

# Critical review of acid in situ leach uranium mining: 2. Soviet Block and Asia

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**Abstract** The technique of in situ leach (ISL) uranium mining is well established in the USA, as well as being used extensively in Eastern Europe and the former Soviet Union. The method is being proposed and tested on uranium deposits in Australia, with sulphuric acid chemistry and no restoration of groundwater following mining. ISL mines in the former Soviet Union generally used acid reagents and were operated without due consideration given to environmental protection. At many former mine sites, the extent of groundwater contamination is significant because of high salinity, heavy metal and radionuclide concentrations compared with pre-mining and changes in the hydrogeological regime caused by mining. After the political collapse of the Soviet Union by the early 1990s, most uranium mines were shut down or ordered to be phased out by government policy. Programmes of restoration are now being undertaken but are proving technically difficult and hampered by a lack of adequate financial resources. The history and problems of acid ISL sites in countries of the former Soviet Union and Asia are presented in this study.

**Keywords** Acid · In situ leach · Solution mining · Uranium plumes

## Background

The unconventional mining technique of in situ leach (ISL) is now the primary producer of refined uranium in

the United States, with a market share of around 95% in the mid-1990s (DoE 1999). ISL mines appear set to assume a greater role in Australia's uranium industry. The commercial ISL uranium mines in the USA use alkaline chemistry compared with the proposed projects in Australia, which are based on the use of acid (Mudd 1998). In contrast, the ISL uranium mines of the former Soviet Union and Eastern Europe have primarily used sulphuric acid, with apparently little consideration given to environmental concerns during operation.

The first trials of uranium ISL were both developed in the USA and the Soviet Union in the early 1960s. It is uncertain who developed the concepts or if they were developed separately (Mudd 1998). The use of ISL uranium mining continued to expand until the collapse in the late 1980s. It is worth noting that an unnamed Russian first suggested ISL mining of gold by as early as 1896 (Mineev and Shutov 1979; Morris 1984).

The majority of countries with uranium mining under the influence or control of the former Soviet Union have undertaken ISL projects, although different countries had contrasting success, from an operational perspective. Bulgaria, for example, experienced a major shift in uranium production from conventional to ISL mines, dramatically reducing the workforce and exacerbating already recalcitrant environmental problems.

Since the reunification of Germany and the collapse of the Soviet Union, the extent of the contamination of groundwater is beginning to come to light, mainly through co-operative programmes of the International Atomic Energy Agency (IAEA), although other agencies are becoming involved. There is also increasing interest in the use of ISL methods for the mining of low-grade uranium ores in other parts of Asia, most notably China and Pakistan.

The resurfacing of the Australian acid ISL uranium mine proposals in 1996, the lack of acid ISL mines in the USA, and the research coming to light through the IAEA concerning the extent of impacts from acid mines in the Soviet block, led to a broad-based review of ISL uranium mining by the author, completed in 1998 (Mudd 1998).

The history and experience of acid ISL in countries controlled by the former Soviet Union is reviewed herein, with sites in Australia and the United States reviewed in a companion paper (Mudd 2001). A compilation of leaching solution quality for most ISL mines reviewed in this paper is compiled in Table 1.

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**Table 1**Leaching solution and groundwater quality at various Former Soviet Union ISL mines (mg/l, except redox in mV and  $^{226}\text{Ra}$  in Bq/l). *T* Total

|                              | Bulgaria      | Stráz Pod Ralskem,<br>Czech Rep. | Königstein, eastern<br>Germany |       | Former Soviet Union |             | Bukinaii,<br>Uzbekistan | Yining, China     |
|------------------------------|---------------|----------------------------------|--------------------------------|-------|---------------------|-------------|-------------------------|-------------------|
|                              |               |                                  | Leach                          | GW    | Acid                | Alkaline    |                         |                   |
| TDS                          | 15,000–20,000 | 50,000–100,000                   |                                |       |                     |             | 33–45                   |                   |
| pH                           | 1.4–2.0       | 0.5                              | 1.9                            | 6.7   |                     |             | 1.0–2.0                 | 1.26              |
| Redox                        |               |                                  |                                |       |                     |             |                         | +652              |
| Acid <sup>a</sup>            |               | 15,000–38,000                    |                                |       |                     |             |                         |                   |
| SO <sub>4</sub>              | 10–12         | 33,000–80,000                    | 1,500                          | 4.3   | 17,000–25,000       | 2,000–3,000 | 46,600                  | 22,800            |
| S <sup>2-</sup>              |               |                                  |                                |       |                     |             |                         | 2.72              |
| CO <sub>2</sub> <sup>b</sup> |               |                                  |                                |       |                     |             |                         | 0.99              |
| HCO <sub>3</sub>             |               |                                  | 5                              | 12    |                     | 500–2,500   |                         |                   |
| NH <sub>4</sub>              |               | 1,000–2,000                      |                                |       |                     | 400–600     | 533                     |                   |
| NO <sub>3</sub>              |               | 200–1,400                        | 63                             | 8.4   |                     |             | 750                     |                   |
| Cl                           |               |                                  |                                |       | 400–600             | 500–1,200   | 300                     | 762               |
| F                            |               | 100–300                          |                                |       |                     |             |                         | 5.2               |
| P                            |               | 50–150                           |                                |       |                     |             |                         |                   |
| PO <sub>4</sub>              |               |                                  |                                |       |                     |             | 580                     | 22                |
| Ca                           | 140–600       | 200–300                          | 201                            | 6.6   | 400–600             | 700–800     | 730                     | 17                |
| Mg                           | 140–330       | 20–30                            | 11.5                           | 1.1   | 300–500             | 100–300     | 2,400                   | 140               |
| Na                           | 30–900        | 10–15                            | 49                             | 3.9   | 50–150              | 200–750     | 650                     | 172               |
| K                            | 30–200        | 40–70                            | 3.1                            | 1.5   | 25–100              | 25–400      | 570                     | 28                |
| Al                           | 310–840       | 4,000–6,000                      |                                |       | 500–800             |             | 2,860                   | 323               |
| Fe <sup>(T)</sup>            | 700–2,200     | 500–1,500                        |                                |       |                     |             |                         |                   |
| Fe <sup>2+</sup>             |               |                                  |                                |       | 400–1,000           |             | 3,300                   | 377               |
| Fe <sup>3+</sup>             |               |                                  |                                |       | 800–1,500           |             | 1,500                   | 440               |
| Mn                           | 6–61          |                                  |                                |       |                     |             | 100                     | 8.4               |
| SiO <sub>2</sub>             | 210–350       | 100–200                          |                                |       |                     |             | 560                     |                   |
| As                           |               |                                  |                                |       |                     |             |                         | 0.1               |
| Ba                           |               |                                  |                                |       |                     |             | 2.05                    |                   |
| Ce                           |               |                                  |                                |       |                     |             | 8.35                    |                   |
| Cd                           |               |                                  |                                |       |                     |             |                         | 0.04              |
| Co                           |               |                                  |                                |       |                     |             | 5.6                     |                   |
| Cr <sup>(T)</sup>            |               | 5–15                             |                                |       |                     |             | 4.05                    | 0.74 <sup>c</sup> |
| Cu                           |               |                                  |                                |       |                     |             | 1.07                    | 0.5               |
| La                           |               |                                  |                                |       |                     |             | 3.15                    |                   |
| Li                           |               |                                  |                                |       |                     |             | 6.2                     |                   |
| Mo                           |               |                                  |                                |       | 10–40               |             |                         | 2.4               |
| Nd                           |               |                                  |                                |       |                     |             | 4.85                    |                   |
| Ni                           |               | 20–30                            |                                |       |                     |             | 12.3                    | 1.3               |
| Pb                           |               |                                  |                                |       |                     |             | 1.13                    | 0.67              |
| Rb                           |               |                                  |                                |       |                     |             | 8.0                     |                   |
| Re                           |               |                                  |                                |       | 0.2–0.5             |             |                         | 3.1               |
| Sb                           |               |                                  |                                |       |                     |             |                         | 0.4               |
| Sc                           |               |                                  |                                |       | 0.15–0.6            |             |                         | 1.86              |
| Se                           |               |                                  |                                |       | 50–60               |             |                         | 0.001             |
| Sr                           |               |                                  |                                |       |                     |             | 8.2                     |                   |
| Ti                           |               |                                  |                                |       |                     |             | 3.44                    | <1                |
| V                            | 1–18          | 10–15                            |                                |       | 10–40               |             | 2.65                    | 4.8               |
| Y                            |               |                                  |                                |       |                     |             | 5.67                    |                   |
| Zn                           | 2.1–7.3       |                                  |                                |       |                     |             | 17.5                    | 0.88              |
| $^{226}\text{Ra}$            | 1–2           | 50–90                            |                                |       | 3.7                 |             |                         |                   |
| U                            | 5–30          | 20–500                           | 46.9                           | 0.009 |                     |             | 86                      | 75                |
| Rare<br>earths               |               |                                  |                                |       | 10–40               |             |                         |                   |

<sup>a</sup>Free acid as H<sub>2</sub>SO<sub>4</sub><sup>b</sup>Free CO<sub>2</sub><sup>c</sup>Cr<sup>6+</sup> 0.1

## Bulgaria

This review is based on Tabakov (1992), Kuzmanov and others (1992), Vapirev and others (1993), Dimitrov and Vapirev (1994), Nedyalkov (1996) and IAEA (1999); more detail is given in Mudd (1998).

The ISL technique was first applied in 1967 to low-grade deposits (ranging from 0.006 to 0.03% U) at Orlov Dol and

Selishte (a former conventional mine). Given the success of these sites, a revolution was perceived whereby many previously uneconomic deposits were exploitable. By 1990, the share of uranium production from ISL had risen to 70%.

All uranium mining and milling in Bulgaria was closed down by government decree on 20 August 1992. Activity since has been aimed at cleaning up and rehabilitating the

numerous mine sites. The total contaminated area as a result of all uranium industrial activity is approximately 20 km<sup>2</sup>, including 6 km<sup>2</sup> from ISL mining and 4 km<sup>2</sup> of contaminated forest.

There has been a total of 19 sites where ISL has been applied, and a further 11 sites where the ISL technique was applied within an underground mine. Most of these sites began operation in the late 1960s to early 1970s, although poor results from initial trials meant some sites were discontinued. The mine sites are all concentrated in the southern and western regions of Bulgaria, shown in Fig. 1. The deposits contain high amounts of organic matter, iron and sulphides.

The ISL mines had a dramatic impact on the workforce in conventional mines, falling from 5,000 workers between 1965 and 1970 to approximately 500 in 1988. The initial chemistry used was sulphuric acid, although this was later switched to sodium-carbonate and ammonium-carbonate leaching chemistry in deposits with a high carbonate content.

The environmental assessment, siting and operation of many uranium mining operations across Bulgaria were often neglected to enable fast tracking of projects and minimize the costs involved in establishing a project. Only one mine was closed because of contamination of drinking water. Almost no preventative or counter measures were implemented during the whole period of mining for the environmental protection of water, soil and air from mechanical, chemical and radioactive pollution. The secrecy of the uranium and nuclear industry was identified as a key reason behind this philosophy.

The leaching of uranium was generally progressed in three stages – first, acid was introduced at levels up to 10 g/l (lasting for a few months); second, the base period of acid leaching at 4–6 g/l (lasting for over 2 years); and third, the closeout period at 0.5–1 g/l. The overall time for a well field was between 3 to 5 years and the recovered uranium was about 60 to 80%. The Ra levels of leaching solutions was generally low. A compilation of typical leaching solution chemistry, summarized from 13 ISL mine sites, is shown in Table 1.



Fig. 1

Location of in situ leach (ISL) uranium mines in Bulgaria (Tabakov 1992)

The pre-mining quality of groundwater in the ore zone aquifers was typically 500–2,000 mg/l TDS, pH of 6.3–8.8, iron 0.1–648 mg/l, and sulphate 24–758 mg/l; which is, good quality groundwater.

There has been significant contamination of groundwater at most ISL sites, with major concerns arising from chemical, radiological and bacterial contamination. For the combined underground-ISL sites, Fe, Cu, Co, Ni, Mo, As and some rare earth elements are several times higher than allowable limits. In the Deveti septemvri ISL mine, Mo reaches 13.4 mg/l (regulatory limit 0.5 mg/l, 27 times higher), and Mn reaches values up to 13 mg/l in groundwater and 4.2 mg/l in the retention pond; quantities of B and Hg have also been detected.

At the Orlov Dol site, after 6 years of monitoring, the acidity of the ore zone aquifer declined from 1,300 to 10 mg/l, although the groundwater still contained elevated levels of uranium despite the associated small increase in pH.

The concentration of sulphate can be very high in surface waters and even in water supply wells of private owners as a result of accidental spilling of solutions at ISL sites. The average chemistry of contaminated groundwaters ranges in TDS from 15–20 g/l, SO<sub>4</sub> is from 10–12 g/l, U is between 5 and 20 mg/l, other salts and the presence of heavy metals and rare earths.

For the Cheshmata (Haskovo) site, in the valley downstream, SO<sub>4</sub> concentration is 1,400 mg/l (limit 300 mg/l), free sulphuric acid 392 mg/l and the pH is 2.2 (over 1,000 times more acidic than the surrounding aquifer). The private wells of residents of the area have also been affected with significantly high concentrations of SO<sub>4</sub> being noted, demonstrating that the leaching solutions have migrated into drinking water supplies.

A similar case has been recorded in Navusen where, in a valley, the SO<sub>4</sub> concentration is 13,362 mg/l and almost 5 g/l of free sulphuric acid, indicating the water is actually a leaching solution. The groundwater quality of such sites has a TDS (salinity) level of greater than 20 g/l, of which SO<sub>4</sub> is 12–15 g/l. Heavy and rare earth elements were detected in some cases, such as V, W, Mo and La, as a result of recycling of the solution.

There were also noted problems because of bacterial contamination, although their exact effects were not able to be predicted and were not studied. It was thought that they were beneficial in the leaching process.

There remains concern that solutions at the various ISL sites could contaminate the deeper groundwater systems, as well as the shallow systems. For the deep systems, which contain the ore being mined, the U content can reach 20–30 mg/l and Ra 1–2 Bq/l, with U content in shallow aquifers of around 3–4 mg/l and Ra 0.5 Bq/l, despite dilution effects during migration through the aquifer.

At some sites, where there were surface spills because of failure of distribution pipes, the U and Ra content of soils is 10 and 2–3 times the background level, respectively.

At most of the ISL sites undergoing restoration, the solutions were continually recycled through the mined aquifer without adding acid, and this led to deposition of

salts within the pipes. These salts contained increased and significant levels of radioactivity. This process has been now been stopped.

A principal problem of the restoration work currently in progress is that the environmental requirements are quite strict, making uranium production unprofitable. It was argued that achieving an acceptable level of environmental protection required that preventative measures, planning and funds are set aside during the early stages of a project, and during the operational phase of a particular project. As this was not done, the necessary funds are not available and restoration work is thus significantly impaired. Only one-third of the land used by ISL operations has been remediated, and as the land will be returned to the original owners for agricultural purposes, there are real concerns for public health and environmental safety. Some of the ISL sites (such as Bolyarovo, Tenevo, Okop and Gorna Trakiiska nizina) are close to areas where potable quality groundwater is extracted by local communities or the groundwater is considered to be an important future water resource.

## Czech Republic

This review is based on Benes (1992), Fiedler and Slezák (1992), Khün (1992), Anděl and Pribán (1993, 1994), Tomas (1996) and IAEA (1999); more detail is given in Mudd (1998).

The Bohemian Massif mining district in the Czech Republic, shown in Fig. 2, has been an important source of uranium for the Russian military and nuclear power programmes, with uranium ore from the Jáchymov mine being used to manufacture the first Soviet atomic bomb. A total of more than 100 uranium mines were developed, including shallow investigation mines.

The Stráz Pod Ralskem district consists of sandstone-type uranium deposits, and acid ISL has been applied as the mining method since 1968 after successful trials during 1967. The associated mineralization is also unusual, with zircon, titanium and phosphorous present.

The Stráz region is characterized by complex hydrogeological and biological conditions that are unfavourable to

the application of ISL, primarily related to the slow dissolution rates of uranium. This causes two principal problems – firstly, large doses of chemicals are required (sulphuric acid at about 5% with nitric acid and nitrate as the main oxidants); and secondly the leaching periods are very long, ranging from 15 to 25 years. The total uranium production by ISL was 16,470 t  $U_3O_8$ .

The Hamr deposit was developed at the Hamr and Luzice mines with both underground and combined ISL techniques. The Hamr mine is only 5 km from the Stráz mine, exacerbating the technical problems at both sites, which led to higher production costs and greater environmental impacts. A principal problem for many of the sites is the density of population across the Czech Republic, with 40,000 people living near the Stráz mine, for example. After detailed evaluation of the negative impact of uranium mining and milling, a progressive programme of declining production from uranium mining has been adopted and an extensive remediation programme implemented by the Czech Government.

### Stráz Pod Ralskem

The hydrogeology of the Stráz region is complex, but can be thought of as two distinct aquifers – the Cenomanian and the Turonian. The Cenomanian is a deep, confined and artesian aquifer, and the Turonian lies above this, separated by up to 100 m of thick low permeability clays and siltstones. A hydrogeological cross section is shown in Fig. 3. The Turonian is designated as an important high quality drinking water reserve with a calcium bicarbonate ( $CaHCO_3$ ) type of water quality, and is known to discharge to the Ploucnice River at about 40 l/s. The Cenomanian aquifer was known to contain elevated levels of radium. For the Stráz deposit, every tonne of uranium (t U) produced:

- 274 t of sulphuric acid injected;
- 7.9 t of ammonia injected;
- 19.3 t of nitric acid injected;
- 1.79 t of hydrofluoric acid injected;
- 0.95 t of sulphuric acid released to the air;
- 1.18 t of nitrous oxides released to the air;
- 53 GBq of radium released to the air;

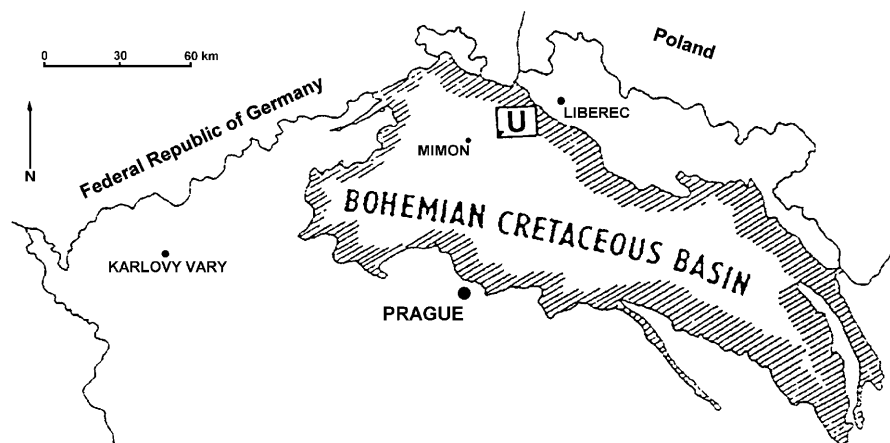


Fig. 2

Location of the Bohemian mining district, Czech Republic (Fiedler and Slezák 1992)

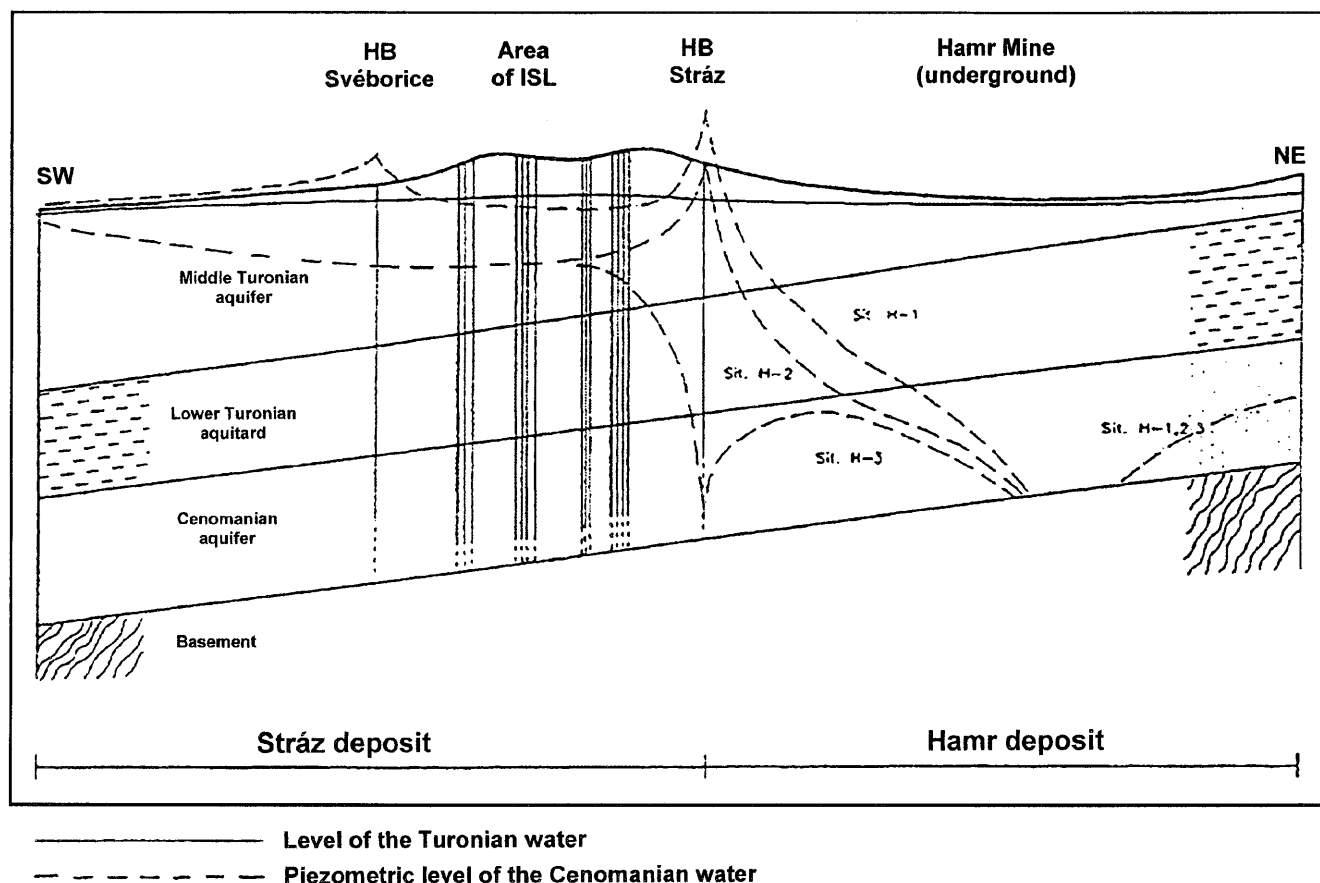


**Fig. 3**  
Hydrogeological cross section of the Stráz Pod Ralskem and Hamr uranium mines, Czech Republic (Fiedler and Slezák 1992)

- 7,260,000 l of contaminated groundwater in the Cenomanian aquifer;
- 1,500,000 l of contaminated groundwater in the Turonian aquifer.

By contrast to experience in the USA, the Stráz ore deposit required 50–70 g/l of sulphuric acid and a leaching period of 15–20 years to reach a yield of 60–80% of the uranium because of the lower permeability of the aquifer materials. By 1994, a total of 32 ISL sites had been commissioned covering a total of 6 km<sup>2</sup> and consisting of 7,000 wells. The Stráz mining district has seen approximately 3,800,000 t of sulphuric acid, 270,000 t of nitric acid, 103,000 t of ammonia and 25,000 t of hydrofluoric acid injected into the well fields. Typical leaching solution chemistry is shown in Table 1. The interactions between the leaching solutions and aquifer sediments are not well defined, and the speciation of many heavy metals and radionuclides remains unstudied. The Stráz site ceased producing uranium on 1 April 1996.

The leaching solutions from the Stráz well fields were not operated with a bleed system to maintain a cone of depression around active well fields and this led to solutions being dispersed widely through the Cenomanian aquifer in the area, as well as vertically into the Turonian aquifer. The

excursions mainly occurred through production bores, but significant excursions also occurred at liquid waste disposal bores. A location plan of contaminated groundwater at the Stráz and Hamr mines is shown in Fig. 4.

The contaminated water in the Turonian aquifer alone is spread over 245 ha (43% of the area of the well fields). A total of 200 billion l of groundwater has been affected, covering a total area of 6 km<sup>2</sup> and the volume of aquifer material affected is thought to be 720 billion l. Approximately 50% of the contaminated water is thought to be residual leaching solutions, with sulphate higher than 20 g/l and salinities between 35–70 g/l. The remaining 50% is thought to be dispersed solutions, formed by migrating leaching solutions mixing with native groundwater, with a salinity level of 4.5 g/l. A comparison of leaching solutions, contaminated and pre-mining groundwater quality is given in Table 2. The urgent need for restoration is governed by the extremely high concentrations of radionuclides and heavy metals in the various solutions and the large volumes of contaminated water involved. The most critical factor is that the Cenomanian aquifer is artesian, and the pressure difference between the Turonian and Cenomanian aquifers will always ensure groundwater flow is vertically upwards, as was the case before ISL mining began.

The presence of known excursions through boreholes highlights the above problem, and if the bores are not effectively sealed during restoration, there will remain a pressure gradient for new excursions of contaminated groundwater from the Cenomanian into the Turonian aquifer.

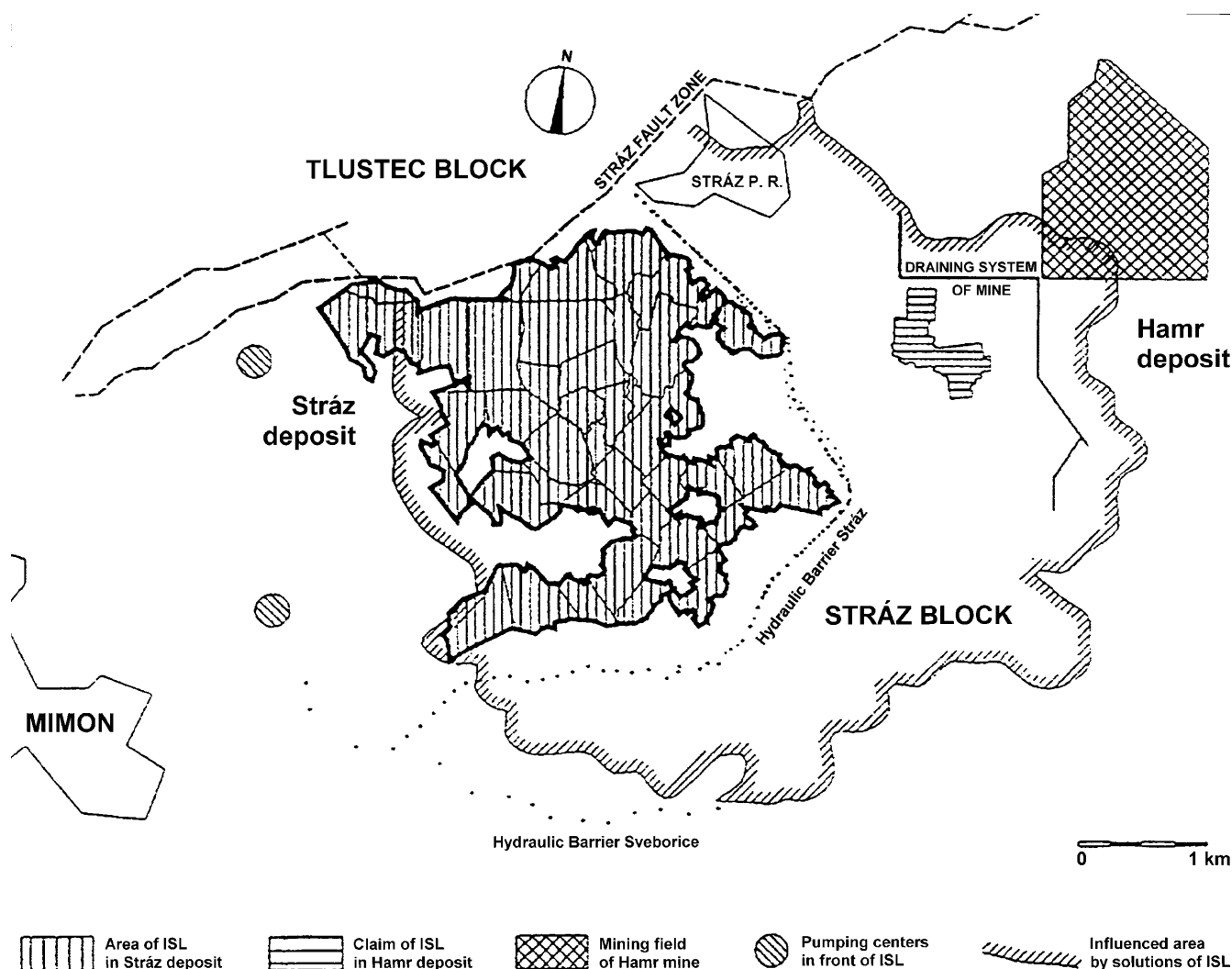


Fig. 4

Location of contaminated groundwater at the Stráž Pod Ralskem and Hamr uranium mines, Czech Republic (Fiedler and Slezák 1992)

The contaminated groundwater in the Cenomanian aquifer is approaching the sanitary protection zone of the Mimon water supply (70 l/s or 6.05 Ml/day). The contaminated groundwater in the Turonian is within 1.2–1.5 km of the sanitary protection zone of the Dolánky water supply

(200 l/s or 17.28 Ml/day). The region presently utilizes 1,500 l/s (129.6 Ml/day) of groundwater for drinking supplies.

The restoration of the groundwater is proving a difficult task, with 1 pore volume of groundwater only removing about 70% of the contaminated groundwater, and 5 pore volumes required for 90% removal. This equates to about 940 billion l of water. While regulators in the USA require a pilot-scale test to demonstrate effective groundwater

Table 2

Groundwater quality before and after ISL mining at Stráž Pod Ralskem

|                     | pH<br>(Units) | TDS<br>(g/l) | SO <sub>4</sub><br>(g/l) | NO <sub>3</sub><br>(mg/l) | F<br>(mg/l) | U<br>(mg/l) | Ra<br>(Bq/l) | H <sub>2</sub> SO <sub>4</sub><br>(g/l) |
|---------------------|---------------|--------------|--------------------------|---------------------------|-------------|-------------|--------------|-----------------------------------------|
| Leaching solution   | 0.5           | 50–100       | 33–80                    | 600–1,400                 | 150–250     | 1–30        | 50–90        | 15–20                                   |
| Cenomanian before   | 6.7           | 0.14         | 0.033                    | <1                        | <1          | 0.02        | 8.74         | n.a.                                    |
| Cenomanian affected | 1.8–2.8       | 5–20         | 3.3–13                   | 5–100                     | 5–50        | 0–15        | 30–70        | 0.5–5                                   |
| Turonian before     | 6.7           | 0.1          | 0.035                    | 5.2                       | <1          | 0.01        | 0.07         | n.a.                                    |
| Turonian affected   | 2.5–7.0       | 0.5–5.5      | 0.05–3.3                 | 5–1,000                   | 0.5–25      | <1          | 0.1–1.0      | <0.5                                    |

quality restoration prior to issuing commercial permits and operation, the Stráz mine only received approvals for liquid waste disposal and restoration requirements in the mid-1990s after three decades of operation.

Currently, restoration programmes are aimed at determining the optimal strategy for long-term remediation of groundwater quality. The technology being used for restoration involves pre-treatment, reverse osmosis, volume reduction by evaporation, crystallization and processing of the concentrated saline solutions or brines. Some components are re-utilized, such as sulphuric acid ( $\text{H}_2\text{SO}_4$ ), aluminium oxide ( $\text{Al}_2\text{O}_3$ ), ammonia ( $\text{NH}_3$ ) and gypsum ( $\text{CaSO}_4$ ). The presence and removal of Ra and other metals of concern is a significant barrier to these programmes.

Further options for groundwater quality restoration are being investigated with a view to a compromise between environmental demands and economic feasibility. It is intended to return the Turonian aquifer to its original groundwater quality (as much as possible). Recent modelling studies indicate that restoring the Cenomanian groundwater to a salinity of 3 g/l can cause undesirable impacts on the Turonian aquifer mainly because of structural and tectonic conditions and the instability of the groundwater regime after returning to natural flows.

For the Cenomanian aquifer, though, it appears impossible to achieve restoration to its original good quality. The philosophy being adopted is to ensure that any escape of Cenomanian groundwater will dilute and disperse to an appropriate quality and not impact on potable or surface waters.

The region, once covered by pine forests, underwent deforestation for mining purposes. This was undertaken hastily with many trunks left in the ground, dead and rotting. The surface soils, devoid of tree cover, were therefore exposed to accelerated rates of weathering, sheet erosion and wash-down of the poorly cohesive sandstones and deep furrows. This was exacerbated by the movement of heavy machinery across the site. In the low-lying areas near the Ploucnice River, the alteration of surface drainage patterns, together with the removal of vegetation, led to a gradual rise in the water table and the formation of lagoons and wetlands.

In the areas where pine trees had been left, to try and preserve some of the remnant forest, it was found that the forest was weakened and unsustainable since it was no longer continuous. This led to increased exposures of the wells and piping systems and high incidences of dead, falling trees. In the hill areas, wells and piping systems were often built partly on benches and partly on platforms. Together with the spills of solutions from pipes and surface runoff, the siting of these parallel to slopes led to significant rates of erosion and the prevention of further vegetation growth because of the lack of suitable soil. Attempts were made from the mid-1980s to address these problems, such as hydromulching, different seed species and other techniques, but they were of varying short duration and thus limited success.

Because of the intransigence of the chemical and physical changes caused by ISL mining at the Stráz site, the

restoration efforts are anticipated to last several decades, or even centuries.

## Germany

This review is based on Hähne and Altmann (1992), Diehl (1995), Ettenhuber (1996), Nitzsche and Merkel (1999) and Biehler and Falck (1999).

The pitchblende uranium deposits of former East Germany were one of the former Soviet Union's first targets for supplying uranium during the late 1940s and 1950s for nuclear weapons and power programmes. Simultaneous work was carried out on all types of uranium deposits in the area. Most uranium mines and mills in East Germany were underground, although Königstein and to a lesser extent Ronneburg, also had underground ISL applied for the extraction of uranium. Underground uranium leaching began in 1968 to take advantage of low-grade ores and increasing conventional costs because in situ leaching costs were only 60–70% of conventional methods. The locations of Ronneburg and Königstein are shown in Fig. 5.

### Ronneburg

Although a less prolific producer of uranium by ISL, repeated attempts were made to increase uranium recovery using ISL from 1970. The leaching solutions used included sulphuric acid or alkaline reagents. Between 40 and 70% of the uranium reserves were recovered at concentrations of sulphuric acid from 3–10 g/l, pH of approximately 1.5–2.5 and uranium content between 20–100 mg/l. A total of 3,203 t of  $\text{U}_3\text{O}_8$  was produced by heap and waste pile leaching, with 106 t of  $\text{U}_3\text{O}_8$  produced by underground ISL techniques.

### Königstein

From 1971, underground and ISL mining (in stopes) were being used until ISL took over in 1984. The extraction of uranium with ISL operated until 1990, with the total uranium production from the life of the mine being 22,711 t of  $\text{U}_3\text{O}_8$ , 6,517 t by sulphuric acid ISL.

The aquifers at Königstein generally contain water quality with a salinity less than 200 mg/l, near neutral pH and low heavy metal concentrations. The uranium mineralization at Königstein is found within the fourth aquifer of a regional groundwater system, shown in Fig. 6. The clay layer separating the third and fourth aquifers was intersected by the underground mine workings. The third aquifer is used by residents of the region for their water supply, as well discharging into the Elbe River 600 m east of the mine site. The dewatering of the fourth aquifer for the mine led to a decrease in water level of the third aquifer.

One of the most difficult problems associated with remediation of the contaminated groundwater is that, at the time of closure, a new underground block had just been prepared for leaching. The prevailing unsaturated condi-

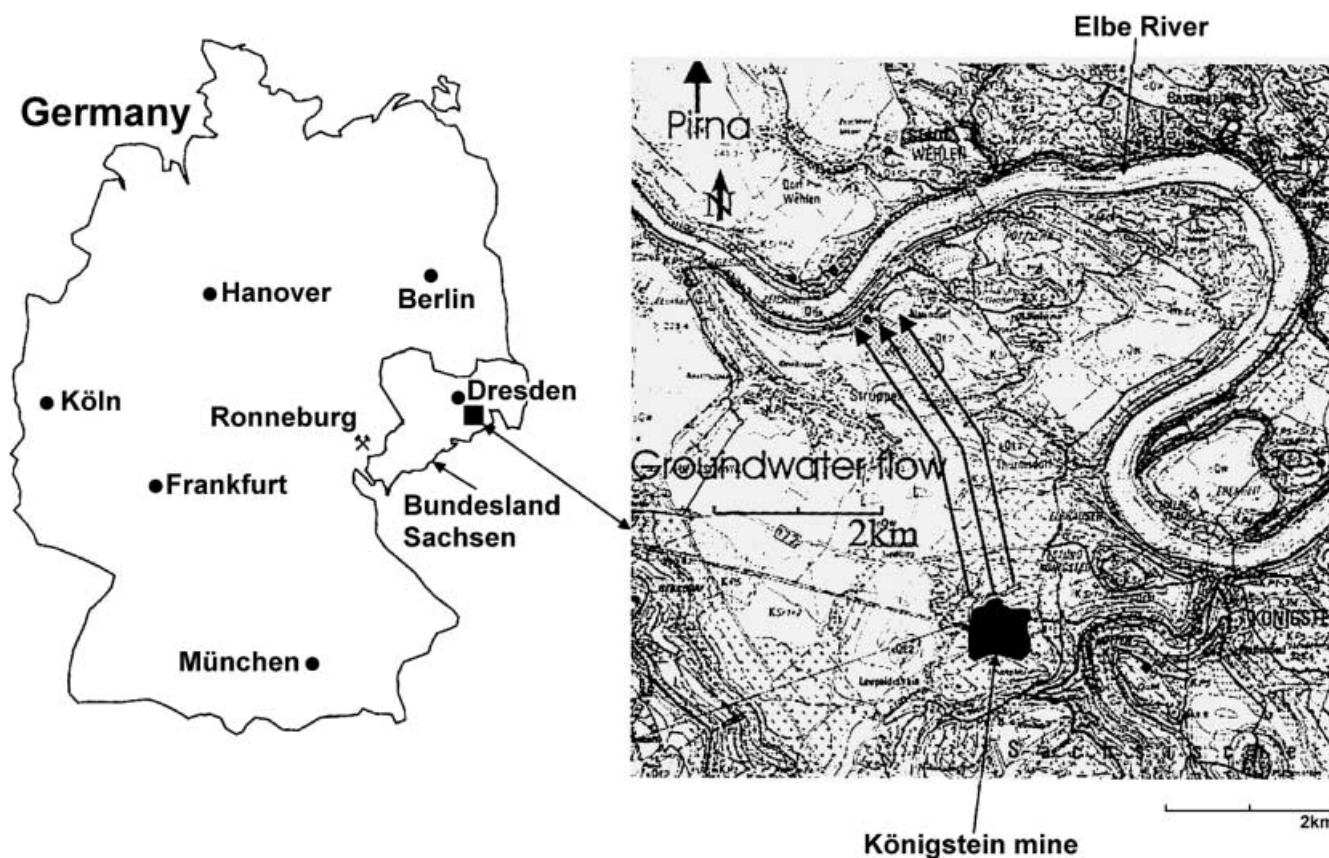


**Fig. 5**  
Location of the Ronneburg and Königstein uranium mines, eastern Germany (Nitzsche and Merkel 1999)

tions allowed the pyrite to oxidize, generating significant quantities of sulphuric acid, which further mobilized heavy metals, uranium and radionuclides and added to the contaminant load to remediate.

The average concentration of leaching solutions was 2–3 g/l sulphuric acid, pH 1.5–1.8, salinity (TDS) 10–14 g/l and uranium 10–150 mg/l. A compilation of groundwater and leaching solution quality is given in Table 1. The uranium recovery was generally about 65–75% within 3 years. A total of 100,000 t of sulphuric acid was injected into the mine. The leaching process has chemically affected a volume of more than 55 billion l of rock and aquifer, while approximately 1.8 billion l contains 1.2–1.7 g/l sulphuric acid and more than 30 mg/l uranium remains circulating or trapped in the pore space of the rocks. A further  $850 \times 10^6$  l are circulating between the leaching zone and the recovery plant. Expressed as multiples of applicable German drinking water standards, the trapped solutions are 400 times higher in Cd, 280 times higher in As, 130 times higher in Ni and 83 times higher in U.

The principal concerns for restoration of the site are centred around the flooding of the underground mine workings that will occur after the mine is closed down. There is potential for contamination of surrounding groundwater and surface water streams with U, Ra,  $\text{SO}_4$ , Fe and heavy metals. Although small-scale flooding trials are currently being conducted, restoration is still not complete

and the mine still represents a threat to the surrounding aquifer, an important potable groundwater resource for the region.

## Former Soviet Union

A general overview of the use of ISL in the former Soviet Union is presented. A more detailed review of each new republic (the 'stans'), however, will be treated separately. This review is based on Skorovarov and others (1987) and Skorovarov and Fazlullin (1992).

There has been active development of ISL-type mines across the former Soviet block countries such as the Ukraine, Uzbekistan, Kazakhstan and Russia since the early 1960s, beginning with tests at the Deviadovskoye site in the Ukraine. After the success of initial tests, ISL technology was rapidly adopted throughout the various territories. The ISL technique was typically applied to low-grade deposits between 0.03–0.05% U. Sulphuric acid was the preferred leaching agent, although alkaline carbonate-bicarbonate agents were used at some sites, depending mainly on the carbonate content of the ore. The well field patterns used were quite variable, including 10×10 m, 10×20 m, 25×50 m and up to 10×100 m spacings. The concentration of sulphuric acid ranged from 2–5 g/l, with the stronger the acidity the greater the recovery of uranium and the shorter the period of leaching. The typical composition of leaching solutions is given in Table 1. The average acid consumption per 1 kg of

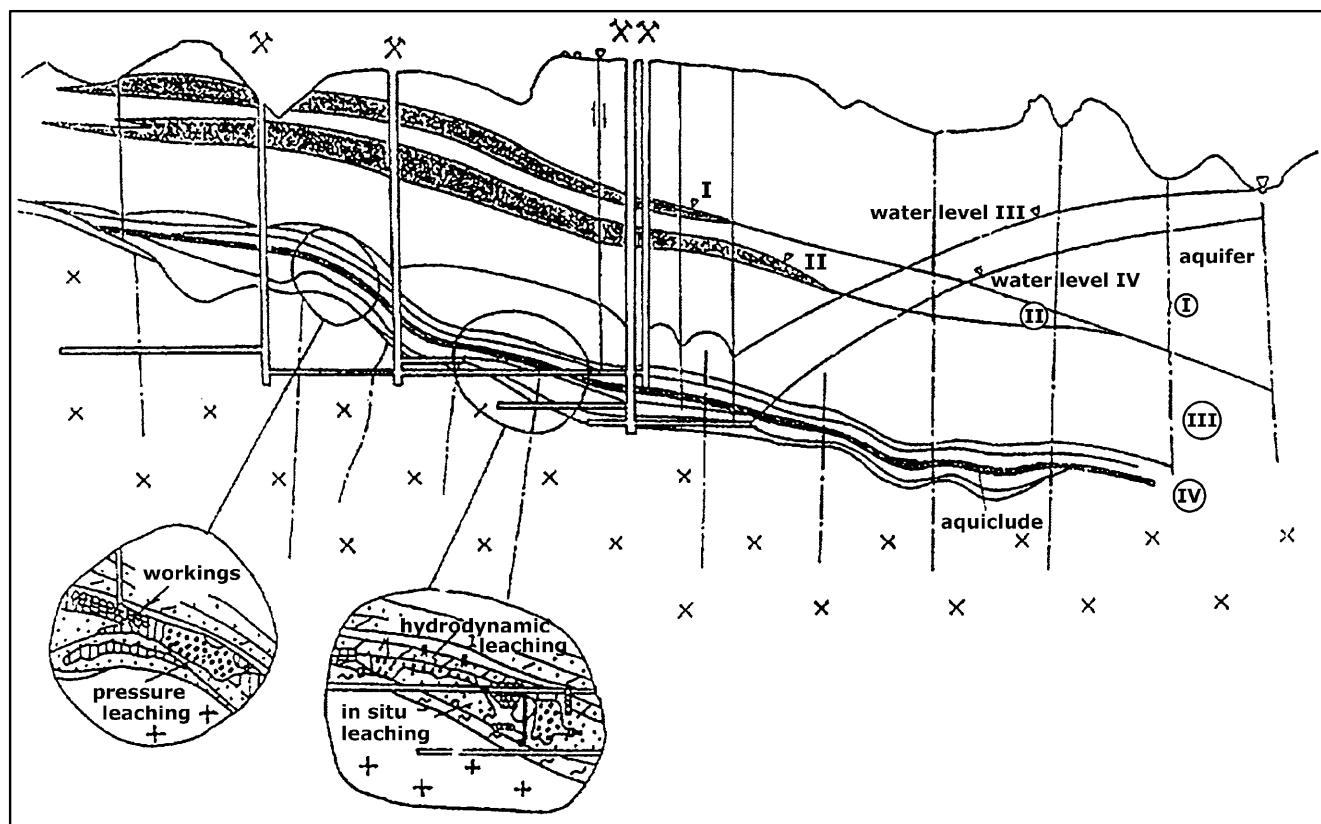


Fig. 6

Hydrogeological cross section of the Königstein uranium mine, eastern Germany (Hähne and Altmann 1992)

uranium recovered as an end product varied widely from 18–150 kg. The recovery rate of uranium was generally between 70 and 90%. No oxidant was needed to ensure dissolution of the uranium. Associated metals were also thought to be extractable, such as V, Re, Se, Mo, Sc, Yt and rare earths.

Some of the main problems of using sulphuric acid was the necessity to use acid-resistant materials and equipment, deterioration of the ore zone permeability because of chemical and gaseous plugging, and the very high salinity levels during mining (ranging from 15–25 g/l).

For ores with a carbonate content higher than 1.5–2.5%, alkaline solutions were used consisting of ammonium bicarbonate with concentrations generally varying from 500 to 5,000 mg/l. Sodium bicarbonate was also applied at some sites with oxygen as an oxidant. The recovery of uranium was generally between 50 and 60%. The use of alkaline agents also tended to show much smaller increases of salinity during mining. The main recognized problems of alkaline ISL were the high degree of solutions escaping outside the mining zone (often caused by gaseous oxygen plugs forming), compulsory pre-treatment to soften the water and restoration difficulties following completion of ISL.

Numerous techniques were being trialled to restore the quality of the groundwater, including lime pulp treatment, hyperfiltration and electrosorption. The success of these

technologies on restoring contaminated groundwater is not known. The production costs of ISL were 40–45% of conventional costs, with significantly lower energy and capital costs and reagent consumption.

## Kazakhstan

This review is based on Yazikov (1993), Akin (1995), Catchpole (1997), Carroll (1997) and OECD (2000). The uranium resources of Kazakhstan are considerable and second only to Australia, of which a large proportion is amenable to ISL extraction (OECD 2000). The location of conventional and ISL uranium mines is shown in Fig. 7. As with many former Soviet-controlled states, the use of ISL in Kazakhstan began in 1970 (after successful testing in the Ukraine) and continued to increase in importance, centred around the large amenable deposits in southern Kazakhstan. The large-scale ISL mines began in 1978. All ISL mines utilize sulphuric-acid-leaching chemistry. By 1990, ISL technology had displaced conventional mines as the predominant method of uranium production. The large ISL-amenable resources are seen as the future of the Kazakhstan uranium industry. There are several operating ISL projects, although an accurate assessment of current and prospective projects is not an easy task. Some operating sites include Stepnoye, Centralia and Chiili. Further sites being assessed and/or developed are the Inkai, Mynkuduk and Moynkum sites. It is known that following acid ISL mining, the aquifer is left to return to its pre-mining state, although timeframes

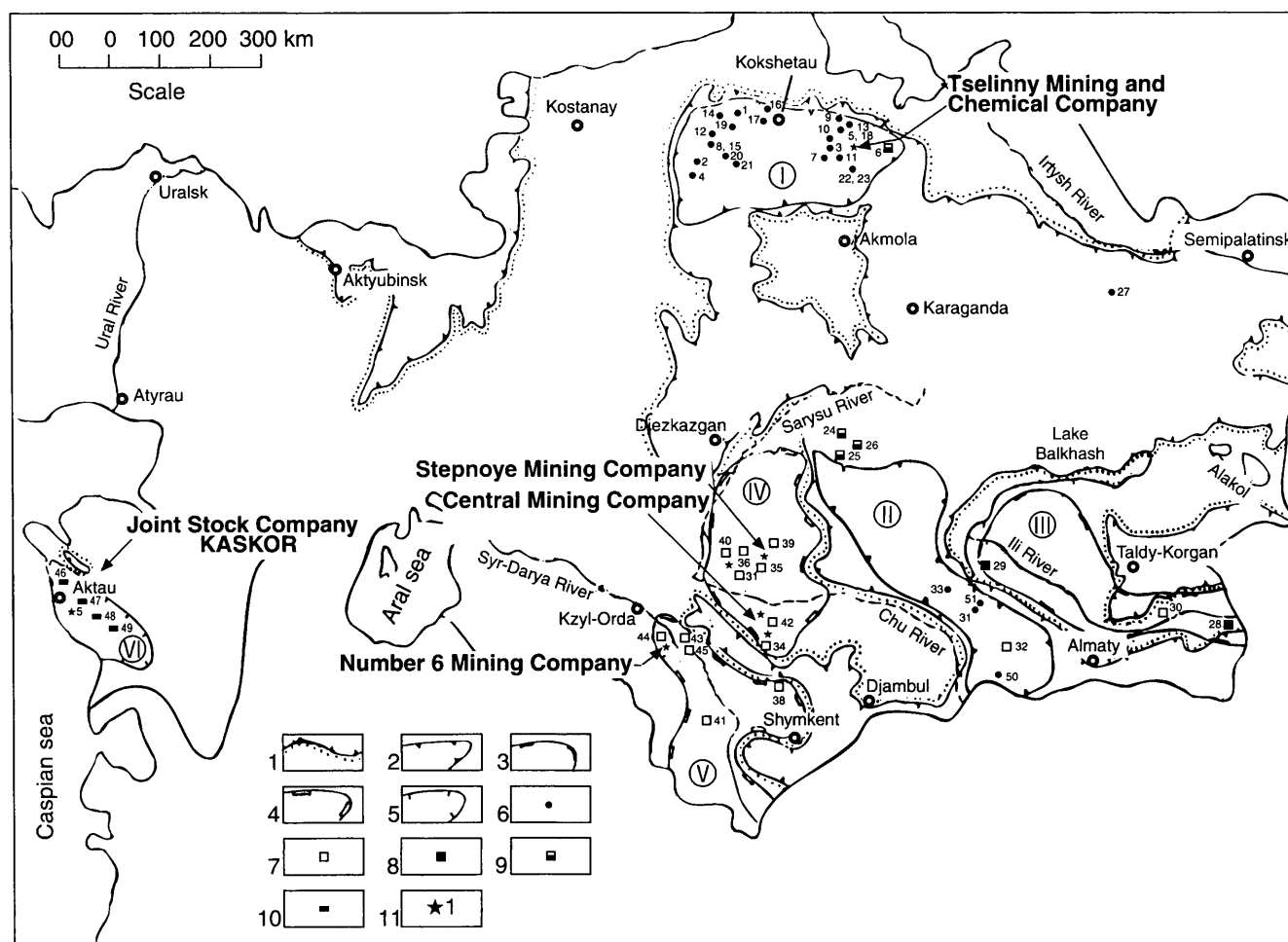


**Fig. 7**  
Location of uranium mines and ISL projects in Kazakhstan (OECD 2000)

are not yet established for this process nor its rate of success. To avoid possible negative impacts from residual mining solutions, an exclusion zone of 150 by 15 km has been declared from which the extraction of drinking water is prohibited. Detailed assessment of the environmental impacts and operational issues are yet to be published, although given the history of the nuclear industry in Kazakhstan, it is likely to be similar to other parts of the former Soviet Union.

## Ukraine

This review is based on Molchanov and others (1995), Rudy (1996) and Chernov (1998). ISL uranium mining was carried out on the Deviadovskoye, Bratskoye and Safonovka deposits.

The Deviadovskoye site was the first ISL mine in the Soviet Union and operated from 1966 to 1983, using sulphuric and nitric acids. The surface area of the mine is 0.12 km<sup>2</sup>,

with a ore body of about 2.18 km<sup>2</sup> and an area for underground storage of 1.2 km<sup>2</sup>. As a result of ISL mining, groundwater was contaminated to a depth of 80 m. The residual solutions are distributed at a distance of 1.7 km along the flow path and for 0.35 km against the upstream hydraulic gradient. The nearest settlement down gradient is 4 km. The volume of residual solutions after the ISL mining of uranium in the Buchak aquifer is 7.09 billion l. The volume of retention pond water is 1 billion l with contaminated silt in pond-collectors of about 40×10<sup>6</sup> l. Leakage from pipelines has contaminated surface soils, totalling about 50×10<sup>6</sup> l.

The Bratskoye ISL mine site operated from 1971–1984, using sulphuric and nitric acids. The orebody area is 0.96 km<sup>2</sup>. At the end of mining, the leaching solutions within the orebody were simply abandoned. The 5.2 billion l of contaminated groundwater is distributed 3 km down-gradient and 1.2 km up-gradient, to a depth of 50 m. The Safonovka ISL site was mined from 1982–1993. The surface area of the mine site covered 0.05 km<sup>2</sup>, although no further information is available on the extent of groundwater impacts and future management and remedial programmes.

The severe lack of financial resources has led to the freezing of restoration activities in 1996.

## Uzbekistan

This review is based on Venatovskij (1992) and Solodov (1998).

The state of Uzbekistan has numerous uranium deposits that host operating or potential ISL mines, concentrated within the large central Kizilkum province. They are generally around 300 m in depth and contain uranium ore in several distinct layers. The ore grades vary from 0.03–0.70% U. Many contain low carbonate content less than 2.5%, although some deposits are rich in carbonaceous matter (higher than 5%). Various leaching agents are used, with sulphuric acid being the preferred acid. A large ISL mine operated at Bukinaii in south-eastern Uzbekistan for about 20 years. The host sands for the roll-front uranium mineralization contain calcite up to 2% and pyrite at 0.2–1.8%. The redox potential in oxidized zones is typically about +150 mV, in the ore zone it varies from –150 to +100 mV and can be as low as –330 mV in more highly reduced sands. The leach solution quality is given in Table 1 and compares with the pre-mining salinity (TDS) of 1,700 mg/l and near-neutral pH of 6.3–6.7.

Six years after ISL mining had been completed on the Bukinaii deposit, a series of field and laboratory investigations were undertaken to assess the migration of the high salinity brine (leaching solutions) abandoned after mining. Field and laboratory testing showed that pH and redox barriers develop as the brine migrates, similar to the natural geological processes that lead to the formation of roll-front mineralization, based on the reactivity of calcite in the aquifer sediments, redox reactions and helped by bacterial activity. Precipitation tended to be in localized zones of thin, fine-grained sands, with a permeability decrease of about one order of magnitude. Medium- and coarse-grained sands only showed limited permeability reductions as a result of precipitation reactions. It was estimated that, assuming an ideal brine lens of 400 m width, it would take about 80 years to achieve complete attenuation and the resulting area would be about half the width of the initial lens (~200 m). No field or laboratory data are presented to demonstrate that this attenuation process returns the levels of all heavy metals and radionuclides to the low levels prior to mining. Importantly, research was still inconclusive on the possibility of redissolution of secondary phases and the remobilization of various heavy metals and radionuclides.

## China

This review is based on Jian and Ning (1992), Xu and others (1998) and (OECD 2000).

Approximately 61% of Chinese uranium is contained within deposits smaller than 3,000 t  $U_3O_8$ , mostly below a grade of 0.2% U. The ISL technique, for underground mines and the more traditional solution mines, has been viewed as the preferential method for economically extracting uranium since the early 1980s with trials on all

types of mineralisation being conducted. Two large ISL projects are currently being actively developed or operated at Tengchong and Yining.

The Tengchong uranium deposit is hosted in sandstone, with gangue minerals including pyrite and carbonaceous matter. A trial of sulphuric acid ISL lasting 42 days was undertaken on a pattern of 31 wells, with the uranium content reaching a maximum of 150 mg/l and an effective yield of 62%. The deposit is being expanded to a commercial facility.

ISL mining at Yining, also known as deposit no. 512, began in 1994 using sulphuric acid and hydrogen peroxide as an oxidant. The deposit is hosted in sandstone being up to 20 m thick and is 0.011–0.17% U in grade. Sulphuric acid levels were initially injected at 2% (20 g/l) and gradually increased to 8% (80 g/l) with hydrogen peroxide concentrations of up to 0.55 g/l. The acid was later reduced to 4–6 g/l. The ISL trial ran for 92 days, with the injection of 9.8 million l of leaching solution, 41.59 t of sulphuric acid, 2.11 t of hydrogen peroxide, and the extraction of 11.7 million l of solutions (18.7% bleed rate) with uranium averaging 300 mg/l. A compilation of leaching solution quality is given in Table 1. No data appear to be available on restoration or if any such activities were undertaken.

## Discussion and conclusions

The experience of acid ISL uranium mining in areas controlled by the former Soviet Union provides a stark contrast to experiences in America and Australia. In most applications of the technique, there have been extreme occurrences of groundwater contamination. At some sites, this contamination has migrated considerable distances to impact on potable drinking water supplies. For other sites, the potential for contamination to reach an undesirable receptor remains significant. The problems at these sites were severely exacerbated by the prevailing paradigm of uranium production without regard for environmental damage. Apart from Asian nations, such as Kazakhstan, Uzbekistan and China, all countries are in the process of closing down and developing remedial action programmes. The restoration of groundwater is proving difficult, both technically but also because of a lack of financial resources within these countries.

Morris (1984) noted that reliance on natural attenuation processes has never been tested for restoration of ISL. The former ISL sites across Eastern Europe and the former Soviet Union allow some insight into the use of 'natural restoration' as a remedial technique.

Buma (1979) argued that natural geochemical processes within aquifers can restore contaminated groundwater from ISL mines, thereby saving valuable chemical, energy and financial resources. The processes include precipitation of reduced compounds; scavenging of heavy metals by pyrite, organic matter, ferric oxyhydroxides and calcite; and adsorption by quartz, feldspars and clays. The key was for active reductants to be present.

At many ISL mines outlined above, there was high organic, iron, carbonate or sulphide content, such as Bulgaria and the Czech Republic. The contamination at these sites, including the high concentrations of major ions, heavy metals and radionuclides, has not attenuated significantly over time, and instead migrates away from the mine sites, up to several kilometres in some instances. The geochemical mechanisms controlling this migration are unclear, although co-precipitation, which may give rise to higher solubilities for species such as  $(\text{Ca,Ra})\text{SO}_4$ , and the complete oxidation of reducing agents during ISL mining with no active agents remaining after mining or coating of possible reactive surfaces with precipitates during or after mining, are likely to be significant, key issues. The extent and ease of remobilization of many elements following migration from the mined aquifer is also poorly understood.

It would appear, therefore, that 'natural restoration' is not a desirable approach in the slightest, even given the complex hydrogeochemical conditions known to exist at most former (and current) ISL sites across Eastern Europe and the former Soviet Union. It would be a more conservative approach to require and provide funds for restoration prior to the commencement of mining, with detailed studies of aquifer geochemistry and physical behaviour to establish likely requirements and potential remedial strategies.

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