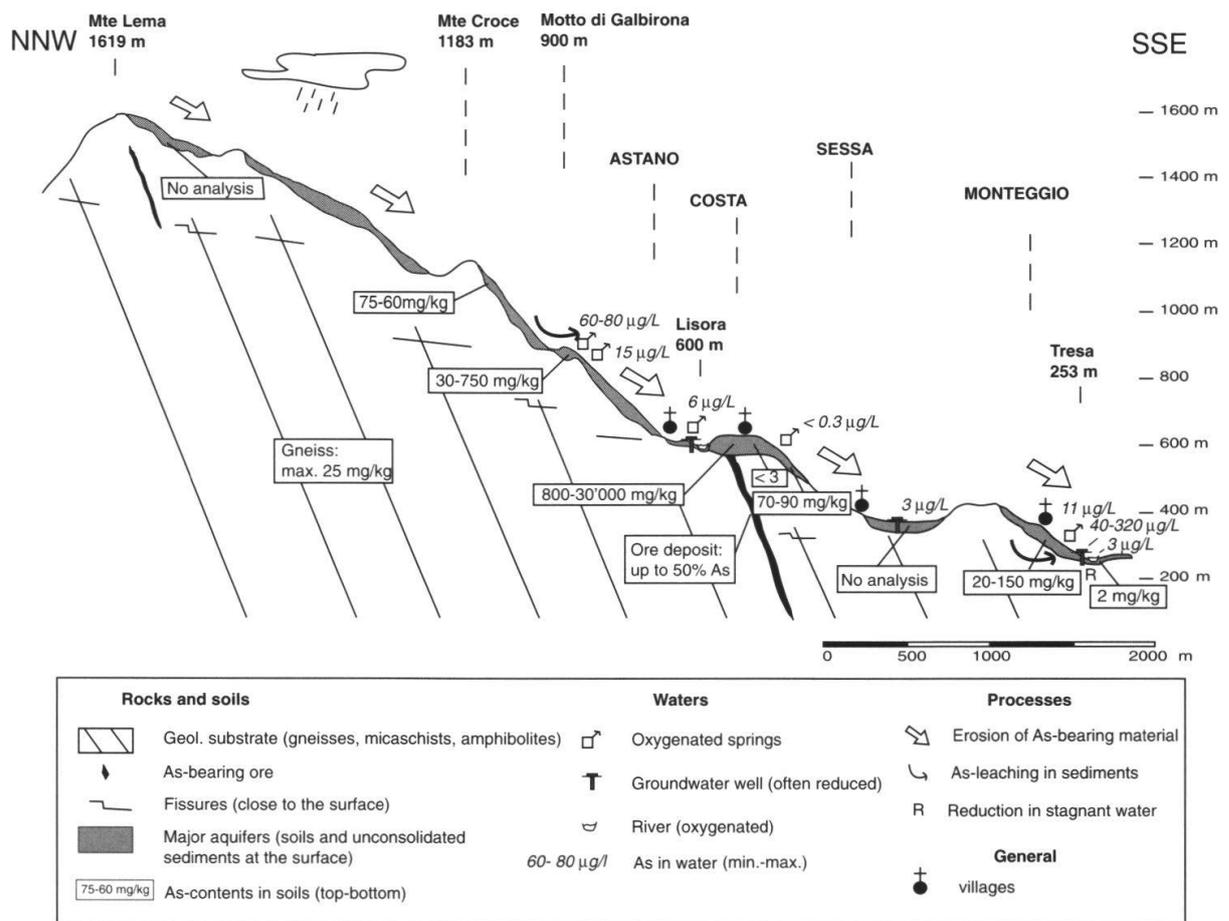


Fig. 8: Cross-section of the Malcantone area, schematically summing up the different situations in which arsenic has been found in rocks and waters: waters rich in As are not directly linked to ore deposits. On slopes oxygenated waters with medium contents dominate, whereas in locally reduced ground waters related to river plains high As-values can be found.

4. VAL ISONE/ VAL COLLA AREA

Although this area partly lies in the same geological rock formation as the Malcantone, it is almost exempt of ore deposits: there are only two known iron mineralizations: pyrrhotine-quartz at Medeglia, and pyrite-arsenopyrite-Fe-dolomite in Val di Bogh, W of Cima di Lago (Fig. 9, cf. also Köppel 1966, p.111). However, the arsenic contents in waters and soils are almost as high as in the latter region (Fig. 9, Schmit 2000).

Glacial deposits are relatively rare, instead, the area is characterized by large deforested and subalpine meadow areas, which cover locally derived rock debris with a thin soil layer on top. The underlying silicate rocks contain very variable concentrations of arsenic (8 to 3'000 mg/kg, the latter found at the bottom of a soil profile at Alpe Rompiago in Val Colla (Schmit 2000). From this, one can conclude that the dominant aquifer is of type 1, i.e. the arsenic must be derived from dispersed sulfide and oxide minerals in the rock. Figure 10 illustrates with the situation in Val Colla, how rain water becomes spring and river water enriched in arsenic, changing systematically its overall loading (conductivity), pH and chemical composition. Another interesting site is located above the village of Camignolo (at the entrance to Val Isonne), where spring water enriched in arsenic (30 to 50 µg/l) flow down the hill slope formed of debris and covered with a soil cover ranging from 20 to 100 cm in depth. Becoming more and more organic this soil shows increasing As-contents from 10 to 400 mg/kg going down the slope. Since the touched area is limited to the area below the springs, it is obviously a zone of secondary As-enrichment (Temgoua et al. 2002). Also in this case, the known cases contain more than 99% of the arsenic as As(V) (analysis 5 and 8, Tab. 1).

Figure 11 schematically illustrates the situation at the iron mine W of Medeglia in the canyon of the Vedeggio-river. Rain and hill slope water enters the old mine adits, gets slightly reducing and enriched in Fe and As and is discharged to the Vedeggio-river. In view of the high discharge of the latter, the few liters of local water rich in arsenic (230 µg/l) do not seriously contaminate the river (analysis no. 7 and 3, Tab. 1). No data is available on sediments downstream.

5. OTHER AREAS

5.1 Introduction

For other areas outlined in figure 1 only limited data is available, i.e. mainly water analyses (Jäggli 1997). Although the geological situation is quite well known, the exact origin of the arsenic needs to be explored in more detail. In the following section we briefly give an overview over the present state of knowledge.

5.2 Barbengo- Figino area

Situated in the delta area of the Scairolo river, up to 1999, the main drinking water of the community of Barbengo stemmed from a 30 m deep well, situated about 50 m from the lake shore close to the village of Figino (schematically shown on Fig. 3). When pumped, the water was rich in Fe(III)-hydroxide with total As-content of 230 µg/l. Upon treatment to de-iron the water and to make it drinkable, its As-content was brought down to 24 to 32 µg/l. When discovered in 1996, despite the fact that a

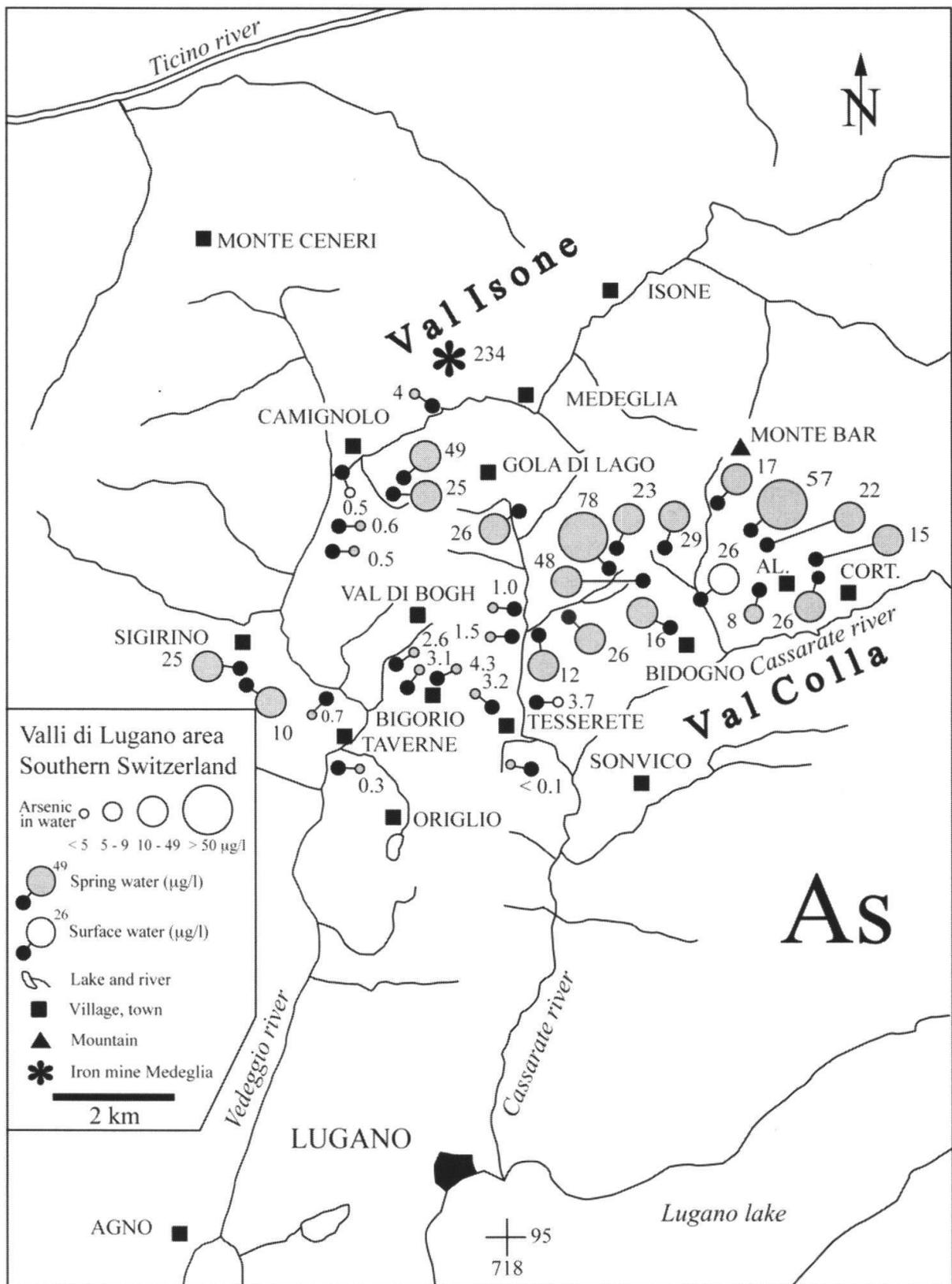


Fig. 9: Arsenic contents in waters of the Valli di Lugano area (Val Isonne- Val Colla). The concentrations are comparable to the Malcantone area, however, Fe-ore deposits which could be at the origin of the elevated arsenic contents are almost absent (only case: pyrrhotine mine of Medeglia). Abbreviations: Cort. Corticiasca, Al.: Albumo.

further treatment would have been possible, the community decided to close the well and now treats lake water to provide for drinking water. Before the well had been constructed in the 70es, in the neighborhood, stagnating CH₄-or H₂S-rich water had been found, indicating reducing conditions, probably related to the proximity of the lake (ROV 1972).

Before the well was closed, the arsenic speciation of the water had been measured: 76 µg/l As(V) and 135 µg/l As(III) (64 % of the total As, analysis 9 in Tab. 1), clearly indicating reducing conditions. Recent short term pumping in a another well of the area only yielded uncontaminated water, indicating a relatively deep origin of the high-arsenic water (30 to 50 m depth). A possible source of the arsenic are the Permian volcanic rocks which outcrop in the area (typical As-contents between 7 and 72 µg/kg). Together with the dolomitic rocks which create a high pH and reduced environment at depth, the two might be responsible for the liberation of arsenic to the ground water.

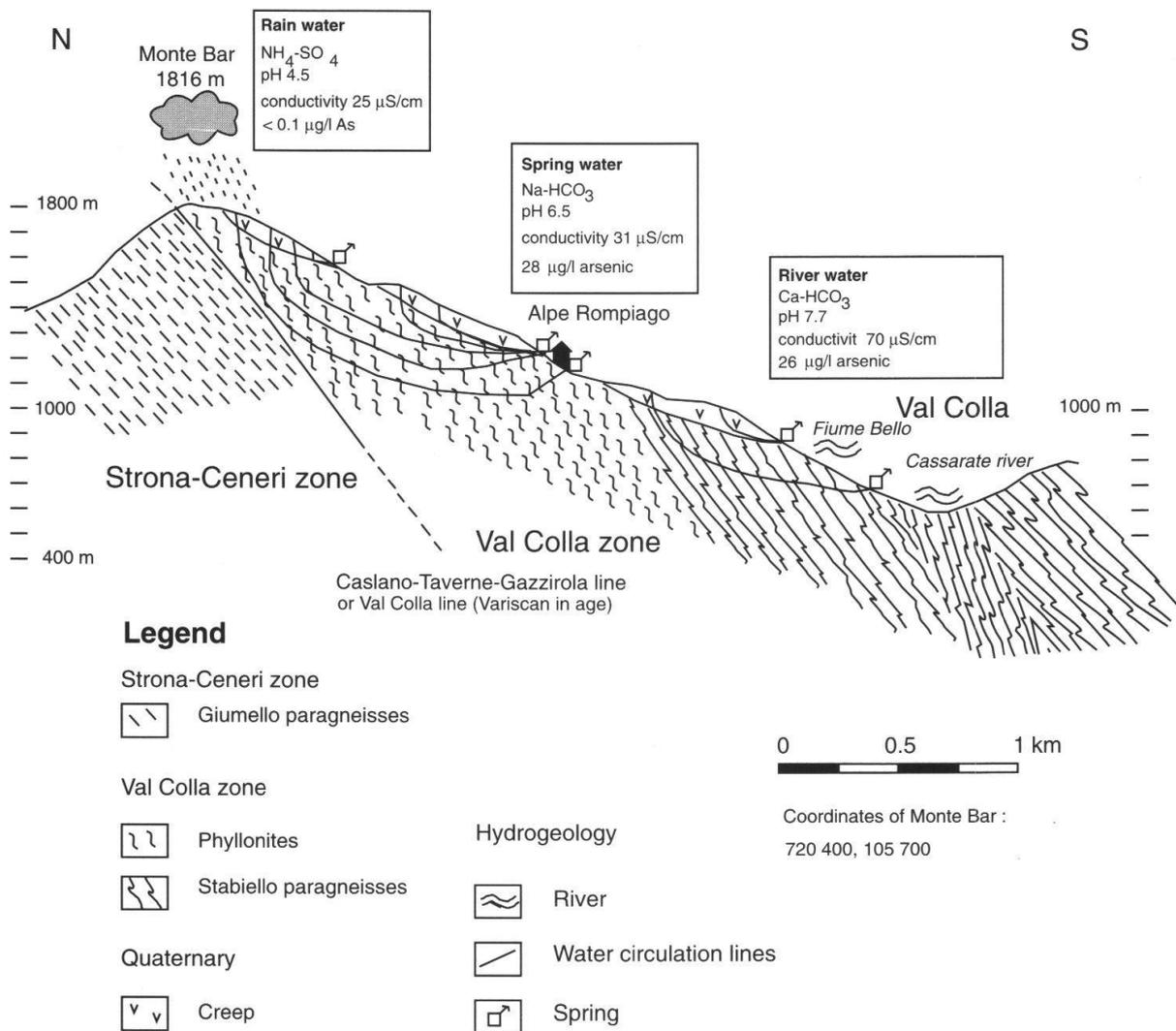


Fig. 10: Schematic hydro-geological cross of the Val Colla area (after Schmit 2000). It shows the systematic changes from rain to soil percolation and river water: the conductivity reflecting the total dissolved matter increases systematically downhill, so does the pH. Once a certain arsenic content is achieved (around 30 µg/l), it remains fixed, however the proportions of other ions change (evolution from a Na-SO₄-type to Ca-HCO₃-type water).

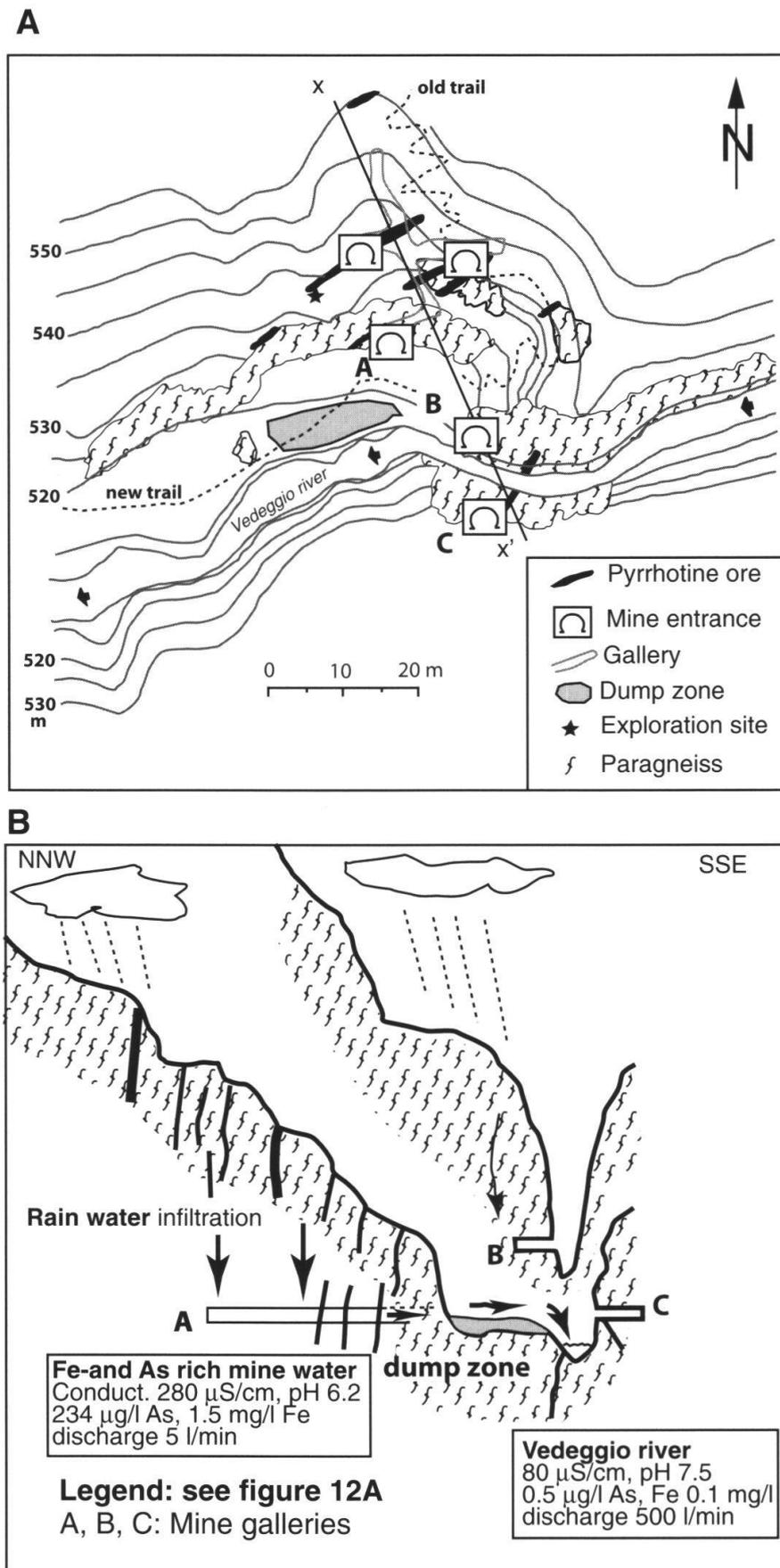


Fig. 11: Example of the release of arsenic by mine galleries and dump material: case of the pyrrhotine mine W of Medeglia (geology updated after Fehlmann 1919 and Kündig et al. 1990).

5.3 Gambarogno area

Situated at the northern end of the Malcantone- Veddasca area, the presence of elevated As-contents in ground waters is not astonishing. In one case, at Vira, in a well, in conjunction with a heating pump, which had been used to heat a school (pers. comm. R. Bregy), arsenic-rich reduced waters have been found, containing about 40 µg/l. The abundant solid Fe(III)hydroxide colloidal phase present, which often blocked the pump, contained 2.3% of arsenic. However, the drinking water of the community has values as low as 2 µg/l As (Jäggli 1997). We suppose that the water belongs to a stagnant fissured aquifer close to the shore line of the lake.

5.4 Val Tresa

In several piezometer tubes along the Tresa river between Ponte Tresa and Monteggio, reducing, Fe-rich waters, again with arsenic values of up to 300 µg/l have been found (Balestra 1996, analysis no. 9 in Tab. 1). The aquifer is of fluvio-glacial origin and rather inhomogeneous, favoring locally reducing conditions (EAWAG 1975, Berthoud 2001). In addition, closeby, there is an important unlined waste repository situated in one of the former gravel quarries SE of Croglio (Nawratil 1997, Balestra 1997), which could possibly create a plume with elevated pH and reducing conditions. Close to this, at low discharge of the Tresa, a seepage zone enriched in ferrihydrite is clearly visible, containing about 0.5% As. Further down the valley, occasional monitoring during the last two years did not bring evidence that the temporarily high As-contents in some of the piezometer tubes (analyses 9 - 11 in Tab. 1), were related to the waste repository, nor that there was a moving As-rich plume present (as we expected originally). The arsenic seems to stem from the adjacent fluvioglacial and glacial deposits (As content up to 150 mg/kg, Berthoud 2001) and seems to flow almost perpendicular from the hills through the river bank towards the Tresa river (Fig. 6 and 7). Under oxidizing conditions, the water only contains around 40 µg/l As, but when stagnating and getting reduced in the vicinity of the steel piezometer tubes, it can contain up to 300 µg/l with about 60 % As(III) (Tab. 1). Fortunately, the main drinking water well for the communities of the area, which lies only at a distance of about 300 m from the contaminated piezometers, has less than 0.5 µg/l As, probably because it drains the river Tresa through the alluvial sediments.

5.5 Morcote area

The only known fact about this area is that local drinking waters contains elevated arsenic contents (29 µg/l As, Jäggli 1997) and that the area is dominated by Permian volcanic rocks. For the moment we assume that the aquifer is of type 1 (cf. Fig. 4-1).

6. CONCLUSIONS

As in many other countries, also in Switzerland, after the discovery of the threat related to anthropogenic lead and cadmium, a new one is coming up, that of natural arsenic. The systematic check in southern Switzerland has revealed several areas in which the concentrations of arsenic in rocks, soils and waters are higher than re-

Limiting value: 50 µg/L

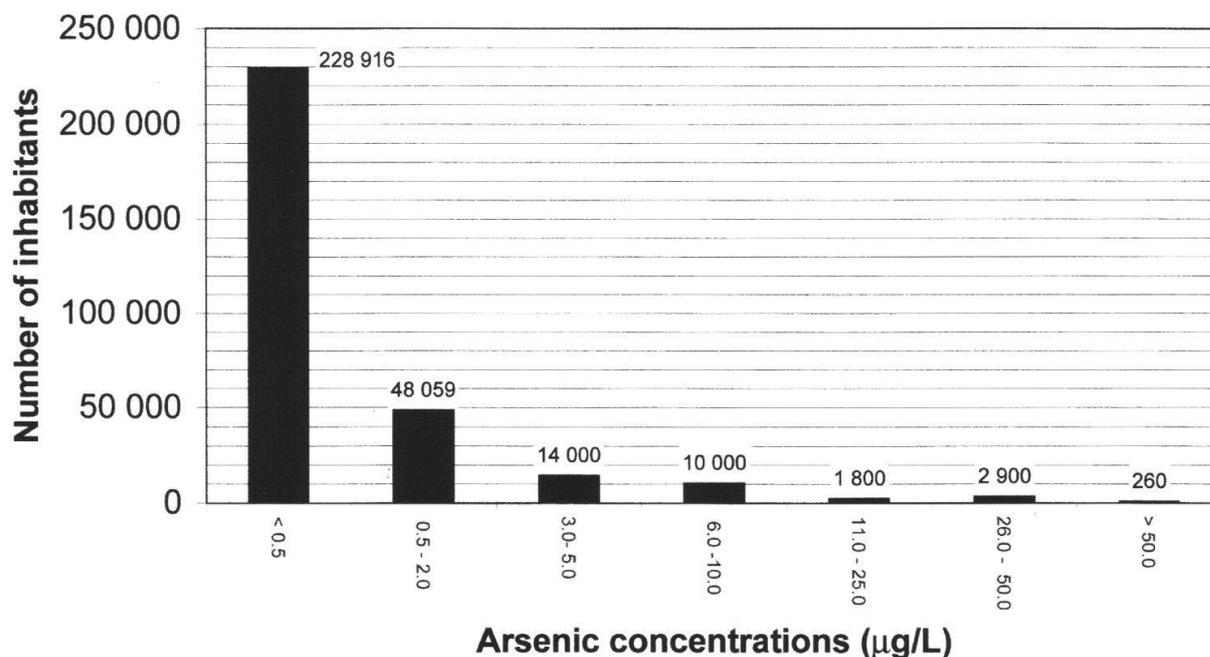


Fig. 12: Number of people touched in the canton of Ticino by elevated arsenic contents in drinking water.

commended guide line values. We estimate that in Southern Switzerland and Northern Italy, at least 6'000 people are touched by this phenomenon (Fig. 12). For the moment the situation is not alarming, but certain spring waters will need an arsenic-removal treatment if one wants to continue to use them for drinking water and new springs or wells will have to be tested for arsenic before they are put in use. Techniques to remove arsenic from waste and drinking waters are in principal known (EPA 2002, Hug et al. 2000), but in detail, the ideal, low-cost, low maintenance procedure has not yet been found (Jäckli 1999a & b). Since the cost-efficiency ratio of such systems, especially for these often small water quantities (typically 100 l/min) is often big, the actual tendency of the touched communities is to shut down the contaminated springs and wells and look for new uncontaminated water in collaboration with the neighbor community.

Provisional results on the vegetation growing in the touched areas point towards increased values in the food chain as well (Nawratil 1998, Pfeifer et al. 2000, Girardet 2001), however this subject will certainly need more research.

Acknowledgements

This research has been supported by the grants no. 21-52758.97 and 2000-06860 of the Swiss National Science Foundation. We thank R. Martini, M. Pillet and U. Balestra all at Geneva, for allowing us to use their data on the Magliasina river and the Tresa area respectively.

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