

FIG. 1. Location map: 1. Daugmales Tomēni; 2. Kaibala; 3. Līcupe; 4. Test drilling No. 21 in the Gulf of Riga; 5. Kolka; 6. Jūrkalne; 7. Kihnu; 8. Elblag; 9. Jonionys; 10. Rybatskoe; 11. Zaton; crosses show localities of *Portlandia arctica* in till (after Dreimanis, 1949).

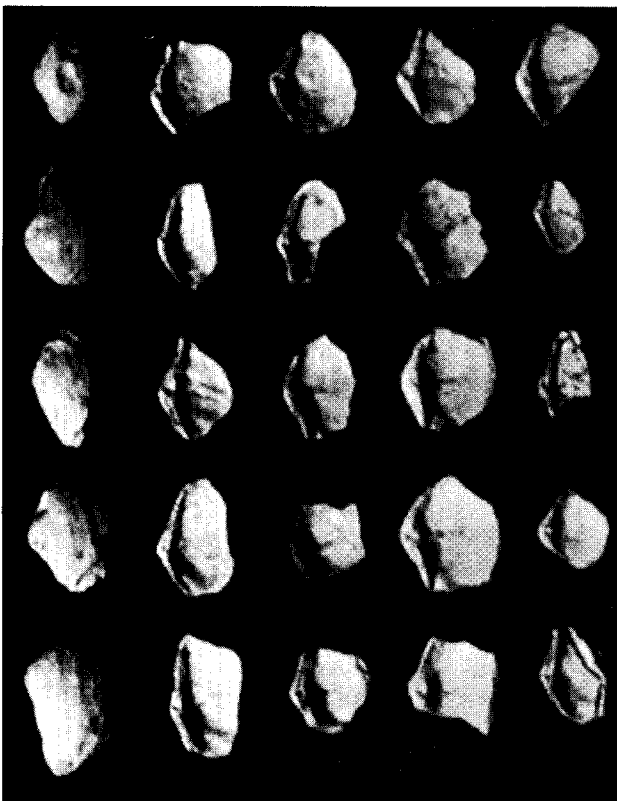


FIG. 2. Reworked abraded *Portlandia arctica* shells from till at the Daugmales Tomēni site (from Zāns and Dreimanis, 1936).

geomorphological observations have led to the same conclusions. Marine fauna, occasionally represented in the Estonian tills, occurs in a redeposited state (Raukas, 1973). Still, because of the controversy about the Līcupe site where *Portlandia arctica* occurs in association with marine microfossils in several types of sediments (see further), we decided to reinvestigate this section in more details.

In previous publications recording the measure of *Portlandia arctica* in Latvia, different ages have been proposed for their source sediments, ranging from Holsteinian to Late Weichselian. It is therefore, important to determine the physical age of the shells collected in Central Latvia by reliable dating methods. The electron spin resonance (ESR) method has been found useful for such purposes. Since Ikeja and Ohmura (1981) first recognised the mollusc shell material as a possible dating object by electron spin resonance, the ESR method has been gradually improved and has become the major tool for mollusc-based chronostratigraphy for Pleistocene to Holocene shell-bearing deposits of various genesis beyond  $^{14}\text{C}$  dating range (see e.g. Gaigalas and Molodkov, 1996; Molodkov, 1989b, 1995, 1996; Molodkov *et al.*, 1992; Molodkov and Raukas, 1996; Skinner, 1989; Shimokawa *et al.*, 1992). In combination with classical geological methods ESR is becoming a promising approach for the study of sea-level changes, sedimentary dynamics, palaeoenvironmental

reconstructions, and correlation of Quaternary deposits over broad geographical areas.

### EARLIER FINDINGS OF *PORTLANDIA ARCTICA* SHELLS IN LATVIA AND THEIR STRATIGRAPHIC ASSIGNMENTS

The first report on *Portlandia (Yoldia) arctica* Grey in Latvia was by Gailitis (1933), erroneously assigning *Portlandia* to the Lateglacial Yoldia sea. He found the shells in gravel in an exploration trench along the bluffs of River Daugava about 3 km S of Salaspils (Fig. 1(1)). Zāns and Dreimanis (1936) noted that these gravels were covered by two till layers of the last glaciation and therefore the *Portlandia arctica* shells could not be of Lateglacial age. They also found numerous partly abraded or broken shells of *Portlandia arctica* (Fig. 2) in tills, varved clays and gravels of last glaciation along the bluffs of River Daugava in the same area and up to 6 km upstream.

Zāns and Dreimanis (1936) concluded that all the above mentioned *Portlandia arctica* shells had been transported and redeposited by glacier ice and its meltwaters during the last glaciation. They suggested that the original marine sediments whence the glacier had picked up the shells had been deposited during the last interglacial. They tentatively correlated these marine sediments with the Boreal transgression of northern Russia (Potulova, 1921) and the Skærumhede transgression of Denmark and Rügen (Ødum, 1933).

Zāns (1936) elaborated on the above correlations and pointed out that the Portlandia Sea existed after the Eemian time, separated from it by a cool climate interval with subarctic flora (Jessen and Milthers, 1928: zone k), indicating probably an advance of the Scandinavian ice sheet. According to the present day stratigraphic terminology (Behre, 1989), the Portlandia Sea would be of Early Weichselian age, correlated with the middle part of oxygen isotope Stage 5.

About 30 more localities of glacially redeposited *Portlandia arctica* shells in Latvia were reported by Dreimanis (1949). They are shown in our Fig. 1. All of them had been found in the Weichselian glaciogene deposits, mainly in the area S, SE and SW of the Gulf of Riga: in reddish brown till or glaciofluvial gravel. Pēr-kons (1957) reported *Portlandia arctica* also from the grey till at Kaibala (Fig. 1(2)) underlying two reddish brown tills. He proposed that these *Portlandia arctica* shells were incorporated by the Moscow (Late Saalian or Warthe) glacier from Odintsovo interstadial marine deposits.

Ulst and Majore (1964) discovered *Portlandia arctica* in primary silty marine sediments of the last interglacial, in a test drilling at Kolka (Fig. 1(5)), in association with arctic-boreal foraminifera. However, according to Meirons and Straume (1979) (p. 227) these marine silts may be a glacial raft, because V. Juškevičs found only till with lumps of fossiliferous marine sediments in the adjoining test drillings at the same depth.

Abundance of *Portlandia arctica* shells was noted in grey till and associated dark grey to black clays at Līčupe (Fig. 1(3)) by G. I. Mironov in 1960 and G. Eberhards in 1965 (unpublished reports), initiating investigation of this site by at least ten other geologists during 1965–1974. Afanasyev (1967, 1968) summarized some of their findings and speculated that all of the Līčupe deposits containing *Portlandia arctica* were marine sediments *in situ*, and, because of their occurrence 110–120 m above the present sea level, assigned them to a marine transgression of the penultimate (Riss) glaciation.

Danilov *et al.*, (1968) linked the formation of the *Portlandia*-bearing deposits in the Līčupe section with a Pleistocene cold-water marine basin. Vegetation on the basin's shores, however, was indicative of moderate climatic conditions.

Konshin *et al.*, (1969) described and discussed the Līčupe section in considerable detail and interpreted the *Portlandia arctica* containing clays and clayey diamictons as interstadial marine sediments deposited at the end of the Frankfurt Stadial of last glaciation, raised by neotectonic uplift to the 110–120 m elevation.

Danilov (1970) (Table 1) proposed, that the Līčupe clays were deposited in the 'Līčupe Lake' during a ice free interval of the penultimate glaciation.

Danilans (1973) (pp. 137–138), referring to published and unpublished reports on the Līčupe site, concluded that its *Portlandia arctica* containing deposits were glacially redeposited marine sediments of the Riss-Würm interglacial.

Hoping to resolve the age problem of the *Portlandia arctica* shells at Līčupe, Krūkle collected shells from the black marine clay in the till unit III of Fig. 3, but in an adjoining section a few metres upstream from ours and submitted them for <sup>14</sup>C dating at the Geochronology laboratory of the University of Leningrad. Two age determinations were published in Krūkle and Arslanov (1977) (p. 191), the most reliable being LU-198B: ≥ 41,160 BP, from the internal shell fraction. They concluded, considering also palaeoecological and lithostratigraphical data from the Līčupe section, that the marine intermorainic deposits were not younger than the first half of a lengthy mid-Valdai (= mid-Weichselian) interstadial complex.

Meirons and Straume (1979) (Fig. 71) published a 400 m long profile section of V. Juškevičs, based upon river bank exposures at Līčupe and test drillings down to bedrock, and agreed with the above mentioned conclusions of Danilans (1973).

'Marinists' Afanasyev *et al.*, (1979), however, disagreed with Danilans (*ibid.*) and proposed that the deposits containing *Portlandia arctica* and/or marine microfossils at Līčupe and several other localities in Latvia were cold-water marine sediments of the Middle Pleistocene Līčupe Formation. They correlated it with the beginning of the Holsteinian Sea transgression, referring also to the presence of *Portlandia arctica* shells in marine inter-till deposits *in situ* in the Holsteinian Ulmale Formation in W Latvia

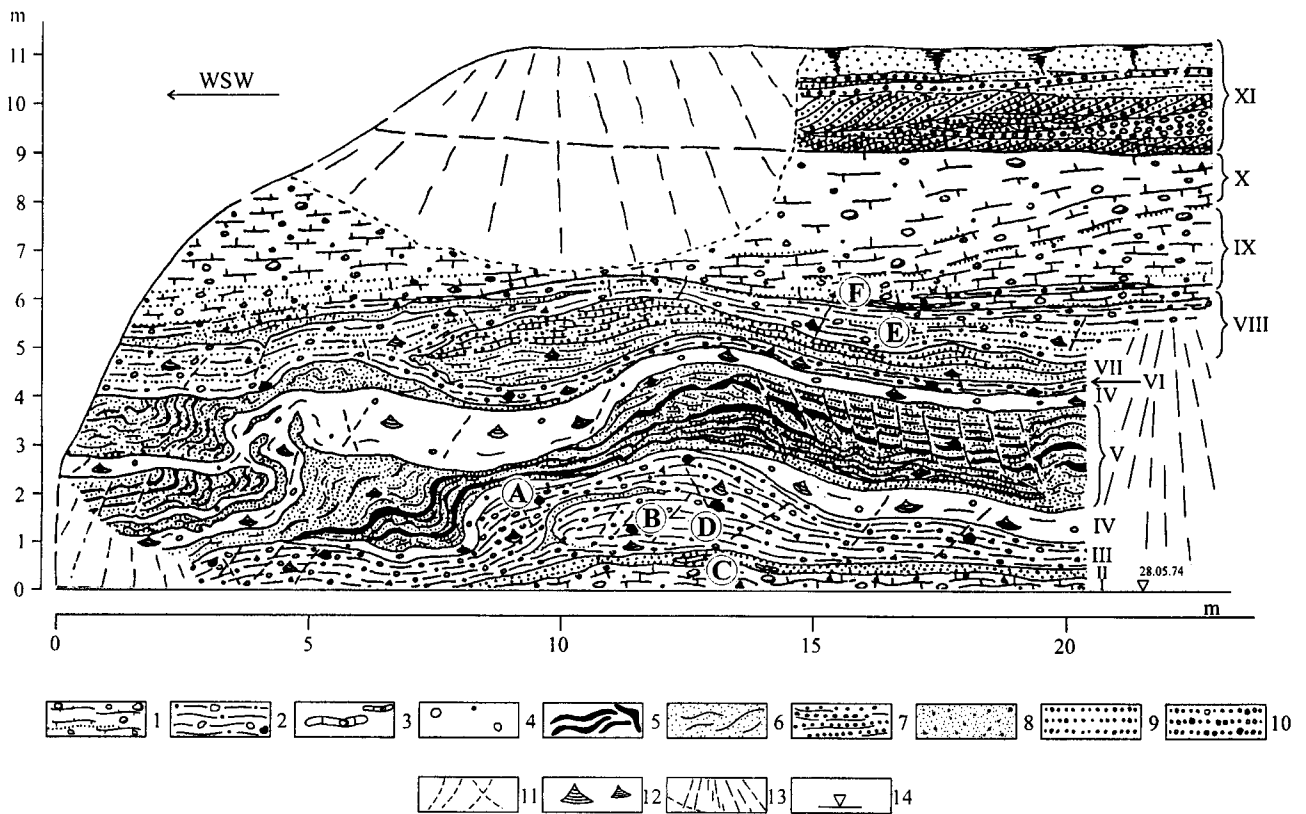


FIG. 3. Līčupe section: 1 — reddish-brown sandy till; 2 — grey (brownish to nearly black) clayey till with grey to black lenses of clay and many shells or fragments of *Portlandia arctica*; 3 — lenses of reddish brown till (1) in sand; 4 — dark grey to bluish or greenish silty clay with occasional pebbles and shells or fragments of *Portlandia arctica*; 5 — thin layers or lenses of dark grey to black silty clay; 6 — light grey or yellowish silty sand with *Portlandia arctica* shells; 7 — medium to coarse grained sand; 8 — sand of various grain sizes; 9 — medium to coarse grained sand with pebbles; 10 — sandy gravel with pebbles; 11 — fractures; 12 — *Portlandia arctica* shells; 13 — slump; 14 — water level in the River Ogre on May 28, 1974; A — our sample 134-086; B — our samples 230-086 and 231-086; C, D, E and F — place of measurements of the orientation of elongated pebbles (Fig. 4); Layers: I — reddish brown loamy till; II — sand with lenses of till; III — multicoloured loamy till with inclusions of clay and *Portlandia arctica* shells; IV — dark grey clay with *Portlandia arctica* shells; V — silty sand with thin interbeds of dark grey silty clay continuing *Portlandia arctica* shells; VI — same as III; VII — stratified sand with some *Portlandia arctica* shells in the silty sand; VIII — same as VI; IX and X — reddish brown fissile sandy till; XI — alluvial gravel and gravelly sand.

(Fig. 1(6)) that were reported first by Konshin *et al.*, (1969). Danilov and Nedesheva (1982) expressed similar views and emphasized that the Līčupe grey stony clays containing *Portlandia arctica* were marine sediments *in situ* deposited in an arctic-type sea of very old age.

#### DESCRIPTION OF INVESTIGATED SITES

Samples for ESR age determinations were collected from Central Latvia at Daugmales Tomēni and Līčupe sites (Fig. 1(1) and (3)). This region is known as an important area of several till deposits with many occurrences of *Portlandia arctica* shells (Fig. 1). Three samples analyzed in this investigation were collected from two stratigraphic levels at Daugmales Tomēni and four samples from black marine sediment lenses and till at Līčupe. Glaciogenic deposits at these sites are believed to be of Weichselian glaciations based on published stratigraphic and geomorphological evidence and our reinvestigation while collecting the *Portlandia arctica* samples.

#### Līčupe section

The Līčupe section (Fig. 1(3)) is on the north side of River Ogre about 300 m downstream from the mouth of its tributary Līčupe. As already mentioned above, the Līčupe section has been extensively studied, described and discussed in numerous unpublished and published reports during the period of 1960–1982. The most complete profile section was published by Konshin *et al.*, (1969) (Fig. 1). Our Fig. 3, measured by Āboltiņš in 1974 and reinvestigated in 1995, presents more details, including glaciotectionic deformations, and till fabric measurements are added on Fig. 4. Most of the lithologic and microfossil data from the Līčupe section were presented by Konshin *et al.*, (1969), Krūkle and Arslanov (1977) and Afanasyev *et al.*, (1979), and we are summarizing some microfossil data in Table 1.

*Portlandia arctica* and marine microfossils occur in the layers III–VIII. Layers III, VI and VIII are silty clay diamictos, grey to brown in colour, with inclusions of grey to black marine clays as lenses or lumps. We interpret these diamictos to be local tills consisting mainly of glacially reworked marine clay, because

megablock of penultimate marine interglacial deposits enclosed in till of the last glaciation. We principally agree with this interpretation, with some amendments. We think that it is a glacial raft or floe, but it consists of interlayered last glaciation local tills (units I, III, VI and VIII) and interstadial marine sediments (units II, IV, V and VII). The raft was transported by the Riga ice stream during the last glaciation (Dreimanis and Zelčs, 1995) from the Gulf of Riga as indicated by the orientation of elongate (Fig. 4) pebbles and shear planes (Fig. 3) in till units VIII and IX. The difference in pebble orientation maxima by  $50^\circ$  in till units I and III suggest that the lower part of the raft may have been shifted by  $50^\circ$  during glacial transport.

Pollen and spore diagrams are available from two Līcupe sections (Konshin *et al.*, 1969, Fig. 2: from samples taken along the lower 4 m of a site close to our section of Fig. 3 and in Krūkle and Arslanov, 1977, Fig. 3: samples taken from the lower 1.5 m in a section a few metres to the right of the previous one). After a close examination of the type of sediments analysed and their pollen spectra, these diagrams appear to contain redeposited pollen. Still, they are useful for concluding the probable source of redeposition.

Most of the samples for both pollen diagrams were taken from a clayey diamicton (mainly unit III of our Fig. 3), interpreted by us to be subglacial till containing incorporated marine sediments (see above). However, six samples of the section investigated by Krūkle and Arslanov (1977) were taken at 3 cm interval from a 20 cm thick lump of black marine clay with many shells of *Portlandia arctica*, and we had a great hope to accurate information about the vegetation and climate during the lifetime of *Portlandia arctica*. The pollen and spore spectra of these six samples are extremely variable, for instance the *Betula* percentages range from 5 to 70, *Pinus* from 30 to 75, *Picea* from 5 to 33, *Carpinus* from 0 to 12 in adjoining samples. As demonstrated by Liivrand (1991) in several pollen diagrams from Estonia, such highly variable pollen assemblages are typical for periglacial clay and silt deposits, with admixture of secondary interglacial pollen. The same may be said about the palynomorph assemblages in tills, where *Pinus*, *Betula* and *Alnus* (in decreasing abundance) dominate the section of Krūkle and Arslanov (1977) and *Betula*, *Pinus* and *Alnus* in the Konshin's *et al.* (1969) section. The cold climate indicator *Selaginella selaginoides* and QM, in low percentages are present in the tills of both sections, and *Corylus* (5–15%) and *Picea* (2–10%) form continuous curves.

Cold climate conditions are indicated also by the presence of arctic sea microfossils, such as the ostracodes listed in Table 1 (Konshin *et al.*, 1969) (p. 40) and the shells of *Portlandia arctica*. The predominance of transitional forms between the subspecies *Portlandia arctica arctica* and *Portlandia arctica siliqua* (see above) suggests a transition from cold to boreal waters.

In the till (unit III) where our *Portlandia arctica* shells were taken, marine foraminifera are very abun-

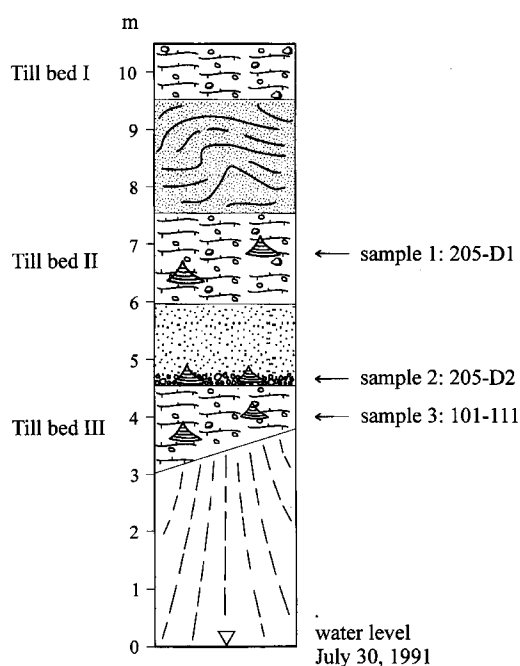


FIG. 6. Vertical profile of the River Daugava bluff section at Daugmales Tomēni, where samples were taken in 1991 for ESR dating. For symbols see Fig. 3.

dant and marine diatoms dominate over freshwater ones (Table 1). However, in Krūkle's section, the sampled sediments are slightly dominated by freshwater diatoms, suggesting influx of freshwater into the marine basin, or incorporation of some freshwater sediments.

#### *Daugmales Tomēni section*

This is a section of River Daugava bluffs (Fig. 6), about 3 km S of the railway station at Salaspils (Fig. 1(1)), described and discussed by Dreimanis (1935, 1943), Zāns and Dreimanis (1936), Dreimanis (1970) and more recently by Dreimanis *et al.* (1996). At least 5 layers of reddish brown tills of the last glaciation have been reported in this section (Zāns and Dreimanis, 1936), all of them containing abraded shells of *Portlandia arctica*. Since the tills and interbedded stratified beds (also containing *Portlandia* shells) are strongly glaciotectonically deformed, some repetition of the layers occurs because of overthrusting. The lithology of tills and the orientation of deformations and clasts in them, as reported in the above publications, indicate that the Riga ice stream or lobe deposited the tills. The proximity to the Gulf of Riga (Fig. 1) suggests that the source of sediments with *Portlandia arctica* shells was the Gulf of Riga (Zāns and Dreimanis, 1936). All till layers of the last glaciation, and also the interbedded sands and gravels had been oxidized by percolating water and their colour was in various hues of brown. The tills are silty or sandy, with low percentage of clay particles ( $< 0.01$  mm), ca. 25%; Dreimanis, 1943).

## DATED SAMPLES

### Daugmales Tomēni

The samples for ESR dating were taken in 1991 from the bluff section (Fig. 6) of River Daugava at Daugmales Tomēni (Fig. 1(1)) near the W end of the old clay pit ('Tomēnu mālu raktuves' in Fig. 3 of Zāns and Dreimanis, 1936).

Two shell fragments of *Portlandia arctica* (Sample 205-D1) were taken from the second reddish brown till. The available amount of shell material was about 105 mg that allowed us to prepare 3 aliquots, 34 mg each for ESR analysis.

One *Portlandia* shell fragment (205-D2) is from the gravel on the top of the third till at the same section. The gravel on the third till is just reworked underlying till material. Three aliquots of 48 mg each were prepared for analysis.

From the third till only a sediment sample (No. 101–111) was available for parallel gamma-spectrometric analysis.

### Ličupe

Three *Portlandia* shell samples from Ličupe site (Fig. 1(3)) were taken from till layer III which is very rich in the original marine sediments, at the section depicted in Figs. 3 and 5, and from an adjoining one discussed by Krūkle and Arslanov (1977). This till was deposited probably by the same Riga ice stream that had transported the Daugmales Tomēni material.

Sample 230-086 was collected from an original marine silty clay lens occurring in the till along NE rising shear planes at locality B (Fig. 3). In this section the multicoloured silty till contained sheared-in lenses of the clay with *Portlandia arctica*, and till itself also contained the shells. Eight aliquots of shell material 120 mg each were prepared for ESR dating.

Many *Portlandia* shells (sample 231-086) were taken from the grey sandy till at the same level as sample 230-086, but about one metre east. Nine aliquots 145 mg each were available for analysis.

A *Portlandia arctica* shell sample 205-L1 was collected by Krūkle in 1970 without surrounding matrix from her Ličupe section from the original blocks of black marine clay enclosed in till. To estimate the age of these shells a similar sample (No. 134-086) was taken in 1995 from dark grey to black silty clay layer IV at site A of Fig. 3. For ESR analysis the *Portlandia* shell sample was divided into 8 aliquots of 72 mg each.

## ESR DATING

ESR-datings of all shell samples were made by the first author at Institute of Geology (Tallinn). ESR measurements were carried out at room temperature using X-band ESR spectrometer (ERS-221) with a 100 kHz field modulation.

## Principle

The ESR dating method is based on a direct measurement of the amount of radiation-induced paramagnetic centres (radiation damages), that have been created in shell material due to natural radiation. At the time of formation the lattice of shell biogenic carbonate has no radiation-induced centres, but radiation from the shell itself and the environment (embedding matrix and cosmic) causes their gradual accumulation with time. A shell sample will therefore have paramagnetic centres the amount of which relates directly to the total radiation dose that the shell has received. The presence of paramagnetic carbonate centres in mollusc shell material can be detected by ESR spectrometry. It produces a differential plot of the microwave absorption spectra where each paramagnetic centre is characterized by a particular  $g$ -value (Fig. 7). Normally five typical peaks are observed in irradiated aragonite shell samples:  $g_1 = 2.0060$ ,  $g_2 = 2.0034$ ,  $g_3 = 2.0022$ ,  $g_4 = 2.0010$  and  $g_5 = 1.9983$ . Under a specific measurement condition (see ESR analysis section below) a strong  $g = 2.0012$  (line-width  $\Delta B_{pp} \approx 0.22$  mT) signal is detected in exoskeletal material of various mollusc species (Molodkov, 1988, 1993), that is most suitable for concentration analysis and dating.

The palaeodose accumulated by the fossil shells is evaluated by comparing the magnitude of the  $g = 2.0012$  signal with the increase induced by laboratory irradiation. If the radiation dose rate is known, then the age of the shell fossils can be derived from the following equation (Molodkov, 1988, 1989a):

$$T = \tau \left[ -\ln \left( 1 - \frac{P_s}{\tau \dot{D}_z(t)} \right) \right]$$

where  $\tau$  is the mean lifetime of the 2.0012 centre in shell carbonate,  $P_s$  is the accumulated palaeodose since the mollusc exoskeleton formation,  $\dot{D}_z(t)$  is the total radiation dose rate as a function of time,  $T$  is the shell age.

The accumulated palaeodose is determined by successive irradiation of the sample aliquots with the calibrated gamma-ray source. It increases the signals in the ESR spectra of the shell sample. The palaeodose is then determined by extrapolation to zero ESR intensity.

The dose rate,  $\dot{D}_z(t)$ , is a sum of the doses due to different radiations

$$\dot{D}_z(t) = \dot{D}_c + W_\gamma \dot{D}_{\text{ext } \gamma} + W_\beta k_\beta \dot{D}_{\text{ext } \beta} + \dot{D}_{\text{int } \alpha, \beta}(t),$$

where  $\dot{D}_c$  is the cosmic dose rate dependent on the sample's latitude, altitude, and burial depth;  $\dot{D}_{\text{ext } \gamma, \beta}$  is the external dose rate depending on radioactive element concentration in the sediment surrounding the shells;  $W_\gamma$  and  $W_\beta$  are the correction factors for water;  $k_\beta$  is the beta-attenuation correction factor;  $\dot{D}_{\text{int } \alpha, \beta}(t)$  is the time dependent component of internal dose rate originating from the uranium incorporated in the shell substance. It may be taken into account according to the method described in Molodkov (1986) or

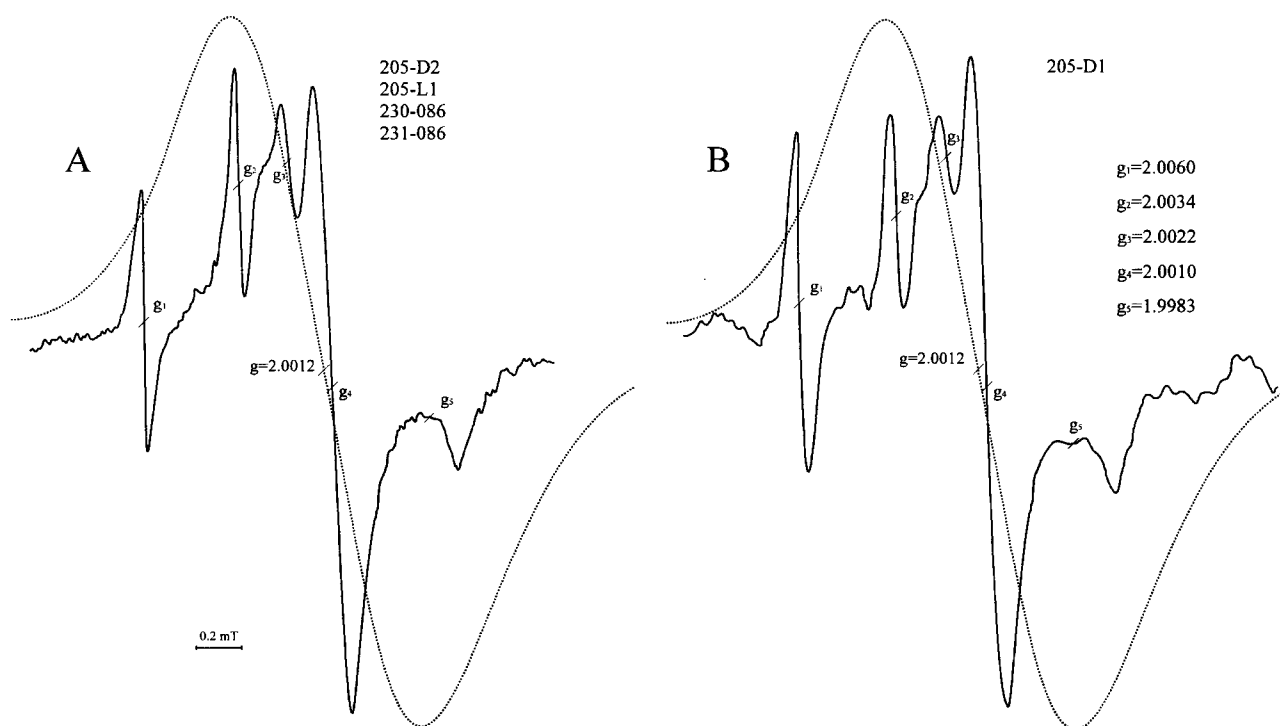


FIG. 7. Typical ESR spectra of the *Portlandia arctica* shell samples (A, solid) from gravel at Daugmales Tomēni (No. 205-D2) and from the till (231-086) and original marine sediments at Līcupe (Nos. 205-L1, 230-086). The spectrum is slightly different from that of the shells from the 2-nd till at Daugmales Tomēni (No. 205-D1) (B, solid). The analytical signal at  $g = 2.0012$  (A, B, dotted) are detected in all shell spectra with microwave power of 2 mW and modulation amplitude of 1 mT [overmodulation (OM) detection method, Molodkov, 1988, 1993]. The signal intensities are normalised in respect to conventional ESR spectra of the *Portlandia* shells.

calculated using following equation (modified from Ikeya, 1982, 1985; see Grün *et al.*, 1987):

$$\begin{aligned} \dot{D}_{\text{int } \alpha, \beta}(t) = & \dot{D}_{238} + \dot{D}_{234}(r_0 - 1)\exp(-\lambda_{234}t) \\ & - \dot{D}_{230}\{\exp(-\lambda_{230}t) - (r_0 - 1) \\ & \times [\exp(-\lambda_{234}t) - \exp(-\lambda_{230}t)] \\ & \times \lambda_{230}/(\lambda_{230} - \lambda_{234})\} + \dot{D}_{235} \\ & - \dot{D}_{231}\exp(-\lambda_{231}t), \end{aligned}$$

where  $r_0$  is the initial activity ratio of  $^{234}\text{U}/^{238}\text{U}$ ;  $\dot{D}_{238}$ ,  $\dot{D}_{235}$ ,  $\dot{D}_{234}$ ,  $\dot{D}_{231}$  and  $\dot{D}_{230}$  are the effective dose rates per concentration unit of uranium (see e.g. Nambi and Aitken, 1986) from  $^{238}\text{U}$  to  $^{206}\text{Pb}$ ,  $^{235}\text{U}$  to  $^{207}\text{Pb}$ ,  $^{234}\text{U}$  to  $^{230}\text{Th}$ ,  $^{231}\text{Pa}$  to  $^{207}\text{Pb}$ , and  $^{230}\text{Th}$  to  $^{206}\text{Pb}$ , respectively;  $\lambda_i$  are the decay constants of respective isotopes.

The total dose rate is derived by radiometric analysis of the naturally occurring radionuclides in the shells and the embedding matrix. An overview of the dating algorithm is shown in Fig. 8.

The basic assumption for ESR dating of the shell samples is that the shell substance did not contain radiation-induced paramagnetic centres at the time of crystallisation of the mollusc's exoskeleton and prior to excavation they were in the same environment.

A fundamental problem in ESR dating of the shells from the glacial sediments is the uncertainty of palaeodoses accumulated since redeposition of the

shells from underlying formations and in original sediment itself. Dose rates in these environments are most likely also different. In such a case the ESR analysis of the shells incorporated in till may not give us reliable age determination, because prior to redeposition shells were in another depositional environment unknown to us. The dating accuracy will depend on the duration of burial state in each of the environments. The longer the shells were situated in the known depositional environment, the closer the obtained dating results are to the true age.

On the other hand, if the shell material is proven to be incorporated in tills with the original surroundings, the age determination of those shells can be reliable. As to the age of the embedding tills, they should be considered as at least younger than the reworked original sediments.

### Sample preparation

The shells for the ESR measurement were washed in water; the remnant clay minerals were removed by ultrasonic bath; the shells were then etched by 10% acetic acid to remove  $\alpha$ -irradiated surface, washed repeatedly in water and dried at room temperature. The dried shell samples were gently ground by pestle in an agate mortar, sieved in order to separate the fraction of 75–400  $\mu\text{m}$ , washed four times with water to remove adhered < 75  $\mu\text{m}$  particles, then allowed to dry at room temperature again.

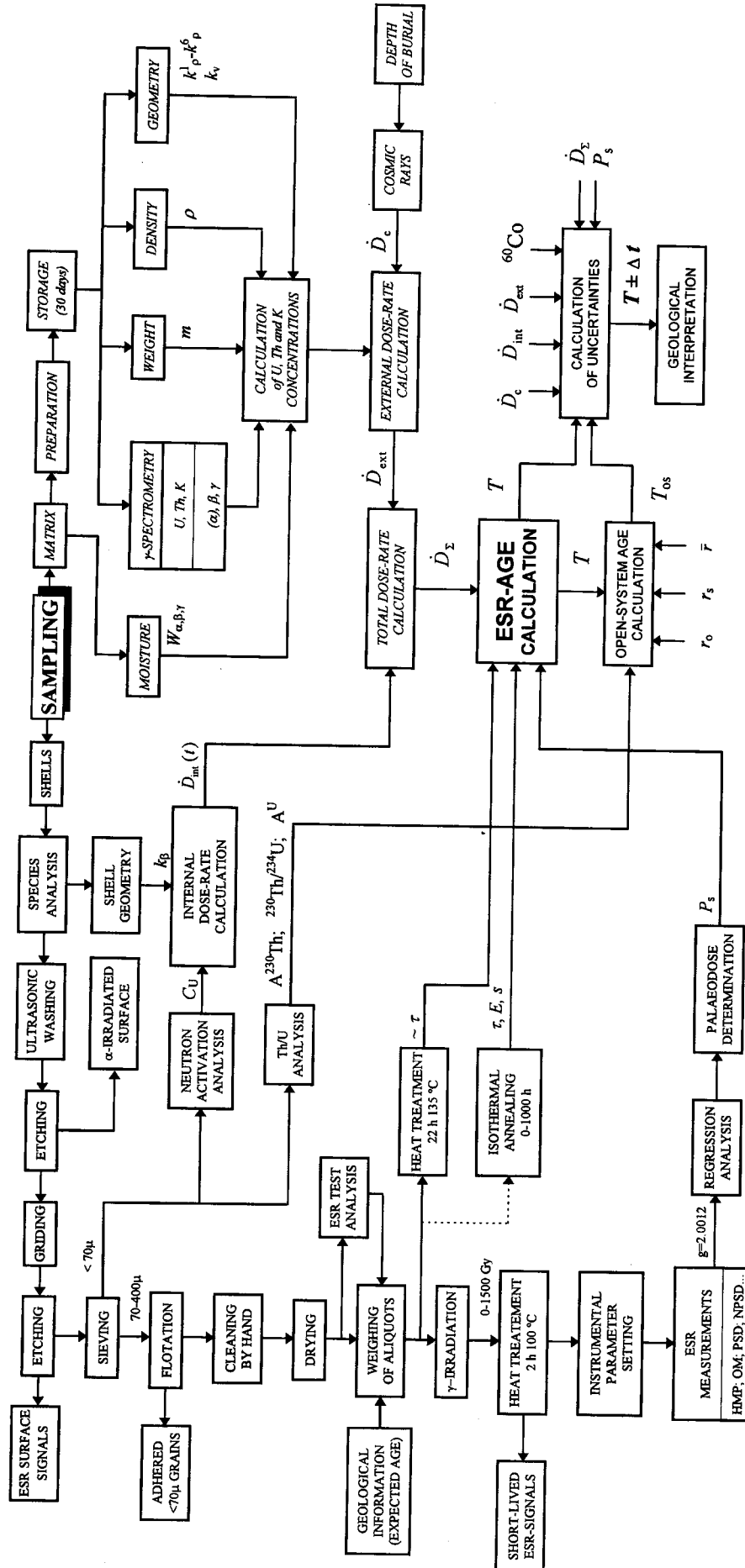


FIG. 8. A chart illustrating the most important steps in the ESR dating procedure of fossil shells.

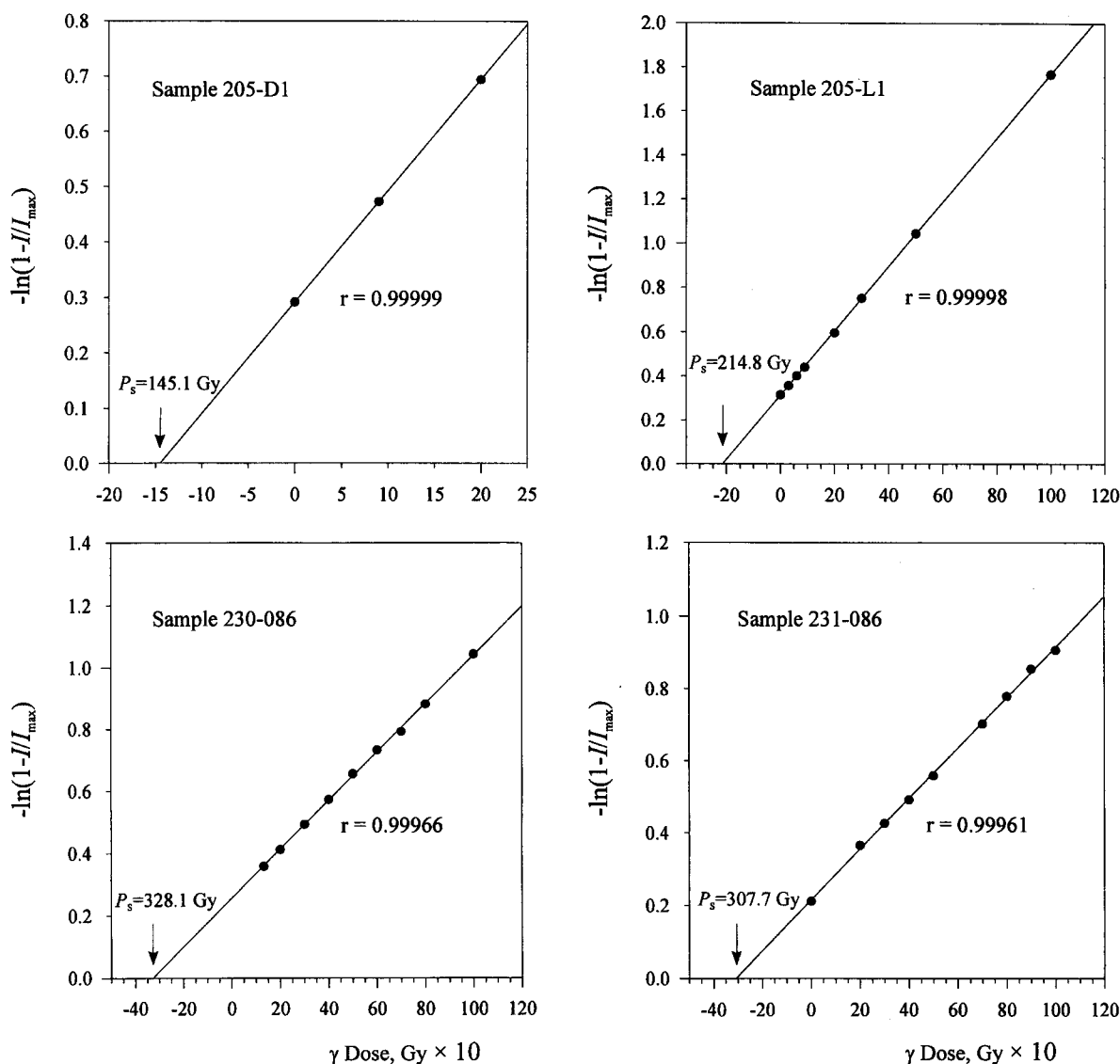


FIG. 9. Mathematical evaluation of the accumulated palaeodose ( $P_s$ ) by logarithmic fitting of the data points obtained by overmodulation (OM) detection method (Molodkov, 1988, 1993) for the 2.0012 carbonate centres in Daugmales Tomēni and Ličupe shell samples. Extrapolation through the added and natural doses to zero ESR intensity defines the palaeodose for the shell samples;  $I$  is the ESR intensity;  $I_{\max}$  is the ESR intensity at saturation dose;  $r$  is the correlation coefficient.

To determine accumulated palaeodose, prepared shell samples were divided into aliquots. One of them was left in the natural state while others were irradiated up to 1000 Gy using a calibrated  $^{60}\text{Co}$  source delivering  $4 \times 10^{-2} \text{ Gy s}^{-1}$ . After irradiation, all shell aliquots were annealed for 2 h at  $100^\circ\text{C}$  to allow fading of any possible short-lived ESR signals induced by laboratory radiation. For reconstruction of the growth curve outside the additive dose section, logarithmic transformation was made of dose - vs - ESR intensity as illustrated in Fig. 9.

#### ESR analysis

Typical ESR spectra of *Portlandia arctica* shells from Daugmales Tomēni and Ličupe sites recorded with a microwave power of 2 mW and a modulation amplitude of  $0.25 \times 10^{-4} \text{ T}$  are shown in Fig. 7. To perform

palaeodosimetric analysis of the *Portlandia* shells the analytical line at 2.0012,  $\Delta B_{pp} \approx 0.22 \text{ mT}$  (Molodkov, 1988) was separated. The dose-response curves with the use of this signal conformed most closely to the single exponential function (Molodkov 1988, 1989)

$$I = I_{\max}(1 - \exp(-\mu(\dot{D}_\gamma - P_s))),$$

where  $I$  is the ESR signal intensity;  $I_{\max}$  is the ESR intensity at saturation dose;  $\mu$  is the sensitivity to radiation;  $\dot{D}_\gamma$  is the radiation dose;  $P_s$  is the accumulated palaeodose.

As an equivalent of the  $g = 2.0012$  centre concentration in the shell material the height of original absorption signal was used. This is possible at conventionally used low microwave power as the amplitude of the dominant 2.0012 signal has practically no contributions from the wings of adjacent peaks at  $g_3 = 2.0022$  and  $g_5 = 1.9983$  (Molodkov, 1993). Quantification of

TABLE 2. ESR results and radioactivity data for shell samples from Daugmales Tomēni and Ličupe site

No.	Lab no.	Field no.	Site	d (mm)	$U_{in}$ (ppm)	U (ppm)	Th (ppm)	K (%)	$\dot{D}_c$ ( $\mu\text{Gy/a}$ )	$\dot{D}_{int}$ ( $\mu\text{Gy/a}$ )	$\dot{D}_{sed}$ ( $\mu\text{Gy/a}$ )	$\dot{D}_T$ ( $\mu\text{Gy/a}$ )	r	$P_s$ (Gy)	ESR-age (ka)
1	205-D1	Sample 1	Daugmales Tomēni (2nd till)	2.00	0.56	1.48	5.96	2.08	168.0	151.0	1436.0	1755.0	0.99999	145.1 ± 7.3	86.0 ± 6.8
2	205-D2	Sample 2	Daugmales Tomēni (gravel)	2.00	1.90	1.81	7.57	2.03	82.5	551.1	1560.3	2193.8	0.99646	219.2 ± 16.2	105.0 ± 9.2
3	101-111	Sample 3	Daugmales Tomēni (3rd till)	—	—	1.76	7.16	1.98	—	—	—	—	—	—	—
4	205-L1	Sample L1	Ličupe (clay lens)	2.00	0.58	2.40	10.82	2.97	87.6	159.0	2232.7	2479.3	0.99998	214.8 ± 11.0	88.5 ± 7.3
5	230-086	Sample 1	Ličupe (clay lens)	1.18	0.92	2.63	13.64	3.49	87.6	251.5	3096.3	3435.4	0.99966	328.1 ± 18.3	97.8 ± 8.2
6	231-086	Sample 2	Ličupe (till)	1.10	1.30	2.91	11.32	3.09	87.6	350.3	2852.7	3290.5	0.99961	307.7 ± 18.5	95.7 ± 8.2
7	134-086	Sample 3	Ličupe (clay lens)	—	—	2.40	10.82	2.97	—	—	—	—	—	—	—

Notes: d is the shell thickness;  $U_{in}$  is the uranium content in shells; U, Th, K are the uranium, thorium and potassium content in sediments;  $\dot{D}_c$  is the cosmic dose rate;  $\dot{D}_{int}$  is the time-averaged internal dose rate;  $\dot{D}_{sed}$  is the sediment dose rate;  $\dot{D}_T$  is the total dose rate;  $P_s$  is the palaeodose; Nos. 101-111 and 134-086 are the sediment samples. Uncertainties: determination of thickness, ± 40 µm; U determination, ± 2-3%; Th determination, ± 3-4%; K determination, ± 1-2%; U determination in the shells, ± 1-3%; gamma irradiation, ± 3-5%.

average of only 25% clay in the Daugmale tills (Dreimanis, 1943). The average Ličupe clay and till radionuclide values are  $2.65 \pm 0.21$  ppm U,  $11.93 \pm 1.23$  ppm Th and  $3.18 \pm 0.22$  ppm K.

At Daugmale, all three samples are either glacial (till) or glaciofluvial (gravel) deposits, with no visible admixture of marine clay. Their U, Th and K content is considerably lower than at Ličupe. Because of the above differences the highest external sediment dose rate,  $\dot{D}_{sed}$ , is in the clayey Ličupe sediments, and the lowest in the Daugmale till.

All three sets of Ličupe *Portlandia arctica* shells have similar ESR ages, ranging from  $88.5 \pm 7.3$  to  $97.8 \pm 8.2$  ka. This is not surprising, because they were taken from similar enclosing materials: from marine clay lenses in till layer III (Fig. 3) and from this till consisting mainly of incorporated marine clay.

All shells from Ličupe and Daugmale Tomēni gravel have identical ESR spectra (see Fig. 7) that may be indicative of the same environments during shell-growth and subsequent burial before shells had been transported and redeposited during glacial advances. It allows us to assume that not only the Ličupe *Portlandia arctica* shells, but also the Daugmale shells from gravel had been derived from the same marine clay unit. To calculate the age of the Daugmale shells taken from the gravel layer we took into account the theoretical modelling of the Weichselian ice sheet advances in the Baltic region by Holmlund and Fastook (1995). The first glacial advance into the Gulf of Riga was probably at about 64 ka, eroding the upper part of the Eemian and Lower Weichselian marine clay containing *Portlandia arctica* shells. Their subsequent transport by the Riga lobe and lying in third till may have lasted for about 8 ka. Then ice-marginal meltwaters redeposited the shells into gravel, and they were in the gravel for the remaining 56 ka. Taking into consideration different surroundings during the burial state (sequentially marine clays, third till and gravel) we have determined for the 205-D2 shells from gravel an ESR age of  $105.0 \pm 9.2$  ka BP.

As suggested in the descriptive section of both sampling sites, the marine clay was deposited most probably in the Gulf of Riga and brought to Ličupe as a glacial raft and to Daugmale as a constituent (*Portlandia arctica* shells) of third till by the Riga ice stream during the first Weichselian glacial advance.

The ESR spectrum of the shells from the second till at Daugmales Tomēni (Fig. 7: B) differs from the ESR spectra of previously discussed shells, probably indicating a different palaeoecologic environment during the growth of these *Portlandia arctica* shells. The second till contains more Ordovician limestone than the underlying gravel and the third till: 63% in the pebble and granule grades of the second till, 35% in the third till (Dreimanis, 1936) (p. 128). It is possible that the *Portlandia* shells from the second Daugmale till have been derived from the Baltic Sea basin north of Saaremaa and Hiiumaa islands, where the lithology of the marine sediments was influenced by the limestone

oceanic substage 5c (see e.g. Behre, 1989; Grüger, 1989; Mangerud, 1989).

To test the correlation with the Brørup interstadial, let us compare the Ličupe pollen data of Konshin *et al.* (1969) and Krükle and Arslanov (1977) with those of the Early Weichselian interstadials south of our area. Both Early Weichselian interstadials, Brørup and Odderade, are considered to be relatively warm (Behre, 1989). Their summer temperature gradients towards the north, however, were very steep, and thermophilous tree species did not reach northern central Europe during the Brørup (Behre, 1989).

The nearest pollen diagram published from interstadial deposits correlated with the Brørup and Odderade are from the Jonionys site in southern Lithuania (Kondratienė, 1996: Figs. 58 and 59; for location see our Fig 1(9)). There the Jonionys-1 is correlated with Brørup and Jonionys-2 with Odderade. The pollen spectrum of Jonionys-1 is dominated by *Pinus* (45–80%); next most common pollen are *Betula* and *Alnus* (up to about 20%); *Picea* and *Corylus* also form continuous curves; *Artemisia* dominates among the NAP, and Sphagnales among spores. A similar pollen spectrum is also at Ličupe. Grüger's (1989) summary of dominant Brørup interstadial pollen in northern Germany is also similar to that of Ličupe section. However, according to Gaigalas and Hütt (1995), the Jonionys 1 and 2 interstadials are dated by optically stimulated luminescence (OSL) dating method as 63–59 ka old, thus placing them into the early part of Middle Weichselian, approximately correlative to the Oerel and Glinde interstadials in NW Germany (Behre, 1989). Palynological investigations (*ibid.*) indicated open treeless shrub tundra during these interstadials in NW Germany that has a warmer climate now than in S. Lithuania. Forest trees (*Pinus*, *Betula sect. Albae*, *Picea* a.o.), however, did grow there during the Jonionys 1 and 2 interstadials as determined by carpologic investigations (Kondratienė, 1996) (Fig. 5(8)), and the 63–59 ka age determinations seem to be underestimated if the presently used oxygen-isotope correlated Late Pleistocene chronology is correct.

The nearest marine interstadial deposit south of our area and correlated with Early Vistulian (Early Weichselian) is in the vicinity of Elblag (Elbing) in northern Poland (Fig. 1(8)): the much debated and glaciotectonically deformed Yoldia clay (Drozdowski, 1988) (and references therein). It has been dated by TL as being  $76 \pm 11$  ka old (*ibid.* p. 53), but this age determination may be too low. This is supported by several underestimated TL age determinations of basin sediments, e.g. the 80 ka TL date (Berger and Eyles, 1994) of the Don Formation at Toronto in Canada which palaeoecologically corresponds to 5e. This underestimation is probably due to anomalous TL fading.

The Yoldia clays of Elblag have been palynologically investigated by Gross (Woldstedt, 1949) (Fig. 3). His pollen diagram shows an upward change from the

*Carpinus* dominated pollen zone (g) to the *Picea* dominated zone (i). While most of this pollen diagram suggests the second half of the Eemian interglacial, the upper part of zone (i) may belong to the Weichselian, with a climatic warming-up indicated at the depth of – 0.5 m, containing *Picea* up to 28% and minor peaks of *Alnus* and QM. This climatic warming-up may be correlated with the Brørup interstadial.

*Portlandia arctica* shells also occur in the upper part of the Elblag Yoldia clays, and Zāns (1936) had already suggested that these Yoldia clays were deposited in the Portlandia-Sea of the Baltic and this sea was separated from the Eemian Sea chronologically by an advance of the Scandinavian ice sheet. According to the present day stratigraphic terminology, Portlandia-Sea occupied the Baltic depression during the Brørup interstadial. Zāns (*ibid.*) proposed also that the Baltic Portlandia-Sea was connected with the North and White seas. Houmark-Nielsen (1989: Fig. 5), however, does not show any connection with the North Sea during the Brørup interstadial. In the North Sea, marine interstadial deposits are recognized in northern Denmark near Kattegat (Fig. 1) in the Skærumhede II boring (Bahnsen *et al.*, 1974) (p. 56).

In the regions SE of the Gulf of Finland of wide distribution are *Portlandia arctica* bearing marine deposits known here as Mga marine bed. The most complete Mga deposits, assigned to the time of last interglacial (*sensu lato*), are represented in Rybatskoe section in vicinity of St.-Petersburg (Fig. 1(10)). The 33 m thick interglacial deposits are sandwiched between two tills and aqueoglacial deposits (at a depth of 43–10 m) related to them. The marine deposits consist mainly of grey to dark-grey clays with *in situ* *Portlandia arctica* shells alongside with the boreal species (*Mytilus edulis*). *Portlandia* are found through the entire marine bed, including its interglacial part (Lavrova and Grichuk, 1960), presumably as a relic of the late glacial transgression (Lavrova, 1967).

Zāns (1936) (p. 238) identified diatoms in the Mga marine clay and concluded that they indicate a relatively deep and salty marine environment. Similar diatoms were encountered also at Elblag. As an example, we mention *Coscinodiscus radiatus* Ehs. that is present at Mga, Elblag and also at Ličupe (Table 1), and prefers marine water with more than 1.25‰ salinity according to Munthe (1892). At a depth of about 27 m all thermophilous trees, characteristic of interglacial climatic optimum (*Quercus*, *Ulmus*, *Tilia*, *Carpinus*, *Corylus* a.o.) decrease considerably (Lavrova and Grichuk, 1960). Pollen and spores of tundra species (*Lycopodium pungens*, *Betula nana*) appear. Seashore oscillations are also observed at that time. At a depth of 20 m *Betula nana* decrease from 16 to about 5%, whereas the content of *Picea* and *Pinus* increases to about 13 and 68%, respectively. Although no data of numerical age are available for this sedimentation level in the above area, we are prone to correlate these *Portlandia arctica* bearing marine clays with those at Ličupe.

It is note-worthy that a wide distribution of tundra vegetation with sharply increasing quantity of *Betula nana* (from 10 to more than 70%) is observed above an erosional boundary at a depth of about 10 m. This cooling can probably be correlated with the end of the oxygen isotope stage 5.

Farther north, in the White Sea basin, intertill deposits of the so called Boreal marine transgression have been commonly correlated with the Mikulino = Eemian interglacial (Devyatova, 1982; Velichko and Faustova, 1986; Molodkov and Raukas, 1988). Molodkov and Raukas (*ibid.*) have determined ESR ages of ten mollusc shell samples from the upper part of the Boreal transgression sediments at Zaton (Fig. 1(11)). Three shell samples from the depth of 5.6–6.4 m are dated as  $105 \pm 10$  to  $120 \pm 8$  ka, but seven boreal and arctic species from the overlying 3.8–5.6 m as  $82 \pm 8$  to  $95 \pm 15$  ka. Though Molodkov and Raukas (1988) had correlated all the deposits studied with the 'long' (125–75 ka BP) Eemian interglacial, it is possible that the upper 3.8–5.6 m part of the Zaton section is of Brørup interstadial age (ca. 105 to 93 ka BP) and correlative to the Līčupe marine sediments. The Boreal transgression was linked, through the system of shallow sounds and lakes Onega and Ladoga, with the Baltic Sea basin (Lavrova, 1961).

### CONCLUSIONS

From the data obtained in this study it can be concluded that all tills investigated are unequivocally Weichselian in age, and that the marine clays containing *Portlandia arctica* shells at Līčupe and Daugmales Tomēni were glacially redeposited by the Riga ice stream or lobe during the Weichselian glaciation.

The five sets of *Portlandia arctica* shells collected from till and glacially transported marine clay give ESR ages ranging from  $86.0 \pm 6.8$  to  $105.0 \pm 9.2$  ka BP and averaging about 94 ka BP that most likely correspond to substage 5c of the oxygen isotope record. Considering the age determination and palynological spectra at the Līčupe site, the source deposit of these *Portlandia arctica* shells is probably of Brørup interstadial age of the Early Weichselian, corresponding to the marine isotope substage 5c. Though some pollen of thermophilous deciduous trees were encountered in the Līčupe sections, their erratic appearance suggest that they had been redeposited from interglacial sediments. The most probable source area of marine clays of Līčupe and the *Portlandia arctica* shells of Daugmale is the Gulf of Riga. Whether the sea water entered the Gulf of Riga from the Atlantic Ocean via North Sea or from the Arctic Ocean via the White Sea and Gulf of Finland, is not certain.

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