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The K–Ar and Rb–Sr Isotopic Systems in Rocks from the Jurassic Terrigenous Complex of the Greater Caucasus

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Abstract—The behavior of K–Ar and Rb–Sr isotopic systems in clayey rocks from the Jurassic terrigenouis complex of the Greater Caucasus sampled along the Terek River is discussed. It is shown that the rocks experienced intense postdiagenetic alterations accompanied by substantial changes in mineral composition and K–Ar and Rb–Sr isotopic systems. Lateral stress is the leading factor responsible for secondary mineral and geochemical transformations of rocks in cleavage zones. Rejuvenation of the radiological age of rocks, relative to their stratigraphic age, is 100 Ma and more. The age estimate of approximately 50 Ma obtained for samples from the southern limb of the anticlinorium reflects the Paleocene–Eocene phase of tectonic activity manifested in both the Caucasus and other areas of the Mediterranean foldbelt.

Large sedimentary complexes of fold zones experienced noticeable mineralogical and geochemical changes during various stages of their evolution (diagenesis and subsidence to significant depths), intense lateral stress, and cleavage formation. Mineral transformations in terrigenous complexes are reflected, first, in changes of mineral associations, decrease in crystallization degree of minerals, changes of their polytype modifications, and so on. The cleavage development is accompanied by the partial dissolution of primary terrigenous minerals and formation of new ones with simultaneous redistribution of elements or their compounds. These geochemical processes result in changes of proportions of parent and daughter isotopes. Such reorganization of isotopic systems can reset radiological clocks, which enables the dating of postdiagenetic rock transformations. Therefore, isotope data are frequently used for estimating the time of rock alterations under the influence of high *PT* parameters and the consequent rock deformation and cleavage formation included. Turner *et al.* (1994) showed that chemical differentiation of material due to cleavage formation restarted Rb–Sr clocks in Upper Proterozoic Adelaidean metasediments and made it possible to date this event. Similar studies of secondary minerals were carried out by for the Rb-Sr system (Muller *et al.*, 2000) and K–Ar system (Dallmeyer and Reuter, 1989; Fergusson and Phillips, 2001; Hailianga *et al.*, 1997; Krauss *et al.*, 1999; Sherlock *et al.*, 2003; Wright and Dallmeyer, 1991, and others).

We studied changes of isotopic systems in clayey rocks of a large terrigenous complex based on the Lower–Middle Jurassic rocks from the central Greater Caucasus. The geological profile across the entire domain of Lower–Middle Jurassic sequences of the Central Caucasus exposed along the Terek River was investigated (Fig. 1). Rock samples were taken from all Sinemurian–Bathonian sequences.

STRUCTURE OF THE JURASSIC TERRIGENOUS COMPLEX AND MINERALOGICAL CHARACTERISTIC OF CLAYEY ROCKS EXPOSED ALONG THE TEREK RIVER

The geological structure of the study area, its tectonic development, and stratigraphy of Jurassic sequences are considered in many publications (Rengarten, 1932; Gushchin and Panov, 1992; *Yura Kavkaza*, 1992; Panov, 2001, 2003; and others). The mineral composition of rocks and their secondary laterations are discussed in (Gavrilov, 2003; Gavrilov *et al.*, 1992, 1999, 2000).

The geological profile along the Terek River crosses Lower–Middle Jurassic structural-facies zones with different types of stratigraphic section, tectonic structure, lithology, and degree of postdiagenetic alterations.

The Lower–Middle Jurassic rocks compose the axial part of the Greater Caucasus meganticlinorium. Their exposure zone is divided by the longitudinal Shaukhokh thrust into two parts (Fig. 1). The northern part corresponds to the western termination of the asymmetrical Dar'yal–Bogos anticlinorium with the Dar'yal and Gvileti massifs of Paleozoic granitoids exposed in the central part. The southern limb of the

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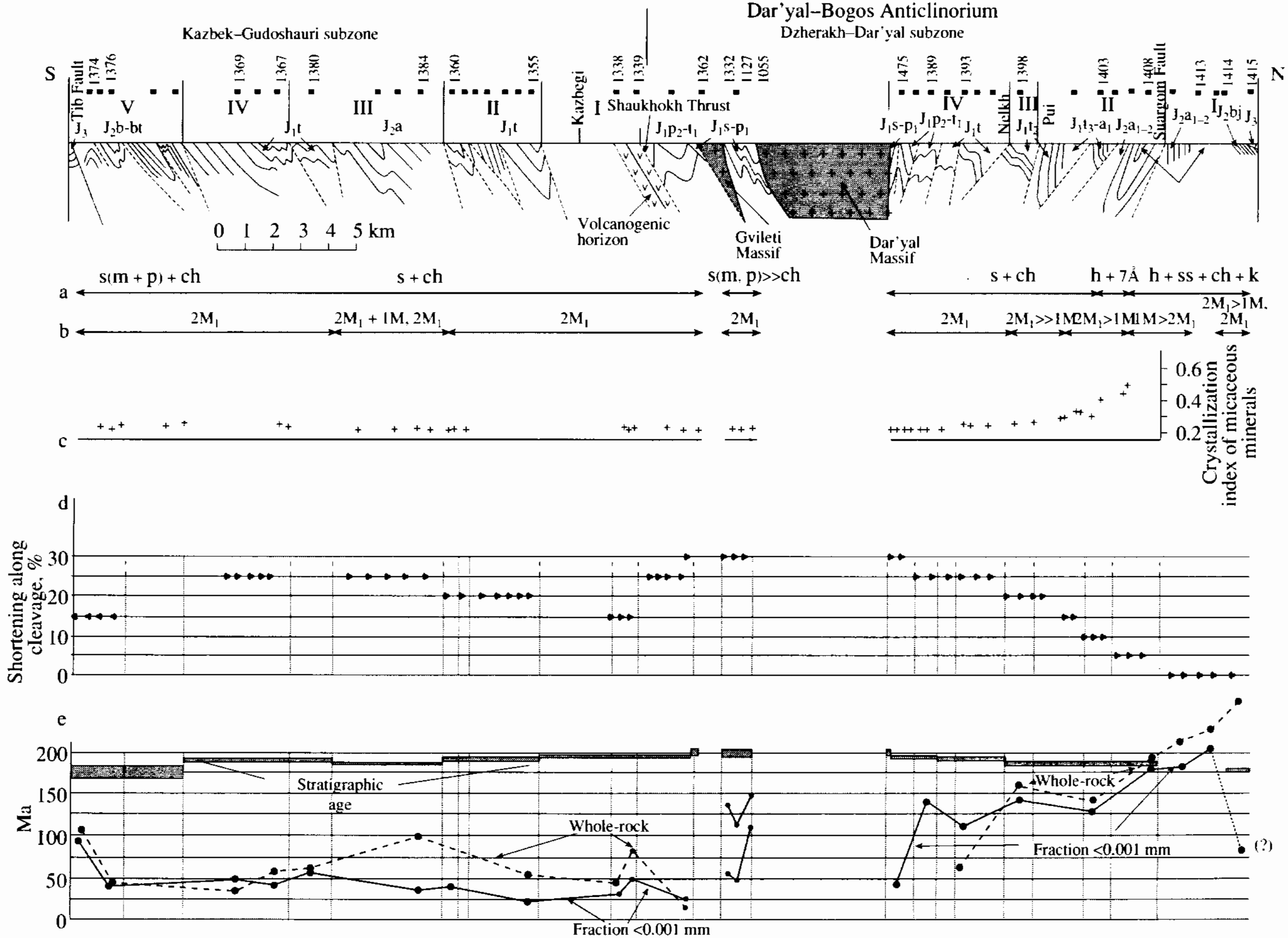

Fig. 1. Geological profile across the Terek River valley and distribution of mineralogical, petrological–structural, and geochemical rock characteristics. (a) Clay mineral assemblages: (k) kaolinite, (ch) chlorite, (ml) mixed-layer mica–smectite mineral, (s) sericite, (m) muscovite, (p) paragonite, (h) hydromica; (b) proportions of polytype modifications of micaceous minerals; (c) crystallization index of micaceous minerals; (d) shortening value along cleavage (%); (e) stratigraphic and calculated K–Ar ages of rocks.

anticlinorium is truncated by the Shaukhokh thrust and fragmentarily preserved. Its wider northern limb represents a locally folded steep homocline divided by longitudinal faults (Suargom, Pui, and Nelkh) into four blocks (nappes). The stratigraphic age in this part of the profile increases in the N–S direction from the Bajocian to Sinemurian.

The southern part of the domain of Lower and Middle Jurassic rocks represents an accretionary nappe–thrust system of the southern slope. It consists of five large nappes separated by northward dipping thrusts. The age of composing nappe rocks gradually decreases from the north (late Pliensbachian) to south (Bajocian–Bathonian). Sediments within the nappes show a normal stratigraphic succession with northward-dipping beds.

The northern and southern parts of the profile generally differ in cleavage intensity. It is intense in the northern part and less developed or missing in the southern part (Fig. 1d). For instance, cleavage is absent in nappe I on the northern limb of the Dar'yal–Bogos anticlinorium. It is observed in rocks of nappe II and its intensity increases southward. Cleavage is intense in nappes III and IV, particularly in the axial part of the anticlinorium. In nappe IV, it is accompanied by the formation of milonite and cataclasite structures. In the southern part of the profile, cleavage is ubiquitous and particularly intense in nappes III and IV where it is developed in both fine- and coarse-grained rocks, indicating strong thrust–fold deformations (Gavrilov *et al.*, 1999, 2001).

The evolution of cleavage from its complete absence to maximum development is accompanied by changes in clay mineral assemblages (particularly noticeable in block II of the northern part of the profile). The hydromica + mixed-layer hydromica/smectite + kaolinite + chlorite assemblage is replaced by the sericite + chlorite assemblage, proportions of polytype mica modifications change from $1M > 2M_1$ to $2M_1 \gg 1M$, and mica crystallization index increases (Figs. 1a–1c).

Jurassic terrigenous rocks locally experienced substantial mineral and petrological–structural transformations, which naturally imply changes in their isotopic systems. Special geochemical studies were carried out to establish the character and degree of such changes.

PREPARATION OF SAMPLES AND ANALYTICAL METHODS

We studied a representative selection of clayey rocks sampled from exposures of Lower–Middle Jurassic sequences along the Terek River valley in the central Greater Caucasus. Location of samples is shown in Fig. 1.

We examined 23 whole-rock samples and fine fractions (<1 μm) of argillites from zones least altered by secondary processes, shales from zones with intense cleavage, and transitional varieties. The rock was crushed and sieved for the whole-rock analysis of the fraction 0.16–0.312 mm. In order to separate fraction <0.001 mm, samples of the fraction 0.16–0.312 mm were grinded by a rubber pestle with the subsequent decantation of the required fraction in distilled water.

K–Ar analysis. The content of radiogenic argon was measured in weighed samples (60–150 mg) on an MI 1201 IG mass spectrometer using the method of isotopic dilution of argon released under the vacuum fusion (1600–1800°C) and subjected to two-step cleaning. The ^{38}Ar tracer purity was 97.5%. The radiogenic argon content was determined with an accuracy of no less than ±1%.

The K content in samples was measured with an accuracy of approximately 1% at the chemical–analytical laboratory of the Geological Institute (I.V. Kislova, analyst) using an AAS-3 atomic absorber.

The calculated age error (2.0–2.5%) was controlled by the convergence of parallel sample measurements and reproducibility of analyses of standard samples. Ages calculations are based on the following constants: $\lambda_e = 0.581 \times 10^{-10}$ yr^{-1}, $\lambda_\beta = 4.962 \times 10^{-10}$ yr^{-1}, and $^{40}K/K = 1.167 \times 10^{-4}$ (Steiger and Jäger, 1977).

Rb–Sr isotopic analysis. The grinded samples (200 mg) were decomposed using the mixture of hydrofluoric and nitric acids with addition of the mixed tracer ^{84}Sr-^{87}Rb at room temperature during 24 h. Then the samples were evaporated with the addition of hydrochloric acid. The Rb and Sr fractions were obtained using the ion-exchange chromatography. The two-step cleaning procedure was applied to fractions for their better cleaning before isotopic analysis. Rb and Sr isotopic measurements were carried out on a MAT-260 mass spectrometer. The accuracy of $^{87}Rb/^{86}Sr$ and $^{87}Sr/^{86}Sr$ measurements was approximately ±1% and ±0.0002, respectively (Vinogradov *et al.*, 1994).

RESULTS

The table and Fig. 1e present results of the study of K–Ar systems in whole-rock samples and their fine fractions, as well as stratigraphic age ranges of formations (after Panov, 2003), and radiometric ages of these stratigraphic intervals (after Harland *et al.*, 1985). As is seen, the timing of the Jurassic terrigenous complex ranges from the Sinemurian to Bathonian (205–170 Ma). The stratigraphic age changes from the Bajocian and Bathonian (~170–180 Ma) in the northern and southern parts, respectively, along the profile to the Sinemurian (~200–205 Ma) in the central part.

Figure 1e shows that the calculated K–Ar ages of rocks also change from the north to south. The comparison of data on whole-rock samples and fine fraction reveals that the latter systemically yields younger ages. This can be explained by both genetic and methodical causes. From the genetic standpoint, clayey rocks (correspondingly, whole-rock samples) contain some admixture of sandy–silty material, which was less subjected to secondary transformations than the clay and

Results of the K–Ar measurement in (s) whole-rock samples and (f) fine fractions from Lower–Middle Jurassic clayey rocks (Terek River area)

Sample no.	Argon rad., mm^3/g	K, %	K–Ar age, Ma	Rb, μg/g	Sr, μg/g	$^{87}Rb/^{86}Sr$	$^{87}Sr/^{86}Sr$	Formation, stratigraphic age
1374s	0.0126	2.90	108					Busarchil, Bajocian–Bathonian (?)
1374f	0.0158	4.02	98					
1376s	0.00606	3.25	47.5	66.15	79.31	2.414	0.71200	
1376f	0.0101	5.77	46	201.5	77.61	7.519	0.71652	
1369s	0.0039	2.81	35.5					Kazbek, uppermost lower–upper Toarcian
1369f	0.00851	4.40	49					
1367s	0.00412	1.82	57					
1367f	0.0093	4.94	47					
1380s	0.00686	3.18	55	141.7	169.7	2.417	0.71473	Gudoshauri, Aalenian
1380f	0.009	4.35	53					
1384s	0.0112	2.82	100	118.2	100	3.423	0.71503	
1384f	0.00641	4.85	34	180.5	112.6	4.639	0.71644	
1360f	0.00689	5.97	38					Kazbek, uppermost lower–upper Toarcian
1355s	0.00913	4.71	51	141.1	64.52	7.678	0.71795	
1355f	0.0051	5.93	22	177.4	119.3	4.303	0.71366	
1338s	0.00343	1.93	45					Tsiklauri, upper Pliensbachian–lowermost Toarcian
1338f	0.00382	3.60	27					
1339s	0.00427	1.35	80					
1339f	0.00239	1.30	48					
1362s	0.0019	2.65	18	81.71	83.21	2.844	0.71735	
1362f	0.00493	5.22	24					
1127/1s	0.0148	2.81	130					Kistinka, Sinemurian–lower Pliensbachian
1127f	0.011	5.21	54					
1132s	0.0162	3.75	115					
1132f	0.00982	4.84	51					
1055s	0.0189	3.18	147					
1055f	0.0196	4.35	113					
1475f	0.00982	5.16	48					
1389f	0.0208	3.90	132					Lars, (t_1^2-t_2)
1393s	0.00591	2.36	63.5					Dzhimara, upper Toarcian
1393f	0.0226	5.08	111					
1398s	0.0143	2.29	154					
1398f	0.0256	4.34	143					
1403s	0.0148	2.70	134					Salgins, lower Aalenian
1403f	0.0352	6.85	128					
1408s	0.0205	2.58	195					
1408f	0.0324	4.35	181					
1413s	0.0220	2.52	212					Koirakh–Fortaukh, upper Aalenian
1413f	0.0321	4.37	179					
1414s	0.0217	2.34	224					
1414f	0.0305	3.67	202					Zgid, Bajocian
1415s	0.0309	2.64	278					
1415f	0.0117	3.61	81 (?)					

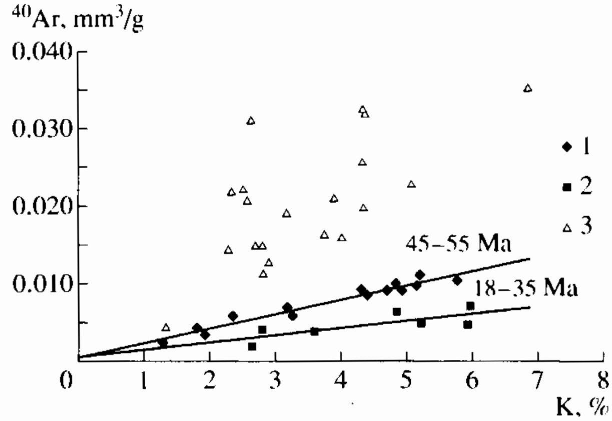

Fig. 2. Results of the K–^{40}Ar isotopic study in K–^{40}Ar coordinates. (1) Samples with K–Ar age of 45–55 Ma; (2) samples with age of 18–35 Ma; (3) other samples with older ages.

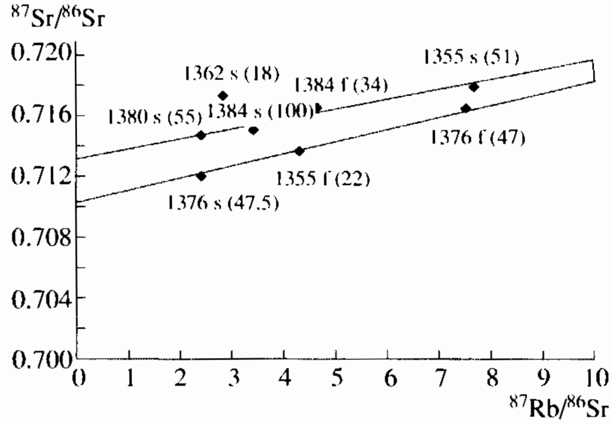

Fig. 3. Rb–Sr isochron plot compiled for (s) whole-rock samples and (f) fine fractions of clays. The K–Ar ages are shown in parentheses.

could retain the age information on older provenances. Methodical reasons for this age difference can be related to the loss of some radiogenic argon during the separation of fine fraction (<1μm) (Gavrilov *et al.*, 2000). Both these reasons could be responsible for age differences of whole-rock samples and their fine fractions. However, it should be noted that three samples show reverse relationships: whole-rock samples are characterized by younger ages than their fine counterparts (table, samples 1362, 1369, and 1393).

The oldest K–Ar age (212–278 Ma) is typical of whole-rock samples from the northern part of the profile (table, samples 1413–1415). This value noticeably (by 30–90 Ma) exceeds the stratigraphic age of corresponding sediments. On the northern limb of the meganticlinorium, the K–Ar age of whole-rock samples and fine fractions gradually and regularly decreases toward the South, although the stratigraphic age of sediments becomes older in the same direction (Fig. 1e). It should be emphasized that the rock age changes against the background of cleavage intensification (Fig. 1d).

Whole-rock samples from the oldest Kistin Formation in the anticlinorium core have an age of 115–147 Ma. The age of fine fractions is 34–76 Ma younger.

In the southern part of the profile, low values of K–Arg age are obtained for both whole-rock samples and fine fractions. The lowest values (18 and 24 Ma, Sample 1362) are registered immediately south of the Gvileti granite gneiss massif. Relative to the stratigraphic age, the radiological age of these rocks is rejuvenated by approximately 170 Ma. Other samples from this part of the profile are also characterized by low measured age values. A group of samples with the age of approximately 45–55 Ma is identified. It is interesting that the difference between ages measured for whole-rock samples and fine fractions is insignificant in (1.5–2.0 Ma samples 1376 and 1380), (Fig. 1e, table) which implies an intense geochemical alteration of rocks.

The obtained results of K–Ar measurements are also presented in K–^{40}Ar coordinates (Fig. 2). This plot is similar to the isochron diagram widely used in the Rb–Sr isotope study. It shows a distinct trend corresponding to the age of 45–55 Ma and a less evident trend of 18–35 Ma. In the upper part of the plot, several data points form a field that mainly corresponds to samples from the northern part of the profile subjected to relatively weak secondary alterations.

The Rb–Sr analysis was only carried out for rocks of the Kazbek–Gudoshauri subzone (Fig. 3, table). It is seen that the obtained data can be approximated on the isochron plane by two straight lines with slopes corresponding to age values of 50 and 60 Ma, which are consistent with the K–Ar data.

DISCUSSION

The established significantly lower radiological ages of rocks, as compared with their stratigraphic ages, results from strong transformations of Lower–Middle Jurassic sequences. Origin and causes of these transformations have a fundamental significance.

As is evident from Fig. 1, the most substantial changes in mineral and petrological–structural properties of rocks are observed in the northern part of the profile at the transition from the northern end to the Dar'yal Massif. The stratigraphic age of sequences also becomes older in the same direction (from the Bajocian to Sinemurian–lower Pliensbachian) in accord with increasing subsidence depth.

Many geological objects exemplify the responsibility of postdiagenetic processes related to the subsidence of terrigenous sequences for both mineral transformations in rocks and changes in isotopic systems. For instance, K–Ar age measurements in rocks from different parts of the Miocene sequence in the Gulf Coast (Louisiana) area showed that their subsidence to the depth of 5.5 km resulted in the rejuvenation of sample ages by several tens of million years (Perry, 1974). Other examples of isotope redistribution in the course of subsidence-related metamorphism and associated mineral transformations are considered in (Vinogradov *et al.*, 1998). The successive reorganization of isotopic systems in different formations of the Riphean sequence in the Uchur–Maya plate is explained by their subsidence and the successive crossing of a certain critical depth and temperature level. Riphean rocks of the Yudoma–Maya Trough in eastern Siberia are substantially more altered (Vinogradov *et al.*, 2000), relative to those in the platform part of the region (Vinogradov *et al.*, 1998), and characterized by the initial stage of metamorphism. Rejuvenation of the K–Ar age is attributed to processes of potassic metasomatism.

Considering postdiagenetic alterations in Jurassic terrigenous rocks of the Greater Caucasus, it should be kept in mind that lower horizons of the sequence were subjected to significant geostatic loads. According to our estimates, upper layers of the Jurassic terrigenous complex were subsided to depths of approximately 2.0–2.5 km. Taking into consideration that the thickness of Liassic–Aalenian sequences is approximately 5.5–6.0 km, the total thickness of the Mesozoic sequence in the axial paleobasin of the Greater Caucasus can be estimated as 7.5–8.5 km. It can be assumed that the subsidence-related geostatic loads and temperatures noticeably influenced mineral transformations and changes in isotopic systems of mineral assemblages in lower parts of the rock complex.

However, it is worth mentioning that coeval Aalenian rocks in the northern and southern parts of the profile, which subsided to approximately similar depths and, correspondingly, experienced similar geostatic loads, substantially differ in mineral compositions and radiological ages. It should be emphasized that rocks in the southern part of the profile are represented by intensely cleaved shales, whereas argillites from the northern part are lacking cleavage. Analysis of obtained isotope data suggests that regular and coordinated changes in the cleavage intensity and mineralogical–isotopic characteristics of sediments along the profile indicates the leading role of the lateral stress in geochemical processes of mineral transformations.

According to geological data, early phases of thrust–fold structure formation in the Greater Caucasus and initial stress loads on accumulated sediments occurred in the pre-Bajocian time. Correspondingly, some associated mineralogical, geochemical, and structural alterations of Liassic–Aalenian rocks, which composed the central uplift system in the axial Greater Caucasus by the beginning of the Bajocian time, are also referred to the same period. As a result of the erosion of these uplifts, sedimentary material with typical parameters of substantially altered rocks (prevalence of micas of the polytype modification $2M_1$) (Gavrilov *et al.*, 1992, 1999), started to enter the Bajocian–Bathonian basin. This was probably responsible for the relatively older measured age relative to the stratigraphic one in lower Bajocian rocks in the northern part of the profile (table, Sample 1414). At the same time, compositional rock transformations under the stress load continued up to the Cenozoic, which is confirmed by the radiological age of rocks. The Cenozoic transformations of sediments significantly changed and masked the age signature of events that occurred during the Jurassic and Cretaceous.

The lowest measured age values are typical of the southern part of the profile with the typical accretionary structure. This structure likely formed during a sufficiently long period under the influence of many stress events. Tectonic nappes separated by large fractures could differently respond to the external load, which probably explains some differences in the behavior of isotopic systems in rocks within individual nappes.

Figures 1 and 2 demonstrate a large group of samples with the age interval of 45–55 Ma. This age, registered by both K–Ar and Rb–Sr isotopic systems, likely reflects a real event in the history of the Greater Caucasus. The event was accompanied by not only the release of radiogenic argon, but also intense redistribution of K, Rb, and Sr. The regression line drawn for the samples with K–Ar age of 45–55 Ma (Fig. 2) almost exactly crosses the point of origin. This means that the K–Ar clocks were set at the zero mark at the end of this geological episode. The Rb–Sr system probably behaved in the similar manner (Fig. 3), but the isotopic strontium ratio had no time to become averaged in the entire rock succession. Therefore, one can see two trends with similar slopes and different strontium ratios.

This time interval coincides with the substantial activation of tectonic processes in both the Caucasus region and other areas of the Mediterranean foldbelt. The middle Eocene was marked by the closure of the southern branch of the Tethys and collision of the Arabian Plate with the accretionary belt of the Eurasian Plate. In the Greater Caucasus, this resulted in the eventual formation of flysch troughs, main orogenic phase, and thrusting on the southern slope (Zonenshain *et al.*, 1987; Koronovskii *et al.*, 1997; Lomize, 1987; Khain and Balukhovskii, 1993; Panov, 2001; and others).

It cannot be ruled out that the group of data points in Fig. 2 corresponding to the Miocene age trend also reflects real processes of the Attic tectogenesis.

Three whole-rock samples of the Kistinka Formation of the Jurassic sequence yielded an age of 115–147 Ma. The relevant fine fractions yielded ages ranging from 51 to 113 Ma. As was mentioned, the relatively older age of whole-rock samples from the Kistinka Formation can be related to the presence of a significant admixture of sandy–silty material, which is an erosion product of Paleozoic granitoid massifs and, correspondingly, carries information on its older origin. In addition, these massifs could serve in the Mesozoic and Cenozoic as a relatively rigid framework, which partly impeded the influence of stress on adjacent sedimentary rocks.

However, Paleozoic granitoids were also sensitive to the lateral stress. Interesting data on the age of the Dar'yal granite gneiss massif are given in (Dudauri *et al.*, 2000a, 2000b). These authors studied the K–Ar system in rock samples from different parts of the massif and obtained a relatively significant scatter of measured age values. Analysis of micaceous minerals from relatively fresh and least deformed rocks of the massif showed the age of 321 ± 6 Ma, which corresponds to the Middle Carboniferous. Similar values were also obtained for rocks from the Dziruli and Lok massifs (Dudauri *et al.*, 1999). However, analysis of biotites from cataclastic granitoids provided different age values (163–182 Ma). Therefore, one can probably assume that the formation of cataclastic zones related to stress load was responsible for the noticeable decrease of radiometric dates for rocks from some parts of the Dar'yal Massif.

Thus, under the influence of secondary processes probably related to stress episodes, both Jurassic terrigenous sediments and Paleozoic igneous rocks were subjected to changes in original isotopic systems with a substantial rejuvenation of their radiological age.

The comparison of rocks exposed along the examined profile clearly shows (Fig. 1) a rapid transition from the relatively fresh argillites in the northern part to cleaved shales in central parts. The maximal gradient of changes in the mineral composition of rocks and their radiological age is observed in relatively narrow intervals a few kilometers in width. In our opinion, this can explained by the following causes.

The central Caucasus was complicated by synsediemntary faults with associated olistostromes (Gavrilov, 2003). One of such faults of the sub-Caucasian strike established in the Gizel'don River basin continues to the Terek River valley and reaches the southern Suargom-don River area approximately corresponding to the zone of most drastic mineral and geochemical rock alterations. The southern basement of the Jurassic trough, which accumulated thick terrigenous sequences, subsided along this fault, as well as other closely spaced and similarly oriented faults, during the Early Jurassic and Aalenian time. Thus, the topography of the basin basement surface in this zone was probably characterized by the stepwise pattern with subsided southern blocks; i.e., the basin basement had a substantially steep bent. During the subsequent compression (stress), this bent (scarp) served as a rigid stamps. Therefore, rocks on the southern side of these faults underwent substantial alterations, whereas rocks on the northern side were only slightly altered. During later deformations related to the formation of the Greater Caucasus, rocks with stress-related cleavage were uplifted to the level of fresh argillites. We assume that in addition to the system of Suargom faults in the Terek River valley, the large Pui and Nelkh faults are also synsedimentary fractures. During compression (stress) episodes at a certain stage of the basin evolution, northern segments of the large faults served as rigid stamps. At the same time, all tectonic nappes in the southern accretionary part of the profile were subjected to more significant stress loads. Consequently, rocks therein underwent strong alterations of the mineral and isotopic compositions.

CONCLUSIONS

(1) The Jurassic terrigenous sequence of the Greater Caucasus underwent intense postdiagenetic transformations accompanied by substantial changes in the mineral composition of rocks and their isotopic systems. Clay minerals in intensely cleaved rocks completely lost the original isotopic signature. Lateral stress was a leading factor responsible for the secondary alteration of rocks in zones of cleavage development.

(2) The age estimate of approximately 50 Ma obtained for rocks from the southern limb of the anticlinorium records the Paleocene–Eocene phase of intense tectonic activity, which is manifested in both the Caucasus and other areas of the Mediterranean foldbelt.

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