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ARTICLE

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Possible uranium sources for the largest uranium district associated with volcanism: the Streltsovka caldera (Transbaikalia, Russia)

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Abstract The uranium deposits of the Late Jurassic Streltsovka caldera (Transbaikalia, Russia) represent the largest uranium field associated with volcanics in the world (280,000 t U) and Russia's largest uranium resources. About one third of the caldera stratigraphic pile consists of rhyolites which are strongly altered. The rhyolitic magma preserved as melt inclusions in quartz phenocrysts corresponds to a mildly peralkaline melt ($1.04 < Na+K/Al < 1.10$), rich in F (1.4–2.7 wt%) and U (15–23 ppm). Fission track distribution shows that U is almost exclusively located in the matrix of the rhyolites. Mass balance calculations demonstrate that the amounts of U and F leached from the rhyolites (26,000 t U and 52 Mt F per km^3) may greatly exceed the amounts of U and F concentrated in the known U and F ore deposits of the Streltsovka caldera. The magmatic fluids expelled from the Streltsovka rhyolitic melts probably contained less than 1 ppm U, according to the low $KD_{fluid/melt}$ of uranium for such F-rich peralkaline melts. Thus, the mass of uranium released by the exsolution of the volatile phase from the extruded rhyolitic melts was of minor importance, in the range of a few 1,000 t U. The Hercynian biotite granites which represent the major part of the caldera basement belong to a sodic-potassic subalkaline magmatic association. An alkaline biotite leucogranite was only identified in the deepest part of the drillings. The pervasive and intensive hydrothermal alteration of U-bearing accessory minerals in the granites shows that uranium was also extracted from these granites. Mass balance calculations for the granites show that zircon and apatite are minor hosts for U and Th compared to thorite and allanite. The alteration of allanite to monazite and REE carbonate may have liberated up to 1,638 t U/km^3 of granite. Beside the efficiency of hydrothermal alteration, the exceptionally large uranium resources of the Streltsovka caldera, compared to other volcanic-related uranium districts in the world, appear to result from the juxtaposition of two major uranium sources: highly fractionated peralkaline rhyolites of Jurassic age in the caldera, and U-rich subalkaline granites of Variscan age in the basement in which the major uranium-bearing accessory minerals were metamict at the time of the hydrothermal ore formation.

Keywords Uranium · Streltsovka · Rhyolite · Granite · Russia

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Introduction

The uranium deposits from the Streltsovka caldera (Transbaikalia, Russia) represent a unique example of a gigantic uranium ore field associated with volcanism. The ore resource estimates grading above 0.2% U reach more than 280,000 t U_3O_8 in the cost category (according to the IAEA classification) lower than 80 US $/kg (Laverov et al. 1992), and are distributed in 20 deposits. About 30,000 t U_3O_8 of additional and possible reserves belong to the IAEA cost category 80–130 US $/kg. Similar uranium mineralisations are known in other parts of the world, such as Dornot in Mongolia (Mironov et al. 1993) which belongs to the same volcanic province as Streltsovka, Ben Lomond in Australia (O'Rourke 1975), Marysvale and Thomas Range in the United States (Cunningham et al. 1982; Christiansen et al. 1983, 1984; Cunningham et al. 1994), Sierra Pena Blanca in Mexico (George-Aniel et al. 1985) and the deposits from the Xiangshan volcanic complex in China (IAEA-NEA 1992; Table 1). However, these deposits are much smaller than the ones from Streltsovka. Drilling to a depth of 2,700 m through the Streltsovka caldera and basement provided a unique opportunity to evaluate the role of the different source

Table 1 Tonnage and grade data on Streltsovka and similar uranium districts in the world (name, location and grade from Georges Aniel et al. 1991; IAEA-NEA 1994; Cunningham et al. 1998; Castor and Henry 2000)

Name	Location	RAR + EAR-I (t U)		Grade
		Cost < US $ 80/kg U	US $ 80 < cost < US $ 130/kg U	
Streltsovka[a]	Russia (Transbaikalia)	167,600 U	15,600 U	0.2% U
Dornot[a]	Mongolia	–	29,000 U + 4,000 U	0.28% U
Xiangshan[a]	China (South)	26,000 U[e]		0.1–0.3% U
Marysvale[b]	USA (Utah)	640 t U_3O_8		
McDermitt[c]	USA (Oregon)	9,500 t U_3O_8		0.13% U_3O_8
Sierra Pena Blanca[d]	Mexico	1,200 t U_3O_8		
Macusani[a]	Peru	1,790 + 1,720 U	140 U	

[a]IAEA-NEA (1994)
[b]Cunningham et al. (1998)
[c]Castor and Henry (2000)
[d]George-Aniel et al. (1991)
[e]Approximately equivalent to the sum of RAR (reasonably assured resources) and EAR-I (estimated additional resources of the first category) according to IAEA definitions. The resources are not classified by production cost

rocks which may be involved in the genesis of the deposits. This paper aims to characterise the possible uranium sources, and to quantify their potential respective contribution to the U deposits.

Geologic setting

The Streltsovka district is located in Transbaikalia (Russia) near the Chinese-Mongolian border (Fig. 1). The caldera (Fig. 1) has a diameter of about 20 km and was filled with a pile of volcanic rocks (basalt, andesite, trachydacite, rhyolite) and interlayered sedimentary horizons. The present thickness of the pile locally reaches more than 1 km. The caldera filling occurred during Late Jurassic times. Rhyolites which correspond to the last erupted volcanics were dated at 142 ± 7 Ma by K–Ar geochronology (Chernyshev and Golubev 1996). They may have represented between 30 and 35% of the volume of the volcanic pile, but at the present time rhyolites are mainly preserved in the centre of the caldera. The basement is mainly composed of Hercynian granites (about 300 Ma).

Uranium mineralisation may be located in any of the different volcano-sedimentary horizons and was also recognised by core drillings down to 2,400 m into the granitic basement. Uranium mineralisation generally occurs as subvertical veins or stockworks but may sometimes expand along stratigraphic levels in sandstones, conglomerates and tuffs of the caldera. Age determinations indicate that the uranium deposits were formed immediately after the end of the volcanic activity (see below). Uranium contents generally amount to about 0.2%, up to 0.6% in large stockworks, and up to 1.0% in veins. Small deposits were also discovered in the surroundings of the caldera. They correspond to remobilization in more recent sediments. The major deposits inside the Streltsovka caldera are the mined-out Tulukuevskoe open pit, and the presently operating Streltsovskoe and Antei underground mines. The Tulukuevskoe deposit presented the most complete volcano-sedimentary sequence (about 800 m thick; Fig. 2). The mineralisation occurred as a stockwork and was restricted to the volcano-sedimentary pile with uranium reserves greater than 35,000 t U (Laverov et al. 1992). The Streltsovskoe-Antei deposit is a vein-type mineralisation hosted in the volcanic rocks and the basement (Fig. 2), with resource estimates greater than 60,000 t U. It is the major deposit of the caldera. Molybdenum, with an unknown grade and tonnage was also produced. Fluorite veins are also common in the caldera and were discovered first. The uranium deposits were discovered more than 10 years later through drilling.

The paragenetic scheme of the Streltsovka uranium mineralisation is mainly based on the mineralogical studies of Melnikov et al. (1980; Fig. 3). The schematic spatial distribution of the alteration zones developed around uranium mineralisation is represented in Fig. 4. Stage I mainly corresponds to local albitisation in the deep part of the deposit. The most intensive, hydrothermal alteration stage II corresponds to the formation of hydromica (Andreeva et al. 1996). In the 2,600-m-deep 7c drilling from the Streltsovkoe-Antei deposit, we have identified the hydromica component as illite, phengite and/or chlorite, along the entire length of the drill core. Illite is dominant in the samples down to 1,800 m and phengite at greater depth. The phengite-illite stage was dated at 139–130 Ma by Andreeva et al. (1996), and at 133 ± 5 Ma by Chernyshev and Golubev (1996) by K–Ar geochronology. In the deepest part of the drill core, the leucogranites are less altered to potassic micas. Chlorites are mainly observed as pseudomorphs after biotite or as newly formed "rosettes". The Al^{IV} chlorite geothermometer (Cathelineau 1988), with the formula $T\ (°C) = -61.92 + 321.98(Al^{IV})$, gives temperatures between 290 and 390 °C for all chlorite types (chlorite pseudomorphs after biotite and rosette).

After this alteration stage, quartz veins with variable quantities of carbonate, pyrite, and hydromica were formed during stage III without uranium mineralisation (Ishukova et al. 1998). Locally high U content was observed with the stage III paragenesis, without visible uranium minerals, particularly at the periphery of the caldera (Ishukova et al. 1998).

Main uranium deposition occurred during the quartz-molybdenite-pitchblende stage IV. The most common uranium mineral, pitchblende, was dated by a U–Pb discordia at 133 ± 4 Ma (Chernyshev and Golubev 1996). Pitchblende occurred as isolated spherolites, as spherolites at the margin of idiomorphic minerals, or as disseminated grains in the cement of breccias. Xenomorphic coffinite was observed at the margin of pitchblende spherolites, associated with pitchblende in the breccia cement, and with pyrite. Fibrous coffinite crystals are locally present at the contact with pitchblende. Brannerite occurred as small idiomorphic, rectangular stick-like crystals deposited earlier than coffinite or pitchblende. Pyrite, galena and molybdenite may be associated with the uranium minerals. Brannerite ores occur at depths of more than 1,300 m in the basement, mixed pitchblende-coffinite-brannerite ores between 800 and 1,300 m, and molybdenite-pitchblende ores occur at higher levels (Ishukova et al. 1998). A first generation of fluorite was deposited during the syn-mineralisation stage (stage IV), and a second generation of fluorite was deposited with calcite during the post-mineralisation stage (stage V).

The rhyolites

Characteristics of the magmatism and its uranium fertility

Rhyolitic rocks were sampled from three deposits in the Streltsovka caldera: Tulukuevskoe and Krasny Kamen open pits, and Streltsovskoe-Antei underground mines

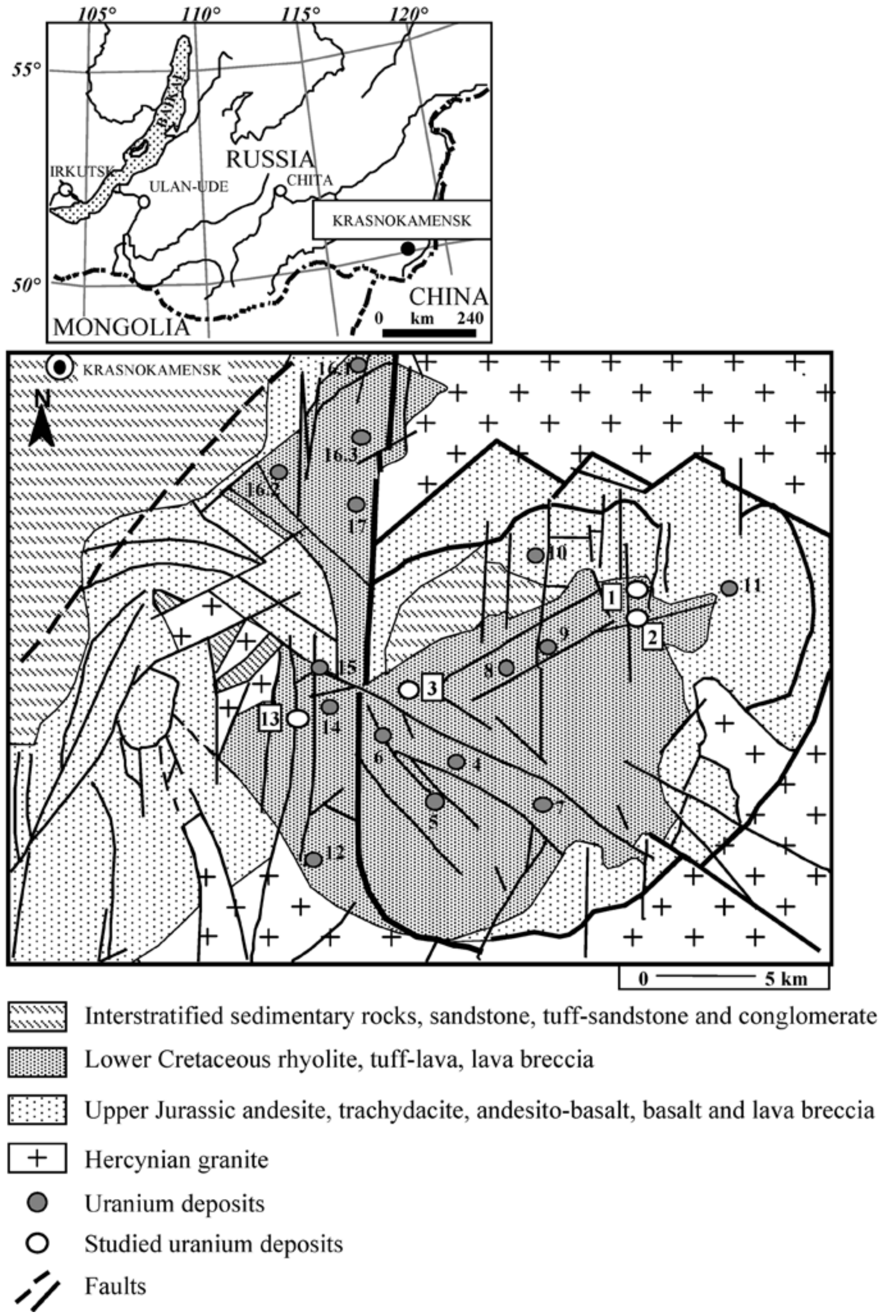

Fig. 1 Geographic map and location of the Streltsovka uranium field and the city of Krasnokamensk, and geological map of the Streltsovka caldera (adapted from unpublished internal report of the Institute of Geology of Ore Deposits, Petrography, Mineralogy and Geochemistry, IGEM). Deposits: *1–2* Streltsovskoe-Antei, *3* Tulukuevskoe, *4* Yubilenoe, *5* Novogodnoe, *6* Vesenee, *7* Malo-Tulukuevskoe, *8* Martovskoe, *9* Lytchistoe, *10* Octiabrskoe, *11* Shirondukuevskoe, *12* Yugo-Zapadnoe, *13* Krasny Kamen, *14* Piatiletnee, *15* Argunskoe, *16* Dalnee, *16.1* Glavny, *16.2* Severny, *16.3* Suknoy, *17* Bezrechnee

(Table 2). They are homogenous rocks with rare phenocrysts. The matrix, which represents about 85 to 90% of the rock, is a recrystallised glass composed of very small quartz and feldspar crystals embedded in a phyllic matrix. Fe-oxide staining is more or less developed. Phenocrysts are feldspars, quartz, zinnwaldite and opaque minerals.

The original chemistry of the rocks was strongly modified by alteration, especially its primary uranium and other easily leacheable constituent (Na, Ca, H_2O, F, Cl, CO_2) concentrations. The rhyolites are characterised by high and variable SiO_2 contents (73–79 wt%), and low FeO, CaO, MgO and TiO_2 contents indicating their high degree of fractionation (Table 3). Because of intense Na-leaching, these rocks are highly peraluminous ($1.12 < A/CNK < 2.05$). LREEs are highly fractionated ($La/Sm_N = 7.3$ to 8.8), HREEs are slightly reversely fractionated ($Dy/Lu_N = 0.6$ to 0.7), and the Eu anomaly is extremely negative ($Eu^* = 1.2$ to 1.5; Fig. 5). Concentrations of Rb (288–475 ppm), Cs (15–43 ppm) and of elements weakly mobile during hydrothermal alteration, such as Nb (53–63 ppm), Y (29–43 ppm), Ta (5–6 ppm), Zr (245–261 ppm) and Th (62–79 ppm), are characteristic of highly fractionated alkaline magmas.

The primary geochemical characteristics of the rhyolitic magma were defined by the analyses of melt inclusions in quartz phenocrysts (Chabiron et al. 2001). The composition of the silicate melt after

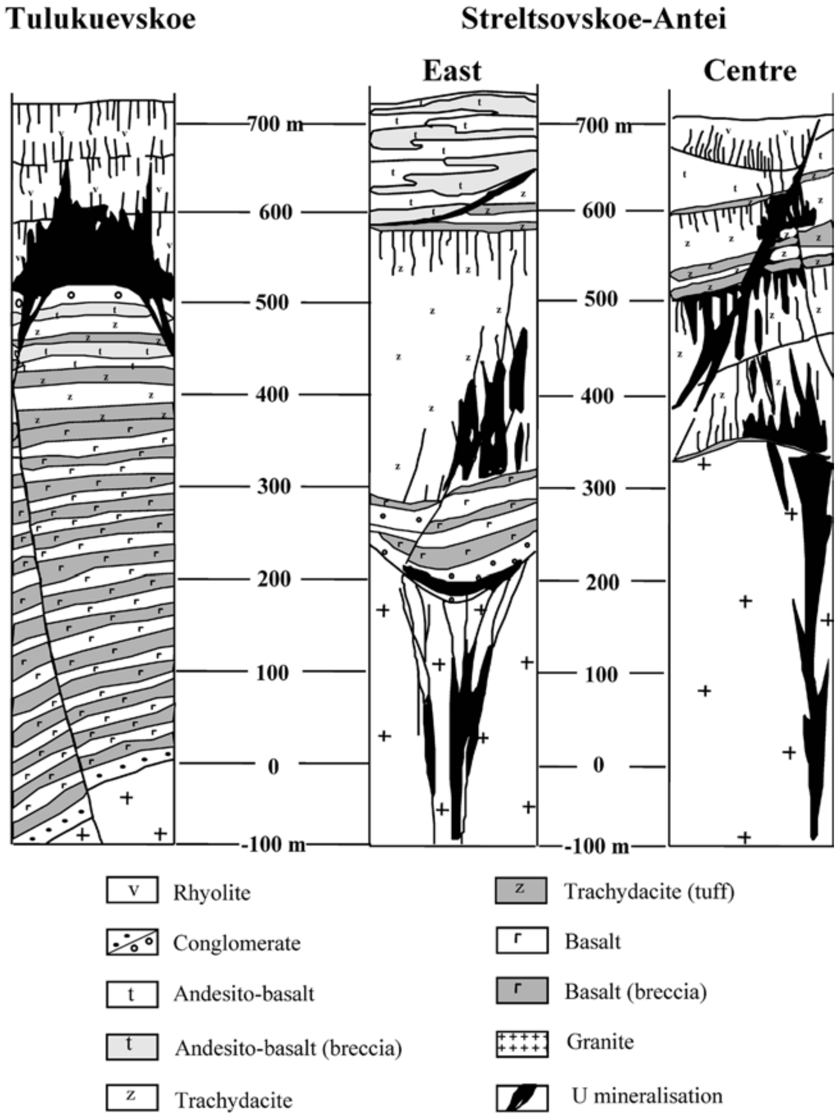

Fig. 2 Cross sections of the Tulukuevskoe and Streltsovskoe-Antei uranium deposits (adapted from unpublished IGEM internal report)

homogenisation (Chabiron et al. 2001) indicates that the Streltsovka rhyolites correspond to mildly peralkaline magmas ($1.04 < (Na + K)/Al < 1.10$), rich in F (from 1.4 to 2.7 wt%) but relatively poor in chlorine (0.2 wt%).

Fission track analyses on melt inclusions indicate uranium contents between 13 and 26 ppm in the rhyolites enclosing different deposits from the Streltsovka caldera (Table 4). The average U content is 19 ± 4 ppm ($n = 46$). Taking into account four additional uranium analyses by SIMS, the average U content is 18 ± 3 ppm (Table 4).

Evidence of uranium mobilisation

After their extrusion, the volcanic rocks in the caldera and particularly the rhyolitic glasses were hydrated, devitrified and altered by hydrothermal fluids. Comparison of altered rhyolite whole-rock compositions with glass-inclusion compositions (Chabiron et al. 2001) shows that easily leachable elements and particularly uranium were strongly leached by hydrothermal fluids. At the whole-rock scale, fission track patterns show that uranium was nearly entirely located in the rhyolitic matrix. Refractory accessory minerals were not observed. This finding is consistent with the peralkaline nature of the melt which favoured a high solubility of the accessory minerals and thus prevented their early crystallisation. Such an uranium distribution is highly favourable for its efficient leaching during subsequent hydrothermal stages (Leroy and George-Aniel 1992).

Quantification of uranium and fluorine leaching

Presently, the Streltsovka rhyolites have an average U content of about 8 ± 2 ppm, obtained on 49 non-mineralised, variably altered rhyolite samples collected over the whole caldera (Ishukova et al. 1998). This U content is

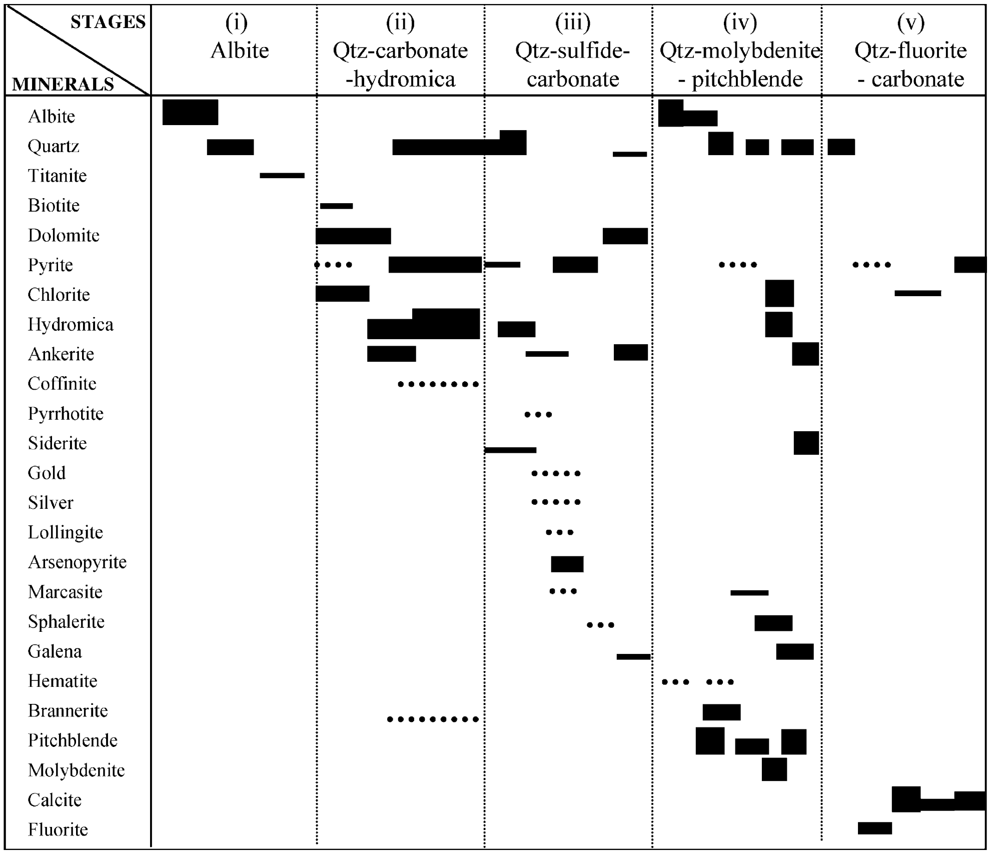

Fig. 3 Paragenetic scheme of the Streltsovka uranium deposits after Melnikov et al. (1980), with the use of unpublished data by M.V. Vampilov, B.V. Brodin, A.A. Zavarzin, I.V. Chernykh, I.V. Dubrova, and V.P. Rugova (IGEM). The *symbol* size depends of the mineral abundance and the length of time of its deposition in the mineralogical sequence

lower than the average U content of the samples from the present study because we have selected the freshest samples from mine open pits to get the best preserved melt inclusions. Also, weak uranium enrichment related to uranium deposition in microfractures cannot be entirely excluded because some of our samples are located some tens of metres from the ore bodies. Compared with the 19-ppm average uranium content of the silicate melt in the magmatic inclusions, 11 ppm of uranium appears to have been removed from the rhyolites by hydrothermal fluids. The rhyolite volume preserved in the caldera, calculated from geologic data (Alyoshin, personal communication), is 10.8 km^3. The total volume of rhyolites was at least 31.4 km^3, assuming an initial 100-m thickness of the rhyolites before erosion and a caldera diameter of 20 km. Consequently, considering the average U content of the pristine rhyolitic magma as representative of the rhyolitic volcanic rocks in the caldera, a loss of 11 ppm corresponds to about 300,000 t U leached. Taking into account the rhyolite volume before erosion, a mass of 900,000 t U is obtained. Thus, the uranium leached from the rhyolites alone may largely explain the 300,000-t total uranium resources of the Streltsovka uranium field. The high Zr contents of the pitchblende (1.3 to 1.5 wt% ZrO_2) in the deposits of the caldera also favour the rhyolites as a major uranium source for the deposits. High Zr contents in pitchblende are characteristic of U deposits associated with acidic volcanism, as observed at Akouta in Niger (Forbes 1989) and Müllenbach in Germany (Pironon 1986), because zirconium located in devitrified volcanic glasses can be easily extracted by hydrothermal fluids.

The rhyolites represent also a major fluorine source for the fluorite deposits of the Streltsovka caldera. The F content of the melt inclusions (1.7 to 2.7 wt%) is much higher than the value recorded in whole-rock samples (only 0.2 wt% F; Chabiron et al. 2001). Therefore, considering an average F content of 2.2 wt% in the pristine melt, hydrothermal alteration of 1 km^3 of rhyolite may have released 52 Mt F or 1.6 Gt F for a 100-m-thick sheet of rhyolite.

Despite the fact that the leaching of the rhyolites may largely account for the total uranium resources from the Streltsovka caldera, we have also to take into account that the efficiency of uranium deposition from the fluids was probably not 100%, and that some other mineralised bodies may have not yet been discovered and/or their resources may have not been evaluated because they were not of economic size or grade. Therefore, other possible uranium sources have to be evaluated: fluids exsolved from the rhyolites and underlying magmatic chamber and deeply altered granitic basement rocks.

Magmatic fluids

Magmatic fluids may have been expelled from extruded rhyolites and from the melts which have crystallised in the underlying magmatic chamber. The experimental

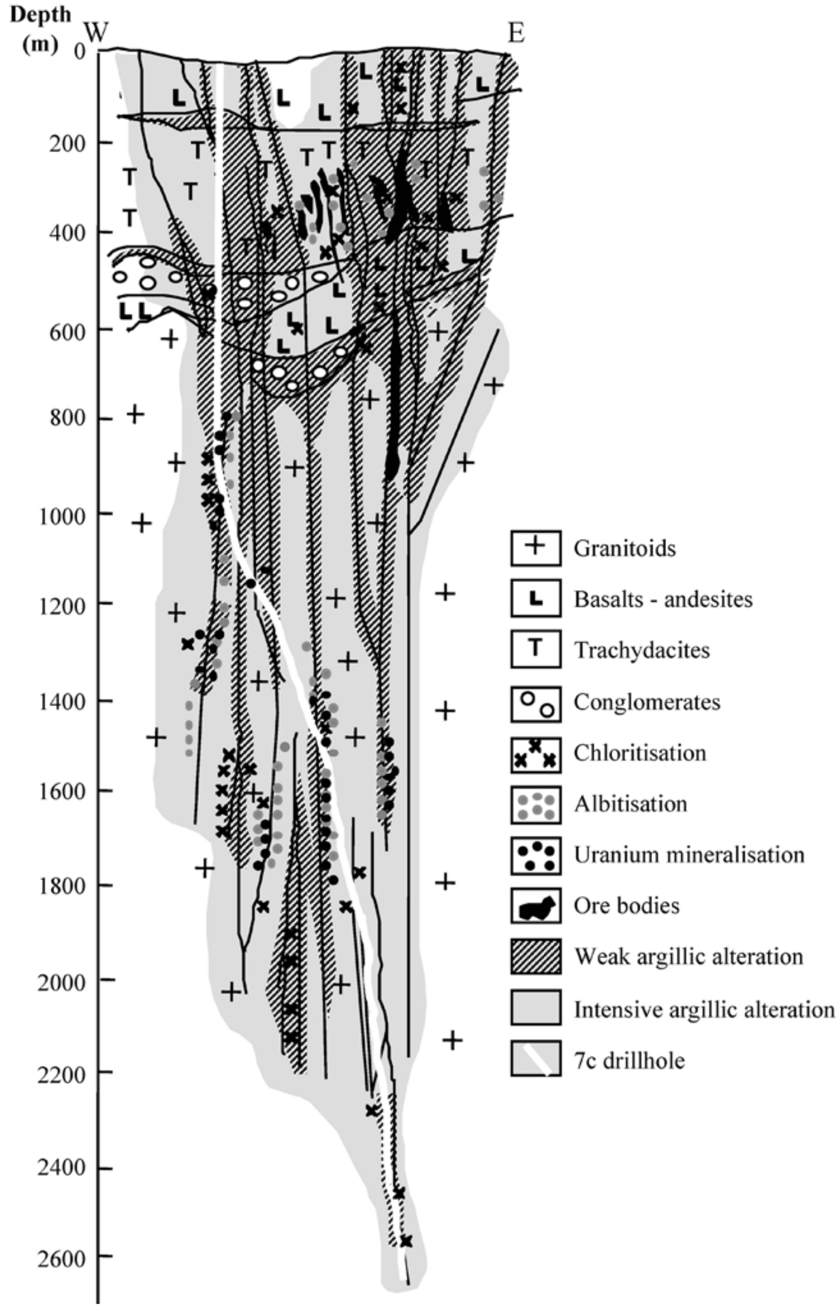

Fig. 4 Schematic geologic cross section of the Streltsovka-Antei deposit showing alteration zones associated with uranium mineralisation (after Andreeva et al. 1996)

data of Peiffert et al. (1996), in the haplogranite–$UO_{2.x}$–H_2O–NaF system for such peralkaline ($1.04 < Na + K/Al < 1.10$) and F-rich (1.4 to 2.7 wt%) compositions, indicate that (1) the Streltsovka rhyolitic melts were strongly undersaturated in uranium: up to 1.1 wt% U can be dissolved for such melt compositions at 800 °C and 2 kbar, and even more at the temperature possibly reached by the Streltsovka rhyolitic magma (1,014 ± 30 °C, Chabiron et al. 2001); and (2) the uranium fluid-melt partition coefficient is strongly in favour of the melt ($K_{DUfluid/melt} = 3.10^{-2}$ to 4.10^{-2}, Peiffert et al. 1996). Based on these K_{DU} values, the uranium content of the aqueous fluids expelled from the Streltsovka rhyolitic melts was less than 1 ppm (0.6 to 0.8 ppm). Such a low uranium content in the magmatic fluids mainly resulted from the high solubility of uranium in highly depolymerised, peralkaline silicate melts. The average amount of water in the melt inclusions was estimated as 2 wt% from ion microprobe measurements on homogenised inclusions (Chabiron 1999). This water content is a minimum estimate of its initial content in the rhyolitic magma, because water may have been lost at several stages during melt ascent (Lowenstern 1994). Therefore, the minimum mass of fluid released by the rhyolite magma corresponds to $31.4(km^3) \times 0.02 \times 2.6$(average rhyolite density) = 1.63 Gt, taking into account the already eroded part of the volcanic pile. Then, the mass of U released is less than

Table 2 Rock samples studied from the Streltsovka uranium district with indication of primary mineral paragenesis, dominant alteration minerals and related uranium deposits. Only the granite samples presenting a significant alteration are described here. *Afs* Alkali feldspar, *Kfs* potassic feldspar, *Qtz* quartz, *Pl* plagioclase, *Zinn* zinnwaldite, *Bt* biotite, *Hem* hematite, *Mag* magnetite, *Ttn* titanite, *Aln* allanite, *Zrn* zircon, *Ap* apatite, *Th* thorite, *Xnt* xenotime, *Mnz* monazite, *Pcl* pyrochlore, *Chl* chlorite, *Ill/Ph* illite-phengite, *Kln* kaolinite, *Fe-ox* iron oxyde, *Mnz** monazite after allanite, *Carb** REE carbonate after allanite

Rock type	U deposit	Mineralogy	Altered samples	Alteration
Rhyolite	Streltsovskoe-Antei	Afs–Qtz–Pl–Zinn–Hem–Mag	STR	Ill, Kln
	Tulukuevskoe	Afs–Qtz–Pl–Zinn–Hem–Mag	207A, F1, F2, F3, F4	Ill, Kln
	Krasny Kamen	Afs–Qtz–Pl–Zinn–Hem–Mag	F6	Ill, Kln
			F7	Ill, Fe-Ox
Granodiorite	Streltsovskoe-Antei	Qtz–Kfs–Pl–Bt–Ttn–Aln–Mag–Zrn	2439.5, 2466.5	Chl, Mnz*, Carb*
Granite	Streltsovskoe-Antei	Qtz–Pl–Kfs–Bt–Mag–Ttn–Ap–Zrn–Th–Aln	891.6, 1068.1, 1231.6	Ill/Ph, Chl
			1348, 1459.3, 1876.5, 2630	Mnz*, Carb*
			1514.2, 1992, 2473.5	Ill/Ph
Leucogranite	Streltsovskoe-Antei	Qtz–Kfs–Pl–Bt(< 5%)–Mag–Ap–Zrn–Xnt–Mnz–Th–Pcl	2664	Chl
			2665	Ph

Table 3 Whole-rock compositions (inductively coupled plasma-emission spectrometry and mass spectrometry analyses) of rhyolites from the Streltsovka caldera sampled in the Streltsovskoe, Tulukuevskoe and Krasny Kamen deposits

Deposit	Streltsovskoe	Tulukuevskoe					Krasny Kamen	
Sample	STR	207A	F1	F2	F3	F4	F6	F7
SiO_2 (%)	78.03	76.28	78.83	77.77	78.67	76.87	73.18	74.80
TiO_2	0.11	0.11	0.09	0.12	–	0.15	0.12	0.09
Al_2O_3	12.13	12.16	11.81	12.54	11.09	11.83	11.85	12.00
FeO	1.18	1.16	1.08	1.08	0.94	0.99	2.69	1.39
MnO	0.04	0.03	0.07	–	–	0.03	0.04	0.05
MgO	0.16	0.12	0.02	0.05	0.08	0.29	0.15	0.20
CaO	0.41	0.47	0.24	–	0.14	0.20	0.04	0.49
Na_2O	0.95	3.08	0.72	3.90	0.15	2.34	1.67	1.43
K_2O	4.38	4.93	4.02	2.96	5.36	4.26	5.47	4.99
F	n.d.	n.d.	–	0.21	0.12	0.26	0.22	n.d.
P_2O_5	–	–	–	–	–	0.18	0.07	0.07
LOI	2.31	1.36	2.42	1.36	2.51	2.09	3.25	3.12
Total	99.7	99.7	99.3	99.99	99.06	99.49	98.75	98.63
A/CNK	1.72	1.08	1.97	1.30	1.76	1.34	1.35	1.39
La (ppm)	51.3	57.5	54.5	53.6	49.1	54.0	54.6	54.4
Ce	95.8	103.1	101.4	99.2	74.9	74.5	74.0	99.5
Pr	9.4	9.2	8.8	8.7	8.2	8.5	8.6	8.7
Nd	24.7	26.3	24.6	23.5	21.9	23.0	24.0	23.8
Sm	4.4	4.6	4.4	4.2	4.0	3.9	4.1	4.1
Eu	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.1
Gd	3.9	4.6	3.9	3.7	3.3	3.3	3.7	4.0
Tb	0.7	0.8	0.7	0.7	0.6	0.6	0.7	0.7
Dy	5.2	5.4	4.8	4.9	3.9	4.4	4.8	4.7
Ho	1.3	1.4	1.2	1.1	0.9	1.0	1.1	1.1
Er	3.6	3.9	3.6	3.7	2.7	3.3	3.4	3.6
Yb	4.8	5.3	4.9	4.7	4.2	4.6	4.5	4.9
Lu	0.8	0.9	0.8	0.7	0.7	0.7	0.7	0.8
Cs	22.4	15.3	20.0	16.5	43.0	21.6	19.9	15.8
Nb	59.4	62.7	52.7	58.3	60.6	62.1	61.7	60.5
Rb	338	403	339	289	448	369	475	394
Ta	5.8	5.7	5.4	5.6	5.5	5.5	5.3	5.4
Th	68.4	79.3	66.7	66.7	63.8	64.3	61.9	63.6
U	21.5	7.4	13.9	7.4	7.1	6.5	28.1	6.5
Y	39.4	43.0	39.6	38.4	29.3	37.4	39.8	42.8
Zr	250	259	261	245	247	259	255	259

1,630 t. Even if the initial water content was significantly higher, assuming 5 wt%, the mass U released by the magmatic fluids represents less than 4,075 t. This amount of U may be somewhat increased if we take into account the fluids expelled from a large underlying magma chamber. Caldera-related magma chambers usually have a volume in the 100–1,000 km^3 range. Thus, taking the same fluid content as in the rhyolites, the volume of fluid released from the whole magma chamber would be 5.2 to 52 Gt. The resulting mass of

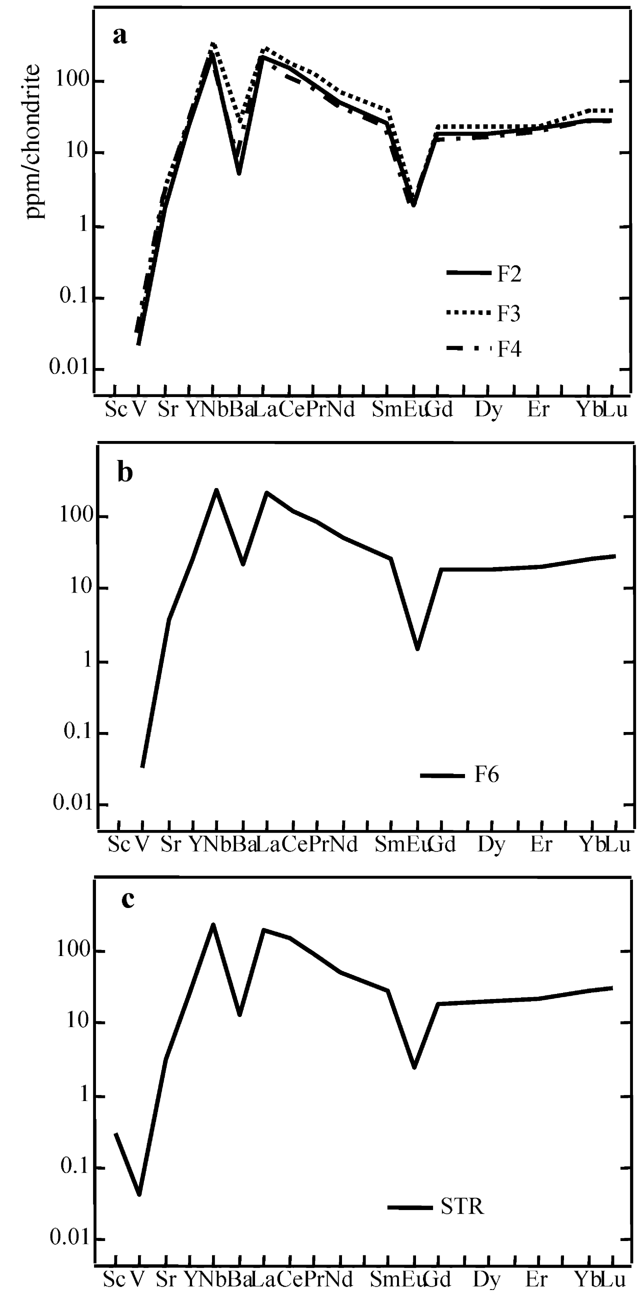

Fig. 5a–c Trace-element (Sc, V, Sr, Nb, Ba) and REE patterns of rhyolites. **a** Tulukuevskoe deposit, **b** Krasny Kamen deposit, and **c** Streltsovskoe deposit

U released by the magmatic fluids, also considering the same U content in the fluid (< 1 ppm), will be in the order of 5,200 to 52,000 t U. These results probably represent an overestimation, because basic and intermediate magmas from the chamber are likely to have lower water and U contents. The quantities of U which may have been released by magmatic fluids remain well below the amounts necessary to explain the estimated resources of the Streltsovka caldera.

The granitic basement

Characteristics of the magmatism

The granites of the basement from the Streltsovka caldera are mainly biotite granites (Andreeva et al. 1996). A biotite leucogranite was recognised in drill core 7c below 2,644 m (Chabiron 1999). The granites are equigranular to porphyritic and characterised by the following accessory mineral paragenesis: magnetite, titanite, apatite, zircon, thorite and allanite, typical of subalkaline granites (Pagel 1982; Cuney and Friedrich 1987; Table 2). The leucogranite is characterised by a texture presenting two stages of crystallisation, with large idiomorphic quartz and K-feldspar crystals and a finer grained matrix mainly composed of plagioclase and microcline crystals with small amounts of biotite. The accessory mineral paragenesis is composed of magnetite, apatite, zircon, xenotime, monazite, thorite and pyrochlore (Table 2).

The granites and leucogranites are variably altered. Despite the numerous drillings studied in thin section and the fact that some of them reached a depth of 2,700 m, fresh plutonic rocks with non-chloritised biotite and non-altered allanite are exceptional. Granite whole-rock analyses are partly disturbed by hydrothermal alteration such as sericitisation, chloritisation, and carbonatation (Andreeva et al. 1990, 1996). A detailed geochemical study (Table 3) was performed on the freshest samples of the 7c drilling which crosscuts the basement from 891 to 2,670 m (Fig. 4), and intersects the most common basement lithologies and some of the major structures of the area. The selected samples present a moderate alteration, as indicated by their low A/CNK index (0.97 and 1.06; Tables 2 and 3; cf. M.Table 5).

Table 4 Average uranium contents of melt inclusions by fission track method and ion microprobe analyses from Streltsovskoe-Antei, Tulukuevskoe and Krasny Kamen deposits

Deposit	Streltsovskoe	Tulukuevskoe	Krasny Kamen	Average U content (ppm)
N	26	15	5	46
U (ppm) fission tracks	17 ± 2	23 ± 3	15 ± 2	19 ± 4
N	–	2	2	4
U (ppm) SIMS	n.d.	14 ± 1	14 ± 0	14 ± 1
Total number of inclusions	26	17	7	50
Average U content (ppm)	17 ± 2	22 ± 4	15 ± 1	18 ± 3

Table 5 Whole-rock compositions (inductively coupled plasma-emission spectrometry and mass spectrometry analyses) of the freshest granitoids of the 7c drilling (Streltsovka caldera)

Rock	Granodiorite				Granite												Leucogranite					
Depth	2,360.5	2,398.0	2,439.5	2,466.5	891.6	1,068.1	1,231.6	1,348.0	1,459.3	1,514.2	1,876.5	1,992.0	2,263.8	2,297.0	2,473.5	2,630.0	2,658.0	2,661.0	2,664.0	2,665.0	2,667.4	2,669.0
SiO_2 (%)	69.26	68.66	66.88	65.87	71.50	70.39	70.91	71.39	70.98	70.62	72.71	71.25	71.42	70.85	73.16	73.85	76.12	76.20	77.16	76.38	75.56	76.95
Al_2O_3	14.90	14.86	16.00	16.18	13.84	14.46	14.48	13.73	14.08	14.43	13.96	14.51	14.37	14.35	13.33	13.28	12.60	12.68	11.95	12.41	12.41	12.10
Fe_2O_3t	2.69	3.24	3.44	4.09	2.34	2.31	2.18	2.44	2.74	2.40	1.98	2.20	2.32	2.97	2.34	1.86	1.22	1.18	1.11	1.16	1.21	1.10
MnO	0.08	0.09	0.09	0.11	0.06	0.08	0.06	0.08	0.08	0.08	0.06	0.06	0.06	0.08	0.06	0.08	0.04	0.05	0.04	0.03	0.05	0.04
MgO	0.65	0.91	0.96	0.91	0.55	0.48	0.48	0.41	0.59	0.54	0.37	0.46	0.58	0.61	0.38	0.29	–	–	0.02	0.02	0.02	–
CaO	2.11	2.44	2.90	2.79	1.79	1.71	1.82	1.36	1.99	1.98	1.41	1.52	1.78	1.75	1.24	1.03	0.28	0.34	0.28	0.39	0.32	0.37
Na_2O	4.74	4.55	5.28	5.24	3.76	4.06	3.85	4.04	4.40	4.41	4.22	4.37	4.26	4.27	4.19	4.33	4.52	4.55	4.10	4.44	4.35	4.24
K_2O	3.34	3.13	2.32	2.52	4.07	4.20	4.49	3.50	3.29	3.71	3.87	4.00	3.75	3.69	3.64	3.97	4.30	4.37	4.28	4.21	4.40	4.24
TiO_2	0.30	0.40	0.40	0.47	0.31	0.30	0.28	0.29	0.34	0.31	0.21	0.25	0.29	0.34	0.21	0.17	0.04	0.03	0.04	0.05	0.05	0.04
P_2O_5	0.11	0.13	0.20	0.17	0.14	0.12	0.03	0.10	0.12	0.13	0.05	0.05	0.07	0.08	0.09	0.03	–	–	–	–	–	–
LOI	0.42	0.88	1.05	1.32	1.32	1.22	1.19	1.38	1.21	0.86	0.99	1.14	0.85	0.69	1.08	0.91	0.72	0.44	0.83	0.77	0.54	0.67
Total	98.59	99.29	99.52	99.67	99.68	99.32	99.77	98.71	99.82	99.46	99.83	99.81	99.75	99.68	99.72	99.80	99.85	99.85	99.81	99.86	98.91	99.76
A/CNK	0.98	0.97	0.97	0.98	1.00	1.01	1.00	1.06	0.98	0.97	1.02	1.02	1.00	1.01	1.02	1.00	1.00	0.99	1.00	0.99	0.99	0.99
La (ppm)	35.2	64.4	43.3	35.3	35.0	39.7	38.5	32.9	41.4	44.9	38.6	41.5	38.9	82.4	42.1	36.7	16.1	19.6	28.0	15.6	20.4	12.2
Ce	71.7	120.3	86.9	68.7	72.3	83.2	81.4	66.9	83.4	91.3	74.5	80.7	79.4	154.4	88.2	78.8	49.9	60.0	79.2	45.6	57.5	40.7
Pr	7.0	11.2	9.1	7.9	7.5	8.9	8.7	7.2	8.8	9.2	7.8	8.5	8.3	15.6	9.9	9.5	7.5	9.0	11.1	6.9	8.5	6.4
Nd	23.4	37.5	31.8	28.9	27.9	29.9	30.1	23.9	31.2	32.3	26.5	29.4	29.4	53.6	32.2	33.7	33.0	38.9	44.0	29.3	34.6	28.8
Sm	4.0	6.4	5.9	6.1	4.9	5.8	5.5	4.6	6.2	5.5	4.5	5.0	5.4	9.3	5.5	7.7	10.9	12.1	13.0	9.1	10.4	9.4
Eu	0.8	1.0	1.1	1.0	1.0	1.1	0.9	0.8	0.8	0.8	0.7	0.7	0.8	0.9	0.5	0.4	0.0	0.1	0.0	0.0	0.0	0.0
Gd	3.9	4.6	5.0	5.7	3.9	4.6	4.4	3.8	5.0	4.2	3.8	4.1	4.4	7.9	4.4	7.0	11.7	13.1	11.6	9.5	9.6	10.0
Tb	0.6	0.7	0.7	0.9	0.6	0.7	0.7	0.7	0.8	0.8	0.6	0.6	0.8	1.3	0.6	1.2	2.1	2.4	2.2	1.9	2.0	1.9
Dy	3.6	3.8	4.4	5.5	3.6	4.4	4.5	4.2	4.7	4.1	3.2	3.8	4.3	7.6	3.6	8.4	13.6	16.0	14.5	12.1	12.3	11.9
Ho	0.7	0.7	0.8	1.2	0.8	1.0	0.9	1.0	1.0	0.9	0.7	0.8	0.8	1.8	0.8	1.9	3.1	3.6	3.2	2.7	3.1	2.8
Er	2.1	2.0	2.4	3.0	2.2	2.4	2.5	2.9	2.9	2.4	1.9	2.0	2.4	4.4	2.1	4.8	7.9	9.2	8.1	7.1	7.2	7.0
Tm	0.3	0.3	0.4	0.5	0.4	0.4	0.4	0.6	0.5	0.4	0.3	0.3	0.4	0.7	0.4	0.8	1.3	1.5	1.3	1.2	1.3	1.1
Yb	2.3	2.2	2.2	3.8	2.5	2.8	2.7	3.9	3.2	2.6	2.2	2.1	2.5	4.8	2.5	5.8	8.2	9.0	7.8	7.3	7.6	7.3
Lu	0.4	0.3	0.4	0.6	0.5	0.5	0.5	0.7	0.5	0.4	0.4	0.3	0.4	0.7	0.4	0.8	1.2	1.4	1.1	1.2	1.1	1.2
Ba (ppm)	801	913	682	653	782	917	796	624	539	661	595	684	826	916	376	258	13	12	15	20	11	12
Nb	13.0	10.6	10.9	15.9	13.7	13.4	14.9	15.7	16.5	12.8	11.4	10.4	11.0	14.1	9.1	20.8	28.4	28.4	12.1	16.2	20.3	18.2
Rb	144	125	114	144	151	167	190	149	156	158	146	131	142	163	186	208	227	238	202	200	213	198
Sr	215	275	329	291	190	211	205	206	220	220	156	175	225	200	105	70	7	6	7	15	9	16
Ta	1.2	0.6	0.6	1.6	1.2	1.4	1.5	2.2	1.6	1.1	0.9	0.9	1.2	2.3	0.7	2.7	1.2	1.4	0.8	1.1	0.6	0.7
Th	18.3	14.6	11.6	14.7	18.8	22.5	22.4	17.6	26.8	21.0	17.0	16.1	15.0	18.9	27.3	29.5	53.4	32.0	37.1	38.9	47.7	34.1
U	6.3	3.4	4.3	6.1	4.8	5.3	5.7	6.4	6.7	6.5	6.8	3.8	4.6	6.0	7.7	9.5	14.1	10.1	8.8	11.8	10.8	9.5
Y	24.5	24.7	24.7	32.5	22.0	26.3	26.3	31.0	33.8	28.0	20.8	21.7	27.3	48.3	21.6	52.7	83.3	94.4	81.7	70.3	79.9	72.0
Zr	225	263	296	343	167	174	165	160	200	190	173	162	186	255	214	173	161	190	132	174	185	216

In the P–Q diagram, most granite samples with ignition loss lower than 1.5 wt% show a quartz enrichment without a major variation of their plagioclase/potassium feldspar ratios (solid squares in Fig. 6). This type of trend is typical of subalkaline granites (Pagel and Leterrier 1980), also called high-potassic calc-alkaline granites (Peccerillo and Taylor 1976). However, the trend occurs for a plagioclase/feldspar ratio located between the potassic subalkaline suites and the sodic subalkaline suites, according to the terminology of Debon and Le Fort (1988). A subalkaline composition is also in accordance with the accessory mineral paragenesis of these granites. A few samples correspond to tonalites of medium-K calc-alkaline composition (inverted solid triangles in Fig. 6). The Streltsovka subalkaline granites are characterised by a moderate LREE fractionation ($La/Sm_N = 3.02$ to 6.33), a low to inverse HREE fractionation ($Dy/Lu_N = 1.17$ to 0.60), and a moderate negative Eu anomaly ($Eu^* = 1.3$ to 1.28; Fig. 5). They have Th contents between 11 and 30 ppm, and U contents between 3 and 10 ppm. They show high Ba (917–258 ppm), Sr (329–70 ppm) and Zr (343–160 ppm) contents.

The leucogranites show a strong quartz enrichment without variation of the K/Na + Ca ratio in the P–Q diagram (solid circles in Fig. 6). LREEs and HREEs are weakly fractionated and the REE patterns present a strong Eu anomaly, characteristic of highly specialised granites (Cocherie et al. 1991; Fig. 6). They are also characterised by low Ca (0.28–0.39 wt% CaO), Ba (10–20 ppm) and Sr (6–16 ppm) contents, and high contents of Th (32–53 ppm), U (8–14 ppm), Nb (12–28 ppm) and Zr (132–216 ppm) for leucocratic granites. The presence of pyrochlore, the vertical trend in the P–Q diagram, A/CNK values lower than 1 and the high Th, U, Nb and Zr contents of the leucogranites are typical of an alkaline to peralkaline composition.

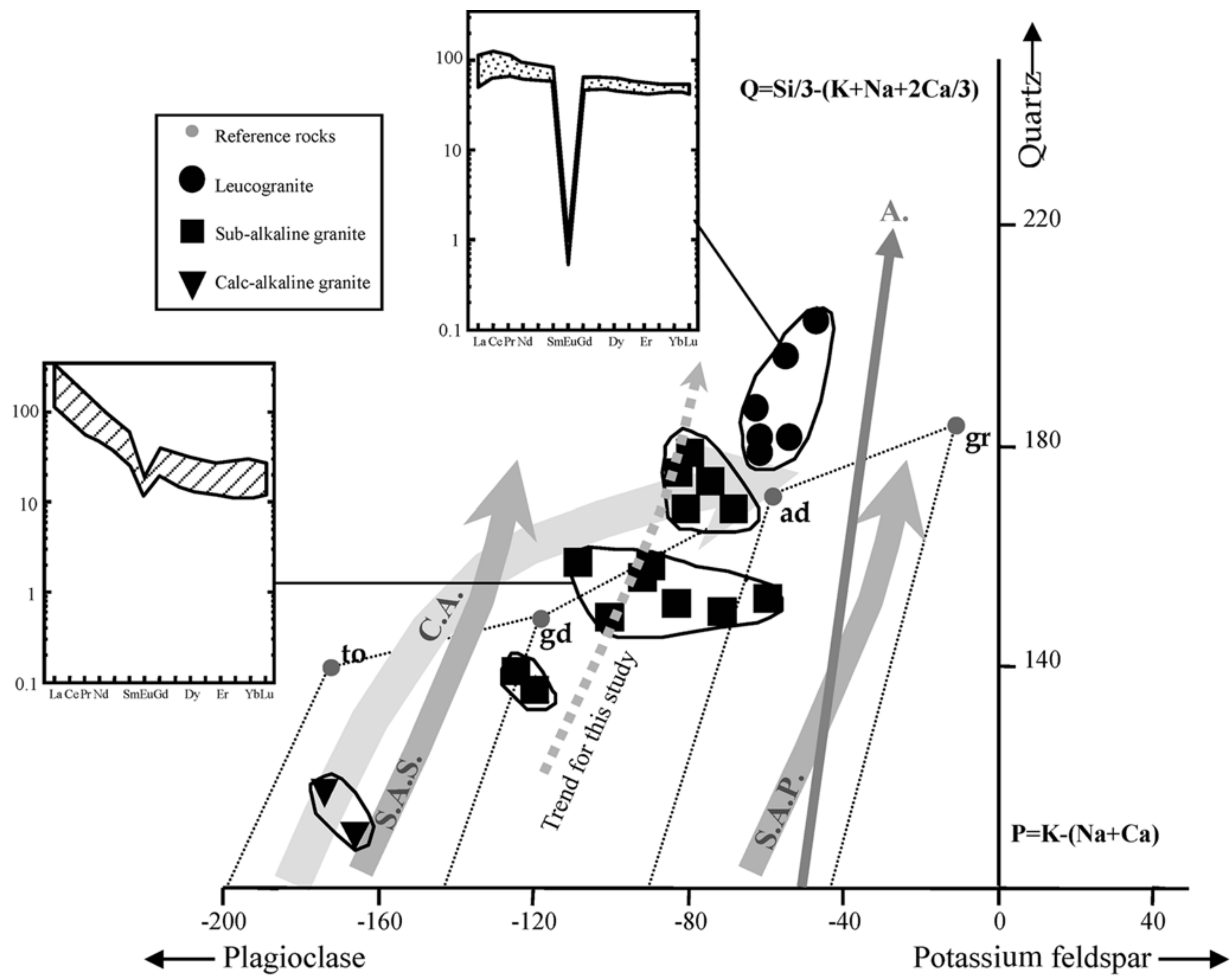

Fig. 6 P–Q diagram (after Debon et Le Fort 1988) with P = K–(Na + Ca) and Q = Si/3–(K + Na + 2Ca/3) in millications, and corresponding rare-earth element patterns spectra of the granites sampled in drilling 7c performed through the Streltsovskoe-Antei deposit. Data are from Table 3. *A* Alkaline reference suite, *C.A.* Calc-alkaline reference suite, *S.A.S.* Sodic subalkaline reference suite, *S.A.P.* potassic subalkaline reference suite, *to* tonalite, *gd* granodiorite, *ad* adamellite, *gr* granite

Uranium distribution

In subalkaline meta-aluminous granites rich in Ca, Th and U, U is mainly located in uranothorite and allanite (Pagel 1982; Cuney and Friedrich 1987). Uraninite is sometimes present. In the Streltsovka granites, allanite and uranothorite have been observed. Uraninite may have been present but leached during hydrothermal alteration of the granite, because of its high solubility in oxidising fluids. Fission track distribution shows that uranium is mainly located in allanite, monazite, zircon, and uranothorite, which largely saturate the detector for the same neutron irradiation as used for uranium determination in the NBS 613 standard (37.38 ppm U). However, uranium is not easily leached by common hydrothermal solutions from such accessory minerals. Metamictisation or specific hydrothermal alteration may alter their structure, and allow the leaching of uranium. Metamictisation more efficiently affects U and Th-rich silicates such as zircon, allanite, uranothorite (Ewing et al. 1988). Phosphates such as monazite and xenotime do not generally become metamict (Ewing et al. 1988). At Streltsovka, hydrothermal alteration occurred during Lower Cretaceous times (Chernyshev and Golubev 1996), ca. 150–200 Ma after crystallisation of the Hercynian granites. Therefore, after such a time span, zircon, thorite and allanite have already undergone a significant degree of metamictisation which enabled the leaching of uranium from their structure.

Evidences of uranium mobilisation

Allanite presents a specific alteration style in the Strelstovska granites (Chabiron and Cuney 2001). Euhedral, fresh allanite crystals have only been identified in a few samples (Fig. 7). Altered allanite consists of an Fe-oxide skeleton which mimics the typical zonation of allanite crystals, filled with newly formed monazite and REE carbonates (Fig. 8). Rare-earth elements, thorium and part of uranium from allanite are trapped in these newly formed minerals. Monazite has a highly variable thorium content (from 0 to 12 wt% ThO_2) and a low uranium content (from 0.1 to 0.2 wt% UO_2). REE carbonates were only detected by SEM because of their small size. They mainly contain LREEs with some yttrium.

Quantification of uranium leaching

Thorite, allanite/monazite, zircon, apatite and possibly uraninite were the major hosts for uranium in the granites, whereas thorite and allanite/monazite were the major hosts of thorium. Microscopic observation shows that about 30% of the allanite volume is filled with newly formed monazite. The same proportion of monazite is obtained by calculation assuming that Th in allanite was not mobile and totally incorporated in the newly formed monazite during alteration. Assuming that the entire Ce content of the whole rock (86.4 ppm on average) was initially contained in allanite (9.8 wt% Ce), the maximum amount of allanite in the whole rock corresponds to 882 ppm. The average Th and U contents in unaltered allanite being 1.05 and 0.10 wt% respectively, the average fresh granite contained 9.2 ppm Th and 0.9 ppm U bound to allanite crystals. About 80% of the Streltsovka granites are characterised by total alteration of allanite to monazite and REE carbonate. Considering Th as an immobile element, the increase of the average Th/U ratio from 10.5 in allanite to 36.3 in monazite shows that 71.1% of the U in allanite was leached during its alteration. Consequently, 882 ppm allanite in the whole rock containing 0.10% U may liberate 0.63 ppm U. Taking an average density of 2.6 for the biotite granite, it results that 1,638 t U/km^3 may have been liberated during allanite alteration. At the caldera scale, using a conservative estimate of the

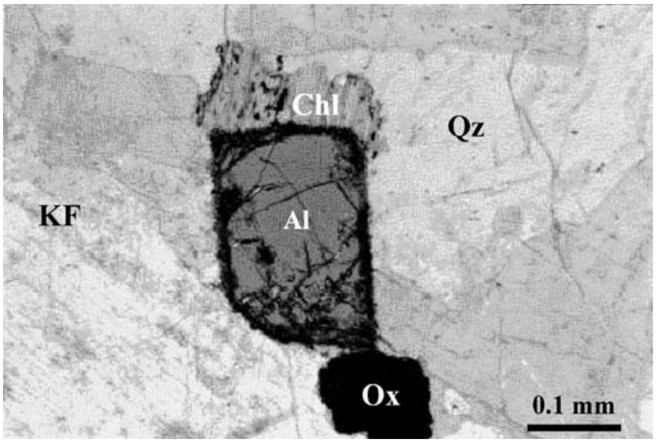

Fig. 7 Microphotograph of a fresh allanite crystal in a granite sample (depth: 1,514.2 m) from the Streltsovskoe-Antei basement. *KF* Potash feldspar, *Ox* oxide, *Qz* quartz, *Chl* chlorite, *Al* allanite

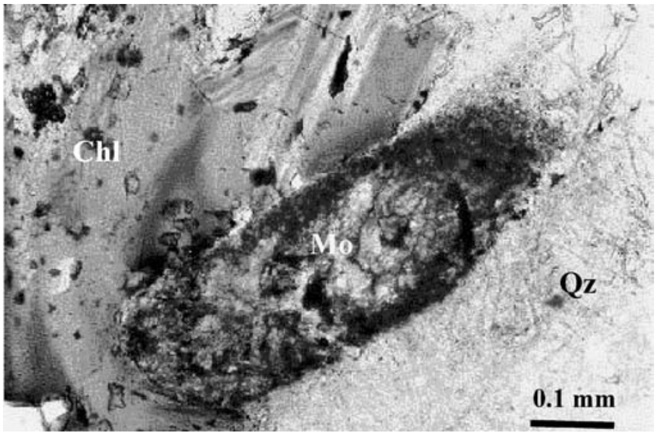

Fig. 8 Microphotograph of an altered allanite crystal in a sample of granite (depth: 2,178 m) from the Streltsovskoe-Antei basement. Allanite shows only a residual structure with a Fe-oxide skeleton filled with newly formed monazite and REE carbonates. *Qz* Quartz, *Chl* chlorite, *Mo* monazite

granite volume of 570 km^3 (considering only the 1,800-m-thick granite layer below the caldera recognised by drillings and a diameter of 20 km) and an alteration of 80% of the estimated volume, a mass of 750,000 t U may have been liberated only from allanite alteration, assuming a 100% efficiency of uranium dissolution and deposition from hydrothermal solutions. Therefore, allanite alteration in the granite below the caldera may be considered as one of the major uranium sources to form the U deposits.

Zircon, apatite and thorite may represent other uranium sources. The Streltsovka granites have an average whole-rock Zr content of 209 ppm. Their zircons contain 47.3 wt% Zr and 0.3 wt% U on average. Thus, the resulting uranium contribution of zircon in the whole-rock U budget is about 1.2 ppm. However, there is no way to determine the initial average uranium content of zircon before its metamictisation, to be able to calculate the proportion of uranium which might have been leached. Apatite with P and U concentrations of 40 wt% and 50 ppm respectively, and with 0.1 wt% of P in whole rock, contributes only 0.13 ppm U to the whole-rock uranium budget. Moreover, apatite is not metamict and thus unlikely to have been a significant uranium source.

Zircon and apatite containing relatively low Th levels, the other main Th-bearing mineral beside allanite is thorite. Allanite hosting 9.2 ppm Th and the average Th content of the biotite granite being 19.5 ppm, 10.3 ppm Th is thought to be in thorite. The thorite from Streltsovka was not analysed because of its very small size and strong metamictisation. Using the Th content of the thorite (52.3 wt% Th) from the Ballons subalkaline granite (Vosges, France; Pagel 1982), 20 ppm thorite in the whole rock is sufficient to explain the Th not bound to allanite in the Streltsovka biotite granites. Additionally, thorite in equilibrium with uraninite at 800 °C contains 30% U (Cuney and Friedrich 1987). Therefore, thorite in the Streltsovka granite may initially have been the host for up to 6 ppm uranium in the whole rock. Because of its strong metamictisation, a large proportion of its uranium may have been leached during hydrothermal fluid circulation. Similarly, uraninite, which may have also existed in these granites, may have been completely leached out. Thus, the calculated 1,638 t U/km^3 liberated from allanite alteration in the granites from the basement is a minimum estimate of the quantity of U liberated during hydrothermal alteration. Taking into account thorite and uraninite alteration, the amount of leachable uranium may have been an order of magnitude higher. The deeper alkaline leucogranite may also have represented an additional uranium source, but its occurrence in the drilling is too limited to be able to make a reliable mass balance calculation.

Allanite alteration was associated with illite formation, especially well developed in the deepest levels of the basement. Andreeva et al. (1996) and Ishukova et al. (1991) have shown that illite alteration was caused by high-temperature (about 300 °C) acidic and reducing fluids which may be partly of magmatic origin. Because of the reducing characteristics of these fluids, U was liberated from the mineral structures during alteration at this stage but probably not significantly mobilised. Uranium dissolution and deposition occurred later, at lower temperature, when the hydrothermal convection cells established above the magma-chamber hydrothermal system were invaded by oxidised meteoric fluids.

Conclusions

Three sources may have contributed to the formation of the uranium deposits in the Streltsovka caldera: (1) the peralkaline rhyolites filling the caldera, (2) the fluids expelled from the volcanic melts or from the underlying magma chamber, and (3) the subalkaline rocks of the basement.

Among the volcanics, the rhyolites alone may have yielded more U than the 300,000-t uranium reserves estimated for the Strelstovska caldera. Mass balance calculations based on the initial uranium content measured on silicate melt inclusions trapped in quartz phenocrysts and average present-day U content of the rhyolites suggest the leaching of up to 900,000 t U.

The magmatic fluids expelled from the extruded rhyolites represent a negligible additional source because of the extremely low $K_{Dfluid/melt}$ between acidic peralkaline melt and Cl-rich fluid. The larger volume of fluids released from the underlying magmatic chamber may have served to slightly enrich the granites of the basement along fractures and may be responsible for the high-temperature brannerite mineralisation occurring in the deepest part of the ore system.

In the basement, subalkaline granites represent the main lithology. Although it was not possible to determine the initial uranium content of the granite magma from melt-inclusion studies, such granites are known for their high U content. Alteration of allanite alone may have liberated 1,638 t U/km^3. Assuming a minimum1.8-km-thick layer of granite below the caldera and allanite alteration in 80% of the granite, the granite may have liberated 750,000 t U. Metamict thorite and uraninite may have also represented still larger uranium sources but their contribution cannot be accurately estimated. Uranium liberated from the accessory mineral structures and other labile uranium sources was mobilised by oxidised meteoric fluids which infiltrated the granitic basement during the development of convection cells above the caldera magma chamber.

Therefore, the juxtaposition of two major uranium sources, e.g. U-rich peralkaline rhyolites and 150- to 200-Ma older, U-rich subalkaline granites, was one of the major parameters contributing to the large size of the uranium resources in the Streltsovka caldera. The acidic hydrothermal solutions, probably partly of magmatic origin, responsible for the pervasive alteration of accessory minerals in the caldera basement, also were essential.

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Appendix

Estimation of allanite (ppm) in the Streltsovka subalkaline granites

- From Ce content:
- Whole rock: 86.4 ppm
- Allanite: 9.8%
- *882 ppm allanite in the granite*

Estimation of Th and U amounts (ppm) in the Streltsovka subalkaline granites originated from allanite

- Whole rock: 19.49 ppm Th, 5.87 ppm U
- Allanite: 1.05% Th, 0.10% U
- *9.2 ppm Th in the granite originated from allanite*
- Then, 10.29 ppm Th in the granite originated from other accessory minerals
- *0.9 ppm U in the granite originated from allanite*
- Then, 4.97 ppm U in the granite originated from other accessory minerals

Estimation of thorite (ppm) in the Streltsovka granites and of U amount (ppm) originated from thorite

- From thorite compositions of the Ballons granite, Vosges, France (Pagel 1982)
- Thorite: 52.25% Th
- 10.2 ppm Th in the granite originated from other accessory minerals
- *20 ppm thorite in the granite*

Estimation of U amount (ppm) released during allanite alteration (Th is assumed to be immobile)

- Allanite: 1.05% Th, 0.10% U
- Monazite: 3.27% Th, 0.09% U
- $Th/U_{allanite} = 10.5$
- $Th/U_{monazite} = 36.3$
- Then, 71.1% U in allanite released during its alteration

Estimation of U amount/km^3 released from allanite during its alteration

- 882 ppm of allanite in the Streltsovka subalkaline granites
- Then, 0.627 ppm U/km^3 released during allanite alteration
- Granite density: 2.6
- *0.627 ppm => 1,638 t U/km^3*

Estimation of zircon content in the Streltsovka granites and of U amount (ppm) originated from zircon

- Whole rock: 209 ppm Zr, 5.87 ppm U
- Zircon: 47.3% Zr, 0.3% U
- ppm of zircon in the granite: 442 ppm
- *1.2 ppm U in the granite is located in zircon*

Estimation of apatite content in the Streltsovka granites and of U amount (ppm) located in apatite

- Whole rock: 0.1% P_2O_5, 5.87 ppm U
- Apatite: 40.0% P_2O_5, 0.005% U
- 2,525 ppm of apatite in the granite
- *0.13 ppm U in the granite is located in apatite*

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