Progress Report #8 (Final)
Project 2.1
Metabolism and Dosimetry of Plutonium Industrial Compounds

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Executive Summary

This the final report for the long-term collaborative research Project between the Dosimetry Registry of the Mayak Industrial Association (DRMIA), operated by Branch N1 of the SRC Institute of Biophysics, and the U.S. Transuranium and Uranium Registries (USTUR), operated by Washington State University. The project was sponsored by the U. S. Department of Energy Office of International Health Programs. It officially began on 1 February 1997 and was scheduled for completion on 30 March 2000.

The primary objective of the Project was to combine the data accumulated by the both Registries, create a joint database, and perform a mutual analysis of the unique information regarding the metabolism and dosimetry of transuranium nuclides, plutonium and americium, in the human body.

Ten tasks to be performed as part of this project were described in the proposal which was approved for funding and the tasks were scheduled for initiation and completion at various times during the three-year period. The tasks included:

Task A--To compare radiochemical analytical methods for actinides currently in use by both Registries with a series of performance evaluations;

Task B--To establish a common database format that can be used by both Registries for completion of tasks F, G, and H, listed below;

Task C-- To coordinate tissue sampling methods used by the two Registries including specific tissues and organs sampled, mass of the sample, and specific structures to be included in a sample, thus improving and making more exact data comparisons;

Task D--To coordinate radiochemical analytical methods used by both Registries to determine actinide contents of tissue samples, including ashing methods, actinide separation techniques, spectroscopy methods, and data recording;

Task E--To characterize workplace aerosols at the Mayak facility and American facilities for the purpose of more accurately predicting their initial deposition and solubility in the lungs of workers;

Task F--To establish transfer coefficients, based on the systemic:lung:lymph node activity ratios measured by both Registries, that describe the transfer of various plutonium and americium compounds from the lungs to the blood and compare the

coefficients with those predicted by the new ICRP-66 (1994) models for the purpose of testing the model directly with human, long-term exposure data;

Task G--To determine the relationships between actinide concentrations of organs of the body and between individual organs and total body burdens in healthy individuals as well as in those with health impairment, specifically those with liver diseases;

Task H--To test the relationships between actinide contents of the lungs and body organs at autopsy and the long-term, temporal pattern of urinary excretion predicted by the current ICRP metabolic models for plutonium and americium (ICRP-67 1994) and to compare actinide metabolism and long-term urinary excretion of the actinides in healthy individuals with that in health-impaired individuals, specifically in those with liver diseases;

Task I--To enhance the sensitivity of the in vivo counter used by the DRMIA and perform calibrations and intercomparisons with other, similar facilities so that it is a more useful tool for characterizing the intake and retention of actinide elements; and;

Task J--To translate previously classified Russian documents into English for submission to peer-reviewed journals or for publication as topical reports, as appropriate.

All planned tasks were accomplished during the project period 1997-2000. Five tasks involving the intercomparison of radiochemical methods (task A), biokinetic modeling (tasks F, G, H) and the modernization of the whole body counter with new detectors (task I) were planned for the final year of the project (1 April 1999 -30 March 2000) and those tasks are described in this final report in detail.

A primary focus of the final year of the project was to complete a very important task concerning the intercomparison of radiochemical analytical methods and instrumentation in use by both Registries for plutonium and americium measurement in autopsy samples (Task A). During the last step of the intercomparison, it was necessary to study the isotopic composition of standard reference materals (SRM), certified by the U. S. National Institute of Standard and Technology. This step was delayed because of changes in the import regulations of Russia; however, radiochemical analyses of the SRM, human lung and liver, were ultimately performed by the DRMIA laboratory. The results of measurements of plutonium and uranium activities in the SRM were in good agreement with their certified values. The measured activity of ^{241}Am was more than its certified value and it is possible that additional activity of ^{241}Am had accumulated since 1982 as result of ^{241}Pu decay. The results of the laboratory intercomparisons (Task A) led to the conclusion that the data accumulated by both Registries are reliable, correct and can be used in joint investigations of actinide metabolism in humans.

Three other high priority tasks involving biokinetic modeling were completed during the third year of the project (tasks F, G, H). The DRMIA lung model used at Mayak PA for dose assessment is thoroughly described in this report with a system of differential equations corresponding to the compartments of the biokinetic model (Task F). Solutions of those equations were used for regression analyses of plutonium distribution in the bodies of workers included in the joint DRMIA and USTUR database. A comparative analysis of the DRMIA model with those proposed in ICRP–66 showed an advantage of the DRMIA dosimetric lung model at long times (10 and more years) after inhalation of actinide elements. Further development of the DRMIA model to increase its effectiveness for early times after exposure will require analysis of data from individuals after acute inhalation exposures. Quantitative descriptions of the processes of actinide deposition and clearance from respiratory tract, both in early and late phases of metabolism, presented by the ICRP-66 model are primarily based on data from animal studies and may not be completely applicable to humans. The DRMIA model is solely based on the human experience.

Data relating plutonium and americium distributions in the body (Task G) confirmed that disease conditions and causes of death have a significant influence on the distribution of actinides in the systemic organs especially in the liver and skeleton. Disease conditions of the liver resulted in significantly reduced plutonium and americium contents in that organ with a significant increase in contents of the skeleton. It was evident from this study that the systemic americium fractions in the liver were consistently lower than those for plutonium, both in healthy individuals and in individuals with health impairments. Such disease conditions were also shown to a significantly increase the urinary excretion of plutonium from the system when compared to the urinary excretion rates of relatively healthy workers (task H).

A whole body counter, formerly in use at the U.S. Rocky Flats Plant, was installed at FIB-1. During the final year of the project, calibration of the high purity germanium (HPGe) and phoswich detectors was performed and preliminary measurements of Mayak personnel were performed.

One manuscript "Metabolism and dosimetry of actinide elements in occupationally exposed personnel of Russia and the United States: a summary progress report", jointly authored by DRMIA and USTUR investigators, was submitted to a peer – reviewed journal and accepted for publication.

Thus, in the three-year term of this collaborative research project (Project 2.1), DRMIA and USTUR combined data were used in metabolic models to describe the retention, distribution and translocation of plutonium in the human body. The information obtained in this Project was used for modification of the DRMIA combined lung clearance and systemic biokinetics and dosimetry models, developed as part of Project 2.4. The results of this research were in direct support of epidemiologic radiation effects studies conducted in Projects 2.2 and 2. 3. Although much was accomplished as a result of Project 2.1, there is much more information to be gleaned from the combined data of the two Registries that

is of vital importance to the epidemiological studies. For example, there are additional bioassay and autopsy information on Mayak workers who worked between 1958 and 1972. These data were not organized in time for use in Project 2.1; however, they could potentially influence the precision of internal dose assessments for the epidemiologic investigations in addition to providing greater numbers of subjects for those studies. Other factors that could influence internal dose assessments include cigarette smoking by large numbers of workers and the administration of the chelating agent, DTPA, to a number of workers. A proposal for the continuation of Project 2.1 is included in Appendix I of this report.

Final Progress Report

This is the final progress report for Project 2.1, "Metabolism and Dosimetry of Plutonium Industrial Compounds". The previous progress report No. 7 was an oral report to the JCCRER Scientific Review Group made by Dr. R. E. Filipy of the USTUR in Bethesda, MD in September, 1999.

Task A Intercomparison of radiochemical analytical methods currently in use by DRMIA and USTUR for determination of actinides in autopsy samples with a series of performance evaluations.

The main purpose of Project 2.1 was the detailed study of actinide metabolism in the human body that was based on the mutual analysis and interpretation of data of both Registries (the DRMIA and the USTUR). These data were obtained by radiochemical analysis of samples of soft tissues and bones collected at autopsy of occupationally exposed personnel. Both Registries have large volumes of such information. During one-year feasibility study (Phase I), differences between the radiochemical methods used by both Registries were determined and so intercomparison of the radiochemical methods was a high priority task for the first year of long-term project (Phase II).

The main differences in radioanalytical methods included:

1. the use of different instrumentation for measurement of plutonium and americium alpha-activity in samples. The DRMIA used the alpha-radiometer, based on ZnS(Ag) scintillation detection, for routine measurements of alpha-activity . The DRMIA also used the SEAM alpha-spectrometer, based on an ionization chamber detector, for low-level activity samples and samples containing significant activity from ^{238}Pu in addition to $^{239+240}$Pu. The USTUR has used silicon surface barrier detectors for many years.

2. the use of different organic reagents for the analytical procedures of plutonium and americium separation and americium extraction.

The characteristics of the radioanalytical and instrumental methods used by laboratories both of Registries are shown in Table A-1. Table A-1 also contains the characteristics of the new alpha-spectrometer system (EG&G Ortec OCTETE), purchased by DRMIA in 1998, and the radioanalytical methods for actinide separation, modernized according to USTUR experience (use of new reagents such as Bio-Rad anion exchange resins and the extractant DDCP). These are now successfully used by the DRMIA for measurement of actinides in samples.

The interlaboratory comparison was a necessary step for the quality assurance of previously accumulated data and it involved a series of performance evaluations through sample exchanges. The capability of the three instruments and radiochemical separation methods used by the two Registries was evaluated to determine if a systemic bias existed in the previous collection of data.

The interlaboratory comparison included three steps:

1. comparison of instrumental methods and equipment currently in use by both Registries for plutonium and americium measurements. This step included the measurement of 18 samples by two laboratories: the DRMIA received, from the USTUR, 5 samples with plutonium and 5 samples with americium electroplated on steel disks and measured by alpha-spectrometer. The USTUR received, from the DRMIA, 8 samples consisting of a plutonium - $BiPO_4$ precipitate mixed with ZnS(Ag) scintillator and measured by alpha-radiometry with aliquots also measured with the DRMIA SEAM alpha-spectrometer for Pu activity. The alpha activity levels of plutonium in the samples varied from 0 to 20 dpm and from 0 to 50 dpm for americium. These results were reported in a previous progress report (Khokhryakov et al. 1998).

2. comparison of radiochemical separation as well as measurement methods for plutonium and americium was performed in October,1997 - February, 1998 and included the radiochemical analysis of 20 samples. Plutonium and americium with alpha activities from 0 to 50 dpm in dehydrated, acid-dissolved samples were measured by both DRMIA alpha-radiometry and SEAM alpha-spectrometry and by
USTUR Ortec OCTETE alpha-spectrometry.

3. analyses, by the DRMIA, of Standard Reference Materials (SRM) of human lung and human liver prepared by the U.S. National Institute of Standards and Technology (NIST). The SRM are routinely used as a part of quality assurance /qualitycontrol program for radioanalytical procedures. For a long time the completion of this step was delayed because of new Russian regulations governing the import of SRMs to Russia. The permission for import was obtained in June, 1999 and the analyses of the SRMs were performed in January, 2000.

Both the USTUR and the DRMIA provided results of analyses of samples exchanged for comparison of radiochemical extraction methods. Analysis of variance was used to test for significant differences in means for the analyses performed by the two laboratories as well as differences due to measurement by radiometry or spectrometry. A randomized complete block design was assumed which provides a statistical analysis equivalent to a paired t-test when comparing two mean values. Both weighted and unweighted analyses of variance were used because of concern about homogeneity of variances. Results of the unweighted analysis are shown in Table A-2. Both the weighted and unweighted analyses indicate good agreement of the analytical results obtained by the two laboratories. Only one significant difference (americium) was found between the two laboratories and this only for the unweighted analysis. The magnitude of this difference was approximately 5% of the mean. All other differences were not significant ($P>0.05$). From these results, it was concluded that the analytical data of the two Registries were compatible for inclusion in a joint database with mutual analyses toward determination of biokinetics parameters.

The final step in the laboratory intercomparison involved radiochemical analysis of two SRM, human lung and human liver, for isotopic plutonium, americium-241, and isotopic uranium. The USTUR radiochemical analytical laboratory routinely analyses SRM as a quality assurance proceure. The results included in this report are those of the DRMIA laboratory. According to NIST requirements the working samples consisted of the entire aliquots provided in bottles. Each of the samples was weighed immediately after the bottle was opened and the weights are shown in Table A-3.

The main steps of the radiochemical analysis of SRM, according to DRMIA radiochemical analytical procedures, were as follows:

1. approximately 1 L of HNO_3 was added to each of the samples, the samples were heated and boiled, and tracer solutions of ^{242}Pu, ^{243}Am and ^{232}U were added to each of the samples;
2. comprehensive ashing process (wet ashing with $HNO_3 + H_2O_2$, muffle furnace, $HF + HNO_3$ fusion);
3. anion exchange separation of Pu, Am, and U with AG 1-X4 resin (an aliqout of the eluate that contained the total plutonium fraction was taken for $BiPO_4$ co-precipitation and measurement by alpha-radiometer);
4. americium isolation by extraction with DDCP and AG MP-1 resin;
5. measurement of Pu isotope activities by two instrumental methods, alpha-radiometry ($\Sigma Pu = {}^{238}Pu + {}^{239,240}Pu$) and Ortec OCTETE alpha-spectrometry (^{238}Pu, $^{239,240}Pu$);
6. measurement of ^{241}Am activity only by Ortec OCTETE alpha-spectrometry because of the low activity level of ^{241}Am in samples and, therefore, the impossibility of using an aliqout for alpha-radiometry measurement;
7. ^{234}U and ^{238}U isotope activities were measured only by Ortec OCTETE alpha-spectrometry; alpha-radiometry measurements of total U in samples were impossible because U (VI) would not be expected to co-precipitate with $BiPO_4$ according to its chemical properties.

The results of SRM analyses along with the certified activities are shown in Table A-3. It is noted that the best results for the plutonium and uranium isotopes were obtained for the SRM of "human liver". A possible explanation for that was the relatively high activity levels of those isotopes. Measured ^{241}Am activity in this sample differed from the certified value. Possibly, additional activity of the ^{241}Am isotope accumulated since the reference time of 1, June, 1982 from uncertified ^{241}Pu present in the sample. The analytical result for ^{239,240}Pu activity in the SRM of "human lung" differed from the certified value; however, it was within the interval of certified value ± certified uncertainty ($1,10 \cdot 10^{-3}$ Bq/g (+110%; -50% for 95% confidence level)). The measured activity of ^{238}Pu in this sample was less than the MDA of the radiochemical analytical methods of the DRMIA, determined as 1 mBq. The DRMIA didn't measure thorium activity in the samples because the radioanalytical procedure for separation of thorium isotopes is still under development. In the near future, the DRMIA plans to complete that development and to certify the radiochemical procedure manual for simultaneous determination of plutonium, americium, uranium, and thorium isotopic activities in autopsy samples.

With the completion of these measurements, Task A was considered to be satisfactorily completed despite the difficulties encountered. The results of the final step indicated that data collected by the two Registries in the future would be compatible for joint analyses.

Task B Establishment of common database formats that will be available to both Registries for studies of actinide metabolism in personnel

The main purpose of this task was to determine the most useful structure of a common database and than to enter data accumulated by both Registries. the DRMIA and the USTUR, for performance of joint analyses of the unique information about plutonium and amercium metabolism and dosimetry in workers. The general database structure was established during the first year of the long-term project and descriptions of the files were presented in progress report for 1997 (Khokhryakov et al. 1997). During the following reporting period (October, 1997 -April, 1998), the database structure was modified to include " State of health" information. This was included on the basis of experience of Russian scientists which showed that actinide metabolism in humans was influenced by health status so disease states and causes of death of DRMIA and USTUR cases were determined. Based on data of the pathologic-anatomic diagnosis and causes of death, three groups of liver pathology were identified and this information was entered into a "health group" file for each individual of both Registries.

Data entry was completed during 1998. The information for selected DRMIA and USTUR cases including data for exposure history (documented data about duration of work and time between end of work and death), health information, and tissue and organ concentrations of plutonium and americium with uncertainty values was merged into the combined database. By the end of 1998, the database contained the records of 295 DRMIA cases and 145 USTUR cases. The DRMIA cases were selected from their

database of more than 1200 cases. Cases were selected on the basis of the following criteria:

1. tissue analyses of major organs for both plutonium and americium,
2. tissue content data for more than eight body organs,
3. body burdens greater than 20 nCi, and,
4. individuals worked in either the plutonium production plant or the radiochemical plant at Mayak.

These criteria excluded most individuals who died before 1975, primarily because tissues from the earlier cases were analyzed only for total alpha activity that included all plutonium and americium.

Data sharing agreements were developed and signed and these data of both Registries were used by Russian and American scientists for performance of planned tasks F, G and H to study actinide metabolism in workers of both countries.

Task C. To coordinate tissue sampling methods used by the two Registries including specific tissues and organs sampled, mass of the sample, and specific structures to be included in a sample, thus improving and making more exact data comparisons;

Similarities and differences in tissue sampling protocols between the two Registries were noted during the one-year feasibility project (Suslova et al. 1996). The coordination of tissue sampling methods was accomplished during the first year of this long-term study. The autopsy protocols of each Registry were modified slightly as a result of the initial comparison. The USTUR added the heart and the aortic arch to the list of tissues collected, largely as a result of reports by Filipy et al. (1994) and Filipy and Kathren (1996) who reported relatively high concentrations of americium in those tissues. The DRMIA added the patellae to their list of bone samples to be collected at autopsy. Lynch et al. (1989) demonstrated that a consistent fraction of skeletal actinide content was present in the patellae and noted that the patellae are well defined bones that are easily removed at autopsy. As the result of the modification each Registry will collect, at autopsy, a similar set of organ samples, both soft tissues and bones. The modified protocols will be used as future autopsies are performed.

Task D The coordination of radiochemical analytical methods used by DRMIA and USTUR for analysis for Pu and Am content in biosamples.

This has been a high priority task, especially for DRMIA. The primary objective of this task was the modification and improvement of DRMIA radiochemical analytical methods for determination of low activity levels of actinide isotopes in biosamples.

The methods formerly used by DRMIA for actinide determination (VP anion exchange resin and alpha-radiometry or alpha-spectrometry based on alpha-spectrometer type SEAM) had satisfactory sensitivity for the actinide activity levels accumulated in personnel during the first years of work at "Mayak". However, these methods had insufficient sensitivity for measurements of low-level activities (mBq) in biosamples. Because of improvement of radiation safety and decreased actinide intake and accumulation levels in personnel at "Mayak", it was necessary for theDRMIA to modernize their radiochemical methods for measurement of very low plutonium and americium levels in biosamples.

This task was accomplished in two steps:

1. Modification of radioanalytical methods of plutonium and americium separation by application of more effective reagents (Bio-Rad resins and DDCP extractant).

During 1997-98, the DRMIA laboratory significantly modernized their radiochemical methods by converting to high-efficiency Bio-Rad anion-exchange resins and the americium extractant, DDCP, for chemical separation of the actinides from biosamples. This decreased the mimimum detectable activity (MDA) to 2 mBq for activities measured in samples with the SEAM spectrometer. To check chemical recovery, the DRMIA performed a comparison of the new resins, AG 1-X4, AG MP-1, and the DDCP extractant (measured with SEAM alpha spectrometry) with the old resin, VP, and the HDEHP extractant on samples (measured with alpha radiometry). The results of plutonium and americium measurments in 18 biosamples are shown in Fig. D-1 and D-2.

Statistical analysis indicated no significant differences ($P>0.05$) in results obtained with the old and new ways (alpha-radiometry and alpha-spectrometry). The average ratios between alpha spectrometry (new method) and alpha radiometry (old method) was equal to 1.027 ± 0.115 for total plutonium and 1.017 ± 0.160 for americium. The average means of differences, normalized to radiometry results, were equal to 0.030 ± 0.118 for total plutonium and 0.022 ± 0.166 for americium. T-tests indicated no statistically significant differences for either total plutonium or americium.

2. The acquisition and installation of and EG&G Ortec OCTETE alpha-spectrometry system with a new method of actinide electrodeposition.

To further increase the sensitivity and efficiency of the methods, the DRMIA acquired a 16- chamber alpha spectrometry system (EC&G Ortec OCTETE), which was installed and put into operation in the summer of 1998. Since that time, DRMIA radiochemists have mastered the procedure for plutonium and americium electrodeposition from H_2SO_4 solution 18-mm diameter stainless steel disks which are currently in use by USTUR. Initially, the DRMIA experienced problems with inconsistent tracer recovery and resolution and the problems were suspected to originate during the electrodeposition

process. During his visit to FIB-1 in July 1999, Dr. S. E. Glover reviewed the process used by the DRMIA and
the problem was resolved by the acquisition of a new power supply.

The two steps of the DRMIA laboratory modernization process was completed in the summer of 1999. Based on Russian Federation regulations "About Ensuring the Uniformity of Measurements" and "About Standardization" the DRMIA provided metrology information needed for certification of both the new alpha-spectrometer and the new radiochemical procedure at the end of 1999. The DRMIA received certification from D. I. Mendeleyev State Institute for Metrology (St. Peterburg) which confirmed the characteristics of the equipment and the radiochemical method shown in Table A-1. These new radiochemical methods have satisfactory sensitivity (approximately 1mBq per sample with uncertainty no more than 60%) (P=0,95) and they are currently in use by the DRMIA for determination of low activity levels of plutonium and americium in biosamples.

Task E. Analysis of physico-chemical properties of workplace aerosols (such as particle size distribution and in-vitro solubility) at the Mayak facility and American facilities for the purpose of more accurately predicting plutonium behavior in the lungs of workers.

The original objective of this task was to investigate the physico-chemical properties of workplace aerosols with respect to particle size distribution and solubility in body fluids. According to ICRP models, knowledge of these properties would be very important to the characterization of aerosol clearance from the lung and dissolution to the systemic circulation.

Investigations of plutonium aerosol solubility have been performed at FIB-1 since the middle of the 1970s. It was shown that plutonium distribution among respiratory tract and systemic organs correlated with the transportability coefficient of aerosol inhaled. Transportability, "S", is the fraction of alpha-activity, expressed as a percent, of the total activity on the air sampling filter that passes through a semi-permeable membrane during two days of dialysis. The dialysis method for measuring the transportability of workplace aerosols and the calculations performed to determine "S" were reported by Khokhryakov et. al. (1998). The dialysis method provides a simple, cost-effective tool by means of which dissolution rates for particles that determine the transfer processes in the lungs can be estimated ss well as by other similar *in vitro* dissolution techniques used by other authors (Eidson and Mewhinney 1983, Mewhinney et. al. 1987, Vashi et. al. 1980, Miglio et. al. 1970, Kanapilly et. al. 1983, Ansoborlo et. al. 1998). Data from many years of tissue analysis, given in the Table E-1, show that the actinide content in the lungs relative to that in the total body is inversely related to transportability.

Table E-1 contains the average geometric mean percents of plutonium content in the body found in the respiratory tract, obtained by grouping the post-mortem data on the basis of the transportability coefficients of aerosols from three different workplaces. The relative nuclide content in the lungs is inversely related to transportability. Therefore, the transportability coefficient can be considered to be a quantitative criterion for industrial aerosol classification especially when personnel were exposed to unknown mixtures of various compounds and classification according to ICRP models is not effective.

Evaluation of air samples with the dialysis method began in 1974 and have continued until the present time. During this period about 300 air samples from workplaces of the uranium reprocessing plant and the plutonium production plant were analyzed. Aerosol transportability varied from sample to sample about a mean value that was typical for a specific workplace. There was a tendency for the transportability coefficient to decrease from the initial stage of the process (dissolution of nuclear fuel elements in nitric acid) to the final stage (work with metallic plutonium) at Mayak PA. The data obtained correspond to the chemical solubility of plutonium compounds from different workplaces.

Thus, knowing the work history of an individual makes it possible to predict the probable properties of alpha active aerosols inhaled by the individual during the time of work with radionuclides, including the first years of Mayak operations. Information about the physico-chemical properties of aerosols in workplaces of USTUR workers is generally not available; however, it has been assumed that USTUR registrants were exposed to mixtures of relatively insoluble aerosols. Data on the relative concentrations of plutonium in the respiratory tract and systemic organs substantiate that assumption (Khokhryakov et al. 1999).

Aerosol transportability studies conducted in 1996 and 1999 in comparison with the old studies conducted in the 1970's show a tendency of the transportability coefficient to increase with time in a given workplace (Table E-2). It is assumed that the reason for the increase was modification of radiation protection technology and improvement in containment, which resulted in decreasing particle sizes of the aerosols that escaped into breathing zones.

As noted in Task F below, lung burdens were strongly dependent on the physico-chemical properties of inhaled actinide compound. Thus, knowledge of aerosol transportability and particle size information is necessary to predict the probable properties and the probable behavior of alpha-active aerosols in the lungs and bodies of the personnel exposed by inhalation.

Task F. Analysis of the dynamics of respiratory tract : systemic concentration ratios from data of the both Registries for the purpose of establishing the lung clearance coefficients for plutonium compounds to the systemic circulation.

The comparison of the biokinetic models – FIB-1, ICRP-30, ICRP-66.

DRMIA (FIB) biokinetic model development, including the schematic diagram with parameters for the description of plutonium behavior in the human body after inhalation of airborne workplace plutonium compound was described in the sixth Progress Report (Khokhryakov et al. 1999). This report includes the system of differential equations that correspond to the compartments of the biokinetic model. The solutions of these equations were obtained using regression analyses of post-mortem plutonium distribution data from Mayak workers. The DRMIA model was used for estimation of body and organ burdens of plutonium, based on the urinary excretion rate, and, ultimately, for the internal dose assessment procedure at Mayak PA. These estimates were also used in Projects 2.2 and 2.3 to calculate risks of plutonium-induced stochastic and deterministic effects. The compartmental DRMIA model differs substantially from ICRP models; therefore, it was necessary to validate its effectiveness if used instead of conventional ICRP models. The validity of a model can be tested by comparing the results of model calculations with actual observations. Data on radionuclide distribution in human body from integrated DRMIA and USTUR database was used for this purpose.

Schematic diagrams of the DRMIA biokinetic model and models defined on the basis of ICRP-30 and ICRP-66 publications are given in Fig. F-1 through F-3. Along with blocks for respiratory tract compartments, they include blocks for systemic plutonium compartments, collectively delineated with a dotted line. Comparative analysis for the models was conducted under the assumption that, in all cases, urine and feces excretion of systemic plutonium was described by Durbin's 10-compartment model (Durbin 1972), modified on the basis of observations relative to long-term transport of plutonium to blood (Khokhryakov et al. 1994). It was also assumed that, t days after a single transfer of Qo Bq of radionuclide to blood, the following equations take place:

$$U_m(t) = Qo\sum_{i=1}^{i=5} a_i \exp(-x_i t) \quad U_f(t) = Qo\sum_{i=6}^{i=10} a_i \exp(-x_i t) \qquad (1)$$

$$Q(t) = Qo\sum (a_i/x_i)\exp(-x_i t) \; i=1,2....10 \qquad (2)$$

$$\sum_{i=1}^{i=10} (ai/xi) = 1 \qquad (3)$$

where Um and U_f = plutonium content in urine and feces, respectively, in a 24 hour period.

Numerical values for the ai and xi parameters, taken from Khokhryakov et al. (1994) satisfy equation (3) and are shown in Table F-1.

Comparison of the analytical results from tissues collected at autopsy of Mayak PA personnel with data of the Mayak Radiation Safety Office showed that rate of plutonium inhalation by Mayak PA personnel steadily decreased since start-up of plants. This reduction was a result of increased efforts to limit radiation exposure. The intake reduction can be described by the exponential equation,

$$V = Vo \exp(-\gamma t), \tag{4}$$

where Vo was the early rate of intake and V represents intake at time, t. It was noted that the constant, γ, was different for the various production sites.

The following symbols for nuclide content in the respective compartments of the DRMIA model (schematic diagram in Fig. F-1) are defined:

$V(t)$ – amount of plutonium inhaled at time, t
Δ - fraction of the inhaled material that is rapidly transferred from the lungs to the system
β - fraction of the intake that goes to compartment, β
K_f – plutonium content in a fixed compartment of the lungs
Q_β-Pu content in compartment β, to be slowly cleared from the lungs
Q_n-content in tracheobronchial (pulmonary) lymph nodes
Q_i-content in i compartment of the system
λ - constant of slow nuclide clearance from compartment β
K_n – nuclide fraction transported from compartment β to lymph nodes

During the period of inhalation intake, which exponentially decreases with time, transport processes from respiratory tract, and excretion from the body can be described in the model by a system of 13 equations as follows:

$$dQ_\beta dt = \beta V \exp(-\gamma t) - \lambda Q_\beta \tag{5}$$

$$dQ_f/dt = K_f V \exp(-\gamma t) \tag{6}$$

$$dQ_n/dt = K_n \lambda Q_n \tag{7}$$

$$dQ_i/dt = (a_i/x_i)\,[(1 - K_n)\lambda Q_\beta + \Delta V \exp(-\gamma t)] - x_i Q_i \quad (i = 1,2,\ldots 10) \tag{8}$$

with the condition that :

$$K_f + \Delta + \beta = 1 \tag{9}$$

After exposure to plutonium is discontinued, metabolism is described by the same system of equations with V=0. Solving the above system of equations at zero intake conditions results in an expression for relative content in lungs and tracheobronchial (pulmonary) lymph nodes. This system of equations was solved using human tissue data of relative body content of plutonium in the respiratory tract from the joint DRMIA-USTUR database by means of regression analyses resulting in numerical values of the lung model parameters that were dependent on transportability (Table F-2). As stated above, the higher the transportability of inhaled aerosol, the faster the lung clearance of plutonium. Aerosol transportability for Mayak workplaces was determined by a method of dialysis of aerosol samples collected at workplaces of the personnel (Task E). Fig. F-4 contains frequency distributions of the ratios of the lung:body plutonium content predicted by the DRMIA model to the measured post-mortem lung:body content for three groups of workers exposed to workplace aerosols of various transportabilities (249 cases from the DRMIA) and for a group that worked with plutonium-containing aerosols of unknown transportability (49 cases from the USTUR). Fig. F-4 shows that the frequency distributions for the modeled:observed ratios of DRMIA cases have a maximum frequency in the area where ratios of predicted to observed relative lung content of plutonium is close to 1. The frequency distributions were lognormal with geometric standard deviations ranging as $2.2 > \sigma_g > 2.46$. A frequency distribution of the predicted: observed relative lung contents for cases of the USTUR also has the maximum frequency at 1; however it has a greater geometric standard deviation ($\sigma_g = 3.28$). This difference can be explained by a greater heterogeneity in the USTUR group that contained individuals exposed to plutonium aerosols of various transportability. Thus, aerosol transportability is an indicator that predicts plutonium behavior in respiratory tract.

Classification of plutonium aerosols by transportability based on dialysis method provides an approach for comparison of the DRMIA model with ICRP-30 and ICRP-66 models. It is of interest to determine which model, allowing for individual intake histories, more accurately represents the post-mortem data on plutonium distribution that is available in both registries. Because of differences in transport mechanisms used by the different models, comparison appears to be possible only by application to the composite lung without consideration of separate depositions in tracheo-bronchial and alveolar compartments.

To test the adequacy of the ICRP-30 model for describing the metabolism of inhaled plutonium, a system of 23 differential equations corresponding to the schematic diagram of Fig. F-2 was established similar to that used in the DRMIA model. Solution of the system of equations under conditions of exponentially decreasing intake rates with time of exposure permitted calculation of relative plutonium contents in respiratory tract for the cases in the joint DRMIA-USTUR database. According to the ICRP-30 publication, plutonium aerosols are subdivided into two classes: class Y, with annual retention (including dioxides and hydroxides) and class W, with weekly retention (including the remaining compounds except for chelated compounds). To compare the model-estimated lung content with the observed post-mortem content, 49 cases were used that included workers who were exposed to low-transportable aerosols ($S \cong 0.3\%$); these were

conditionally referred to as class Y. The remaining 200 cases from the DRMIA, exposed to higher transportability aerosols, (S≅1-3 %) were referred to as class W. USTUR cases (43), who were exposed to aerosols of unknown transportability, were referred to as class W as well.

Table F-3 contains the results of the comparison between model-predicted lung:body plutonium contents (ICRP-30) and observed lung:body contents. The comparisons indicate that actual rate of lung clearance is considerably lower than that proposed by ICRP-30. Even for class Y compounds, characterized by maximum half-life within the model, the geometric mean of ratios are in tenths of a percent. An even greater difference between predicted and observed values results for class W compounds. Thus, the comparison showed that ICRP-30 model inadequately describes retention in the respiratory tract at long times after inhalation exposures and that may result in underestimation of dose from protracted lung exposures.

To quantitatively compare observed data on post-mortem distribution with modeled estimates from ICRP-66, a system of 40 differential equations (14 describing resorption, 14 describing mechanical nuclide clearance of respiratory tract, and 12 describing accumulation and excretion of systemic plutonium) was established corresponding to the schematic diagram (Fig. F-3). Using the values for metabolic parameters recommended by ICRP-66, the system of equations was solved for two aerosol classes (S, M). These calculations were performed allowing for a non-uniform inhalation rate. The computer program, written to solve the set of equations, was, in fact, a modification of the well-known LUDEP program used within the framework of the ICRP-66 model for dose assessments for two modes of intake, acute inhalation and chronic uniform inhalation. On the basis of the solutions of the differential equations, allowing for the exponentially decreasing intake with time, the nuclide content of the respiratory tract relative to the total body burden for each individual of joint USTUR/DRMIA database was calculated. Then, as done with the ICRP-30 model, results were compared with post-mortem data.

Table F-3 contains comparisons of the ICRP-66 model-predicted lung:body content and the observed lung:body content and frequency distributions of those ratios are shown in Fig. F-5. For workers who were exposed to slightly soluble plutonium aerosols (S=0.3%), the ICRP-66 model generally results in close agreement with observed values with a geometric standard deviation equal to σ_g=2.54 for compounds of class S (slowly dissolving). Mean ratios for cases of the USTUR database, assuming that they were also exposed to class S aerosols result in model-predicted lung:body ratios that are 2.95 times higher than the observed ratios (Table F-3) and that reflects the heterogeneity of the group in terms of exposue to aerosols of various transportabilities. However, if ICRP-66 class M (moderately soluble) aerosol parameters are used, the model predicts virtually no lung content relative to body burden, substantially different from observed post-mortem data (Table F-3). Thus, the ICRP-66 model quite properly describes post-mortem data on plutonium distribution between the respiratory tract and the system in the case of plutonium dioxide, but apparently requires some revision of parameters for compounds of class M.

This comparative analysis indicates the advantage of the DRMIA dosimetric lung model that is based on the dialysis method, an objective means of classifying alpha-active aerosols to predict the behavior of a wide spectrum of inhaled compounds in the respiratory tract. At the same time, it should be noted that predictions by this model are valid only for long times (10 or more years after initiation of exposure to the actinides). It is also worth noting that the DRMIA model doesn't allow for mechanical clearance (nuclide removal by mucociliary clearance with excretion through the gastrointestinal tract) reducing systemic deposition. The impact of this clearance mechanism is substantial during early times after inhalation and it is important for dose evaluation in those early times. Because mucociliary clearance is not considered in the DRMIA model, it can be assumed that, for long times after a single or chronic intake, the DRMIA model might lead to underestimation of the accumulated lung dose. A more detailed and physiologically correct description of the processes of nuclide deposition and clearance from the respiratory tract, in both early and late phases, is presented in the ICRP-66 model. However, quantitative description of the early metabolism phase in this model is substantially based on data from animal studies and, therefore, requires further development and verification by analysis of the respiratory tract dynamics in individuals subjected to a single inhalation exposure.

Task G. Determination of the distribution of plutonium and americium between systemic organs in healthy individuals as well as in those with health impairment, specifically those with liver diseases.

An objective of this task was the improvement of biokinetic models for workers and it included determination of retention fractions of plutonium and americium in the systemic in the organs of major deposition, the liver and the skeleton.

The ICRP models describing retention, translocation and excretion of systemic plutonium were largely based on extrapolation of data from animal experiments and on limited data from a few documented cases of accidentally-exposed humans. These data demonstrate a great individual variablity in the partitioning of the plutonium between the skeleton and the liver depending on exposure time, the amount present, the physico-chemical form of the radionuclide injected, and the age of the human or animal. Previosly published data of DRMIA scientists indicated that the health state is another important factor affecting plutonium metabolism in workers. Impairment of health, particularly serious chronic diseases or malignant tumors can cause a consistent increase in urinary excretion of plutonium and americium [Khokryakov et al. 1994; Suslova et al. 1994].

The purpose of Task G was to compare plutonium and americium systemic fractions in the skeleton and the liver and to use those comparisons as a basis for reconstructing the models used by the DRMIA and those proposed by the ICRP [1979,1986,1993]. Another aspect of Task G was the investigation of the effect of disease on the systemic distribution of actinides in the transfer compartment (blood).

This task was conducted during 1998-1999 and the results were presented in a previous progress report (Khokhryakov et al. 1999). Investigations were directed toward studies of the partitioning of actinide contents between the liver and the skeleton two organs of major concern for health effects. Primary attention was given to the study of systemic distribution of plutonium and americium in relatively healthy workers who died suddenly and those who died from serious diseases of the liver. The liver is known to react quickly to many pathological states of the body. As indicated, the DRMIA determined that some chronic diseases can cause destructive processes in this organ and lead to radionuclide redistribution from the liver to the skeleton and to other systemic organs as well as causing an increase urinary excretion of plutonium. To investigate the effect of liver pathology on the distribution of actinide elements in the body, all DRMIA and USTUR cases were classified into three "Health Groups". The groups were:

1. Group 1 included relatively health worker who died suddenly from accidents, suicides or acute cardiovascular diseases.
2. Group 2 included cases of death from malignant tumors of any organs (except liver) without multiple metastases to the liver.
3. Group 3 included workers who died from liver diseases such as cirrhoses, cancer of the liver, or tumors of other organs with multiple massive metastasis to the liver.

This same grouping was indicated by morphological analyses of livers. Microscopic sections of livers of individuals in group1, at autopsy, usually showed only slight signs of protein dystrophy of hepatocytes. Livers of group 2 were generally characterized by moderate fatty degeneration of hepatic cells. Morphologica changes in the livers in cases of group 3 were generally characterized by marked fatty degeneration of liver tissue.

Table G-1 contains data showing the mean distribution of plutonium and americium in the systemic organs of individuals of the three Health Groups. From TableG-1 it appears that a strong influence of liver pathology on distribution of actinides between the liver and the skeleton was more evident for DRMIA cases. It appears likely that systemic fractional deposition of plutonium and americium in the liver and skeleton were inversely related and disease conditions of the liver resulted in significantly reduced plutonium and americium fractions in the liver with significantly increased fractions in the skeleton. The redistribution involved a considerable shift of the actinides from the liver to the skeleton, especially for the 3rd group (those individuals who died from the diseases of the liver). The DRMIA mean systemic plutonium deposition fractions in the liver were 37.5, 30.2, and 12.7 percent and in the skeleton they were 49.2, 56.3, and 74.0 percent in Health Groups 1, 2, and 3, respectively. According to the increase of the fraction in skeleton it appears that all of the systemic fraction leaving the liver enters the skeleton. During the formation of pathological processes in the body, the fraction deposited in the liver was decreased by 24.8% (37.5-12.7 = 24.8 %), and that amount of plutonium was redistributed to the skeleton, where the systemic deposition fraction was increased by 24.8 % (74.0- 49.2 = 24.8 %). It is noted that this shift would probably only affect organ doses during the last few years of life.

It is evident from Table G-1 that the systemic fractions of americium in the liver were consistently lower than those for plutonium. In the relatively healthy DRMIA cases (Health group 1), the average americium fraction in the liver (17%) was 2 times lower than that for plutonium. The fractions of americium deposited in the skeleton were consistently much higher in the relatively healthy workers (62.6%) in DRMIA cases. During the formation of pathological processes in the liver, the systemic fraction of americium in the liver was reduced appreciably and, in individuals of Health Group 3, 13.6 % of the deposited amount that was released from the liver apparently recycled to the skeleton (the liver fraction was reduced by 17.4-3.8 = 13.6 %, while the skeleton fraction was increased by 76.7-62.6 =14.1 %).

In USTUR cases, the systemic fractions of actinides in the liver were higher in all three Health groups than they were in DRMIA cases. In the relatively healthy individuals, the average plutonium fraction in the liver was 50 % and the fraction in the skeleton was 36,7%. With liver disease, the systemic fractions of plutonium in the liver also decreased appreciably. In those individuals who died with a marked liver impairment (Health group 3), the mean systemic fraction of plutonium in the liver decreased by 18.9 % (50%-31.1 %) and the systemic fraction in the skeleton increased by the same amount (55.6%-36.7 % = 18.9 %). The mean values in Table G-1 show that, in USTUR cases, skeleton:systemic fractions of americium increased only minimally between groups (the USTUR had only one case in Health Group 1) indicating that less of the americium than plutonium mobilized from the liver and was transferred to the skeleton as a result of the disease conditions.

The coefficients of variation, CV, (Table G-1) for the mean systemic fractions of plutonium in the liver and in the skeleton in Health Group 1 in DRMIA and USTUR cases did not exceed 27 %; the maximum CV for americium was 48%. This was evidence of uniform partitioning among relatively healthy individuals. However, in Health Groups 2 and 3, the CV varied for plutonium fractions in liver between 52% and 60% and, for americium fractions in liver, between 68% and 74%. Such variation in the CV can be explained by the progression of pathologic processes in unhealthy individuals and the resulting marked destructive processes of different degrees in the livers in those groups. The results of statistical analyses indicated statistically significant differences between the mean systemic fractions of plutonium in the skeletons and the livers of each Health Group for cases of both Registries (Table G-2). For americium, however, the differences between Health Groups 1 and 2 (DRMIA) and between Health Groups 2 and 3 (USTUR) were not statistically significant.

Possible explanations for the differences in the magnitude of changes in liver deposition fractions observed by the DRMIA and the USTUR might be:

1. differences in classification into health groups of DRMIA and USTUR cases (DRMIA classification was performed by a pathologist on the basis of microscopic liver sections while USTUR classification was largely based on autopsy reports written by a variety of pathologists);

2. differences in the method of estimating actinide content in the skeleton (different bones collected at autopsy and the skeletal content was based on actinide concentrations in wet bones of USTUR cases and ashed bones in DRMIA cases);
3. a possible effect of body burdens on the distribution of actinides in systemic organs (plutonium concentrations in the livers of DRMIA cases were, on average, 250 times greater than those in USTUR cases; and
4. higher uncertainties in measurements of low levels of activity in the bones of USTUR cases.

Thus, the health status of workers is shown to have a statistically significant influence on the systemic distribution of plutonium and americium in the liver and the skeleton. These results are expected to have a significant impact on dose assessments made on the basis of bioassays for urinary excretion of plutonium and americium. The present ICRP models (ICRP 1986; 1989) recommend ratios of skeletal burden to liver burden as 50:30% for calculation of the plutonium or americium doses, based on bioassay methods. It is noted that the model recommendations were not contradicted by observed data from autopsy cases of the DRMIA and the USTUR who died from various diseases other than diseases with heavy liver impairment. The weighted average value of the skeleton:liver ratios for groups 1 and 2 in both DRMIA and USTUR cases was 51:36% and that was in close agreement with the 50: 30 ratio proposed by ICRP.

The distribution of americium in the bodies of relatively healthy individuals differed considerably from that proposed by the ICRP. The observed americium fraction retained in the liver was approximately in two times lower than the plutonium fraction and the americium skeleton:liver ratio was 62.6:17.4%. As a result of liver disease conditions, the americium fraction in the liver was three times lower than that for plutonium and the skeleton:liver ratio was 76.7: 3.8.

Based on this information, it is noted that the doses for liver and skeleton predicted by current ICRP biokinetic models will be different from the actual doses estimated for individuals with serious liver diseases for long periods of time. The health status of workers should be considered in models for more exact dosimetric estimations for plutonium or americium.

The information obtained in this investigation was used for modification of the combined lung clearance and systemic model developed for Project 2.4 to obtain the dose assessments for the Mayak cohorts studied for Projects 2.2 and 2.4.

Task H. Quantitate the relationships between actinide contents of the lungs and the body organs at autopsy and the long-term, temporal pattern of urinary excretion in healthy individuals and in health-impaired individuals.

In addition to a redistribution of actinide elements between the skeleton and the liver, marked liver impairment also results in an increased rate of urinary excretion of the actinides. Data supporting this observation were presented in a previous progress report (Khokhryakov et al. 1999).

The relationship between plutonium excreted in urine and that contained in body organs depends on the transportability of the aerosol inhaled and on the heath state of the individual. There were 55 cases of former Mayak and U. S. workers in DRMIA-USTUR database of 430 cases for which urinary bioassays were performed in the last year of life. Data on the plutonium distribution in the bodies of these cases were also available and the workers were exposed to plutonium containing aerosols of low transportability (0.3 to 1.0%). The cases were divided into two groups: Health Group 1 (defined in Task G, above) and a group with chronic diseases (Health Groups 2 and 3). Linear regression equations were used to describe the relationship between plutonium concentrations in the urine and concentrationtions in the lungs as well as between urine concentrations and systemic concentrations for each group. The regression equations contain an intercept term to improve the fit in the range of interest and to allow for a nonzero predicted urine concentration at the origin do to measurement error:

$$q_u = a_l + k_l \, X \, q_l \qquad (1)$$

$$q_u = a_s + k_s \, X \, q_s \qquad (2)$$

where q_u, q_l, q_s are the plutonium concentrations in urine (measured during the last year of the life), lung concentrations and systemic concentrations of plutonium (measured in autopsy samples), respectively. The values a_l and a_s are the intercept terms and k_l and k_s are the regression coefficients or slopes of the regression lines.

Table H-1 represents the results of regression analyses. According to these results, the plutonium concentration in urine is related to lung and to system contents. The correlation coefficients between urine and lungs are 0.78 and 0.33 for the relatively healthy group and the diseased groups, respectively. The correlation coefficients between urine and systemic contents were higher, 0.89 and 0.80, for both groups indicating a better fit of the regression line to the urine-systemic content data. It follows from the table H1 that both coefficients, k_l and k_s, for Health Groups 2-3 are higher than for the healthy group. This may be the result of increased nuclide excretion due to losses from the liversof people with liver diseases.

This interesting observation has importance for dosimetry purposes only if a bioassay were performed during the last few years of the life of an individual with marked liver impairment. In such cases, it would be prudent to base dose assessments on bioassays performed prior to the onset of the liver disease.

Task I. To enhance the sensitivity of the in vivo counter used by DRMIA and perform calibrations and intercomparisons with other, similar facilities so that it is a more useful tool for characterizing the intake and retention of actinide elements.

In 1997, the United States Department of Energy (DOE), Branch No. 1 of the Institute of Biophysics (FIB-1), United States Transuranium and Uranium Registries (USTUR) and Lawrence Livermore National Laboratory (LLNL) started the installation of a low background in vivo counting (WBC) facility at the FIB-1 site in Ozersk, Russia. This task included the construction of a new addition to the existing WBC facility, refurbishing and transferring the WBC to FIB-1 site after its disassembly at the Rocky Flats facility. Electronic Counter Corporation (ECC) performed refurbishing of the counter equipment.

The new room for the WBC was constructed at FIB-1 as an addition to the existing facility between March and October 1998. Installation of the shield was conducted between October 25 and November 3, 1998 with the collaboration of personnel from LLNL, ECC and FIB-1. The detectors, electronics, and some additional mechanical parts of the WBC arrived at Ozersk in May 1999. In July 1999, the following tasks were performed: installation of detectors and associated electronics, verification of the operability of the installed equipment and in vivo measurement software, and conduction of preliminary measurements of Mayak personnel. The initial checkout procedures and calibration of the new WBC was performed between September and December 1999. From October 1999 to March 2000, measurements of "Mayak" employees were conducted.

I-1. Description of newly installed counter

DRMIA investigations have shown that the ^{241}Am content of the body, measured by in vivo counting, correlates well with the ^{239}Pu body burden estimates based on urinalyses and with the analyses of tissues collected autopsy. The new WBC facility at the DRMIA was used for routine measurement of plutonium in the lungs, liver, and skeleton with calibrations based on ^{241}Am.

The counter includes the shielded room with a measurement chair and detector positioning equipment, high purity germanium (HPGe) detectors, phoswich detectors, associated electronics, and software for spectrum analysis. The shield is a room of 3x3x3 m size constructed of (from outside to inside) 6 inches pre-WW II steel, 0.125 inches Pb, 0.25 inches of Sn, followed by 0.1 inches of Zn. The counter is equipped with two arrays of 4 (total 8) planar HPGe detectors from PGT and two phoswich detectors from Harshaw. The guaranteed system resolution of the HPGe detectors, at 122 keV, is not greater than 750 eV. Each detector has approximately a 200 mm^2 surface area.
The acquisition, analysis and storage of spectral data are accomplished using a multichannel analyzer (MCA) and a computer with GENIE-2000 and ABACOS-2000 software for in-vivo analyses. The program, GENIE-2000, is a comprehensive tool set for

acquiring and analyzing spectra from multichannel peak analyzers. Its function includes MCA control, spectral display and manipulation, basic spectrum analysis and reporting. ABACOS-2000 is a "shell" above GENIE-2000 that provides a complete set of operating procedures allowing the reliable counting and analysis subjects, as well as performing operating functions such as calibration, quality control and background counting. GENIE-2000 is not able to process the spectral data obtained by the phoswich detectors; therefore, the calibration and processing of results of measurement from the phoswich detectors must be performed manually. For the evaluation of an individual background in the area of 59.5 keV, the method utilizes the background count at a higher energy region to predict the background count in the lower energy region (59.5 keV). In order to obtain the correlation between two energy regions a series of measurement of background counts were performed in each of the measurement geometries.

Patients are measured in a supine position for 30 minutes. The lung measurement, is accomplished by placing two arrays of HPGe detectors or two phoswich detectors as close as possible to the upper anterior thoracic surface. Usually the detectors are angled to conform to the natural contour of the chest. Estimation of skeletal contents are performed by skull measurement. The skull is most convenient site and it is known to contain about 14% of the total skeletal activity. For the skull measurement using HPGe detectors, the patient lies on a measuring chair with the detectors positioned over the facial area of the head. The phoswich detectors, for this measurement, are centered on opposite sides of the head. The liver measurement is performed with one array of HPGe detectors or one phoswich detector positioned over the liver area adjacent to the bottom rib of the rib cage.

Calibration of the newly installed counter was performed using phantoms with ^{241}Am contents shown in table I-1. Calibrations in lung and liver geometries were performed using a LLNL-type torso phantom that includes removable organs and interchangeable chest overlays to simulate a wide range of chest-wall thicknesses (CWT). Calibration points for lung and liver measurement were obtained for each chest wall thickness, and the dependence of efficiency on CWT was calculated. The efficiencies for skull measurements were obtained with an RSD skull phantom. The measured efficiencies are shown in tables I-2 and I-3 for HPGe and phoswich detectors, respectively. CWT of individual subjects are calculated by the ABACOS-2000 program, based on anthropometric data. The wall thickness over the liver area assumed to be the same as the thickness over the lung area.

For measurement of activity in a certain organ, it is necessary to consider the contribution of activity contained in an organ other then the one being measured (organ cross – talk factors)). For this purpose, six calibration measurements were performed:

1. determination of the contribution of the skeleton and liver activity to the lung count,
2. determination of the contribution of the skeleton and lung activity to the liver count, and
3. determination of the contribution of the liver and lung activity to the skull count.

The USTUR torso skeletal phantom was measured in the lung and liver geometries to investigate the skeletal contribution to lung and liver counts. Measurements of the USTUR torso phantom were conducted with only one absorbing CWT because the LLNL-type torso phantom interchangeable plates were not useful with the USTUR torso phantom. Efficiencies determined from these measurements for the 59,5 keV energy are shown in tables I-2 and I-3 for HPGe and phoswich detectors, respectively.

The results of determination of organ cross – talk factors support the conclustion that, for lung counting, it is necessary to make a correction for the contribution from activities in the skeleton and liver

To verify the calibrations and the evaluations of reproducibility of results, measurements of phantoms as subjects with the unknown contents were performed and the results of the measurements were compared with the certified activity of phantoms. The results, thus obtained, indicate acceptable calibration and reproducibility of the measurements. The relative bias of the measured activity compared with the certified activity of the RSD skull phantom does not exceed $1 \pm 2.4\%$ for the HPGe detectors and $1.75 \pm 1\%$ for the phoswich detectors. For the HPGe detectors in the lung geometry, the average relative bias does not exceed 1 % and, for liver geometry, 2.1%; average variances for lung and liver were 2 % and 1.3%, respectively. For the phoswich detectors in the lung mesurement, the average relative bias does not exceed 4.7 % and, for liver, 5%; average variances for lung and liver were 1.5 % and 2.7%, respectively

I-2. Comparison of the various WBCs, used by the DRMIA

There are three in-vivo counters in use by the DRMIA at the present time:

1. Skull Counter, an NaI(Tl) counter intended for measurements in the skull geometry to evaluate the contents ^{241}Am in the skeleton at long times after exposure of workers. The skull is the most convenient site to measure and it is known to contain about 14% of total skeletal activity. This counter is equipped with two NaI(Tl) detectors, each with a diameter of 150 mm and a thickness of 100 mm and it is positioned over the facial and cervical surfaces of the head. Initially, a tissue-equivalent skull phantom (DRMIA skull) with a content 320±20 Bq of ^{241}Am, uniformly distributed in the volume of the skull, was measured for calibration of this counter. A new calibration factor was obtained by measurement of the RSD skull phantom; this and the historic calibration factor are shown in Table I-4. The new calibration factor was approximately about 20% greater than the old factor.

2. Canberra counter, equipped with germanium detectors, is used for measurements in any geometry. Two of detectors are low energy detectors, GL3825, with an active area of 3800 mm^2 each and they are designed to measure photons with energies up to 500 keV. A wide-energy coaxial detector, GX3019, with an active diameter of 51 mm is used for radionuclides that emit high-energy photons (up to 2.5 Mev). The shielded room for this

counter is made of 50 mm thick Pb blocks and 0.5 mm thick Cd. Calibrations for lung and skull measurement for americium are currently underway in this counter..

3. Rocky Flats counter, the counter installed as part of Project 2.1, is used for measurements in lung, liver, and skull geometries with phoswich and HPGe detectors. This counter is described in detail above.

Until June, 1999, an in-vivo counter equipped with 4 NaI(Tl), 3 mm-thick and 140 mm-diameter detectors was used by the DRMIA on a routine basis. Measurements, with it, were conducted in lung, liver, and skeletal geometries simultaneously. The output of each detector was adjusted with a calibration factor and spectral data were summed to provide the contents of ^{241}Am in the whole body. This counter was replaced with the Canberra counter described above.

To verify the new calibrations and to compare the results obtained on Skull NaI(Tl), Rocky Flats (HPG detectors) and Canberra counters, multiple measurements of the USTUR skull phantom were conducted. The results of these measurements are shown in Table I-5 and the results obtained on the three counters are in close agreement. The smaller uncertainty in the measurement from the Skull NaI(Tl) counter is a result of the configuration of the detectors in this counter; it was designed specifically for measurement in the skull geometry.

The minimum detectable activities (MDA) for the various organs and for the whole body (where possible) for all WBCs with a measurement time of 30 minutes are given in Table I-6. The MDA was calculated by using Currie method:

$$MDA = \frac{1.645^2 + 2\,(1.645\,\sqrt{B + \sigma_B^2}}{y\ X\ \varepsilon(E) T}$$

where

B is the background continuum under consideration;
σ_B is the standard deviation of the background measurement;
y is the branching ratio of the gamma energy under consideration;
$\varepsilon(E)$ is the efficiency ; and
T is the collection live time in seconds.

The smaller MDA of the HPGe detectors for skull measurements, despite the higher efficiencies of the Skull NaI(Tl) counter and phoswich detectors for this measurement is result of lower background associated with the HPGe detectors.

I-3. Results of measurements of human subjects

A total 100 persons were measured using HPGe detectors: 99 in lung geometry, 33 in liver geometry and 49 in skull geometry. The total number of measurement was 181 and 31 subjects were measured in all three geometries for determination of the ^{241}Am contents in the body. The results of those measurements with uncertainties (2σ), obtained directly

by measurement of certain organs (no cross-talk factors) and the lung result, with corrections for the skeletal and liver contributions (with cross-talk factors) are shown in Table I-7. Those analyses show that the difference between the two methods of data processing for lung measurement was an average 42 %. The evaluation of the contents in the whole body, without consideration of organ cross-talk factors, overestimated the body burden by an average of 7%. Such insignificant differences in the evaluation of total body contents indicate that cross-talk factors can be neglected for routine measurements.

Thirty subjects were measured in skull, lung, and liver geometries with the phoswich detectors and the results of those measurement with 2σ uncertainties (and assuming no interference between measured organs) are given in table I-8.

A number of subjects, measured with the HPGe and phoswich detectors were also measured with the Skull NaI(Tl) counter and the Canberra counter. A total of 49 subjects were measured in the skull geometry using the HPGe detectors. Contents in the skull, above the MDA, were detected on 36 of those cases while the Skull NaI(Tl) counter indicated contents above the MDA in only 13 cases from this group of subjects. This is a good indication of the improvement of sensitivity of the WBC facility resulting from the installation of the HPGe detectors. Table I-9 shows correlation coefficients (with number of cases) of the positive results for comparison of the HPGe and phoswich detector results with results obtained by measurement on Skull NaI(Tl) and Canberra counters.

In conclusion, the newly installed in-vivo counter greatly improves the quality of monitoring of "Mayak" workers and it expands the range of research that can be conducted by the DRMIA WBC facility. In vivo monitoring can be a useful tool in establishing a new and improved dosimetric model for "Mayak" personnel. The new counter permits measurements in lung, liver, and skull geometries with either HPGe or phoswich detectors and to determine the ^{241}Am contents of individual organs as well as the total body content. The MDA of the HPGe detectors is the lowest of all DRMIA in-vivo counters and the high energy resolution of the HPGe detectors allows the determination and identification of nuclides with high probability, providing precise quantitative analyses. Application of phoswich detectors together with the shape analyzer reduces the background contribution of compton scattering in the 59.5 keV area. Calibration of the counters, using generally recognized phantoms, was an innovation to the in-vivo facility of more precise determination of the radionuclide content in the lung. FIB-1. The calibrations, with compensation for organs cross-talk factors, resulted in a more precise determination of the radionuclide content of the lung.

Task J. To translate previously classified Russian documents into English for submission to peer-reviewed journals or for publication as topical reports, as appropriate.

A manuscript, Metabolism and dosimetry of actinide elements in occupationally –exposed personnel of Russia and United States: a summary progress report. Khokhryakov, T. F.; Suslova, K. G.; Filipy, R. E.; Alldredge, J.R.; Aladova, E. E.; Glover, S. E.; Vostrotin V.V., was accepted for publication in Health Physics. This paper was a result of the studies conducted during of the long-term collaborative project. It is included in the list of publications below (Khokhryakov et al. 2000).

Two other Russian papers were prepared in partial fulfillment of task J with the objective of translating previosly Russian papers into English for publication. These documents were selected by the U. S. National Council on Radiation Protection and Measurements (NCRP). They were considered to have direct applicability to the work of NCRP Committee 57-17, Radionuclide Dosimetry Model for Wounds. The authors and translated titles of the reports are:

The risk of incorporation of Pu and ^{241}Am into bodies of radiochemical facility personnel through injured skin. Khokhryakov, V. F.; Kudryavtseva, T. I; Shevkunov, V. A.; Filipy, R. E.

Injuries and Burns of Skin with Alpha Activity Contamination in Mayak Workers Bazhin, A. G.; Khokhryakov, V. F.; Shevkunov, V. A.; Filipy, R. E.

Other relevant information, including relevant trip reports, obstacles to completion of work outlined in FY work proposal, unexpected costs.

Two USTUR scientists, Dr. S. E. Glover and Dr. R. E. Filipy, visited the DRMIA facilities in July 1999. The primary purpose of the visit was to prepare the seventh semi-annual progress report and to assist the DRMIA with calibration of their newly-installed in vivo counters. Because of unexpected shipment delays, the calibration phantoms did not arrive at FIB-1 during the visit; therefore, the calibration of the in vivo systems could not be accomplished at that time. However, calibration procedures were reviewed with the Russian scientists and they performed the calibrations later in 1999.

During the current reporting period in February, 2000, four DRMIA scientists, Dr. V. F. Khokhryakov, Dr. K.G. Suslova, V.V. Vostrotin, and E.E. Aladova and Dr.R.E. Filipy, Principal Investigator of the project from USTUR, visited Dr. S.C. Miller, U. S. Principal Investigator of Project 2.4, in Salt Lake City, Utah. The final report for Project 2.1 and the progress report for Project 2.4 were discussed and drafted during these meetings. Possible continuation of Projects 2.1 and 2.4 for the purpose of making improvements in plutonium dosimetry in the lungs, was discussed with Dr Ruth Neta, U. S. DOE representative. Much attention was given to the discussion of questions regarding the modifying effects of such factors as smoking, age, and nonuniformity of lung distribution of plutonium on the deposition and retention of inhaled plutonium compounds in lung that may influence the dose accumulation. It was decided that modifying effects of these

factors should be studied during the next stages of project 2.1 and this is a part of the proposed continuation in Appendix I of this report.

Publications and Preprints

This Final Report for Project 2.1 (15 March 1999 –30 March 2000) was submitted in English and in Russian.

Manuscript: Metabolism and dosimetry of actinide elements in occupationally –exposed personnel of Russia and United States: a summary progress report. Khokhryakov, V. F.; Suslova, K. G.; Filipy, R. E.; Alldredge, J.R.; Aladova, E. E.; Glover, S. E.; Vostrotin V.V. Accepted by Health Physics.

Oral Presentation: Combination of the actinide analysis data of two human tissue analysis programs: triumphs and problems. Filipy, R. E.; Alldredge, J. R.; Glover, S. E.; Khokhryakov, V. F.; Suslova, K. G.; Aladova, E. E.; Vostrotin, V. V. At the international symposium, Chronic Radiation Exposure: Possibilities of Biological Indication, in Chelyabinsk, Russian Federation on March 14-16, 2000.

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Appendix I

Proposal for Continued Collaborative Research on
Project 2.1—Metabolism and Dosimetry of Plutonium Industrial Compounds

By

The Dosimetry Registry of the Mayak Industrial Association (DRMIA)
K. G. Suslova, Project Leader

And

The United States Transuranium and Uranium Registries
R. E. Filipy, Director

Purpose: The primary objective of this proposed research project is to provide additional and more specific dosimetry information that can be used in epidemiologic and molecular genetics studies conducted under the auspices of the Joint Coordinating Committee for Radiation Effects Research.

Task Objectives

1. Complete, for submission to peer-reviewed scientific journals for publication, five manuscripts including results of Project 2.1 collaborative research and translations of Russian documents.
2. Establish, within the DRMIA, a transuranium registry of living Mayak workers and former Mayak workers who will receive internal dosimetric evaluations and who will contribute tissue samples to the newly established Russian Human Tissue Repository.
3. Compile internal dosimetric data on approximately 1200 deceased Mayak workers, many of whom were examined by bioassay during the period, 1955-1972. This is in direct support of epidemiology studies currently underway.
4. Further evaluate the internal dosimetry of DRMIA cases in terms of uncertainties of measurements, age at exposure, gender, chelation therapy, and plutonium body content, in support of Project 2.4.
5. Investigate the influence of cigarette smoking on the deposition, clearance, and retention of actinide elements in the lungs.

Background and Approach

1. Preparation of a number of manuscripts was begun during the 3-year duration of Project 2.1 and there was not sufficient time to complete them for submission to scientific journals. Some of those manuscripts and authors are:

An interlaboratory comparison of radiochemical analytical methods for actinide elements in human tissues and bioassay samples. Filipy, Glover, Suslova, Alldredge, Orlova, Stuit, Chernikov, Khokhryakov

Injuries and skin burns with alpha activity contamination among Mayak workers. Bazhin, Khokhryakov, Shevkunov, Filipy

The risk of intake of plutonium and americium through skin injuries of radiochemical facility personnel. Khokhryakov, Kudryavtseva, Shevkunov, Filipy

Lung clearance of inhaled plutonium compounds at long times after intake. Khokhryakov, Suslova, Vostrotin,

Systemic plutonium distribution at long times after intake in relatively healthy individuals at the Mayak radiochemical plant. Suslova, Khokhryakov, Tokarskaya, Nifatov, Kudryavtseva,

It is anticipated that the majority of these papers will be submitted within six months of initiation of the work.

2. Branch No. 1 of the Federal Research Center Institute of Biophysics (FIB-1) has identified a large group (> 1000) of occupationally-exposed workers that still reside in Ozyorsk. A cohort of those workers, to be selected by the DRMIA, will be the subject of this project. The cohort will be selected on the basis of actinide exposure histories, sex, smoking histories, and willingness to participate. These individuals will be requested to consent to the special studies described below.

Individuals of the cohort will be asked to submit periodic urine samples that will be analyzed with the new DRMIA methods and equipment acquired as a result of Project 2.1. Probable body burdens of plutonium will be estimated by the use of established urinary excretion models.

Performance of in vivo counting on the same individuals will be used to further characterize the actinide body burdens by identifying the body organs (respiratory tract, liver, skeleton) that contain the actinides. As a result of the previous collaborative research, the DRMIA has acquired an in vivo counting system, formerly used at the Rocky Flats Facility. It was refurbished, installed, and calibrated during the last two years.

Individuals will be asked to submit blood samples to be collected by members of the clinical staff. These samples will be included in the newly established Russian Human Tissue Repository along with the results of bioassays and in vivo counts as well as exposure histories.

Upon the death of an individual from the cohort, an autopsy will be performed to collect tissue samples that will be divided for radiochemical analyses and for submission to the Russian Human Tissue Repository.

This task will be performed primarily by the DRMIA. The USTUR will act in an advisory capacity and provide assistance when needed; however, the DRMIA and USTUR radiochemical laboratories will collaborate to establish a quality assurance program. This program will include periodic analyses of split tissue samples and Standard Reference Materials provided by the U. S. National Institute of Standards and Technology to verify the DRMIA methods used to analyze bioassay and tissue samples. The USTUR also has a Policies and Procedures Manual governing its operation and many of those policies and procedures are expected to be readily adaptable for use by the DRMIA.

3. As part of a collaborative research project (Project 2.4), the DRMIA calculated internal doses for 5300 workers on the basis of bioassays, in support of Projects 2.2 and 2.3. There are autopsy data for approximately 1200 cases in the DRMIA laboratory, most of which were not included in the 5300 cases. Only 300 cases were included in the DRMIA-USTUR joint database for Project 2.1. The cases that were used in Project 2.1 were selected on the basis of the following criteria:

a. tissue analyses of the major organs including lung, liver and skeleton for both plutonium and americium,
b. plutonium tissue content data for more than eight body organs
c. plutonium body burdens greater than 20 nCi, and,
d. individuals that worked in either the plutonium production plant or the radiochemical plant at Mayak.

These criteria excluded most individuals who died before 1975, primarily because tissues from the earlier cases were analyzed only for total alpha activity that included all plutonium and americium. During 1999-2000 an additional 600 cases were added in the DRMIA database. As part of the continuation of Project 2.1, internal dosimetry based on autopsy tissue analytical data will be included for 900 cases. These cases, plus the 300 cases which also will be added to the DRMIA database during the continuation of Project 2.1 (thus, a total 1200 cases), will be available for the epidemiological studies. Many of these individuals worked at Mayak in the early days of operation so they might be appropriate for studies of both stochastic and deterministic effects. Assessments of plutonium body burdens, with bioassay methods, were conducted for approximately half of the 1200 cases. It will be necessary to develop methods for assessment of internal doses based solely on autopsy tissue analytical data with autopsies generally long after exposures. This will add considerable numbers to the cohorts studied in Projects 2.2 and 2.3. It is expected that evaluation of doses, based on the results of tissue analyses, will result in less uncertainty than doses based on the bioassay method. Many of the tissues from those early cases were ashed, dissolved in acid, archived in bottles, and they are available for analyses for isotopic plutonium and americium with the new, improved DRMIA analytical capability.

This task is primarily a DRMIA task and it will be conducted concurrently with the establishment of the transuranium registry (Task 2).

4. During the course of Project 2.1, it became apparent that there could be several modifying factors that might influence internal dose assessments and increase the uncertainties associated with the dose assessments. A goal of this task is to investigate some of the modifying factors with the objective of decreasing those uncertainties. Some of the potential dose modifying factors include: age at exposure, gender, and chelation therapy which was used on a few hundred cases included in the dosimetry databases.

There is a wealth of information available on animal experiments with chelation therapy with only limited data from a few studies of DTPA treatments following the contamination of workers with actinide elements. DRMIA has approximately 700 Mayak workers that were examined by two methods: they received the DTPA injections and urine samples were collected for bioassay purposes both before and after treatment with the DTPA. These data will be used to determine the effectiveness of DTPA treatments on the rate of urinary excretion of plutonium. This study is especially important for a large group of Mayak workers examined in the early years, before 1972. Most of this cohort (more than 800 workers) was treated with DTPA injections. This study will be in support of Project 2.4 with the purpose of extending the DRMIA database with additional internal exposure doses for the cohort of Mayak workers studied in projects 2.2 and 2.3.

Another factor, such as the plutonium body burden, can also result in an increase in the uncertainties associated with dose assessments. During the collaborative research in Project 2.1, it was determined that DRMIA registrants had median plutonium body burdens more than 200 times those of USTUR registrants. Between the two Registries, there was a range of body burdens over five orders of magnitude; maximum values were on the order of 10^5 Bq in the DRMIA cases and minimum values of USTUR cases are near background levels. Animal experiments have demonstrated changes in distribution and retention of plutonium as a result of different masses of injected plutonium isotopes (Bair, 1974; Guilmette, 1978; Lloyd 1984).

5. Most epidemiological studies of lung cancer from inhaled radionuclides list cigarette smoking as one of the leading confounding factors. This was especially true in a study of the Mayak workers where Tokarskaya et al. (1993) found that more than 85% of workers exposed to plutonium by inhalation were also smoked cigarettes at some time during their lives. Cigarette smoke is an established carcinogen but it has also been shown to affect deposition and retention of plutonium in the lungs. Filipy et al. (1981) reported significant differences in lung clearance of plutonium between cigarette smoke-exposed and sham-exposed (control) rats as early as 2-3 weeks after inhalation of plutonium. At six weeks after plutonium exposure, the smoke exposed rats had retained approximately 65% of their initial lung burdens (ILB) while the controls and retained only 40% of the ILB. Filipy et al. (1982) reported similar impaired lung clearance in beagle dogs that had been exposed to cigarette smoke and plutonium. Sham-exposed dogs had cleared 30% of

inhaled plutonium at 60 weeks after plutonium exposure while smoke-exposed dogs had cleared only 15% of the ILB. A consequence of the greater retention of plutonium by smokers would be higher lung doses from the plutonium. Finch et al. (1998) calculated lifetime lung doses from plutonium inhaled by rats that had been exposed to cigarette smoke or sham-exposed and the doses were 3.8 Gy in sham-exposed (control) rats, 4.4 Gy in rats exposed to a low concentration of cigarette smoke, and 6.7 Gy in rats exposed to a high concentration of smoke. They speculated that those doses would result in an increase in lung neoplasms by approximately 20% for the low concentrations of smoke and 80% for the high concentrations of smoke when compared to the control group. They also noted that smoke influenced the amount of plutonium initially deposited in the lungs; more was deposited in the lungs of smokers.

Data regarding the influence of cigarette smoke on lung clearance in humans are limited; however, Cohen et al. (1979) demonstrated retarded lung clearance in smokers and Kathren et al. 1993) compared lymph node:lung plutonium concentrations in tissues of human donors to the USTUR. They reported significant differences between smokers and non smokers in the lymph node:lung concentration ratios for Pu-239+240 and for Am-241. Greater fractions of the radionuclides remained in the lungs of smokers indicating impaired clearance and most of the individuals had died between 10 and 40 years after chronic low-level exposure to plutonium. The expansion of the DRMIA autopsy database by up to 1200 cases, which included a considerable number of smokers, will provide the subjects for a study of the influence of smoking on lung dose accumulation.

The performance of this task will involve compilation of the smoking histories of DRMIA and USTUR cases and including them in the joint DRMIA-USTUR database. Plutonium distributions in the body, particularly those in the respiratory tract relative to other body organs, will be investigated with the intent of verification or modification, as necessary of lung dosimetry models. Improvement of plutonium dosimetry as a result of this study is expected to provide refinement of the individual organ doses for the Mayak cohorts studied in the Projects 2.2 and 2.3, to determine risk estimates of plutonium-induced effects in Mayak workers.

This task will be conducted jointly by the DRMIA and the USTUR and will run concurrently with the establishment of the DRMIA transuranium registry.

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Table A-1. Characteristics of the measurement and radiochemical analytical methods for actinides currently in use by the DRMIA and the USTUR.

Index	DRMIA			USTUR
Measuring method:	Co-precipitate with $BiPO_4$, radiometry	Alpha-spectrometry		Alpha-spectrometry
- type of device	Radiometer RIA-05	Alpha-spectrometer SEAM	Alpha-spectrometer EG+G Ortec OCTETE	Alpha-spectrometer EG+G Ortec OCTETE
- type of detector	photo-electronic multiplier scintillation with ZnS(Ag)	Ionization chamber	Silicon surface-barrier	Silicon surface-barrier
- background, s^{-1}	$9 \cdot 10^{-4}$	$8 \cdot 10^{-5}$	$2 \cdot 10^{-5}$	$2 \cdot 10^{-5}$
- efficiency, %	95±3	45	25	no less than 25
- resolution, keV	-	50	25	25
Radiochemical analytical method:	Anion exchange isolation			
	anion exchange resin ВП 1-АП (50-100 mesh)		anion exchange resin AG 1-X4 (100-200 mesh)	anion exchange resin AG 1-X4 (100-200 mesh)
- background, 5-1	Pu 1.66 ± 1.00 Am 2.00 ± 1.71	$^{239+240}Pu$ 0.69±0.28 ^{241}Am 0.79±0.55	$^{239+240}Pu$ 0.061±0.065 ^{241}Am 0.172±0.172	-
- MDA, mBq/sample	5.0	2.0	1.0	0.7
- accuracy, %*)	45	50	60	-

*NOTE: The mean of accuracies calculated for measuring times of 5 days.

Table A-2. The results of the first and second steps of the intercomparisons of instrumental and radiochemical analytical methods of both Registries.

Steps	Samples[1]	Methods of measurement	n[2]	Isotope	Difference[3] $\Delta_i \pm SD$	Analysis of variance[4] (unweighted P value)
Step 1	DRMIA	Radiometry DRMIA- α-spectrometry USTUR	5	ΣPu[5]	0.040±0.269	0.828
	USTUR	α-spectrometry USTUR - α-spectrometry DRMIA	3	$^{239+240}$Pu	0.170±0.276	0.529
		α-spectrometry USTUR - α-spectrometry DRMIA	4	^{241}Am	-2.378±4.835	0.537
Step 2	DRMIA	Radiometry DRMIA - α-spectrometry USTUR	10	ΣPu[5]	-1.588±5.425	0.775
		α-spectrometry DRMIA -- α-spectrometry USTUR			-1.423±5.425	-
		Radiometry DRMIA - α-spectrometry USTUR	9	Am	-0.797±1.389	0.259
		α-spectrometry DRMIA -- α-spectrometry USTUR			-1.085±1.389	-
		α-spectrometry DRMIA - α-spectrometry USTUR	10	$^{239+240}$Pu	-1.435±5.007	0.538
		α-spectrometry DRMIA - α-spectrometry USTUR	6	^{238}Pu	0.011±0.077	0.817
	USTUR	α-spectrometry USTUR - α-spectrometry DRMIA	9	$^{239+240}$Pu	8.866±16.741	0.267
		α-spectrometry USTUR - α-spectrometry DRMIA	6	^{238}Am	0.576±0.879	0.308
		α-spectrometry USTUR - α-spectrometry DRMIA	9	^{241}Am	-0.581±0.505	0.041

[1] - The laboratory that prepared the samples for intercomparison.
[2] - The number of measured samples.
[3] - The mean of differences between measured results from each laboratory.
[4] - Weighted and unweighted analysis of variance were used. Only p-values for unweighted analysis are presented.
[5] - The result of DRMIA radiometry for total Pu: Σ Pu = ^{238} Pu + $^{239+240}$ Pu.

Table A-3. Results of radiochemical analysis of Standard Reference Materials (SRM).

SRM	Isotope	Certified activity, Bq / g (± uncertainty, %)	Measured activity, Bq / g (± uncertainty, %)	
			Alpha-spectrometry Ortec OCTETE	Alpha-radiometry
4351 human lung m = 44.31g	^{234}U	$1.00 \cdot 10^{-4}$ (± 25 %)	$9.24 \cdot 10^{-5}$ (± 22 %)	-
	^{238}U	$1.01 \cdot 10^{-4}$ (± 11 %)	$9.38 \cdot 10^{-5}$ (± 24 %)	-
	$^{239,240}Pu$	$1.10 \cdot 10^{-3}$ (+110 %; - 50 %)	$7.15 \cdot 10^{-4}$ (± 13 %)	$7.42 \cdot 10^{-4}$ (± 34 %) [1]
	^{238}Pu / $^{239,240}Pu$	$1.5 \cdot 10^{-2}$ (±18 %)	- (^{238}Pu < MDA)	
	^{241}Am [2]	$1.10 \cdot 10^{-4}$ (±64 %) [2]	$7.90 \cdot 10^{-5}$ (± 20 %)	-
4352 human liver m = 42.07 g	^{238}Pu	$5.50 \cdot 10^{-5}$ (± 44 %)	$5.73 \cdot 10^{-5}$ (± 30 %)	$1.94 \cdot 10^{-3}$ (± 34 %) [1]
	$^{239,240}Pu$	$2.6 \cdot 10^{-3}$ (± 19 %)	$2.06 \cdot 10^{-3}$ (± 8 %)	
	^{241}Am	$1.50 \cdot 10^{-4}$ (± 37 %)	$2.14 \cdot 10^{-4}$ (± 16 %)	-
	^{234}U [2]	$1.00 \cdot 10^{-4}$ (± 11 %) [2]	$9.81 \cdot 10^{-5}$ (± 20 %)	-
	^{235}U[2]	$9.00 \cdot 10^{-6}$ (± 113 %) [2]	(^{235}U< MDA)	-
	^{238}U[2]	$8.80 \cdot 10^{-5}$ (± 12 %) [2]	$1.05 \cdot 10^{-4}$ (± 21 %)	-

[1] - the measured activity of total Pu: $\Sigma Pu = {}^{239,240}Pu + {}^{238}Pu$;
[2] - uncertified values of isotope activities;

Table E-1. Geometric means of plutonium content in the respiratory tract as a percent of (the total body content at death) related to the transportability(s) of workplace aerosols

Workplace	Reprocessing of uranium fuel	Plutonium fuel production workplace 1	Plutonium fuel production workplace 2
Transportability, S %	3.21(1.69)*	1.0(3.00)	0.2(1.55)
Quantity of cases	281	112	45
Lung content, %	2.56(2.26)	6.5(2.47)	22.6(1.81)
Lung and lymph nodes content, %	3.60(2.30)	13.2(2.47)	43.2(1.80)

- geometric standard deviation is given in the parentheses

Table E-2. Dynamics of transportability of aerosols from plutonium fuel production

Year of investigation	1977	1996	1999
Number of air samples	5	6	5
Content of Am, %	14.3±3.9	87.8±5.9	58.0±6.7
Transportability of Pu, %	0.13±0.01	1.26±0.3	1.18±0.6
Transportability of Am, %	0.48±0.08	2.58±1.5	4.5±3.9
Total transportability, S %	0.18±0.05	2.42±1.3	2.85±2.7

Table F-1. Parameters of the plutonium excretion models for urinary and fecal excretion

Urine			Faeces		
I	a_i	x_i, day^{-1}	I	a_i	x_i, day^{-1}
1	4.1×10^{-3}	0.5634	6	6.0×10^{-3}	0.3465
2	1.2×10^{-3}	0.126	7	1.6×10^{-3}	0.105
3	1.3×10^{-4}	0.0165	8	1.2×10^{-4}	0.0124
4	3.0×10^{-5}	0.00231	9	2.0×10^{-5}	0.0018
5	1.3×10^{-5}	2×10^{-5}	10	5.2×10^{-6}	2×10^{-5}

Table F-2. Parameters of the DRMIA plutonium lung model for DRMIA and USTUR cases.
(Values of the parameters are given as the arithmetic mean ± standard deviation, except the values for K_f which are given as geometric mean and geometric standard deviation).

	S(%)	N	K_f(%) , GSD *)	β (%)	λ (y^{-1})	K_n (%)
DR MIA	0.3	49	5.0 , 3.52	63.0 ± 35.9	0.099 ± 0.032	22.0 ± 3.9
	1.0	101	3.0 , 2.61	25.7 ± 9.8	0.106 ± 0.023	23.0 ± 1.4
	3.0	99	1.9 , 2.1	17.8 ± 10.3	0.190 ± 0.065	6.0 ± 0.68
UST UR	-	43	2.6 , 3.86	53.0 ± 26.0	0.172 ± 0.065	4.0 ± 1.1

* geometric mean and geometric standard deviation

Table F-3. Comparison of the predictions of different lung models with observed measurements

Model	Chemical compounds	Class of aerosol	Transportability (%)	Ratio of Model /Observed[1] (geometric mean)	Geometric standard deviation	Number of cases	Database
DRMIA	Dioxide, Metal	-	0.3	1.0	2.47	49	FIB-1
	Mixed : dioxide, nitrate, chloride, оксалат	-	1.0	1.04	2.21	101	FIB-1
	Nitrate	-	3.0	1.08	2.24	99	FIB-1
	Unknown Mix	-	-	1.01	3.29	43	USTUR
ICRP-30	Oxides, hydroxides	Y	0.3	2.4×10^{-3}	82.1	49	FIB-1
	Others	W	1.0 – 3.0	3.5×10^{-37}	3.93×10^{25}	200	FIB-1
	Oxides, hydroxides	Y	-	0.19	15.6	43	USTUR
	Others	W	-	6.2×10^{-13}	2.8×10^{14}	43	USTUR
ICRP-66	Oxides, Metal	S	0.3	1.2	2.54	49	FIB-1
	Others	M	1.0 – 3.0	6.9×10^{-15}	2.2×10^{9}	200	FIB-1
	Oxides, Metal	S	-	2.95	3.33	43	USTUR
	Others	M	-	4.6×10^{-6}	2.8×10^{-5}	43	USTUR

[1] Ratio of model-predicted/observed fractions of total plutonium body burdens in the lungs

Table G-1. Relative distribution of plutonium and americium in systemic in liver pathology groups, % systemic fraction in the organ (arithmetical mean ± standard deviation, M ±σ)

Organ	DRMIA				USTUR			
	Plutonium		Americium		Plutonium		Americium	
	N[1)]	M ±σ	N	M ±σ	N	M ±σ	N	M ±σ
	Health group 1							
Liver	74	37.5 ± 10.1 CV = 27 % [2)]	14	17. 4 ± 8.4 CV = 48 %	7	50.0 ±7.4 CV =15 %	1	22.6
Skeleton	74	49.2 ± 10.1 CV = 21 %	14	62.6 ± 8.4 CV = 13 %	7	36.7 ±7.4 CV = 20 %	1	57. 6
Other organs [3)]	74	10.9 ±31.7	14	2. 8 ± 3.2	-	-	-	-
	Health group 2							
Liver	01	30.2 ± 15.7 CV = 52 %	17	16.2 ± 12.0 CV =74 %	80	40.5 ± 21.4 CV = 54 %	37	18.1 ±13.0 CV = 72%
Skeleton	101	56.3 ±15.6 CV = 10 %	17	63.7 ± 12.0 CV = 19 %	80	46.2 ±21.4 CV = 46 %	37	61.9 ±13.2 CV = 21%
Other organs [3)]	101	9.3 ± 7.7	17	4.7 ± 15.8	-	-	-	-
	Health group 3							
Liver	83	12.7 ± 7.4 CV = 58 %	29	3.8 ±2.7 CV = 71 %	43	31.1 ± 18.7 CV = 60 %	23	15.5 ±10.5 CV = 68 %
Skeleton	83	74.0 ± 7.4 CV = 10 %	29	76.7 ± 2.8 CV = 4 %	43	55.6 ±18.7 CV = 34 %	23	64.5 ±10.5 CV = 16 %
Other organs [3)]	83	7.4 ± 12.6	29	5.2 ±6.3	-	-	-	-

NOTE: [1] N – number of cases;

[2] CV – the coefficient of variation, calculated as σ/ X, %;

[3] Sum of the relative content in other organs analyzed (muscle, heart, kidneys and spleen).

Table G-2. Level of significance of differences between systemic fractions of plutonium and americium in liver (and in skeleton) for the three pathology groups. Based on the T-test with determined confidence level, P.

Pathology group	DRMIA			USTUR		
	1	2	3	1	2	3
	Plutonium					
1	-	t =3.62 p > 0.99	t =17.28 p> 0.999	-	t = 2.48 p > 0.95	t = 4.54 p > 0.99
2	-	-	t =10.00 p > 0.999			t = 2.49 p > 0.95
	Americium					
1	-	t = 0.30 *	t = 5.67 p > 0.99	-	-	-
2	-	-	t = 4.16 p > 0.99	-	-	t = 0.84 *

* The differences were not statistically significant.

Table H-1. Intercepts (Bq) and regression coefficients k_l and $k_s \pm$ S.E. (10^{-5} day^{-1}) in regression equations: Pu concentration in urine – Pu concentration in lung or system.

Pathology Group	Lung			System		
	$a_l=k_l \pm s_{kl}$	N[1]	r[2]	$a_s+k_s \pm s_{ks}$	N	r
1	0.03 + 5.17 ± 1.1	17	0.78	0.02 + 1.28 ± 0.17	17	0.89
2-3	0.15 + 21.4 ± 10.1	38	0.33	0.05 + 1.68 ± 0.21	38	0.80

(1) Number of cases
(2) The correlation coefficient

Table I-1. Types of calibration phantoms and activities in each phantom

Type of phantom	Lung set for LLNL torso	Liver set for LLNL torso	Skull phantom manufactured by RSD	USTUR skull phantom	USTUR torso phantom
Activity, Bq	19184±170	31942±396	2940±12	621±8	734±19

Table I-2. Summary of values for efficiencies and cross-talk factors obtained with HPGe detectors

Geometry of measurement	Active organ				
	Lung		Liver		Skeleton
	CWT, cm	Efficiency, cps/Bq	CWT, cm	Efficiency, cps/Bq	Efficiency, cps/Bq
Lung	1.64	1.38E-02	1.64	3.67E-03	2.95E-03[1]
	2.27	1.11E-02	2.27	3.07E-03	
	2.64	9.66E-03	2.64	2.78E-03	
	3.34	7.52E-03	3.34	2.30E-03	
	4.12	5.84E-03	4.12	1.79E-03	
Liver	1.32	7.41E-04	1.32	1.18E-02	1.84E-04[1]
	1.93	6.86E-04	1.93	9.30E-03	
	2.25	6.58E-04	2.25	8.34E-03	
	3.00	5.66E-04	3.00	6.42E-03	
	3.78	5.01E-04	3.78	4.87E-03	
Skull	1.64	3.64E-04	1.32	1.17E-04	2.35E-02[2]
	2.27	3.03E-04	1.93	9.71E-05	
	2.64	2.89E-04	2.25	8.11E-05	
	3.34	2.62E-04	3.00	6.17E-05	
	4.12	2.35E-04	3.78	5.49E-05	

[1] value obtained by measurement of USTUR torso
[2] value obtained by measurement of RSD skull

Table I-3. Summary of values for efficiencies and cross-talk factors obtained with phoswich detectors

Geometry of measurement	Active organ				
	Lung		Liver		Skeleton
	CWT, cm	Efficiency, cps/Bq	CWT, cm	Efficiency, cps/Bq	Efficiency, cps/Bq
Lung	1.64	$(38.41\pm0.88)\cdot10^{-3}$	1.64	$(10.51\pm0.34)\cdot10^{-3}$	$(8.93\pm0.51)\cdot10^{-3}$ [1]
	2.27	$(32.95\pm0.58)\cdot10^{-3}$	2.27	$(9.22\pm0.39)\cdot10^{-3}$	
	2.64	$(30.16\pm0.62)\cdot10^{-3}$	2.64	$(8.60\pm0.56)\cdot10^{-3}$	
	3.34	$(25.58\pm0.39)\cdot10^{-3}$	3.34	$(7.39\pm0.427)\cdot10^{-3}$	
	4.12	$(21.15\pm0.38)\cdot10^{-3}$	4.12	$(6.23\pm0.41)\cdot10^{-3}$	
Liver	1.32	$(2.31\pm0.23)\cdot10^{-3}$	1.32	$(42.85\pm1.35)\cdot10^{-3}$	$(0.90\pm0.41)\cdot10^{-3}$ [1]
	1.93	$(2.28\pm0.25)\cdot10^{-3}$	1.92	$(35.95\pm1.42)\cdot10^{-3}$	
	2.25	$(2.24\pm0.24)\cdot10^{-3}$	2.25	$(34.01\pm1.14)\cdot10^{-3}$	
	3.00	$(2.22\pm0.24)\cdot10^{-3}$	3.00	$(28.61\pm0.59)\cdot10^{-3}$	
	3.78	$(2.13\pm0.34)\cdot10^{-3}$	3.78	$(22.83\pm0.89)\cdot10^{-3}$	
Skull	2.64	$(0.61\pm0.04)\cdot10^{-3}$	-	-	$6.81\cdot10^{-2}$ [2]

[1] value obtained by measurement of USTUR torso
[2] value obtained by measurement of RSD skull

Table I-4. Calibration factors for the Skull NaI(Tl) counter

Phantom	Calibration factor, Bq/cps
RSD skull	41.92±4.67
DRMIA skull	34.7±5.5

Table I-5. Comparative measurements of the USTUR skull phantom with three counters used at FIB-1

Measured activity, Bq			Certified activity of the phantom, Bq
Skull NaI(Tl) counter	WBC 7.7 (HPG detectors)	Canberra counter	
607±12	609.5±24.5	620.7±12.7	621±8

Table I-6. Minimum detectable activity (MDA) for all WBC used at FIB-1, Bq

Organs	Counter				
	Historic-NaI(Tl)	Canberra counter	Skull NaI(Tl)	Rocky Flats HPGe detector	Rocky Flats phoswich detector
Total	74			54.02	73.7
Lung		8.95		11.47	16.25
Liver				10.61	11.8
Skull		5.64	9.66	3.92	6.4
Skeleton[1]		40.28	69.06	28.02	44.93

[1] Obtained from MDA for skull geometry

Table I-7. Results of worker measurements on HPGe detectors of the former Rocky Flats counter

	NO CROSS-TALK FACTORS								WITH CROSS-TALK FACTORS					No Cross / With Cross	
#	LUNG, Bq	UNC %	LIVER, Bq	UNC %	Skeleton, Bq	UNC %	Total, Bq	UNC %	LUNG, Bq	LIVER, Bq	Skeleton, Bq	Total, Bq	UNC %	Lung/Lung	Total/Total
1	13.20	40.21	<MDA		83.26	20.51	96.45	18.54	8.65	<MDA	83.26	91.91	18.96	1.53	1.05
2	159.26	7.53	14.36	40.25	644.37	6.52	817.99	5.39	120.92	14.36	644.37	779.65	5.56	1.32	1.05
3	207.58	6.33	29.81	21.74	583.94	6.93	821.33	5.24	173.29	29.81	583.94	787.04	5.39	1.20	1.04
4	262.19	6.12	<MDA		327.38	9.18	589.57	5.78	242.62	<MDA	327.38	569.99	5.88	1.08	1.03
5	17.14	32.65	11.98	43.53	24.21	49.85	53.32	26.80	12.50	11.98	24.21	48.69	28.27	1.37	1.10
6	26.44	27.15	<MDA		186.01	12.65	212.46	11.58	14.63	<MDA	186.01	200.64	11.89	1.81	1.06
7	26.36	23.88	30.54	23.33	43.64	30.60	100.53	16.30	14.27	30.54	43.64	88.44	17.54	1.85	1.14
8	471.62	4.60	327.50	6.04	793.57	5.91	1592.70	3.47	337.34	327.50	793.57	1458.41	3.65	1.40	1.09
9	87.36	12.14	32.79	24.52	609.44	6.77	729.60	5.94	41.71	32.79	609.44	683.95	6.19	2.09	1.07
10	44.37	17.70	36.18	21.15	90.69	18.40	171.24	11.66	28.42	36.18	90.69	155.30	12.26	1.56	1.10
11	45.99	20.51	44.58	20.67	76.57	21.28	167.14	12.54	24.52	44.58	76.57	145.66	13.31	1.88	1.15
12	81.93	10.17	142.67	8.21	133.75	15.12	358.36	6.92	42.01	142.67	133.75	318.43	7.46	1.95	1.13
13	95.30	9.30	51.16	14.41	80.63	20.68	227.09	8.93	78.67	51.16	80.63	210.46	9.33	1.21	1.08
14	76.42	11.80	18.62	31.36	138.96	14.76	234.01	9.89	64.08	18.62	138.96	221.67	10.21	1.19	1.06
15	199.71	6.47	143.17	8.41	281.07	9.89	623.95	5.28	151.04	143.17	281.07	575.28	5.53	1.32	1.08
16	29.16	21.83	17.80	32.53	66.61	25.12	113.56	16.57	20.98	17.80	66.61	105.39	17.35	1.39	1.08
17	69.14	11.49	27.58	22.60	162.43	14.15	259.15	9.69	55.10	27.58	162.43	245.10	10.05	1.25	1.06
18	23.10	26.64	<MDA		56.66	27.47	79.76	20.98	19.97	<MDA	56.66	76.64	21.46	1.16	1.04
19	37.29	18.00	8.11	56.02	91.97	18.90	137.37	13.96	31.18	8.11	91.97	131.26	14.34	1.20	1.05
20	31.99	19.41	<MDA		29.06	47.52	61.05	24.80	30.71	<MDA	29.06	59.77	25.16	1.04	1.02
21	417.58	4.58	206.99	6.75	252.77	10.53	877.34	4.06	357.63	206.99	252.77	817.39	4.19	1.17	1.07
22	22.08	25.09	11.95	43.01	85.92	19.70	119.95	15.45	14.04	11.95	85.92	111.91	16.12	1.57	1.07
23	15.15	32.28	<MDA		95.81	18.61	110.96	16.66	10.86	<MDA	95.81	106.67	17.04	1.40	1.04
24	61.01	15.69	24.68	29.49	238.95	11.10	324.64	8.97	38.11	24.68	238.95	301.74	9.33	1.60	1.08
25	78.27	9.76	40.22	15.21	590.08	7.15	708.57	6.11	47.47	40.22	590.08	677.76	6.33	1.65	1.05
26	46.79	14.54	49.71	14.92	121.22	16.15	217.72	10.11	28.89	49.71	121.22	199.82	10.69	1.62	1.09
27	39.09	19.81	19.25	32.90	40.77	34.17	99.11	17.30	30.52	19.25	40.77	90.54	18.17	1.28	1.09
28	36.66	19.27	27.87	22.59	91.43	21.68	155.96	14.08	25.64	27.87	91.43	144.94	14.75	1.43	1.08
29	40.78	18.64	20.52	30.59	54.65	27.48	115.95	15.49	31.57	20.52	54.65	106.75	16.22	1.29	1.09
30	107.77	8.83	104.95	9.81	368.40	8.38	581.12	5.83	63.02	104.95	368.40	536.36	6.16	1.71	1.08
31	47.07	14.41	25.86	22.33	158.99	13.79	231.92	10.20	33.63	25.86	158.99	218.48	10.61	1.40	1.06

Table I-8. Results of worker measurements with phoswich detectors of the former Rocky Flats counter

№	Lung, Bq	UNC %	Liver, Bq	UNC %	Skeleton, Bq	UNC %	Total, Bq	UNC %
1	40.61	15.64	18.57	23.61	194.42	11.74	253.59	9.50
2	23.97	24.56	18.96	23.49	<MDA		42.93	17.20
3	19.16	30.92	30.16	16.05	<MDA		49.32	15.51
4	21.60	36.72	<MDA		<MDA		21.60	36.72
5	21.34	32.97	26.98	20.15	<MDA		48.33	18.40
6	48.01	15.69	18.80	27.42	51.10	42.24	117.90	19.87
7	65.56	12.01	24.29	22.24	<MDA		89.84	10.63
8	22.85	28.29	34.43	14.90	<MDA		57.27	14.41
9	40.25	18.37	37.33	15.29	<MDA		77.58	12.04
10	28.94	25.70	17.20	31.49	<MDA		46.15	19.94
11	68.31	10.36	35.40	14.32	<MDA		103.70	8.39
12	49.90	14.83	37.54	14.83	<MDA		87.44	10.59
13	16.63	44.01	22.19	24.87	<MDA		38.82	23.61
14	18.29	37.90	20.09	25.84	<MDA		38.38	22.57
15	49.80	17.28	33.77	19.37	<MDA		83.57	12.93
16	592.08	2.28	211.83	4.07	733.76	4.08	1537.67	2.21
17	62.42	12.62	50.05	11.79	46.13	45.91	158.60	14.72
18	29.35	25.87	20.48	26.67	<MDA		49.83	18.77
19	23.57	28.08	21.57	22.76	<MDA		45.13	18.25
20	21.14	31.66	19.40	25.69	<MDA		40.54	20.58
21	36.27	20.88	38.61	15.22	<MDA		74.87	12.80
22	160.34	6.15	151.71	5.40	257.28	9.69	569.34	4.92
23	142.75	6.92	<MDA		128.85	18.00	271.61	9.28
24	704.01	1.96	180.32	4.34	359.77	7.17	1244.09	2.43
25	342.28	3.39	265.34	3.52	428.00	6.22	1035.62	2.94
26	93.18	8.33	49.50	11.22	122.97	18.52	265.64	9.30
27	28.83	22.05	39.34	13.81	<MDA		68.17	12.27
28	409.43	2.74	451.17	2.32	490.56	5.60	1351.16	2.33
29	245.19	3.58	65.04	8.33	184.44	12.24	494.67	5.02
30	1873.81	1.09	224.34	3.62	1204.13	2.85	3302.28	1.24

Table I-9. Comparison of results obtained with the two detector systems of the former Rocky Flats counter with the Skull NaI(Tl) counter and the Canberra counter by regression

Detectors system of Rocky Flats counter	Skull NaI(Tl) counter		Canberra counter			
	Skull measurement		Skull measurement		Lung measurement	
	$R^{2(1)}$	$n^{(2)}$	R^2	n	R^2	U
HPGe	0.86	13	0.74	87	0.96	26
Phoswich	0.96	9	-	-	-	-

(1) Correlation coefficient of the regression
(2) Number of measurements

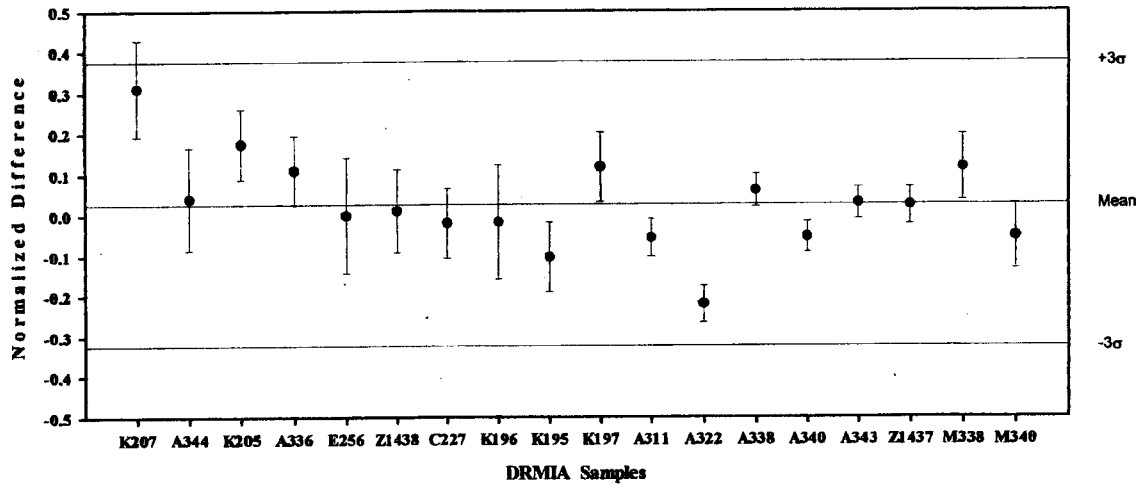


Fig.D- 1. Relative bias (Δ) for DRMIA alpha spectrometry and radiometry results
for total Pu (^{238}Pu + ^{239,240}Pu) in 18 biosamples
(alpha spectrometry results normalized to radiometry).
$\overline{\Delta} = 0.030 \pm 0.118$ $T = 1.077 < 2.086$

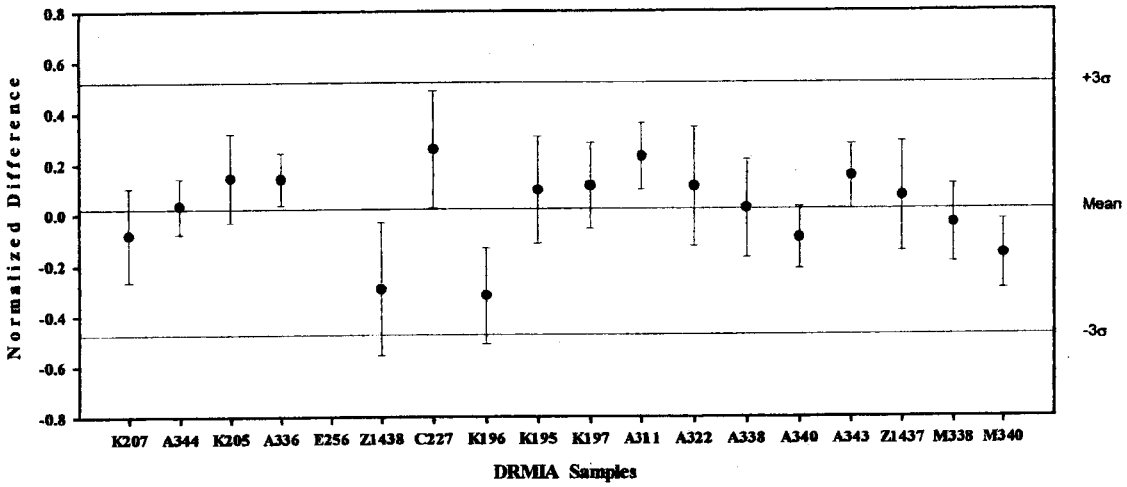


Fig.D- 2. Relative bias (Δ) for DRMIA alpha spectrometry and radiometry results for ^{241}Am in 18 biosamples
(alpha specrtrometry results normalized to radiometry)
$\bar{\Delta} = 0.022 \pm 0.166$ $T = 0.551 < 2.131$

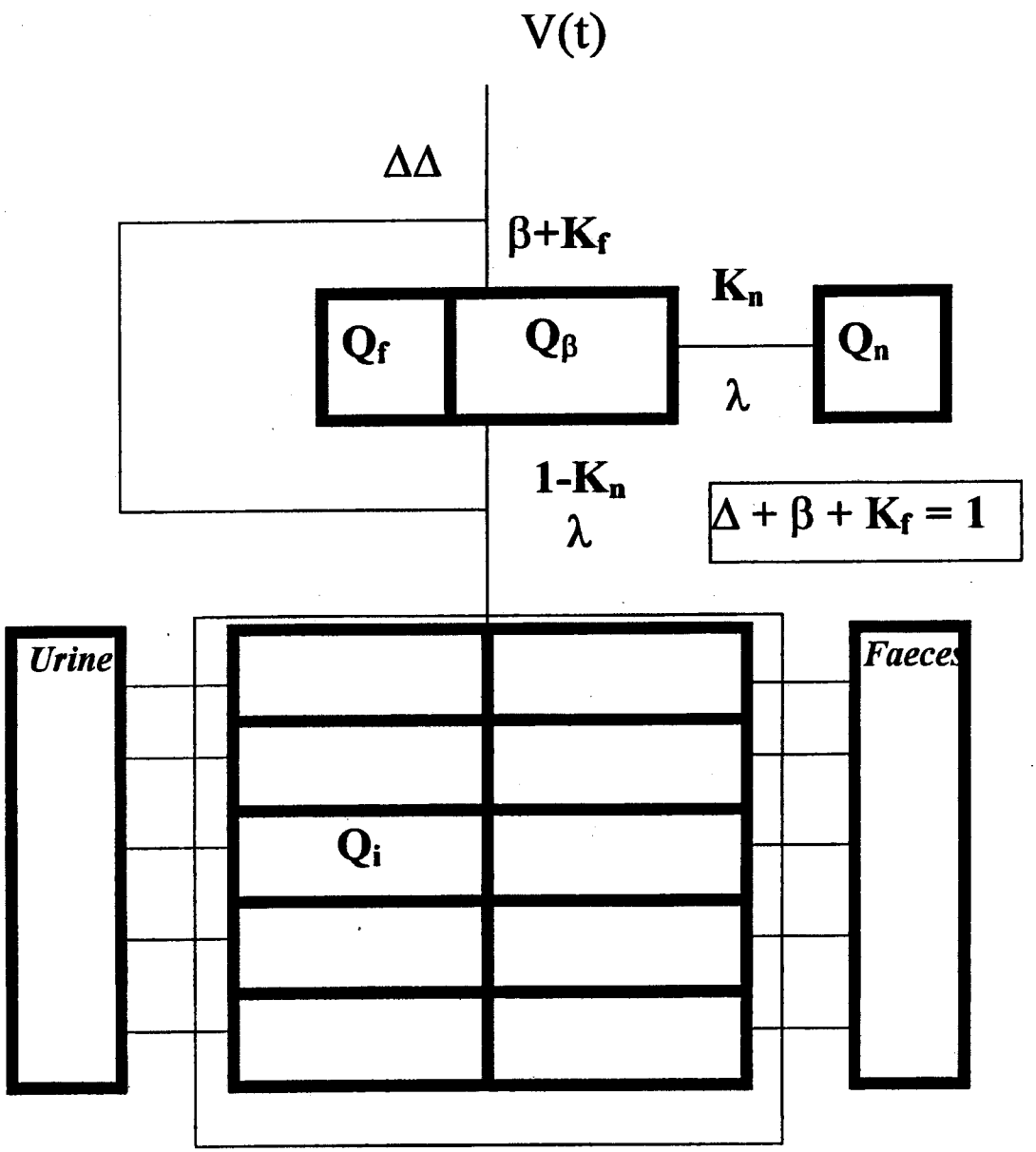


Fig. F- 1. Lung model of DRMIA

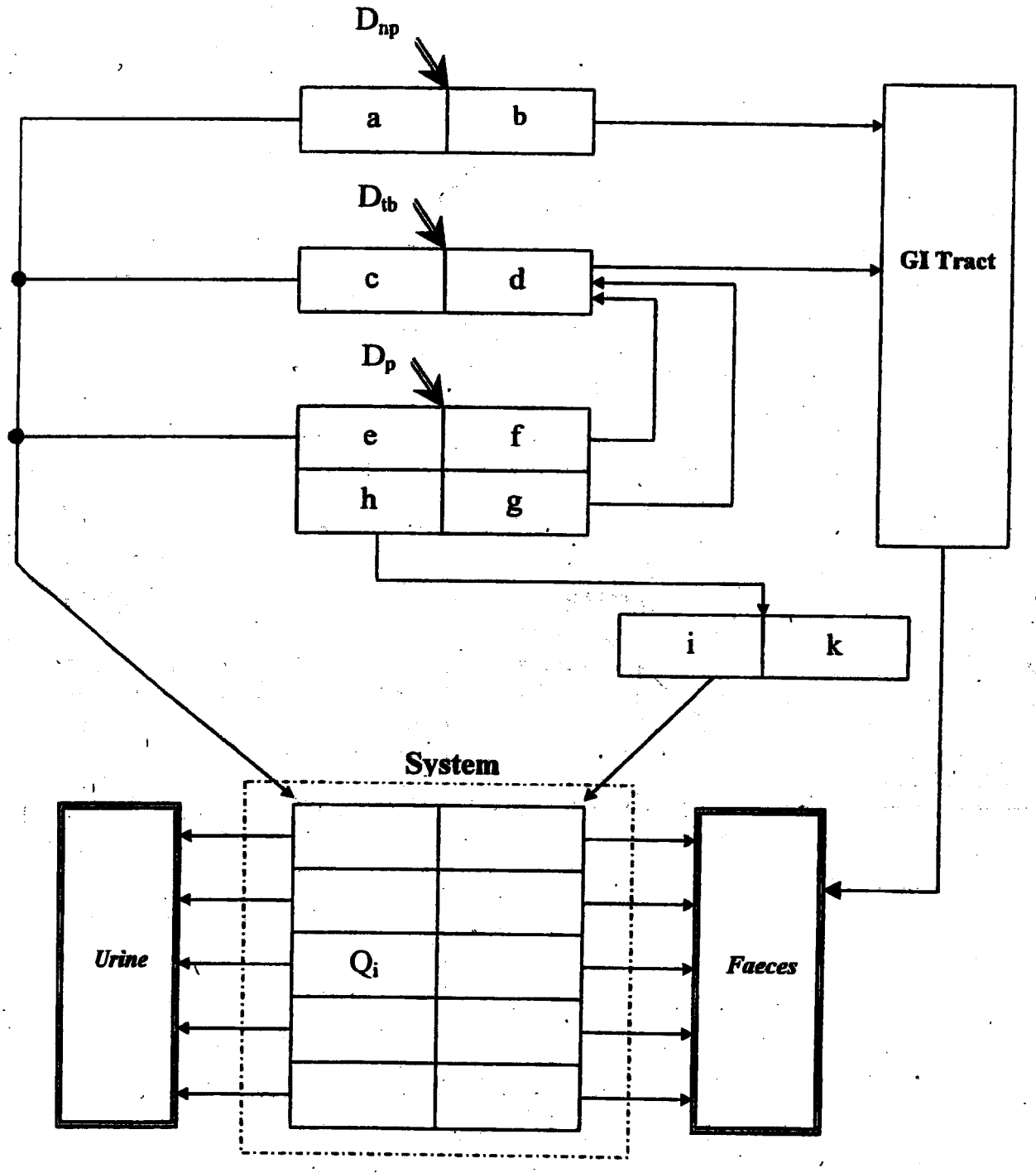


Fig. F-2 Lung model of ICRP-30 with the modified model of excretion of 239Pu by Durbin and Schmidt (1985)

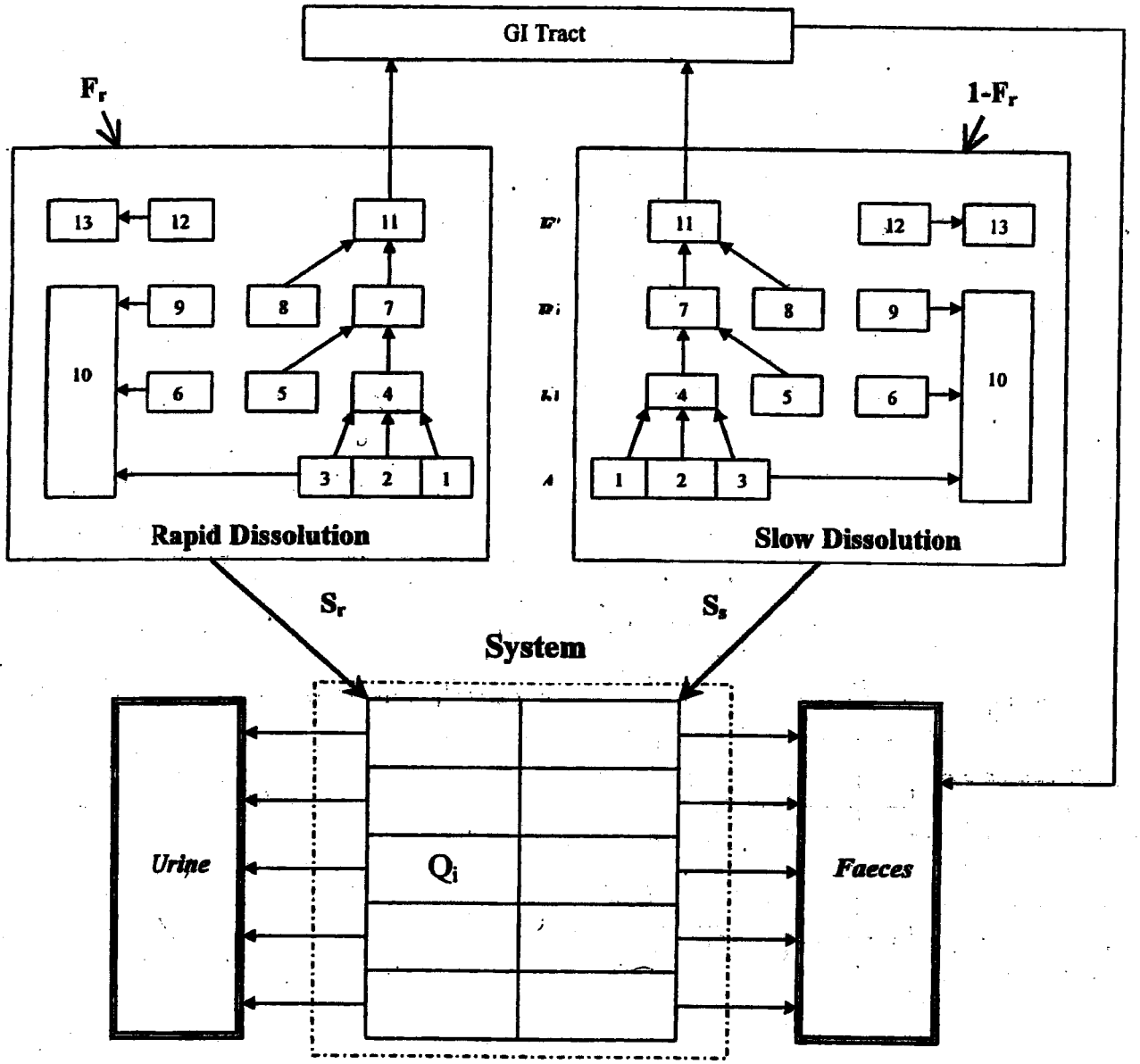
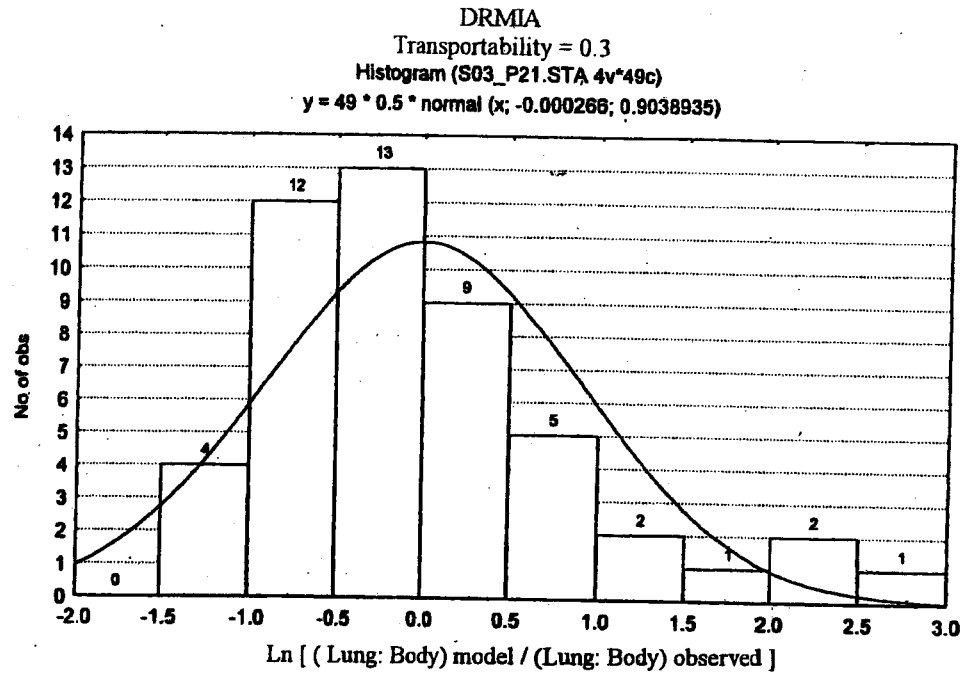


Fig. F-3 Lung model of ICRP-66 with the modified model of excretion of 239Pu by Durbin & Schmidt (1985)



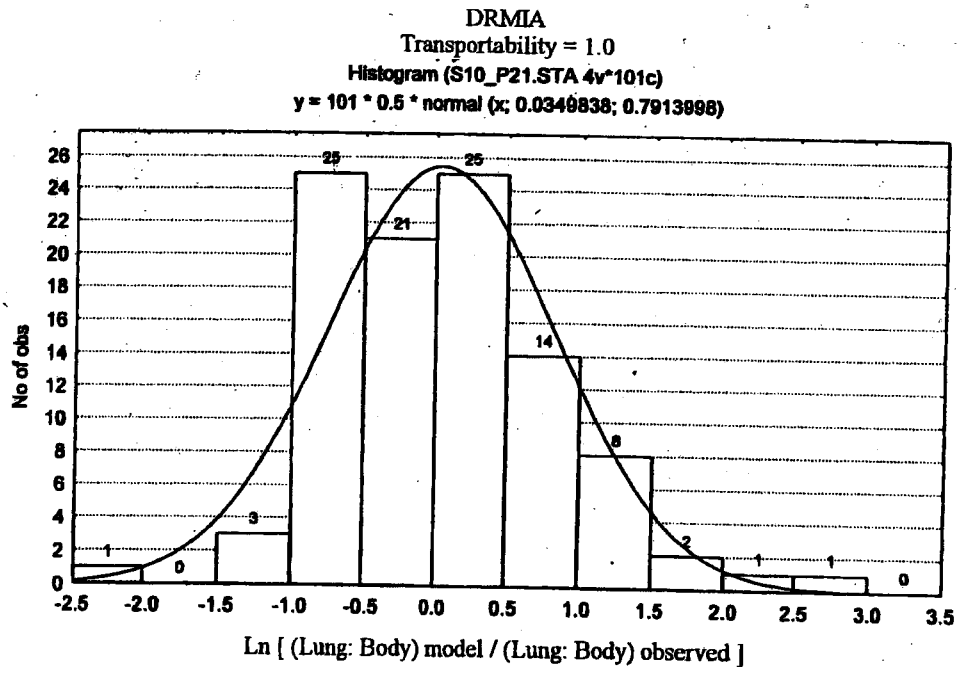
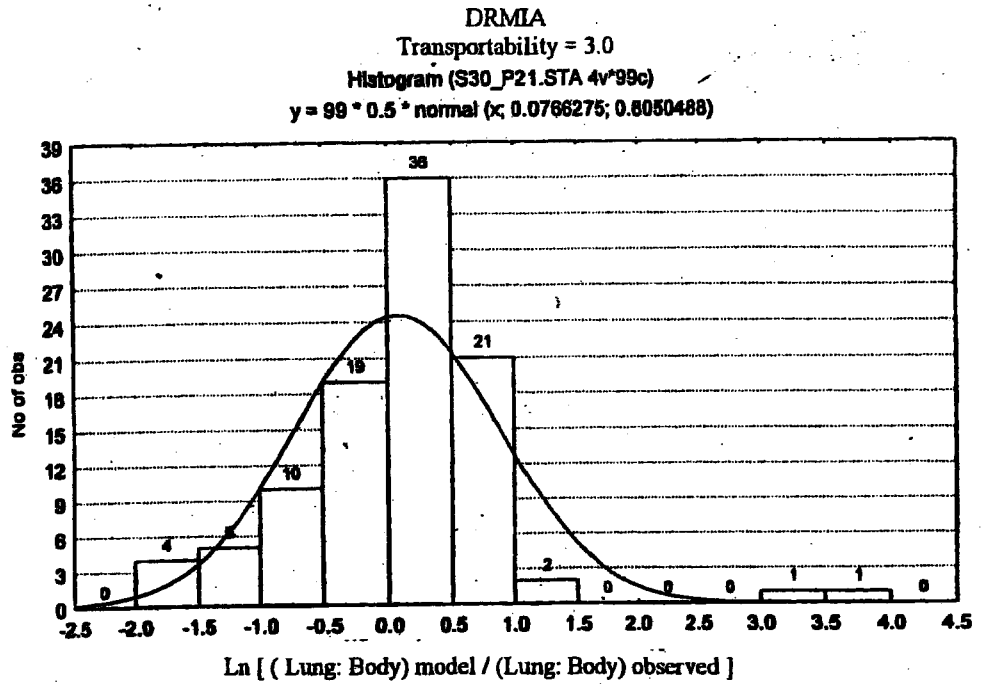


Fig. F-4 Comparison of the Lung Model of the DRMIA with observed measurements



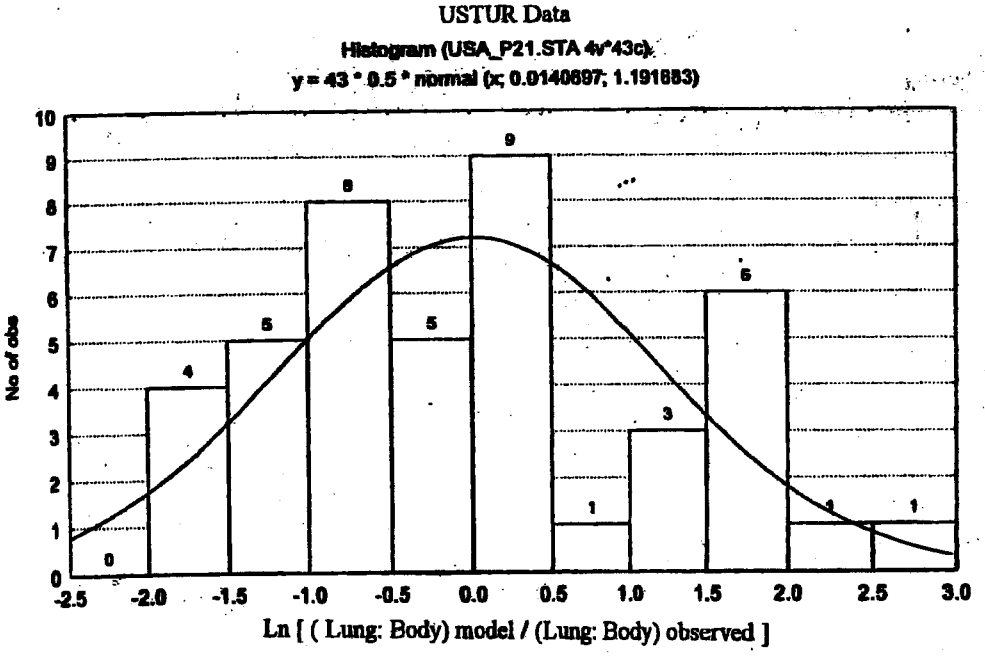
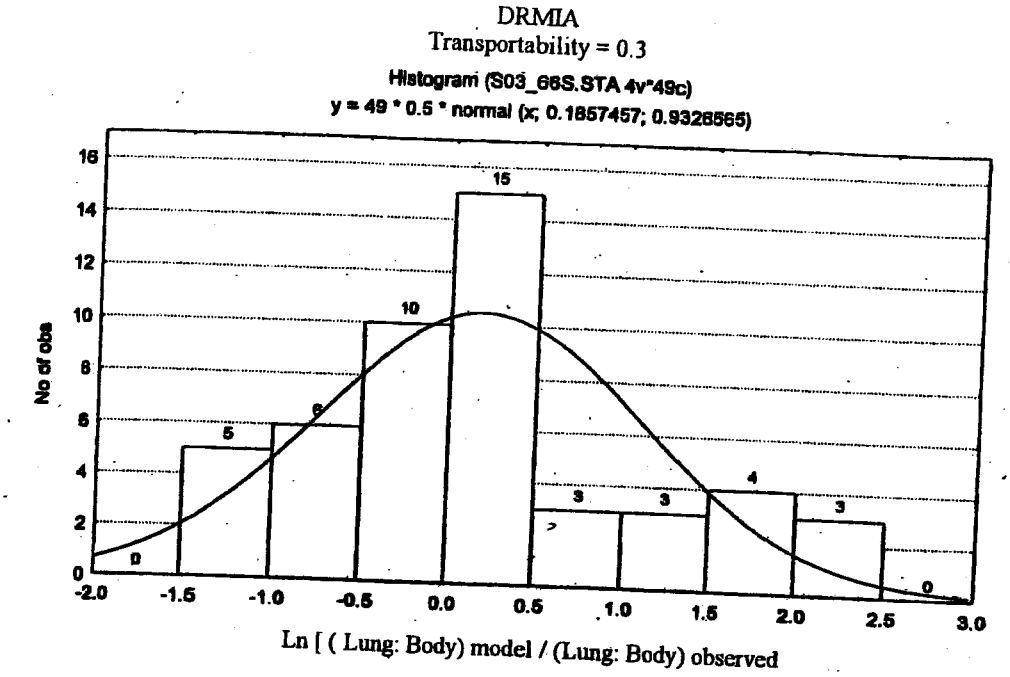


Fig. F-4 (continued). Comparison of the lung model of the DRMIA with the observed measurements



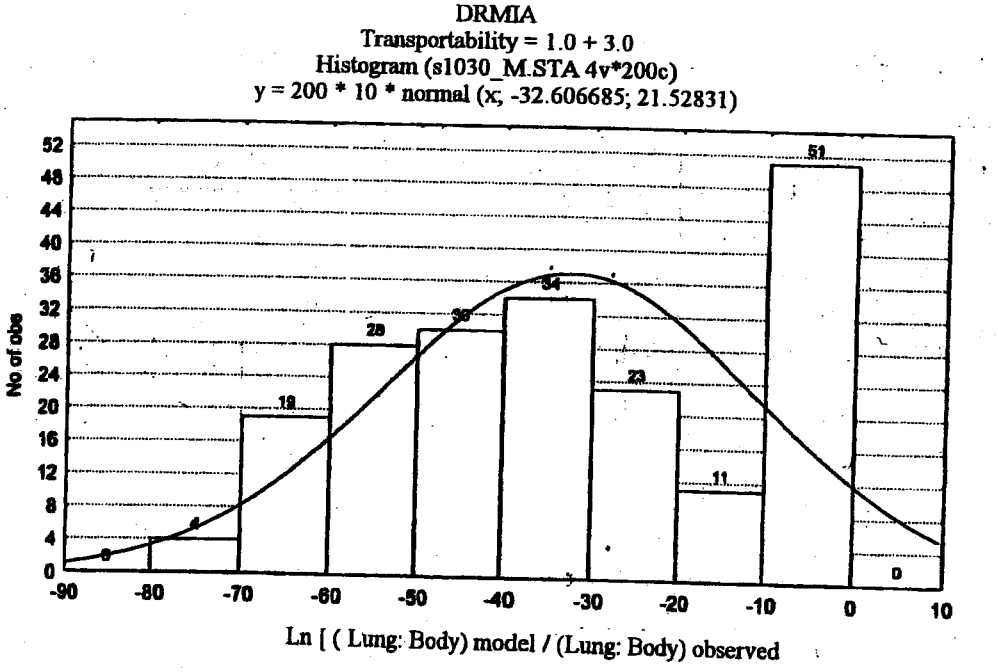


Fig. F-5 Comparison of the lung model of ICRP-66 with the observed measurements

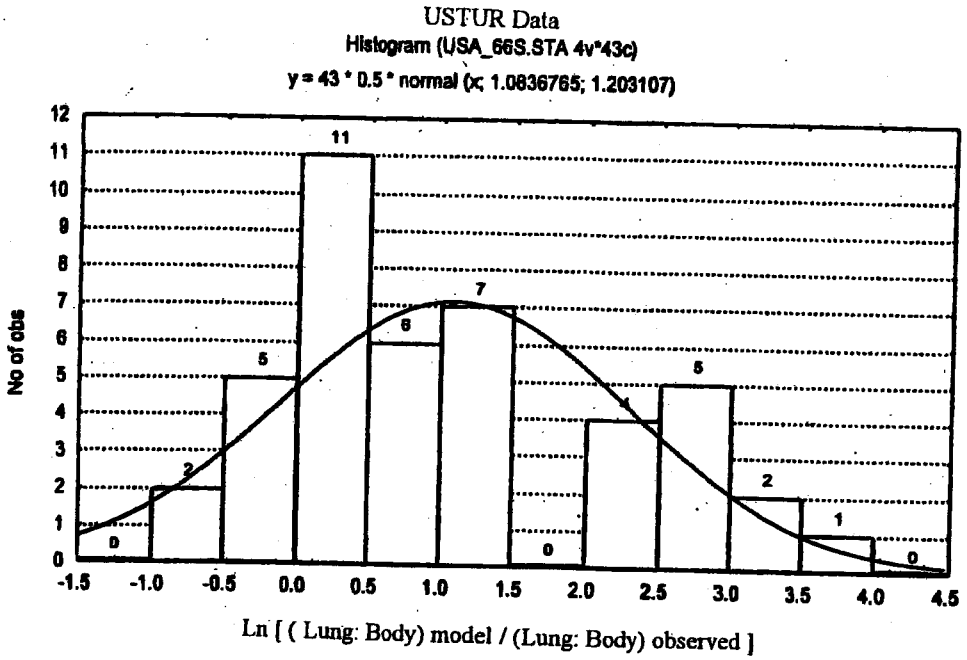


Fig. F-5 Comparison of the lung model of ICRP-66 with observed measurements (continued)